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Toughening strategies for co-cured composite joints

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List of acronyms and symbols

★ List of acronyms

CFRP	Carbon Fibre Reinforced Polymer Composite
CZM	Cohesive Zone Modelling
DCB	Double cantilever beam
FE	Finite element
FTIR	Fourier-transform infrared spectroscopy
GSIF	Generalized Stress Intensity Factor
IPN	Interpenetrating network
MPS	Maximum Principal Stress criterion
NCF	Non-crimp fibre
PA	Polyamide
PC	Polycarbonate
PEEK	Polyetheretherketone
PEI	Polyetherimide
PEO	Poly(ethylene oxide)
PES	Polyethersulfone
PET	Polyethylene terephthalate
PI	Polyimide
PMMA	polymethylmethacrylate
PP	Polypropylene
PPO	Poly(phenylene oxide)
PPS	Polyphenylene sulfide

PS	Polystyrene
PSU	Polysulfone
PTFE	Polytetrafluoroethylene
PVDF	Polyvinylidene fluoride
PVP	Poly(vinylpyrrolidone)
RED	Relative Energy Difference ratio
RTM	Resin transfer moulding
SEM	Scanning electron microscope
SENB	Single edge notch bend
SENT	Single edge notch tension
TAST	Thick adherend shear test
TP	Thermoplastic
UD	Unidirectional laminate
UHMPE	Ultra-high-molecular-weight polyethylene

★ **List of symbols**

Symbol	Definition	Units
a	DCB: crack length	mm
	SENT, SENB: crack length	mm
	Centred crack: half crack length	mm
a_c	Centred crack: half critical crack length	mm
a_{edge}	Crack length of an edge crack	mm
A	Ch.2: Amplitude of the pattern	mm
α	First Dundur's parameter	-
B	DCB width	mm
β	Second Dundur's parameter	-
C	Ch.8: Cost	€
C_a	Mass cost of the adhesive	€/kg
C_i	Constant intervening in equation 5.1	-
C_m	Joint manufacturing cost	€/mm ³
d	Debonding length	mm
D	Wedge thickness	mm
D_i	Constant intervening in equation 5.1	-
δ	CZM: Nodal separation or opening displacement	mm
	DCB: Displacement	mm

δ^N	CZM: Normal opening displacement	mm
δ_c^N	CZM: Normal critical opening displacement	mm
δ^T	CZM: Tangential opening displacement	mm
δ_c^T	CZM: Tangential critical opening displacement	mm
δ_c	CZM: Critical opening displacement	mm
δ_{di}	Ch.2: Energy from the dispersion forces between the molecules of i	$\sqrt{\text{MPa}}$
δ_{hi}	Ch.2: Energy from the hydrogen bonds of molecules of i	$\sqrt{\text{MPa}}$
δ_{pi}	Ch.2: Energy from the dipolar intermolecular forces between the molecules of i	$\sqrt{\text{MPa}}$
Δa	Crack propagation length	mm
Δa^{SS}	Distance over which R-effect takes place	mm
Δh_1	Distance between the crack and the closest adherend	mm
E	Young's modulus	GPa
E^f	Young's modulus of bridging ligament	GPa
E_i	Young's modulus of material i	GPa
ε	Strain	-
ε_f	Tensile elongation	-
F_i	Bridging force acting on ligament i	N
F_σ	Tensile force	N
F_τ	Shear force	N
F^f	Tensile strength of bridging ligament	N
G_0	Fracture toughness in the absence of any toughening mechanism	J/m^2
	Ch.5: Fracture toughness in the absence of bridging	J/m^2
G_c	Joint fracture toughness	J/m^2
G_I	Mode I energy release rate	J/m^2
G_{Ic}	Mode I fracture toughness	J/m^2
G_{Ic}^a	Mode I fracture toughness of the adhesive	J/m^2
G_{Ic}^{joint}	Mode I fracture toughness of the joint	J/m^2
G_{Ic}^{max}	Maximum fracture toughness attained with bridging	J/m^2
G_{Ic}^{min}	Minimum fracture toughness attained with bridging	J/m^2

G_{Tip}	Crack tip driving force	J/m ²
G^{SS}	Steady-state fracture toughness	J/m ²
$\Gamma^{bridging}$	Contribution of crack bridging to G_{Ic}^{joint}	J/m ²
$\Gamma^{deflection}$	Contribution of crack deflection to G_{Ic}^{joint}	J/m ²
Γ^p	Contribution of plastic dissipation to G_{Ic}^{joint}	J/m ²
Γ_c	CZM: Cohesive energy	J/m ²
H	Ch.3: Sample height	mm
H	Ch.6: Channel height	mm
h	DCB: beam thickness	mm
h_{bot}	Thickness of the lower beam	mm
h_s	Adherend thickness	mm
h_{top}	Thickness of the upper beam	mm
I	Second moment of inertia	mm ⁴
K_F	Interface corner stress intensity factor	MPa $\sqrt{m}$
K_I	Mode I stress intensity factor	MPa $\sqrt{m}$
K_{Ic}	Critical mode I stress intensity factor	MPa $\sqrt{m}$
K_{II}	Mode II stress intensity factor	MPa $\sqrt{m}$
κ_i	Shear coefficient of material i	-
l_b	Bridging zone length	mm
l_{coh}	Length of cohesive element	mm
l_p	Process zone length $1/\lambda_w$	mm
l_s	Fracture process zone length	mm
l^f	Tensile elongation of fishing line	mm
L	Sample length	mm
L_a	Adhesive length	mm
L_i	Position of the bridging ligament i	mm
L_{s1}	Top adherend length	mm
L_{s2}	Bottom adherend length	mm
λ	Ch.2: Wavelength of the pattern	mm
	Ch.3,7 - CZM: Adimensional separation	mm
	Ch.4 - Fishing line spacing	mm
	Ch.8: Singularity order	mm
λ_{c1}	Spacing above which stick-slip is found	mm
λ_{c2}	Spacing above $G_{Ic}^{min} = G_0$	mm
λ_w	Elastic foundation number	1/mm
m	Joint mass	kg
m_a	Adhesive mass	kg

m_{TC}	Objective function: mass and tensile strength constraint	kg
m_{SC1}	Objective function: mass and shear stiffness constraint	kg
m_{SC2}	Objective function: mass and shear strength constraint	kg
M	Bending moment	N · mm
μ_i	Shear modulus of material i	MPa
n (n_i)	Hardening exponent (of material i)	-
	Ch.5: number of fishing lines in the wake of the crack	-
P	Applied load (DCB)	N
ρ	Crack tip radius	mm
r	Fishing line radius	mm
ρ_i	Density of material i	kg/m ³
R_a	Distance between two molecules A and B	$\sqrt{\text{MPa}}$
R_0	Hansen model: interaction radius of the dissolved molecule	$\sqrt{\text{MPa}}$
R_0 (R_{0i})	Mechanics: plastic zone size (of material i)	mm
S	Ch.6: Channel spacing	mm
	Ch.8: Surface area of the joint	m ²
S_{archi}	Spacing between the stop holes	mm
S_{holes}	Surface area occupied by stop holes when orthogonally projected	m ²
σ	CZM: traction	MPa
σ_0 (σ_{0i})	Yield stress (of material i)	MPa
σ_a^f	Adhesive tensile strength	MPa
σ^{f*}	Effective tensile strength	MPa
σ_f	Tensile strength	MPa
σ_c	CZM: Cohesive strength	MPa
	MPS: critical maximum principal stress	MPa
σ^∞	Remote tensile stress	MPa
σ_y^l	Lower yield stress	MPa
σ_y^u	Upper yield stress	MPa
t	Adhesive/film thickness	mm
t_{max}	Maximum adhesive thickness for adhesive selection	mm

t_{min}	Minimum adhesive thickness for adhesive selection	mm
T	T-stress	MPa
T_{dwell}	Dwell temperature	°C
t_{dwell}	Dwell time	min
T_g	Glass transition temperature	°C
τ_a^f	Adhesive shear strength	MPa
τ^{f*}	Effective shear strength	MPa
τ_{archi}^f	Architected joint shear strength	MPa
τ^∞	Remote shear stress	MPa
u_τ	Shear displacement	mm
u_τ^{max}	Maximum shear displacement	mm
ν_i	Poisson ratio of material i	-
w_i	Deflection at the bridging ligament position	mm
	L_i	
W	Sample width	mm
	Ch.6: Channel width	mm
W_{ik}	Guess for the iteration in position i at iteration k	mm
X_c	MPS: critical distance	mm
X_k	Guess for iteration k	-
$y_{K_{II}=0-i}$	Position in layer i such that $K_{II} = 0$	mm
Y	Geometric factor to compute K_I	-
ψ	Phase angle	°

★ List of subscripts

- a Ch.5,6: Adhesive
- s Ch.5,6: Adherend
- 1 Ch.7: Layer 1 = soft layer
- 2 Ch.7: Layer 2 = hard layer

Abstract

Carbon fibre reinforced polymer composites are broadly used in complex structures. Manufacturing of these structures requires joining composite parts with each other or with other materials. Adhesive bonding offers many advantages compared to mechanical fastening such as lightweight and homogeneous stress transfer. However, adhesive joints suffer from low fracture toughness and time- and labour-intensive processes preventing intensive use by the industry.

The first objective pursued by this thesis is to investigate composite co-curing through a thermoplastic interlayer to tackle the process intensity issue. PEI, PES, PSU, PA, PC and PMMA are the thermoplastic materials investigated for this purpose. Interdiffusion is key to achieve a good toughness but under certain conditions a weak interface can develop along the morphological gradient, resulting in a brittle joint or interphase.

Secondly, toughening strategies are investigated. First, polyethylene filaments are inserted to trigger significant bridging. As an alternative, a TP film is manufactured through 3D printing to introduce channels that can be filled or left empty during the co-curing process. This controlled filling allows fine tuning the properties of joints in terms of toughness and shear resistance. The last investigated strategy is the addition of a dissipative layer which proves to be efficient if this layer is soft and stacked between two thin brittle layers.

Strategies to improve the joint toughness also affect other properties such as the shear strength and stiffness of the joint. To compare toughened joints with their classical counterpart, a selection methodology is developed to determine under which loading conditions the toughened solutions perform the best.

Résumé

Les composites polymères renforcés de fibres de carbone sont largement utilisés dans les structures complexes. La fabrication de ces structures nécessite l'assemblage de pièces composites entre elles ou avec d'autres matériaux. Le collage offre de nombreux avantages par rapport à la fixation mécanique, tels que la légèreté et le transfert homogène des contraintes. Cependant, les joints collés souffrent d'une faible ténacité et de procédés de mises en oeuvre complexes, empêchant leur utilisation intensive par l'industrie.

Le premier objectif de cette thèse est d'étudier la co-cuisson des composites par le biais d'une couche intermédiaire thermoplastique pour réduire l'intensité du procédé. Différents matériaux sont étudiés metant en avance l'importance de l'interdiffusion pour obtenir une bonne ténacité. Cependant, dans certaines conditions, une interface fragile peut se développer, résultant en un joint ou une interphase fragile.

Deuxièmement, des stratégies de ténacification sont étudiées. Tout d'abord, des filaments de polyéthylène sont insérés pour déclencher un pontage. Dans un second temps, un film TP est fabriqué par impression 3D pour introduire des canaux pouvant être remplis ou non pendant la co-cuisson. Ce remplissage contrôlé permet d'ajuster les propriétés du joint. La dernière stratégie étudiée est l'ajout d'une couche dissipative qui s'avère efficace si cette couche est souple et empilée entre deux fines couches fragiles.

Les stratégies visant à améliorer la ténacité des joints affectent également d'autres propriétés telles que la résistance au cisaillement et la rigidité du joint. Pour comparer les joints trempés avec leur équivalent classique, une méthodologie de sélection est développée pour déterminer dans quelles conditions les solutions architecturées sont les plus performantes.

1

Introduction

Carbon fibre reinforced polymer composites (CFRPs) are broadly used in complex structures such as in airplanes, automotive, marine structures or windmills [1, 2]. Indeed, CFRPs offer strength to weight and stiffness to weight ratios superior to metals, allowing for lighter structures. In addition, CFRPs offer good corrosion and fatigue resistance. Weightsaving is an important parameter in airplanes or cars as it results in lower fuel consumption and lower CO_2 emissions. Consequently, the amount of composite parts in airplanes drastically increased since the 70's. Today, CFRPs represent up to 50% of the total weight of some airplanes such as the Boeing B787 or the Airbus A350 (Figure 1.1) [2]. In order to manufacture these complex structures, the CFRP parts must be joined to other CFRPs or to other materials such as titanium or aluminium. This requires the development of efficient and reliable joining techniques.

Three main methods exist to join materials together, namely: mechanical fastening, adhesive bonding and welding [1]. Mechanical fastening corresponds to the joining through rivets or bolts. Holes must be drilled into the adherends to insert the fasteners, resulting in stress concentrations reducing the structural integrity potential [3, 4]. CFRPs are very sensitive to holes due to their brittleness and anisotropy. Consequently, thicker parts are used to guarantee structural integrity, resulting in extra weight in addi-

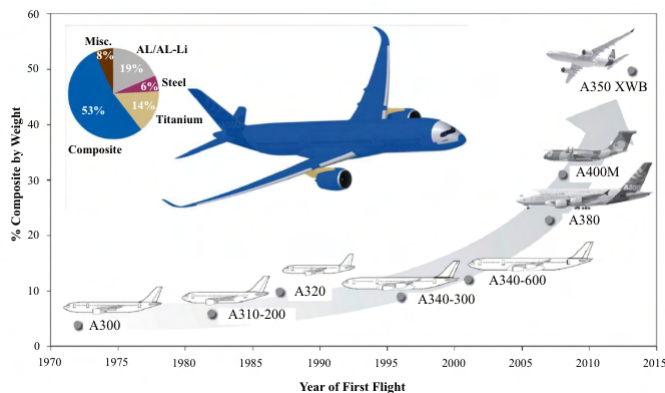


Figure 1.1 Evolution of the relative composite weight in airplanes over the years. Taken from [2].

tion to the weight already coming from the insertion of the fasteners, playing again the expected weight gains. Welding can be achieved between two metals or if one of the parts to join is a thermoplastic (TP) matrix composite [3, 5, 6] but not with thermosetting composites which still represent the largest share of the CFRPs used in advanced structures. The last option is adhesive bonding which does not suffer from stress concentrations resulting from holes drilling and which is thus suitable to join CFRP parts. Consequently, adhesive bonding is used in many applications involving CFRPs such as airplanes, cars, windmills, building, marine structures or sport equipments as shown in Figure 1.2.

Adhesive bonding is achieved through the intermediate of an adhesive interlayer [1]. An adhesive joint is thus made of two substrates referred to as the adherends assembled to each other through the adhesive. Similar and dissimilar materials can be joined. Opposite to mechanical fasteners, it offers homogeneous stress transfer across the joint. If properly designed, it is even possible to benefit from the full strength of the adherends. In addition, it results in weightsaving. Adhesive joints also offer good sealing performance, improved damping properties and improved fatigue resistance. Finally, adhesive bonding allows the assembly of complex structures. For example, in windturbine blades or marine structures, the use of adhesive allows to bond parts separated by large gaps of varying thickness resulting in joints with an adhesive thickness up to 30 mm [7]. Although, adhesive bonding presents a large number of advantages, some disadvantages pre-

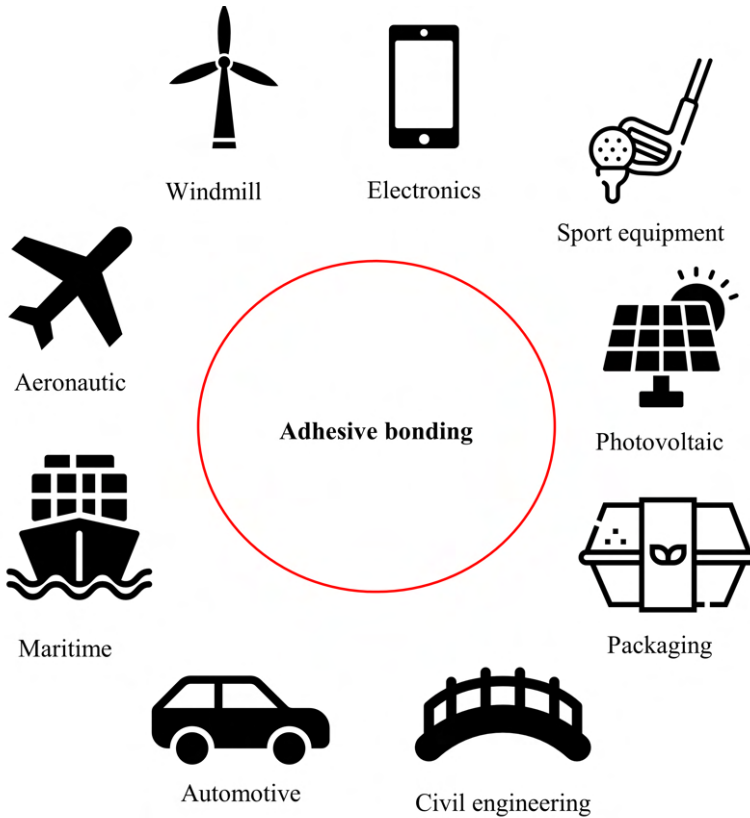


Figure 1.2 Applications involving adhesive bonding. Taken from FlatIcon.

vent its use at a larger scale and in critical structural components. Indeed, adhesive joints are often limited to the bonding of secondary structures and not to load-bearing primary structures. The first main problem is the formation of adhesion defects due to surface contaminations. This type of defects cannot be detected by non-destructive testing techniques to ensure the joint strength which is a critical point of the certification regulation process in aeronautic [8]. In addition, the adhesive bonding process involves many steps such as composite curing, surface treatments and finally bonding resulting in a time- and labour-intensive process. The last main problem is the limited fracture toughness G_{Ic} of the adhesive joints. Hence, defects can easily lead to joint failure and adhesive joints are sensitive to

peel stresses. Consequently, adhesive joints are mainly loaded under shear (lap-shear joints) or under tension (butt joints). To prevent this issue, many studies focus on adhesive toughening by using rubber particles, a thermoplastic phase or organic and inorganic fillers in the adhesive [9, 10]. More recently, it has been suggested to develop extrinsic toughening strategies involving crack blunting or crack deflection for example (see [11] for a review).

Objectives

The objective of the thesis is to tackle the two following issues encountered with adhesive bonding: (i) the time- and labour-intensive process and (ii) the limited G_{Ic} of the joints. This led to the following research questions:

Q1 - *How can we manufacture composite joints in a single step to achieve simultaneous composite curing and bonding?* The so-manufactured joints are referred to as the co-cured joints.

Q2 - *How can we toughen the co-cured joints based on extrinsic dissipation mechanisms?*

The last question has been extensively addressed during the thesis. Many strategies have been developed to extrinsically toughen adhesive joints. However, not only the G_{Ic} of the joint matters, but also its resistance under shear. This led to the following question

Q3 - *How do toughened joints compare to classical adhesive joints depending on the loading configuration, objectives and constraints of the application?*

Outline

The thesis is divided into 9 chapters (including this introduction and Chapter 9 being the conclusion) divided into three parts, each part corresponding to one of the above-mentioned questions. The outline is schematically shown in Figure 1.3

Chapter 2 reviews the state of the art. It is divided into three parts. Section 2.1 briefly introduces joining methods for CFRPs. Section 2.2 reviews the state-of-the-art regarding co-cured adhesive joints and an alternative to it using a thermoplastic interlayer exhibiting good chemical interactions with the CFRP matrix. The toughening of adhesive joints through extrinsic

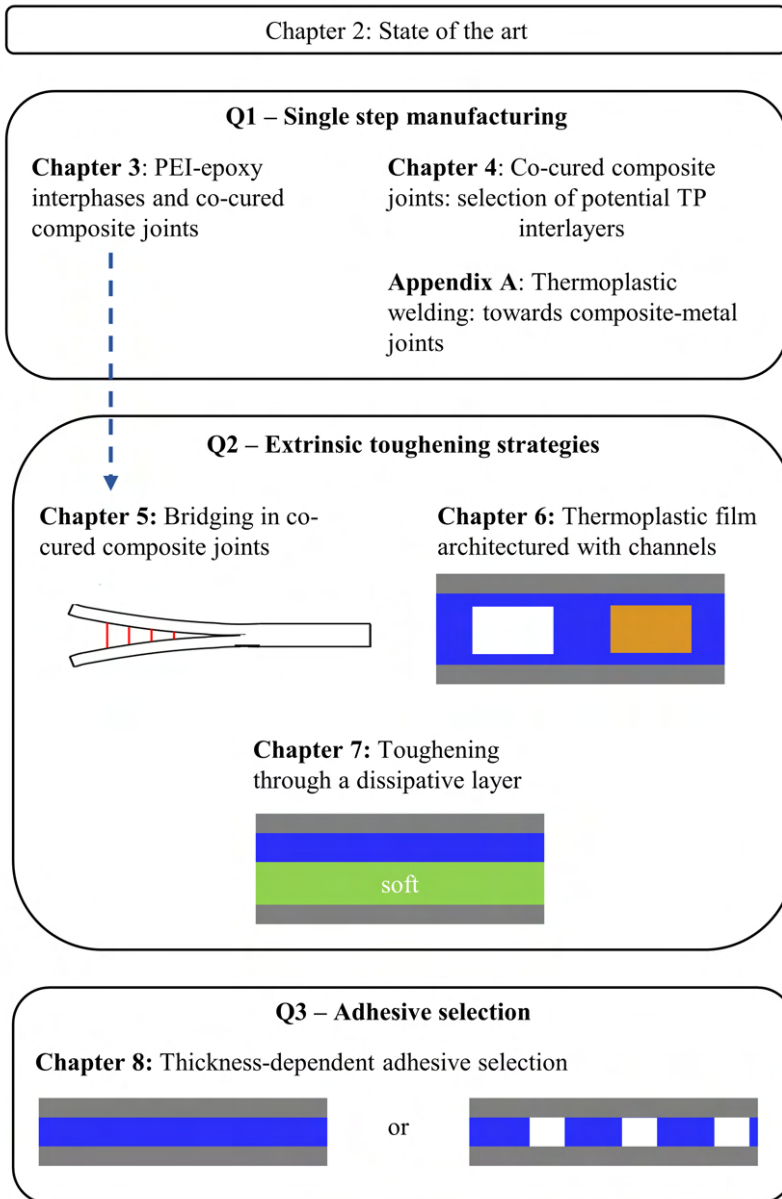


Figure 1.3 Schematic of the logic and organization of the thesis outline.

mechanism is reviewed in Section 2.3.

The first part focuses on Question 1. It investigates the manufacturing of composite joints through co-curing using a thermoplastic interlayer.

Chapter 3 investigates the use of a polyetherimide (PEI) interlayer for the manufacturing of co-cured joints. Voleppe et al. [12] reported an interesting crack trapping mechanism in PEI-epoxy interphases resulting in a high fracture toughness ($\approx 880 \text{ J/m}^2$). The origin of this crack trapping mechanism is first investigated by understanding the failure mechanisms taking place in PEI-epoxy blends and interphases. Then, the G_{Ic} of PEI co-cured composite joints is investigated.

Chapter 4 investigates other thermoplastic materials such as PSU, PES, PA, PC and PMMA as interlayer for the manufacturing of co-cured composite joints. It is a preliminary study which only focuses on the G_{Ic} of the joints and not on the formation of the interphase and the deep understanding of the failure mechanisms taking place in each of them.

Appendix A is linked to this part. As CFRPs are not only joined together but also to metals. As first step towards the manufacturing of co-cured composite-TP-metal joints, it investigates the welding conditions leading to the strongest titanium-TP-titanium joints.

The second part focuses on the investigation of extrinsic toughening strategies for the thermoplastic co-cured composite joints (Question 2). It remains the core of this thesis.

Chapter 5 investigates the addition of fishing lines between the PEI film and the composite adherends to trigger crack bridging. This study investigates the influence of the spacing between the fishing lines and their tensile strength on the joint fracture toughness. This is assessed through experiments and an analytical model.

Chapter 6 focuses on the design of an architected 3D printed PA film containing channels that can be either filled with resin or left empty during the co-curing process. The mode I fracture toughness is determined experimentally while a finite element (FE) model allows to further understand the failure mechanisms taking place in the joint. The FE model is also used to predict the lap-shear strength depending on the amount of filled/empty channels and their position in the joint. This chapter is linked to the patent application number EP23161330.8.

Chapter 7 studies the potential toughening provided by the addition of a soft layer in a joint in which plastic deformation takes place. It investigates the crack propagation locus depending on the different material properties and geometric parameters. This is achieved using the steady-state code implemented by Martiny et al. [13].

The third part focusses on Question 3 corresponding to the comparison of toughened joints with classical adhesive joints.

Chapter 8 presents a selection approach for adhesive joints based on the influence of the adhesive thickness on the joint properties. The developed method allows to compare adhesive together but also to compare toughened joints with their classic counterpart.

2

State of the art



This chapter covers the literature results dealing with the co-curing of adhesive joints and toughening strategies for adhesive joints. Part of this information will sometimes be repeated in the results chapters but, this current state of the art chapter is generally more detailed.

Carbon fibre reinforced polymer composites (CFRPs) are broadly used in advanced structures such as in airplanes, wind turbine blades or marine structures, owing to a high strength and stiffness to weight ratio. In many structures, CFRPs are joined together or to other materials such as metals. Section 2.1 briefly describes available techniques to join dissimilar materials with a focus on structures including CFRP parts. Among these, adhesive bonding is an interesting solution but its use remains limited due to intensive bonding processes and to the limited fracture toughness (G_{IC}) of the adhesive. Section 2.2. presents bonding through co-curing as an alternative to common adhesive bonding processes which can be implemented with an adhesive (subsection 2.2.1) or with a thermoplastic (TP) film (subsection 2.2.2). This last strategy requires good interactions between the composite matrix and the thermoplastic (see subsection 2.2.3). Section 3

focuses on the improvement of the fracture toughness of adhesive joints either by chemical modification of the adhesive (subsection 2.3.2) or by triggering extrinsic toughening mechanisms (subsections 2.3.3 to 2.3.8).

2.1 Joining of dissimilar materials

CFRPs are broadly used in advanced structures such as in airplanes, wind turbine blades or marine structures. The manufacturing of these structures requires to join the CFRPs parts. Several techniques exist and among them: mechanical fastening, adhesive bonding and thermoplastic welding. These techniques are introduced here-below.

Mechanical fasteners have been used to join isotropic materials for a long time and their use has been extended to the joining of composite materials in the 70's [3, 4]. Mechanical fasteners include screws, bolts, nuts, washers or rivets. Mechanical fastening requires to drill holes in the adherends. Due to their brittleness and anisotropy, CFRPs are sensitive to holes. Upon mechanical fastening, stress concentration develops close to the holes and the inserted fasteners, reducing the structural integrity [10]. In order to prevent premature failure, thicker adherends can be used. However, this results in additional weight while today the trend is to decrease the weight of structures of airplanes and cars for example. As advantage, mechanical fasteners can be removed allowing to repair the adherends and to control their integrity. In addition, stiff assemblies are obtained. Although they present major disadvantages, mechanical fasteners are still widely used in aerospace because they constitute a trustworthy solution which has proven to be efficient.

The second technique is adhesive bonding [1, 4]. In this case, an adhesive is inserted between the two parts to join and is then cured to achieve bonding. In structural applications, adhesives are mainly made of thermoset polymers such as epoxy, urethane, phenolic or acrylic resins that can be toughened by adding rubber particles, a thermoplastic phase, silica particles, carbon nanotubes and many others [10]. Adhesive bonding is a labour- and time-intensive process. Indeed, the two parts to join must first be manufactured involving some relatively long processes if composite parts must be joined together. Then, the surfaces to bond must be treated. Depending on the adherends material and on the adhesive physico-chemical properties, the optimum surface treatment may vary [1,

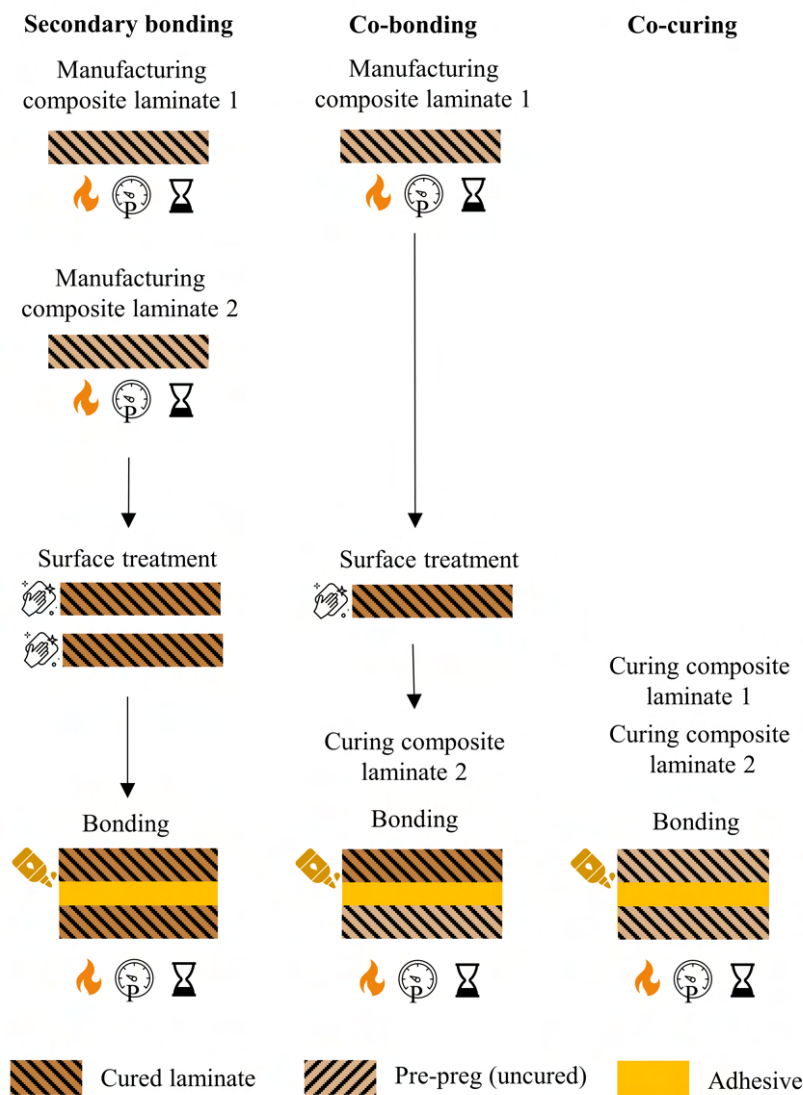


Figure 2.1 Summary of the common adhesive bonding process (secondary bonding) and its alternatives: co-bonding and co-curing.

14]. The parts can finally be bonded using the adhesive as shown in Figure 2.1. This process is referred to as secondary bonding. In order to reduce the number of steps and the manufacturing time, co-bonding and

co-curing [15] are two alternatives as shown in Figure 2.1. One considers two composite adherends. In co-bonding, one of the two adherends is already cured while the other one is a uncured pre-preg. An adhesive is inserted between the two adherends and is cured simultaneously with the second composite adherend. In the case of co-curing, the two composite adherends are uncured pre-pregs and prior to their curing, an adhesive is inserted inbetween in order to achieve composite curing and bonding simultaneously. Co-curing and co-bonding require to have matching curing cycles for the composite and adhesive. Hence, these techniques can be applied only with a limited number of adhesives. In addition, during the bonding process, although the surfaces have been carefully treated, every joint contains some adhesion defects degrading the mechanical performances of the joint and that cannot be detected upon bonding. Such defects constitute a major issue because adhesives suffer from low to moderate fracture toughness and cracks can thus easily initiate and propagate in the joint. Due to their limited fracture toughness, adhesive joints easily fail under tensile and peel loads. Hence, they must be preferably loaded under shear. Adhesively bonded joints are also difficult to repair and are irreversible. However, adhesive joints lead to an homogeneous stress transfer across the joint and in lighter structures compared to structures joined using fasteners. Adhesive joints also offer improved fatigue life and are suitable to join shock-sensitive surfaces. This leads to an increased demand from high performance industries to improve adhesive joint performances and processes to replace mechanical fasteners. Adhesives and mechanical fasteners have been combined to design strong, reliable joints. According to Lehman and Hawley [16], combining adhesive bonding and mechanical fastening joint results in enhanced strength. However, it can also lead to a strength reduction. The company Haviland used this combination in 1949 to introduce windows on the ceil of the airplane [17]. However, this combination led to three fatal accidents five years later.

Polymer composites can be either produced with a thermoset or a thermoplastic matrix. Thermoplastic composites have the advantage of melting above a certain temperature T_m allowing to weld them with another material. In this case, one refers about thermoplastic welding [3, 5, 6]. The main limitation is that thermoset composites still represent the largest share of composites used in structures. Different technologies have been developed to perform thermoplastic welding (Figure 2.2) such as ultrasonic welding [18], laser welding [21], friction welding [19], resistance heating [6] or in-

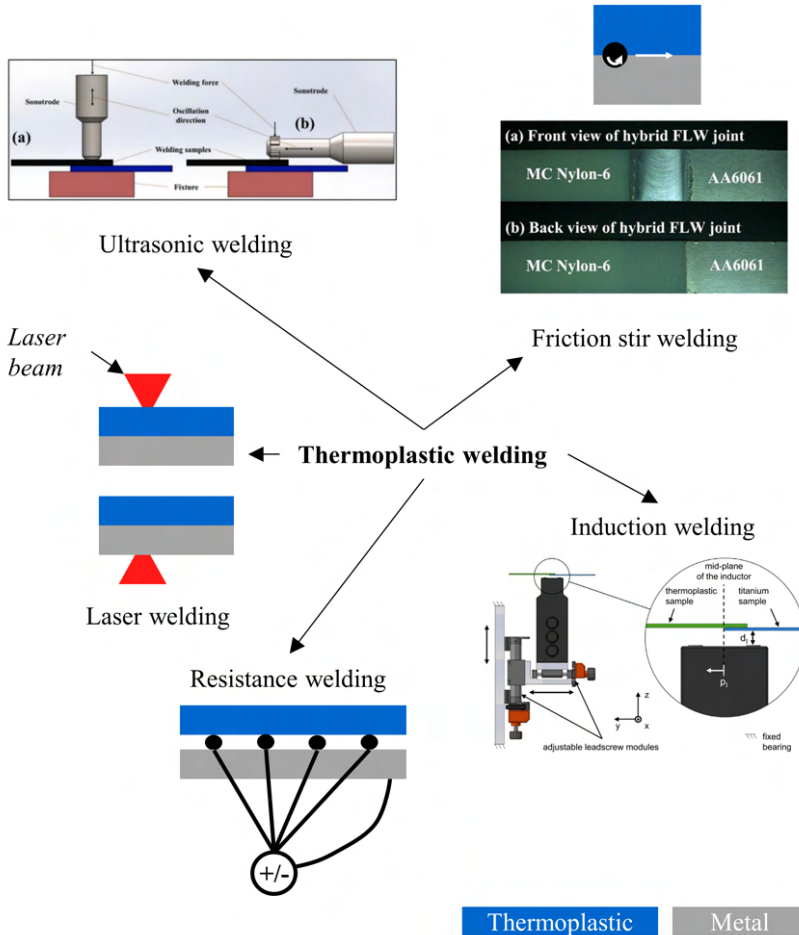


Figure 2.2 Thermoplastic welding technologies: ultrasonic welding, laser welding, induction welding, resistance welding and friction welding. Images taken from [18–20].

duction welding [20]. In any case what matters is the amount of heat provided to the thermoplastic material in order to melt it without degrading it. Hence, the achieved temperature is important, but also the applied pressure. Indeed, the pressure must be high enough to induce penetration of the thermoplastic into the surface roughness to create intimate contact but must be also low enough to prevent to squeeze out a large amount of thermoplastic material. Thermoplastic welding works to bond either two ther-

moplastic based adherends together or one with another material such as a metal.

Adhesive bonding is thus an interesting solution for the joining of dissimilar materials when welding is impossible as adhesive joints offer lightweight and homogeneous stress transfer. However, due to the time- and labour-intensive process, the low fracture toughness of joints and the high probability of defects in the bondline, adhesive bonding remains limited in high performance applications. Section 2.2 deals with solutions to improve the bonding process while Section 2.3 summarizes investigated toughening strategies for adhesive joints.

2.2 Co-curing of composite joints

Adhesive bonding refers to three different manufacturing processes: (1) secondary bonding, (2) co-bonding and (3) co-curing as shown in Figure 2.1. Co-curing and co-bonding allow to improve the bonding process efficiency for composite joints by simultaneously bonding composite laminates and curing at least one of them. Co-curing has already been applied to the joining of composite-composite, composite-metal and composite-sandwich structures (subsection 2.2.1) [22–26]. As the co-curing of composite laminates and adhesive results in lower joint fracture toughness, it has been suggested to replace the adhesive by a thermoplastic film (subsection 2.2.2) having good interactions with the composite matrix (subsection 2.2.3).

2.2.1 Co-curing of composite joints with adhesive

Co-curing of composite pre-pregs and adhesive has already been applied to the joining of composite-composite, composite-metal and composite-sandwich structures. The defects coming from the co-curing process, the dimensional stabilities of the so-produced parts and their mechanical properties have been investigated. According to Kim et al. [22], the co-curing of hat-stiffened panels results in high geometric accuracy and improved mechanical properties compared to co-bonding and secondary bonding that leads to the lowest accuracy. Although, in co-cured composite sandwich structures, a higher level of defects is sometime reported such as increased level of porosity and poor bonding between the composite facesheets (pre-pregs) and the sandwich core due to pre-preg resin flowing into the adhesive and the sandwich core [23–25]. Mohan et al. [27, 28] and Brito

al. [26] compared the mechanical performances of co-cured and secondary bonded joints. Both reported a lower mode I fracture toughness for co-cured composite joints than for secondary bonded joints. For co-cured joints, interfacial failure tends to take place instead of cohesive failure as in secondary bonded joints, reducing G_{Ic} . The main origin of this difference is coming from the presence of voids developing at the adhesive-composite interface due to the release of free and bound water from the pre-pregs and the adhesive during the co-curing process. For secondary bonded joints, the composite laminate being previously cured, the joints are only affected by free and bound water from the adhesive reducing the amount of voids. Another consequence of this phenomenon is the lower glass transition temperature T_g of the adhesive in the co-cured assembly compared to the secondary bonded composites. Mohan et al. [28] also reported a decrease of the Mode II and mixed-mode fracture toughness for co-cured joints. Brito et al. [26] also investigated the influence of hygrothermal ageing on the properties of co-cured and secondary bonded joints. Co-cured joints are less affected by hygrothermal ageing than secondary bonded joints. In secondary bonded joints, hygrothermal ageing leads to adhesive failure instead of cohesive failure as the absorbed moisture tends to weaken the fibre-matrix interface. Consequently, the joint fracture toughness is more negatively affected than in co-cured joints in which adhesive failure already takes place in un-aged systems. To conclude, in addition to increasing process efficiency, co-curing improves the geometrical accuracy of bonded parts but it also leads to a larger number of defects such as voids resulting in a lower fracture toughness compared to secondary bonded joints. For this reason, Quan and co-workers [15, 29–32] suggested to replace the adhesive by a thermoplastic film for co-cured joints as introduced in the following subsection 2.2.2.

2.2.2 Co-curing of composite joints with thermoplastic interlayers

Due to the lower G_{Ic} of the co-cured composite joints, Quan and co-workers [15, 29–32] suggested to replace the adhesive in co-cured composite joints by a thermoplastic film, a methodology already suggested to increase composite delamination fracture toughness [33, 34]. In addition, replacing the adhesive by a thermoplastic film removes the need for selecting only adhesives cured at a temperature similar to the one of the composite. A good bonding strength between the composite matrix and the thermoplastic can be achieved if they show a good compatibility [5]. These interactions can

be of different type and will be further developed in subsection 2.2.3. Compared to adhesives, thermoplastic films have an infinite shelf-life, will not overflow during the curing process, do not require low storage temperature and the bonding thickness can be easily controlled. Here is a number of examples from the literature.

Van Velthem et al. [33] inserted phenoxy and polyethersulfone (PES) films. They reported an improvement in G_{Ic} of +100% when inserting 40 μm thick phenoxy film and +150% with 80 μm thick films while the PES film led to a decrease in G_{Ic} . It is believed that this difference arises from the better compatibility between epoxy and phenoxy than between PES and epoxy, resulting in the complete dissolution of the phenoxy film but not of the PES film. On the contrary, Cheng et al. [34] reported complete dissolution of porous PES film with thicknesses ranging from 20 to 100 μm . This leads to an increase of the mode I and mode II delamination fracture toughness that depends on the film thickness. An improvement of the interlaminar shear strength is also reported. Cohesive propagation in the interdiffusion layer and formation of microcracks due to the presence of PES spheres are at the origin of the improvement in fracture toughness.

Surface treatments have been applied to TP film to promote adhesion. Using UV-irradiation, Quan et al. [29] investigated the co-curing of polyetheretherketone (PEEK) and polyphenylene sulfide (PPS) composites with an epoxy-based composite. Both the mode I fracture toughness and the lap-shear strength of the co-cured joints are lower than for the secondary bonded joints. Without UV-irradiation treatment, no adhesion could be achieved. This study highlights the importance of a surface treatment at the thermoplastic side to achieve bonding using a thermoplastic film.

In later studies, Quan and co-workers [30, 31] investigated the co-curing of epoxy-based composites with a PEEK or a polyetherimide (PEI) film. The PEEK film requires a surface treatment while bonding can be achieved with PEI without any surface treatment. Regarding the co-cured PEI-composite joints, they are characterized by a lower mode I fracture toughness and a larger mode II fracture toughness at room temperature compared to secondary bonded joints while at 130°C, the PEI co-cured composite joints better perform in both cases than the adhesively bonded joints. During co-curing, interdiffusion between the epoxy resin and the PEI takes place. The interdiffusion layer exhibits a nodular morphology with epoxy nodules embedded in PEI. Particle debonding and void growth are observed at

room temperature while at 130°C, the crack tends to propagate at the interface with the carbon fibres where it can be pinned by PEI particles. Under mode II loading, discontinuous cracks develop ahead of the crack tip in the PEI film due to the ductility and high failure strength of PEI. Co-cured lap-shear joints also exhibit a shear strength larger than the adhesively bonded systems at both room temperature and 130°C. For all these properties, a maximum is reported when the PEI film is 200 μm thick. Then, for the PEEK-composite co-cured joints, a higher fracture toughness is reported in both mode I and in mode II at room temperature and 130°C compared to the secondary bonded joints. Under mode I, the high fracture toughness results from the cohesive crack propagation in the PEEK film and large plastic deformations of PEEK. Under mode II loading, discontinuous cracks develop as for PEI co-cured joints. Regarding the lap shear strength, it is equivalent to the one of secondary bonded joints at room temperature while at 130°C, PEEK co-cured joints perform about 3 times better than secondary bonded joints. Regarding the fracture toughness, it is maximized for 250 μm thick PEEK films (the thicker tested films) while the thickness has no influence on the lap-shear strength. Liu et al. [32] inserted PEEK meshes and a PEEK tape with carbon fibres instead of a plain film resulting in a fracture toughness larger than the reference laminate, but lower than co-cured joints with a plain PEEK films due to either adhesive failure between the PEEK mesh and the composite or modest PEEK fracture limiting the fracture toughness. The largest fracture toughness was achieved with the PEEK tape containing carbon fibres. Using the same PEEK tape, Quan et al. [15] reported improved mode I and mode II fracture toughness and lap shear-strength compared to secondary bonded joints. Composite co-cured joints with PEI and PEEK films surpass thus the mechanical performances of secondary bonded joints or adhesively co-cured joints, except for the mode I fracture toughness of PEI co-cured joints at room temperature. In addition, all conditions led to higher performances than the reference laminate (composite without adhesive).

Composite laminates have been co-cured with thermoplastic films not only to produce co-cured joints but also to generate a thermoplastic layer at the surface of thermoset composite to perform thermoplastic welding [5, 35–39] or to improve some surface properties such as erosion resistance [40]. Upon co-curing, two composite-thermoplastic laminates can be welded together. To meet this objective, several thermoplastics have been used as surface layers: PEI [35, 36], PEEK [35, 36], poly(methyl methacrylate)

(PMMA) [37], PPS [36], polyvinylidene fluoride (PVDF) [36], polyamide 12 (PA12) [37], polypropylene (PP) [39], polysulfone (PSU) [41] and phenoxy [37]. Zweifel and Brauner [37] reported interphase formation in PMMA-epoxy and phenoxy-epoxy systems but not in PA12-epoxy systems prior to welding. Upon welding two composite-thermoplastic laminates together, the system made of the phenoxy layer is characterized by the highest lap-shear strength due to high diffusion between epoxy and phenoxy resulting in cohesive failure instead of interphase failure. The PA12 welds exhibited a higher lap shear-strength than the PMMA welds even in the absence of any interphase due to cohesive failure. PMMA welds failed adhesively although they presented a good weld quality. Regarding PEI, PEEK, PPS and PVDF, Brauner et al. [36] reported the formation of an interphase only for the composite laminates co-cured with a PEI surface layer. Upon welding of two composite-PEI laminates, a lap-shear strength of 38 MPa is reached with cohesive failure indicating a strong interphase between PEI and epoxy. Fernandez Villegas and van Moorlegheem [35] welded epoxy composites with a PEI surface layer to PEEK composites. Upon welding, the PEI-epoxy interphase is almost unaffected and the joints are characterized by a lap-shear strength about 25 MPa. Replacing the PEI surface layer by a UV-treated PEEK layer led to similar performances but spontaneous peeling has been reported after few months of storage. Rubino et al. [39] used PP as surface layer material. However, no interphase is reported in this case but the excellent properties of the welded co-cured joints are achieved by hot-pressing the PP film with one fibre ply so it partially impregnates the surface ply of the composite laminate. Finally, Erartsin et al. [40] co-cured a polyester composite with both ABS and PC films for applications to leading edge protection. Better mechanical properties are reported when the composite is co-cured with ABS than with PC due to a better compatibility between the two polymers. In PC-polyester systems the crack is found to propagate at the interface between the developed morphological gradient and the composite while in the ABS system, the crack propagates through the morphological gradient.

The co-curing of thermoset composite joints with thermoplastic films has already been investigated during the last decade to toughen composites, to replace adhesives or to enable thermoplastic welding with thermoset composites. The studies [29–31, 33, 35–39] show that good interactions between the composite resin and thermoplastic layer material can lead to the formation of a strong interface. Although, even in the absence of interactions,

some strong joints can be manufactured. The following section discusses the interactions between different thermoplastics and epoxy resins.

2.2.3 Thermoplastic-epoxy interactions

Interactions between thermoplastic and epoxy can be of two different types: (i) interdiffusion of the polymers resulting in a chemical gradient and (ii) chemical reactions between functional groups to create, for example, covalent bonds. These interactions depend upon many parameters such as the curing cycle, the molecular weight of the thermoplastic, the viscosity of the resin, the solvent used to prepare blends and, the hardener. In what follows, interactions between epoxy and several thermoplastic used in the present work (see Chapter 3 and 4) will be discussed for blends and some interphases. The influence of the processing parameters and the mechanical performances of blends and interphases are discussed after introducing the Hansen solubility parameter, a criterion for polymer compatibility.

Criterion for polymer interactions

The Hansen solubility parameter is often used [42] to determine the compatibility of two polymers. One of the two polymers must be a solvent. In the case of TP-epoxy blends, the uncured epoxy resin can be considered as the solvent. For each molecule/polymer, one needs three parameters:

- δ_{di} : the energy from the dispersion forces (intermolecular forces such as Van der Waals forces) between the molecules of polymer i .
- δ_{pi} : the energy from dipolar intermolecular forces between molecules i .
- δ_{hi} : the hydrogen bonds energy of molecules i .

They can be seen as coordinates in a 3D space. The closer the molecules from each other, the more likely they will interact with each other. The distance between two molecules A and B is given by

$$R_a = \sqrt{4(\delta_{dB} - \delta_{dA})^2 + (\delta_{pB} - \delta_{pA})^2 + (\delta_{hB} - \delta_{hA})^2}, \quad (2.1)$$

A second required value is the interaction radius R_0 of the molecule being dissolved (the thermoplastic). This is the radius of a sphere of which the centre corresponds to the coordinate $(\delta_{di}, \delta_{pi}, \delta_{hi})$. R_0 has been computed for many molecules and can be found in tables. Knowing R_a and R_0 , the relative energy difference ratio (RED) can be computed and is given by

$RED = R_a/R_0$. Two molecules present a high-affinity if it is lower than 1 while if insolubility corresponds to a RED larger than 1. TP-epoxy blends can thus be processed if the RED ratio is lower than 1. For example, for epoxy blended with PEI, PSU and PES, the RED value is about 0.8, 0.8 and 0.4, respectively [43].

Epoxy and Polyetherimide (PEI)

Polyetherimide is one of the thermoplastics used to toughen epoxy resins owing to its high stiffness, strength, toughness and temperature resistance.

□ PEI-epoxy blends

PEI-epoxy blends are characterized by different morphologies mainly depending on the PEI content [12, 44–48]. It is often reported that below 15 wt%, the blends exhibit a sea-island morphology due to a nucleation and growth mechanism (Figure 2.3a). Above 15 wt%, phase inversion takes place leading to a nodular morphology (Figure 2.3c). Around 15 wt%, a co-continuous morphology is observed with regions characterized by the one or the other morphology (Figure 2.3b). The PEI molecular weight has also an influence on the observed morphologies. Indeed, Wang et al. [48] observed ribbon-like morphology for blends containing 5 to 20 wt% of PEI by increasing the PEI molecular weight, as shown in Figure 2.3d.

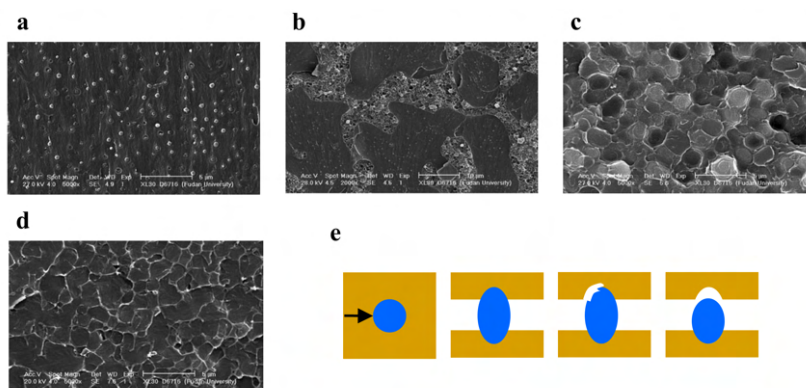


Figure 2.3 Morphologies of PEI-epoxy blends containing a) 10 wt%, b) 15 wt% and c) 20 wt% of PEI. d) morphology for a blend containing 10 wt% of PEI with a higher molecular weight. e) Failure mechanism of low PEI content blends: deformation of PEI domain bridging the crack and subsequent interfacial decohesion. Taken from [48].

The blend morphology influences the mechanical properties and the failure mechanisms taking place in the blends. Regarding, the Young's modulus [44, 45, 47, 48], no influence of the PEI content and of the morphology is reported. However, a general increase of the yield and the tensile strength is found when the PEI content is increased with a sharper increase when phase inversion takes place [44, 45, 47, 48]. Some authors reported a small decrease of strength at around 15 wt% [12, 44, 48]. It is believed that the origin of this decrease is due to the large number of weak PEI-epoxy interfaces in these blends exhibiting a sea-island morphology [44]. Indeed, weak adhesion between PEI and epoxy domains is often reported due to their different polarity and the lack of interacting functional groups [47]. In addition, increasing the PEI content improves the blend ductility [44, 45, 47, 48]. The main investigated property is the fracture toughness of the blends [12, 44–48]. An increase of fracture toughness is reported as the PEI content increases with a slight increase for low PEI content and a sudden increase at the transition between the sea-island and the nodular morphology. For example, by increasing the PEI content from 15 to 20 wt%, Kim and Kim and Harismendy et al. [45] reported a doubling of G_{Ic} . The evolution of the fracture toughness with the PEI content is related to the failure mechanisms taking place in the blends depending on the morphology [44, 49, 50]. At low PEI content, fracture is brittle. The slight toughening effect results from the deformation and bridging of the crack by the PEI domains. The crack can further propagate due to the decohesion between the epoxy matrix and the deformed PEI domains as shown in Figure 2.3e. For blends exhibiting a nodular morphology, ductile tearing of the PEI matrix and decohesion between the epoxy domains and the PEI matrix are reported.

□ PEI-epoxy interphases

The development of PEI-epoxy interphases has also been investigated depending on the curing cycle, curing agent and PEI film thickness. During the curing, the epoxy resin swells the PEI and both polymers are able to diffuse in each other, resulting in a chemical gradient and thus in a morphological gradient. The morphological gradient exhibits generally three zones [12, 44, 49]: (i) close to the PEI film, a region with a nodular morphology referred to as PEI-rich zone, (ii) a zone exhibiting a co-continuous morphology, referred to as transition zone and, finally, (iii) a region characterized by a sea-island morphology away from the PEI film referred to as epoxy-rich zone. These regions are shown in Figure 2.4a and c. The transition zone is not always observed [12, 49, 51]. Kim and Kim [44] re-

ported a reduction of the interphase length as the amount of curing agent is increased as the reaction tends to dominate over the diffusion of the two polymers. The curing cycle can also have an influence on the interphase as shown by comparing Figure 2.4a (fast cycle with dwell stage) and Figure 2.4b (slow cycle).

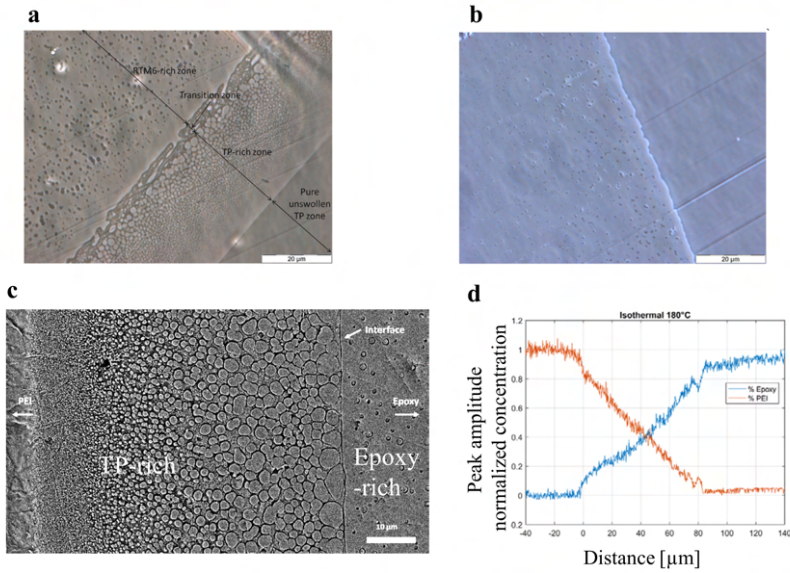


Figure 2.4 Morphological gradient observed by a)-b) Voleppe et al. for a fast curing cycle with a dwell stage at 160°C and b) a slow curing cycle without any dwell stage. c) Morphological gradient observed by Teuwen et al. and d) the associated chemical gradient similar to the one proposed by Lestriez. Taken from a)-b) [12] and c)-d) [52].

Some authors cure their PEI-epoxy interphase in two steps. The first step is the dwell stage during which the species diffuse and occurs after increasing the temperature up to a certain temperature T_{dwell} and holding it for a certain time before increasing the temperature up to the final curing temperature. Zweifel et al. [53] investigated diffusion during the dwell stage. They found that the diffusion length of the species increases as T_{dwell} increases. In addition, they reported Fickian and non-Fickian diffusions to occur simultaneously. Farooq et al. [51] investigated the influence of the dwell stage in fully cured epoxy-PEI interphases. The interphase gets thicker as T_{dwell} increases up to 160 °C. For higher T_{dwell} the interphase

thickness starts to decrease. In addition, the duration of the dwell stage, referred to as the dwell time t_{dwell} also plays a role, leading to a thicker interphase if the dwell stage finishes at the onset of phase separation than when ending when 80% of conversion is achieved. In addition, T_{dwell} and t_{dwell} influence the size of the observed features along the morphological gradient. As T_{dwell} increases, the size of the epoxy domains in the PEI-rich zone close to the epoxy-rich zone increases. t_{dwell} does not influence the size of these large epoxy domains, but well influences the frequency of small epoxy domains close to the PEI film.

Teuwen et al. [52] reported concentration profiles upon curing similar to the profile of Lestriez in the same systems as Farooq et al. [51] (Figure 2.4d). Voleppe et al. [12] also investigated different curing cycles resulting in different morphological gradient (Figure 2.4a-b). The PEI and epoxy-rich zones are also reported when diffusion dominates (Figure 2.4a) but not if the curing cycle is very low and that curing reaction is favoured (Figure 2.4b). However, the features observed at the transition between the epoxy-rich and PEI-rich zone differ from what is commonly found. Indeed, the transition zone is more tortuous as shown in Figure 2.4a compared to the classical observation shown in Figure 2.4c.

The failure mechanisms taking place in specimens containing PEI-epoxy interphases have been recently investigated. Vadugappatty Srinivasan and Vassilopoulos [54] and Farooq et al. [49] ran crack perpendicular to the interface. Vadugappatty Srinivasan and Vassilopoulos [54] initiated the crack in the epoxy resin and observed that when approaching the PEI film, the crack is deflected along the PEI film and does not propagate through it, significantly improving the fracture toughness. Farooq et al. [49] investigated the failure mechanisms of PEI-epoxy stacks cured following several curing cycles and with PEI films of different thicknesses. For low T_{dwell} , short cracks were deflected along the interphase before propagating through the PEI film (Figure 2.5b) while for high T_{dwell} and/or long crack, no deflection was observed, but only propagation through the stacking (Figure 2.5c). Mactotoughening is found if the remaining PEI layer is thicker than the PEI plastic zone (62 μm) upon curing. If it is thinner, only microtoughening due to the presence of large epoxy particle (larger than 3 μm) in the PEI-rich part of the morphological gradient is generated, see Figure 2.5a. Depending on the investigated system, fracture toughness even larger than the one of PEI were obtained.

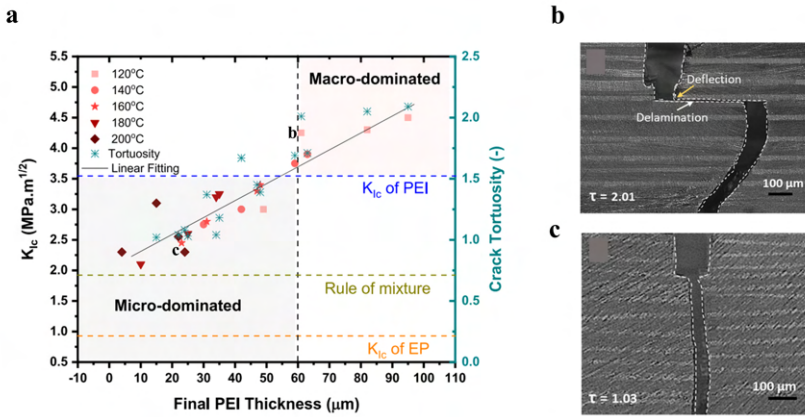


Figure 2.5 a) Variation of the fracture toughness perpendicular to PEI-epoxy interphase with the residual PEI film thickness. The shades of red correspond to different dwell temperatures. b) Deflection of a short crack in a sample cured at a dwell temperature of 120°C with an initial film of 60 μm and c) no deflection for T_{dwell} equal to 180°C. Taken from [49].

Opposite to these works, Voleppe et al. [12] propagated cracks parallel to the interface and reported a toughening effect associated with a crack trapping mechanism taking place in the PEI-rich zone of the morphological gradient shown in Figure 2.4a. The different failure scenarios are shown in Figure 2.6 with the trapping mechanism corresponding to the scenarios 1, 2 and 3. This mechanism is not well understood but they tentatively explain that this crack trapping mechanism originates from a softening (lower yield stress) in the transition zone as shown in Figure 2.6b. In addition, they reported an interphase fracture toughness dependent upon the curing cycle with the highest values about $880 \text{ J}/\text{m}^2$ if the temperature is increased at a rate of $2^\circ\text{C}/\text{min}$ until the dwell stage occurring at 160°C for 90 minutes and if the final curing stage takes place at 180°C for two hours.

Epoxy and Polyamide (PA)

Polyamide is a potential candidate for co-curing owing to its ability to form covalent bonds with epoxy. PA-epoxy interactions have been investigated in the context of the development epoxy-nylon adhesives. Most of the studies focuses on the cure kinetics and the different phases present in PA-epoxy blends. Blends can be obtained by either dissolving PA and epoxy

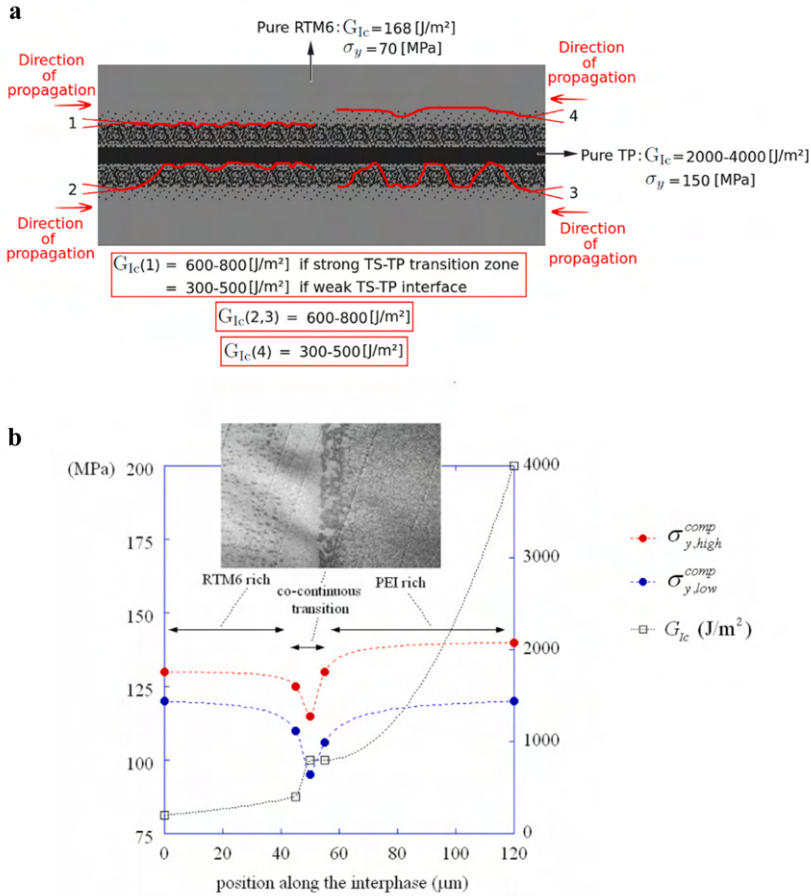


Figure 2.6 a) Failure mechanisms taking place in PEI-epoxy interphases for cracks running parallel to the PEI film observed by Voleppe et al.[12]. Mechanisms 3 and 4 correspond to the trapping mechanism that is believed to originate from soft transition zone. b) Variation of yield strength and fracture toughness along the interphase. Taken from [12].

in an alcohol based solvent or by adding fine polyamide particles in the epoxy matrix, leading to different results. Wang and Chen [55] and Zong and Guo [56] both used the dissolution technique to process their blends. Wang and Chen observed three phases in PA-epoxy blends containing 60 to 100 wt% of PA: a semi-crystalline PA phase, an amorphous epoxy phase and an amorphous PA-epoxy phase. On the contrary, Zong and Guo did

not add any curing agent forcing the epoxy pre-polymers to react with the PA chains and resulting in a single amorphous PA-epoxy phase. Kim et al. [57] added nylon powder into the epoxy and mixed the blends, all containing 5 wt% of PA6 at different temperatures. Different morphologies were found depending on the mixing temperature. For low temperatures, the PA particles and the epoxy resin can well be distinguished from each other while when mixing takes place at temperatures higher than 215°C, the interface between the PA particles and the epoxy is less distinguishable due to the reaction between the primary amine end group and the epoxide group. Zong and Guo also reported an additional reaction for blends containing at least 63 wt% of epoxy as the products of the first mentioned reaction become the reactants.

The mechanical properties of PA-epoxy blends have been investigated in the literature [55, 57, 58]. Wang and Chen [55] reported a maximum tensile strength for blends containing 90 to 95 wt% of PA due to stress-induced crystallization. However, by performing lap-shear tests, the maximum shear strength is attained by adding 70 wt% of Nylon into epoxy. Gorton [58] also reported an increase in lap shear strength by adding 15 wt% of epoxy in PA from 19.7 MPa to 46.7 MPa. Blends containing 5 to 10 wt% of epoxy are characterized by the highest tensile strength. According to Gorton this originates from the double effect of the epoxy resin that simultaneously plasticizes and crosslinks nylon. Crosslinking tends to increase the tensile strength while plasticizing tends to decrease the yield strength. The combination of both leads thus to a maximum. Kim et al. [57] investigated the influence of the mixing temperature on PA-epoxy blends containing 5 wt% of Nylon. A maximum fracture toughness was found for blends mixed at 215 °C, just below the melting temperature of PA resulting from a combination of crack path altering, primary crack bridging, initiation of secondary cracks and ductile fracture of the nylon phase. In addition, increasing the mixing temperature promotes the development of tiny crystal lamellae forcing the crack to proceed as a series of fingers and not as a straight crack.

Epoxy and Polysulfone (PSU)

Polysulfone has been used as additive in epoxy resins due to its high T_g about 190 °C, high strength, good electrical properties and its high resistance to many solvents. Furthermore, adding PSU to epoxy resins proved to have a toughening effect.

□ PSU-epoxy blends

Depending on the functional end groups and on the molecular weight of PSU, different morphologies can be observed. With unreactive PSU, blends containing less than 15 wt% of PSU exhibit a sea-island morphology with PSU domains embedded in the epoxy matrix (Figure 2.7a-b) while a nodular morphology resulting from phase inversion is observed above 20 wt% of PSU (Figure 2.7d) [59–61]. Around 15 wt%, a co-continuous morphology with some regions exhibiting the nodular morphology and others the sea-island morphology is reported (Figure 2.7c). These morphologies are not always reported. For example, for low molecular weight PSU, Hedrick et al. [62] only reported a single phase morphology in which PSU and epoxy cannot be distinguished from each other.

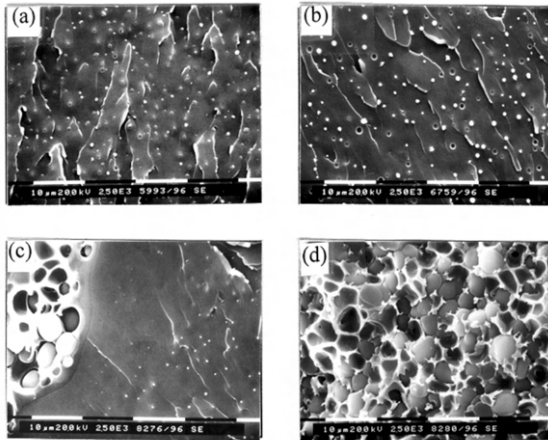


Figure 2.7 Morphologies of PSU-epoxy blends containing a) 5, b) 10 c) 15 and 20 wt% of PSU. Taken from [60].

Regarding the evolution of the glass transition temperature T_g with the PSU content, Martinez et al. [61] reported a decrease of the T_g from 15 wt% of PSU while PSU is characterized by a higher T_g than the used epoxy resin. This is believed to be due to the phase inversion taking place for these blends. This agrees with the results of Min et al. [59] who reported a decrease in epoxy conversion as the PSU content increases.

Regarding the mechanical properties, due to the high stiffness of PSU, the Young's modulus of PSU-epoxy blends is known to not be drastically af-

affected by the increasing amount of PSU and can even be increased [59–62]. The same accounts for the tensile strength. Increasing the PSU content improves the blend fracture toughness as already reported for PEI-epoxy blends due to changes in morphology. At low PSU content, G_{Ic} slightly increases with the PSU content and the toughening mechanism is crack path deflection and crack pinning by the PSU domains that can also plastically deform [59–62]. However, decohesion between the PSU domains and the epoxy matrix is reported in the absence of reactive end-groups due to low interfacial adhesion between the two polymers. Above 15 wt% of PSU, fracture toughness is significantly improved due to phase inversion as the matrix now corresponds to the tough PSU in which the epoxy domains are embedded. Adhesion between epoxy and PSU can be increased by adding some functional groups, resulting in a significant improvement of the fracture toughness. As reported by Fu and Sun [63], in that case, the toughening effect depends on the end-groups with a higher effect being obtained with hydroxyl-terminated PSU (reactive) than chlorine-terminated PSU (unreactive). Finally, by increasing the molecular weight of PSU, fracture toughness can also be improved [62].

□ PSU-epoxy interphases

Min and Kim [60] inserted a PSU (no reactive end groups) film in an epoxy resin prior to curing. Upon curing, a morphological gradient forms as shown in Figure 2.8, similar to the one observed for PEI-epoxy interphases. Close to the PSU film, a 240 μm thick PSU-rich zone exhibits the nodular morphology (Figure 2.8b-c) while away from the film, the sea-island morphology corresponds to a 260 μm thick epoxy-rich zone (Figure 2.8e). This originates from the the interdiffusion of the two polymers, leading to a chemical gradient. In addition, a clear boundary region shows up between the two zones which does not exhibit a co-continuous morphology due to the steep chemical gradient (Figure 2.8d).

Epoxy and Polyethersulfone (PES)

PES is another thermoplastic used as epoxy resin toughener.

□ PES-epoxy blends

As for PEI and PSU, the curing cycle, the PES molecular weight or the end-groups influence the observed morphologies and the mechanical properties of PES-epoxy blends. At low PES contents [64–66], a sea-island morphology can be observed. Increasing the PES molecular weight increases

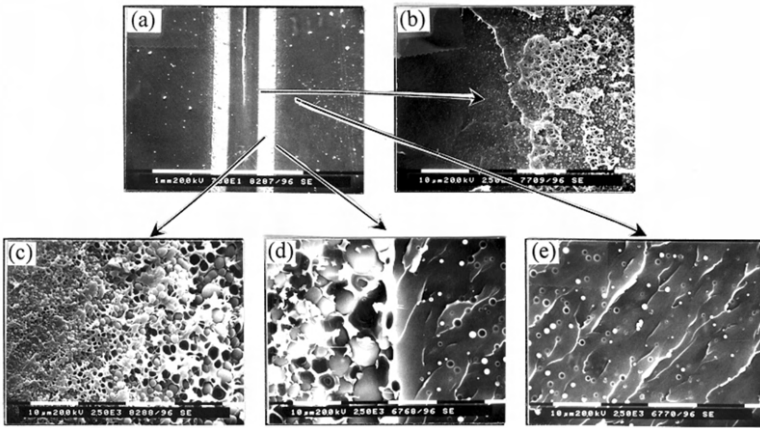


Figure 2.8 Morphological gradient of PSU-epoxy interphase: a) whole interphase, b) PSU-rich zone close to PSU film, c) PSU-rich zone with nodular morphology, d) boundary region between the PSU and epoxy rich zones and d) epoxy-rich zone. Taken from [60].

the PES nodules size. The curing cycle is also reported to have an influence [66]. It is often reported that the observed nodules are mainly made of PES but also of crosslinked epoxy resin as they cannot be dissolved due to the reaction of the hydroxyl end-groups of PES with the epoxy [64]. At larger PES content, a co-continuous morphology (20 wt% of PSU) or a phase-inverted morphology (minimum 25 wt% of PSU) are found [67]. The phase inverted morphology corresponds to epoxy-rich domains embedded in a continuous PES-rich phase. With some epoxy resins, phase separation does not occur [64].

Different trends have been observed regarding the mechanical properties of PES-epoxy blends, depending on the PES reactive functional end groups. The Young's modulus is not affected by the addition of PES into the epoxy resin [64]. Bucknall et al. [64] reported a limited change in fracture toughness by adding PES in epoxy resin. This slight increase is reported to be a consequence from the deviation of the crack front around the PES-epoxy nodules. According to Raghava [65], the blend G_{Ic} is also dependent on the PES molecular weight with a higher molecular weight leading to a higher toughness due to the higher ductility of PES. The influence of PES on fracture toughness also depends on the functional end-groups. According to MacKinnon et al. [63], non-reactive PES leads to an improved toughening

effect compared to reactive PES. Opposite, Yoon et al. [67] did not report any difference between reactive and unreactive PES at low PES contents while at high PES contents, the toughening effect is enhanced for reactive PES.

□ PES-epoxy interphases

Voleppe et al. [68] inserted a PES chlorine-terminated (unreactive) film in the RTM6 epoxy resin. The resulting morphological gradient can be decomposed into two zones: a PES-rich zone exhibiting a phase-inverted morphology and an epoxy-rich zone exhibiting a sea-island morphology, as shown in Figure 2.9a. However, this gradient is dependent upon the curing cycle. Adding a dwell stage at 160°C leads to a longer penetration front (66 μm). In the absence of this plateau, a fast curing cycle also results in a longer interdiffusion length (33 versus 3 μm). The interphase resulting from the lowest curing cycle is shown in Figure 2.9b while the morphological gradient in Figure 2.9a is representative of a fast curing cycle with a dwell stage. As for PEI-epoxy interphases, Voleppe [69] investigated the failure of the PES-epoxy interphase parallel to the PES film. Two populations of samples were produced with different G_{Ic} around 270 and 600 J/m². The brittle population corresponds to a crack propagating in the epoxy-rich zone or at the interface between the PES and epoxy-rich zones while the tough population is associated with crack propagation in the PES-rich zone. The low fracture toughness of samples for which the crack propagated at the interface between the two zones of the morphological gradient originates from the low mechanical interlocking between the two regions and the lack of interactions between PES and epoxy.

Epoxy and polycarbonate (PC)

Polycarbonate is another potential thermoplastic toughener added in epoxy resins. Studies focused on the interactions between PC and epoxy [70–72], but not on the mechanical properties of these blends. In many PC-epoxy blends, a single homogeneous phase is observed and during the curing of the blend, reaction between epoxy and PC takes place. Don and Bell [70] and Li et al. [71] reported chain scission during the curing reaction at 150°C resulting from a transesterification reaction between the secondary hydroxyl groups of the epoxy and the carbonate groups of PC. Opposite to common results, Lee et al. [72] reported a slightly heterogeneous morphology associated with a rough fracture surface indicating some potential toughening. Although, compared to other blends, this morphology is al-

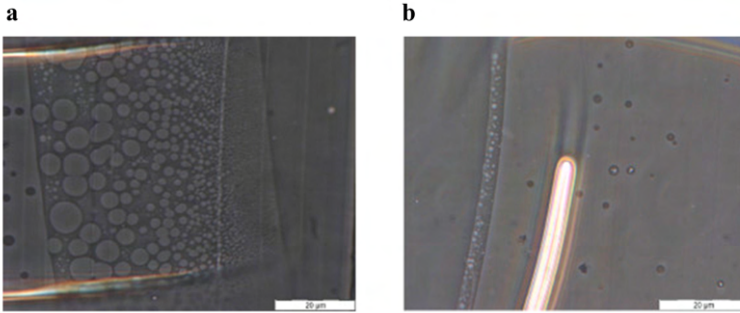


Figure 2.9 Morphological gradient of PES-epoxy interphase depending on the curing cycle: a) 90 min at 160°C and 2h at 180°C with a heating rate of 2°C/min, b) 2h at 180°C with a heating rate of 0.2°C/min. Taken from [68].

most homogeneous due to the degradation of PC during curing, increasing its miscibility with the epoxy resin.

Epoxy and poly(methyl methacrylate) (PMMA)

□ PMMA-epoxy blends

Poly(methyl methacrylate) has been incorporated into epoxy resin as toughener. The properties of PMMA-epoxy blends have not been widely investigated. Studies more focused on the influence of the hardener and, of the PMMA molecular weight on the observed morphologies. Ritzenthaler et al. [73] Rastegar et al. [74] reported an influence of the epoxy resin hardener on the morphology. For a given PMMA concentration, depending on the hardener, a sea-island, a co-continuous morphology or a nodular morphology can be observed. Olmos et al. [75] investigated the influence of the PMMA molecular weight on the observed morphology. Increasing the PMMA molecular weight results in a change in morphology going from a sea-island morphology (34 kg/mol) to a phase-inverted morphology (360 kg/mol). Increasing the molecular weight from 34 to 76 kg/mol results in an increase of the size of the PMMA rich domains dispersed in the epoxy matrix. In addition, PMMA domains debond from the epoxy resin indicating low interfacial adhesion in agreement with the results of Rudresh and Mahorti [76]. In addition, Rudresh and Mahorti reported a sea-island morphology for blends containing at least 6 wt% of PMMA while lower PMMA content blends only involve a rough surface. In addition, the frac-

ture toughness increases with the PMMA content from 0 to 6 wt% owing to shear deformation preventing crack propagation. Above 6 wt%, particle debonding can take place and as the number of PMMA domains increases, the fracture toughness slightly decreases due to particle debonding. Finally, the modulus and flexural strength decrease as the PMMA content increases in agreement with the results of Galante et al. However, Galante et al. [77] reported a co-continuous morphology for blends containing both 3 and 7 wt% of PMMA (hardener = MHTPA). The hardener seems thus to have a strong influence on the observed morphologies of PMMA-epoxy blends.

□ PMMA-epoxy interphases

Zweifel et al. [37] investigated the formation of an interphase in PMMA-epoxy systems containing a PMMA film. They reported the formation of such interphase with a PMMA-rich region exhibiting a phase-inverted morphology as shown in Figure 2.10. This interphase has a thickness about 20 μm .

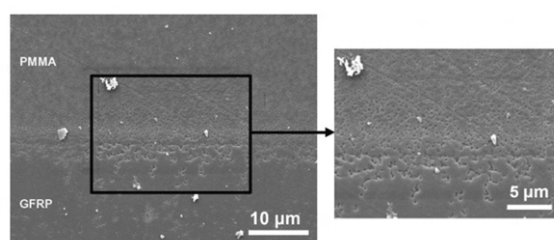


Figure 2.10 Interphase observed at the interface between a PMMA film and glass fibre reinforced composite with an epoxy matrix. Image from [37].

Other thermoplastics

The interactions of epoxy with several thermoplastics have been discussed in the former subsections. However, this is not a comprehensive list and interactions are possible with other thermoplastics, but which are not used in Chapter 4 as potential interlayer materials allowing the manufacturing of co-cured composite joints. Epoxy has been reported to have a good compatibility with phenoxy [33], poly(phenylene oxide) (PPO) poly(ethylene oxide) (PEO), poly(vinylpyrrolidone) (PVP), PEEK, polystyrene (PS) or polyimide (PI) [5, 78].

2.3 Toughening of adhesive joints and of other assemblies

One of the main disadvantage of adhesive bonding is the inherent presence of flaws. These defects originate from the manufacturing process (kissing bonds due to contamination, voids) or appear during the service-life due to the mechanical loading or due to the exposure to the environment (solvents, thermal changes, UV, ...). In order to avoid joint failure, the crack propagation resistance, and thus the fracture toughness, is an important properties of adhesives. However, most structural adhesives used today in the industry are made of thermosets which are strong and stiff but brittle. Consequently, increasing the joint fracture toughness has been a topic of particular interest for the last decades. Two main strategies can be considered: (i) the chemical modification of the adhesive and (ii) the development of extrinsic toughening mechanisms. The chemical modification strategy is first briefly introduced. As the goal of the present work is to increase the joint fracture toughness by investigating different extrinsic toughening strategies, strategies and designs already suggested in the literature are presented, as summarized in Figure 2.11. Finally, strategies to improve other joint properties such as the shear strength are briefly summarized.

2.3.1 The toughness of adhesive joints

The adhesive joint fracture toughness is dictated by the competition or co-operation between several mechanisms taking place during crack propagation. Each adhesive is characterized by a cohesive energy G_0 which corresponds to pure mode I crack propagation in the neat adhesive and in absence of any extra toughening mechanisms. For adhesives which do not contain a secondary phase, this corresponds to the fracture toughness of very thin joints. When toughening mechanisms take place, the total fracture toughness is the sum of G_0 with other contributions:

$$G_{Ic}^{joint} = G_0 + \Gamma^p + \Gamma^{deflection} + \Gamma^{bridging} + ... \quad (2.2)$$

with Γ^p the energy dissipated by plastic deformation, $\Gamma^{deflection}$ additional energy dissipated by mode II crack propagation when a crack is deflected and $\Gamma^{bridging}$ the contribution of crack bridging. Other contributions are possible, as explained in the next subsections.

In addition, if the failure is cohesive, G_0 is the one of the adhesive while if

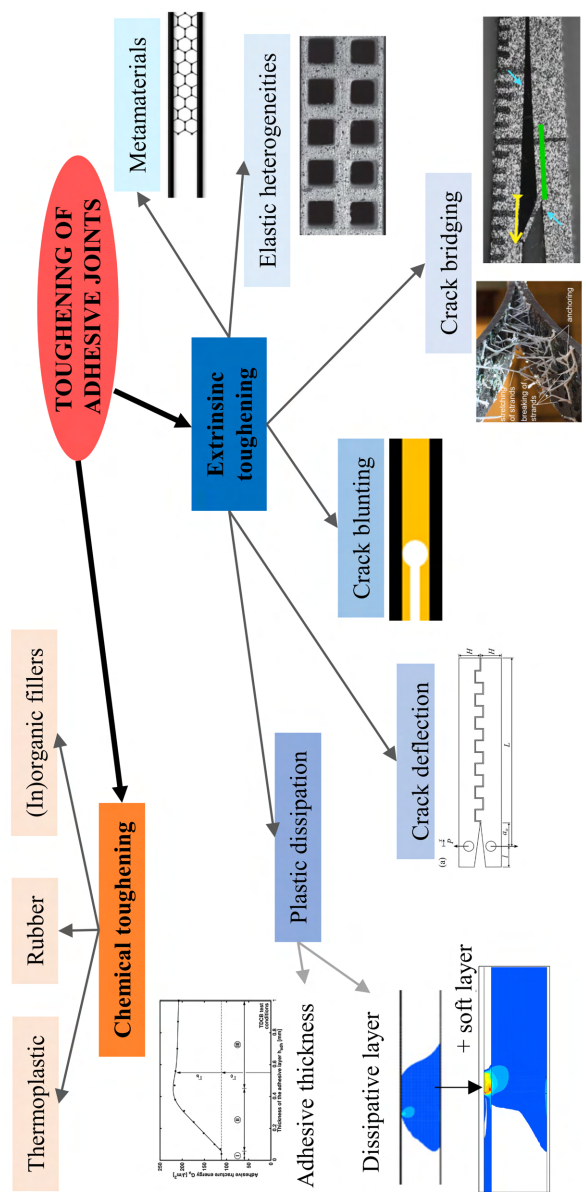


Figure 2.11 Summary of the strategies available to toughen adhesive joints. Images taken from [13, 79–84].

adhesive failure takes place, it corresponds to the resistance of the interface between the adhesive and the adherend. In order to produce tough joints, cohesive failure must be favoured requiring a good adhesion between the adhesive and the adherends.

Modelling joints toughness

Joint fracture toughness can be predicted using finite element models. Several techniques exist to consider crack propagation such as the virtual crack closure technique or cohesive elements [85]. Some models allow to determine the contribution of different mechanisms. For example, this is the case using a cohesive law of energy G_0 embedded in an elastoplastic material to determine Γ^p [13]. To consider bridging, multilinear cohesive laws are often used. In what follows, cohesive zone modelling and the maximum principal stress criterion are introduced.

Cohesive zone modelling (CZM) Cohesive elements are interface elements [86]. In general, the cohesive law is characterized by an elastic part, with a stiffness K , until the traction reaches a critical value σ_c . This part is followed by a softening regime which represents intense damage accumulation. The traction vanishes when the displacement reaches a critical value δ_c . The area under the curve is the cohesive energy Γ_c . The cohesive elements can either represent the whole bondline or two crack faces initially bonded together. In the first case, the cohesive parameters change with bondline thickness while in the second case, they are kept constant and $\Gamma_c = G_0$ and the joint behaviour depends on the surrounding materials. For example, if the cohesive law is embedded in an elastoplastic material, it is possible to predict the influence of the adhesive thickness on the joint fracture toughness [13]. The cohesive law can follow different shapes: trapezoidal, linear, exponential ... For brittle elastic adhesives, the shape does not influence the response. For ductile, plastically deforming adhesives the cohesive law may have an influence and a trapezoidal law must be preferred [87–90]. However, the trapezoidal law often results in more convergence issues than linear and exponential laws. Crack path prediction based on cohesive zone modelling suffers from mesh dependency in the sense that the crack path is determined by the position of the cohesive elements and the mesh topology (structured VS unstructured).

Maximum principal stress criterion (MPS) In this kind of model a crack propagates when the maximum principal stress ahead of the crack tip reaches

a critical value σ_c at a critical distance X_c from the crack tip. This criterion does not suffer from mesh dependency as long as the mesh is sufficiently refined. Hence, joint fracture toughness can be predicted, but also the crack propagation direction. Such criterion has not been extensively used in adhesive joints but well in metals [91]. However, Martiny et al. [92] showed that for some adhesives, cohesive zone models fail to accurately predict the influence of adhesive thickness on the joint fracture toughness. Indeed, the MPS criterion better captures the high level of constraint and of triaxiality ahead of the crack tip in thin bondlines resulting in a more accurate prediction.

2.3.2 Toughening through blending

The joint fracture toughness can be improved by modifying the adhesive response through the chemical modification of its formulation. Four types of second constituents can be added: rubber, thermoplastic, inorganic filler or organic filler [10].

Toughening by the addition of liquid-rubber or rubber particles is a well known strategy [10, 93]. When liquid-rubber is added to the adhesive, depending on the rubber concentration and the processing conditions, different morphologies, as observed in thermoplastic-epoxy blends, are found: sea-island, co-continuous and phase-inverted morphology. However, the addition of rubber into the adhesive results in a decrease of thermal resistance (lower T_g) and of strength and stiffness. Hence, it is preferred to introduce low amounts of rubber to achieve a sea-island morphology with spherical rubber domains embedded in the adhesive. A similar morphology is achieved when introducing rubber particles. Adhesive toughening results from the cavitation due to the debonding between the rubber particles and the adhesive that results in microvoids shielding the stress field ahead of the crack tip.

An alternative to rubber is the use of a thermoplastic phase that has good interactions with the adhesive material (see subsection 2.2.3 for epoxy adhesives) [10]. The advantage of the addition of thermoplastics such as PEI, PES or PSU is their high T_g , stiffness and strength compared to rubber [78]. Opposite to rubber-modified adhesives, in this case, large amount of thermoplastics are added in order to obtain a co-continuous morphology or a phase-inverted morphology as these morphologies are associated with a large improvement in fracture toughness compared to the sea-island mor-

phology obtained with low amounts of thermoplastic. Depending on the added thermoplastics, different toughening mechanisms take place: crack pinning, micro cracking or plastic deformation of the thermoplastic phase. Thermoplastics can also be inserted in the form of filaments and short fibres such as kevlar short-fibres or PEEK fibres [9].

Inorganic rigid particles such as silica particles can also be added into adhesive but are less effective than rubber to toughen adhesives. However, the stiffness is improved, curing shrinkage is reduced and they have a limited effect on the T_g . Kinloch and Taylor [9] also added mica that proved to be efficient for adhesive toughening with crack deflection being the main toughening mechanism.

As organic fillers, carbon nanotubes and graphene derivatives have been widely incorporated into adhesives. They not only provide a toughening effect but also other functional properties such as improvement in thermal and electrical conductivity, their sensing capability to detect some defects.

2.3.3 Enhanced plastic dissipation

The first strategy to improve the fracture toughness of the joint is by enhancing plastic dissipation. Indeed, the higher the energy dissipated by plastic deformation, the larger the fracture toughness [94]. In bulk materials, the dissipated energy due to plasticity depends on the cohesive strength σ_c and on the plastic response of the material (yield stress σ_0 and strain-hardening behaviour). There is a threshold σ_c/σ_0 ratio above which significant plasticity yielding takes place [94]. For a material exhibiting a strain-hardening-law modelled using a power-law given by

$$\varepsilon = \frac{\sigma_0}{E} \left(\frac{\sigma}{\sigma_0} \right)^{1/n} \quad (2.3)$$

with ε being the strain corresponding to the stress σ , E the Young's modulus and n the strain-hardening exponent. An improvement in G_{Ic} takes place when σ_c/σ_0 is typically above 3 if $n = 0.1$ as shown in Figure 2.12a. For a perfectly plastic material ($n = 0$), this threshold ratio is decreased and if the strain-hardening exponent is increased, this threshold ratio is higher. In addition, there exists a σ_c/σ_0 ratio above which the toughening effect is infinite and the crack tip blunts indefinitely. For hardening exponent equal to 0, 0.1 and 0.2, this upper ratio is about 3, 3.75 and larger than 5, respectively. The larger the σ_c/σ_0 ratio, the longer the crack advance over

which the R-effect takes place. When, an elastoplastic material is bonded to a perfectly elastic material as shown in Figure 2.12b, the fracture toughness of the interface for a given σ_c/σ_0 is enhanced by increasing the elastic mismatch between the two materials with the elastoplastic one being the less rigid [95]. If now the elastoplastic material corresponds to a single layer of thickness t between two elastic solids (Figure 2.12c), the interface fracture toughness depends on the elastic mismatch, on σ_c/σ_0 and the ratio between t and the plastic zone size R_0 , t/R_0 [95, 96].

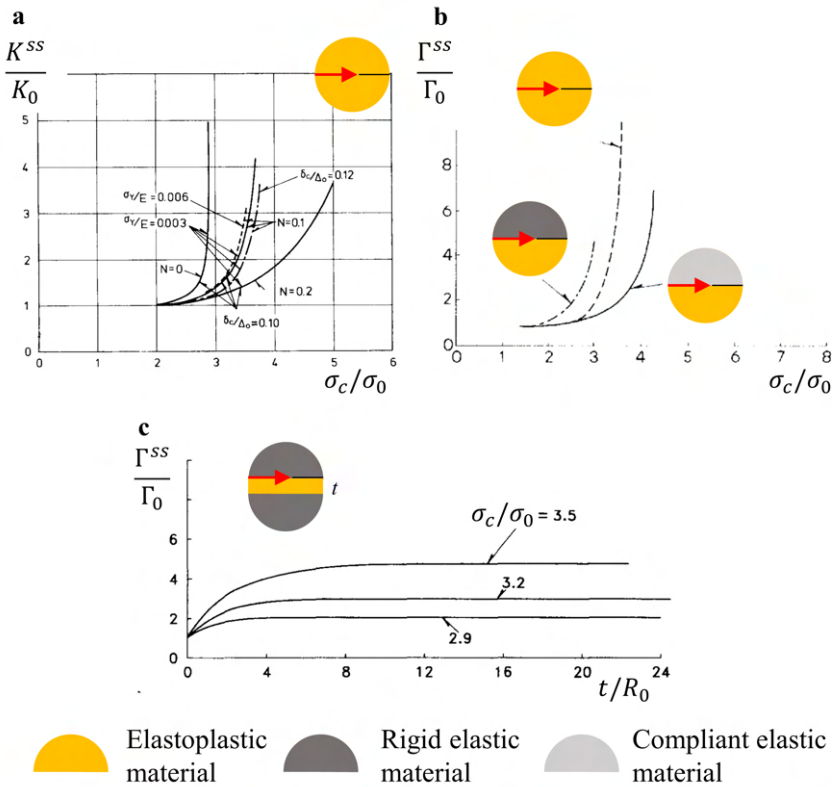


Figure 2.12 Evolution of the steady-state toughness with the cohesive strength ratio σ_c/σ_0 for three situations: a) in an elastoplastic material, b) at the interface between an elastoplastic and an elastic material, c) an elastoplastic layer between two elastic materials. Adapted from [94–96].

In adhesive joints, the thickness t of the adhesive layer also influences the joint fracture toughness as explained here-below. Another option to in-

crease an assembly fracture toughness, is to add a softer layer in the assembly in which plastic dissipation can take place [79, 97]. This strategy will be introduced after the thickness effect in adhesive joints.

Influence of adhesive thickness

The influence of the thickness on the joint toughness is a well known effect in adhesive joints [13, 92, 93, 98–100]. The following lines are very much inspired from [101]. In general, the toughness increases with thickness until reaching a maximum at a thickness t^* corresponding approximately to the plane stress plastic zone size of the bulk adhesive equal to $2G_{Ic}E_a/(\pi\sigma_{0a}^2)$ where E_a and σ_{0a} are the adhesive Young's modulus and yield strength, respectively [13, 99, 102]. With increasing thickness, the degree of constraint in the bondline decreases, and the plastic zone can expand. Not only plastic dissipation explain the changes in fracture toughness. Indeed, for thin adhesive layers, the high degree of constraint leads to a high triaxial stress state ahead of the crack tip [103], which may lead to a change in failure mechanism [93, 98]. With increasing t , the plastic zone can spread over a larger volume and more energy can be dissipated by plastic deformation. For large thickness, two different behaviors are reported. In some adhesive systems, once the maximum G_{Ic} is reached at $t = t^*$, the fracture toughness does not vary anymore with t , corresponding to adhesive A in Figure 2.13a [13, 104]. However, in other systems, after the optimum thickness has been attained, G_{Ic} decreases (adhesive B in Figure 2.13a) [13, 99, 102]. Martiny et al. [13] investigated the different contributions to plastic dissipation. They observed three main contributions: (A) plastic dissipation around the crack tip due to crack-tip stress field, (B) reverse plastic loading due to unloading before the crack tip and (C) plastic dissipation due to shear stresses developing close to the interface with the adherends. The evolution of these contributions are shown in Figure 2.13b. The energy dissipated by contribution A increases with thickness until reaching a plateau. The contribution of phenomenon B is negligible. Regarding the contribution of C, it first increases with thickness upon reaching a peak and decreases then to zero. This leads to the peak followed by a decrease and finally a plateau.

Addition of a dissipative layer

In order to improve the interfacial fracture toughness of some assemblies, it has been suggested by Bertholet et al. [97] and Dépinoy et al. [79] to introduce a soft elastoplastic interlayer. Bertholet et al. [97] pursued the

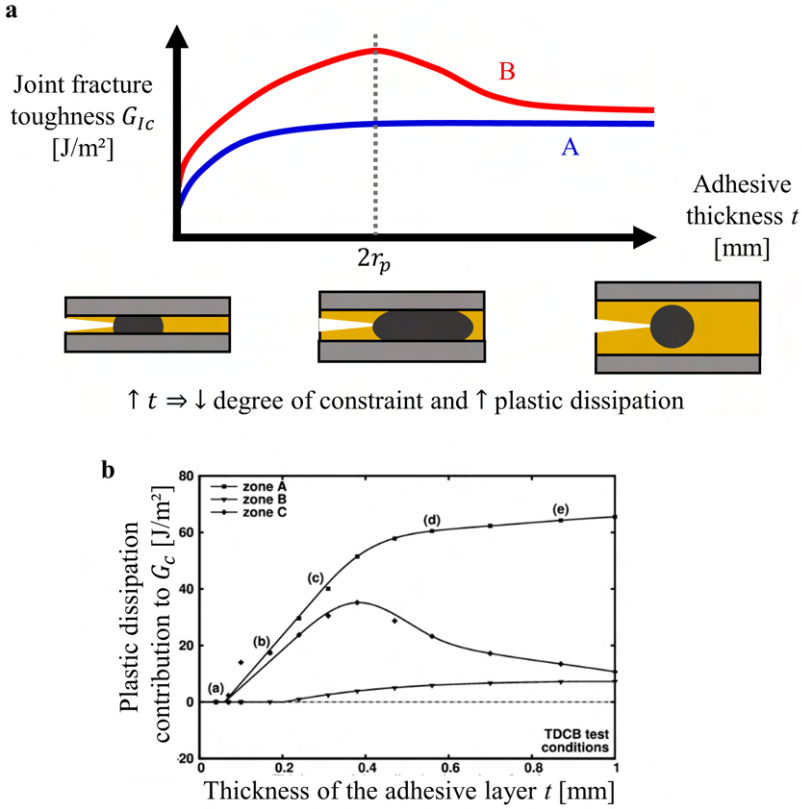


Figure 2.13 a) Influence of the adhesive thickness on the joint fracture toughness with two types of adhesives A and B. b) Evolution of each contribution to the fracture toughness of the joint: crack tip stress-field (A), unloading (B) and shear stresses at interface (C). Images taken from [13, 101].

objective of increasing the toughness of bonded wafers made of a SiO_2 layer deposited on a Silicon substrate. Between the two layers they inserted a ductile alumina layer resulting in a $\text{Si-Al}_2\text{O}_3\text{-SiO}_2/\text{SiO}_2\text{-Al}_2\text{O}_3\text{-Si}$ multilayer. Dépinoy et al. [79] investigated a system similar to the one tested in the thesis of F. Strepenne [105] consisting of an elastoplastic material deposited on a stiff elastic substrate and bonded to the other substrate using an adhesive, softer than the deposited layer. The two configurations are shown in Figure 2.14a-b. These authors investigated the influence of

the thickness of the two layers and their relative properties.

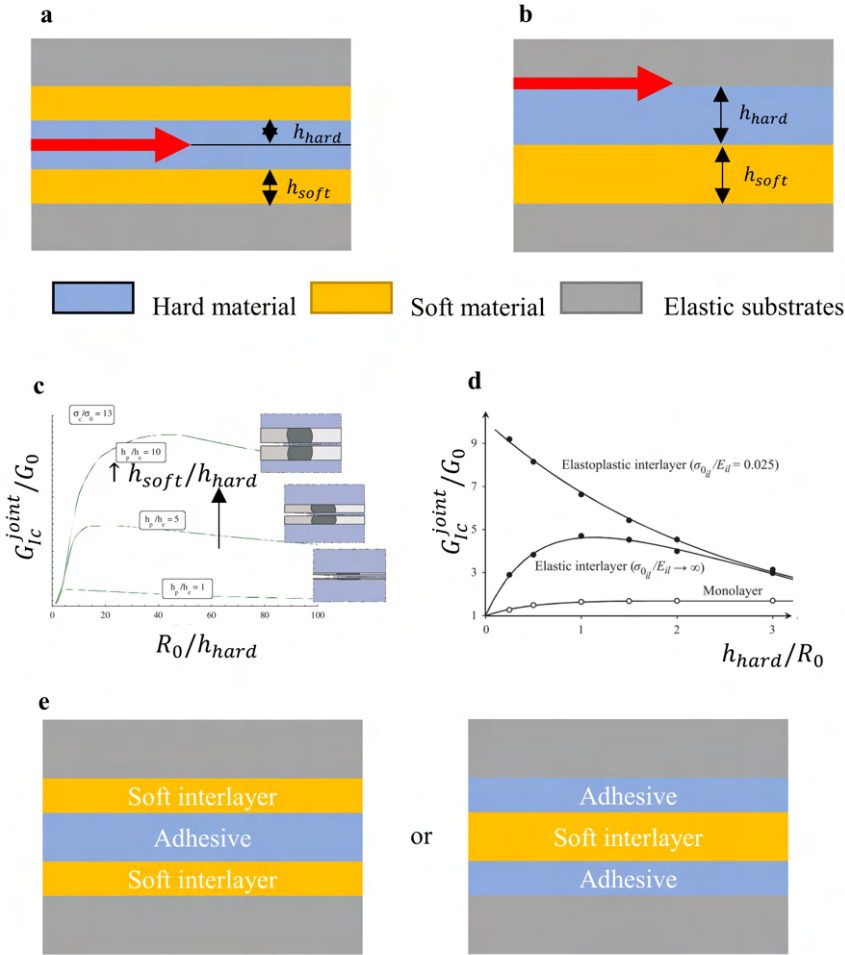


Figure 2.14 Configurations with a dissipative interlayer investigated by a) Bertholet et al. [97] and b) Depinoy et al. [79]. c) Influence of h_{hard} and h_{soft} in Bertholet et al. [97] and d) influence of h_{hard} in Depinoy et al. [79].

The two systems can be generalized to a case of a four-layer material with the two external layers being the stiff elastic adherends and in between these two adherends, there is a hard elastoplastic layer of a thickness h_{hard} and a soft/ductile interlayer of thickness h_{soft} . The crack propagates at the interface between the hard layer and the bonded adherend. Bertholet et

al. reported an optimum thickness h_{hard} below which the plastic dissipation in the hard layer increases with the thickness and above which, the plastic dissipation in the interlayer decreases due to less interactions of the crack tip stress field with the interlayer (Figure 2.14c). Depinoy et al. [79] reported a similar behaviour in the presence of an purely elastic interlayer. However, if the interlayer has an elastoplastic behaviour a decrease in G_{Ic} as h_{hard} increases (Figure 2.14d) is found. In all cases, increasing h_{soft} results in a increase in G_{Ic} .

Depinoy et al. [79] also investigated the mechanism leading to this toughening effect. Depending on the presence or not of the interlayer and on the mechanical response of the interlayer material, plastic dissipation takes place at different places. In the absence of any interlayer, similar zones as those observed by Martiny et al. [13] are reported. If an elastic interlayer is added, plastic dissipation takes place ahead of the crack tip due to crack stress field, at the interface with the interlayer behind and in front of the crack tip and at the interface with the adherend behind the crack tip. If an elastoplastic interlayer is introduced, plastic dissipation takes place in both layers and is the highest at the interface between them. It is important to note that these studies did not consider the fracture toughness of the hard layer and the interlayer. If one of these two layers is weaker than the interface, the crack could be deflected in this layer reducing the toughness of the assembly.

This could be extended to adhesive joints as shown in Figure 2.14e and f by inserting a soft interlayer either close to the adherends or at the middle of the joint [11]. This is investigated in chapter 7.

2.3.4 Crack deflection

Crack deflection is another mechanism that can enhance the fracture toughness of adhesive joints [11]. One considers crack propagation in the middle of an adhesive joint propagating under pure mode I. If the crack encounters an obstacle through which it cannot propagate, it will be deflected. If one consider the geometry in Figure 2.15a, behind the obstacle, the crack propagates in the adhesive under pure mode I while when approaching it is deflected following the boundaries of the obstacles. Although, pure mode I is applied to the joint, locally some mode mixity (mode I/II) appears depending on the slope of the edges of the obstacle as shown in Figure 2.15a. If the obstacle edge is perfectly perpendicular to the crack

propagation path, pure mode II applies. It is well known that the most critical mode is the mode I and that mode mixity generally enhances the apparent fracture toughness when a crack propagates along an interface. This can be applied at the micro and macroscale.

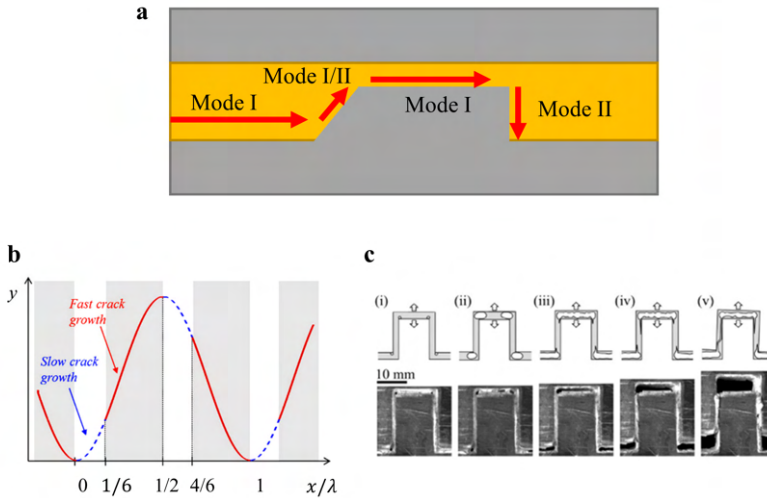


Figure 2.15 a) Schematic picture of the deflection toughening mechanism driven by forced crack deflection to favour mode II, depending on the architecture of the adherends. b) Sinusoidal interface of Cordisco et al. [106] and Zavattieri et al. [107] with the locus of stable and unstable crack propagation. c) Failure mechanism taking place in the square-waved interface of Maloney and Fleck [80, 108] with horizontal interfaces loaded under mode I failing first and then the vertical edges loaded under mode II.

At the macroscale, Zavattieri et al. [107] and Cordisco et al. [106] designed sinusoidal interfaces to benefit from the toughening effect induced by crack deflection. Compared to a flat specimen, the crack surface area is increased with a patterned specimen for a given horizontal crack propagation length. This increase in surface area cannot solely explain the toughening effect which also comes from the local mode-mixity at the crack tip due to crack deflection along the interface. The increase in surface area and the mode-mixity depends on the geometrical parameters: the wavelength amplitude A and the period λ . The larger the A/λ ratio, the higher the toughening effect. In addition, it does not only affect the maximum propagation toughness reached in the specimens, but also the initiation fracture toughness in

a similar way. During crack propagation, stable and unstable crack propagation events take place. These are shown in Figure 2.15b and depend on the relative crack tip position (x) with respect to λ . One considers a crack located in a valley. It will propagate in a stable way until reaching an adimensional distance $x/\lambda = 1/6$ before unstable crack propagation takes place until the crack reaches the following peak ($x/\lambda = 1/2$) as shown in Figure 2.15b. The crack then again propagates in a stable way over a distance equal to $x/\lambda = 1/6$ before being unstable until reaching the following valley. Following a similar idea, Maloney and Fleck [80, 108] designed square-waved adhesive joints. In this case, the adhesive is subjected to pure mode I at the top and bottom interfaces of the square while mode II loading is applied along the vertical edges. By increasing the amplitude of the squares (length of vertical edges), the energy dissipated in DCB and tensile butt joints is enhanced, indicating an improvement of the effective fracture toughness of the joint. As shown in Figure 2.15c, debonding first takes place at the corners and then propagates leading to the failure of the mode I loaded zones. Subsequently, failure is initiated along the edges loaded under mode II until final failure of opposing faces.

Patterning the interface at the microscale has also been investigated. Surface roughness can induce some mechanical interlocking resulting in a toughening effect for interfacial cracks [109–113]. Hence, the performance of adhesive joints with various surface treatments such as grid blasting and sandblasting have been compared. For each adhesive, there exists an optimum surface roughness depending on its mechanical properties and physico-chemical properties such as viscosity. More recently, researchers patterned interfaces using other techniques such as lithography (on the substrates [114, 115] or to manufacture a mold for composite adherends [116–118]). Matsuzaki and co-workers [116–118], Reedy [119], Hamilton et al. [115] all designed square-waved interfaces. Suzuki et al. [116] and Reedy [119] both reported an increase in apparent fracture toughness as the interface area increases. This corresponds for example to deeper channels or to an increase of the aspect ratio. However, Suzuki et al. [116] observed that for channels deeper than 40 μm and for a given aspect ratio, the fracture toughness was not longer affected. As observed by Maloney and Fleck [80] the horizontal edges are subjected to mode I and first fail while the vertices are loaded under mode II, improving the apparent fracture toughness. Kim et al. [114] and Yukimoto et al. [117] also observed an improvement in mode II fracture toughness using patterned adherends

while Hamilton et al. [115] reported an improved lap-shear strength.

2.3.5 Crack tip blunting

Crack tip blunting is known to enhance the apparent fracture toughness. Considering a single edge notch tensile specimen with a crack of radius ρ , the tensile stress acting ahead of the crack tip at a distance x is given by

$$\sigma_{yy} = \sigma^\infty \sqrt{\frac{a}{2x}} \frac{1 + \rho/x}{(1 + \rho/2x)^{3/2}} \quad (2.4)$$

where σ^∞ is the remote tensile stress and a the crack length [120]. As the crack tip radius increases, the stress at a distance x decreases, reducing the crack tip driving force. Consequently, blunting leads to toughening of the material. However, this is only true if the crack tip radius is above a certain critical radius $\rho_c \approx K_{Ic}^2 / E\sigma_0 \approx G_{Ic} / \sigma_0$. Adhesives being brittle, the critical radius is rather limited. Adhesive joints can thus be toughened by introducing an architecture allowing for crack tip blunting. Following this idea, Heide-Jørgensen and Budzik [121, 122] and Maloney and Fleck [123] inserted holes in the adhesive bondline.

Heide-Jørgensen and Budzik [121, 122] investigated the influence of the void length and the spacing between the voids with respect to the process zone length l_p equal to the inverse of the elastic foundation number λ_w (see subsection 6.7.1 and section 8.5). If the spacing between the stop holes is too short, the process zone length expands over several bonded and unbonded regions resulting in a weakening effect as shown in Figure 2.16b. At the opposite, if the bonded region is long enough ($\approx l_p$), the process zone will expand over a distance encompassing maximum one void and an oscillatory behaviour is observed with improved overall fracture toughness compared to the previous case (Figure 2.16a). A bonded length ranging from l_p to $1.5l_p$ seems to be sufficient.

Regarding the length of the voids, two behaviours can be observed. For small holes (Figure 2.16c), as the crack starts to propagate, a slight R-effect is observed. However, when the crack tip is located at a distance equal to l_p from the stop hole, the fracture toughness decreases and unstable crack propagation takes place. The crack is then trapped by the stop hole and must be reinitiated. The shorter the void, the smoother the process (less instability). However, the reinitiation fracture toughness does not exceed the joint fracture toughness in the absence of void. As shown in Figure 2.16d,

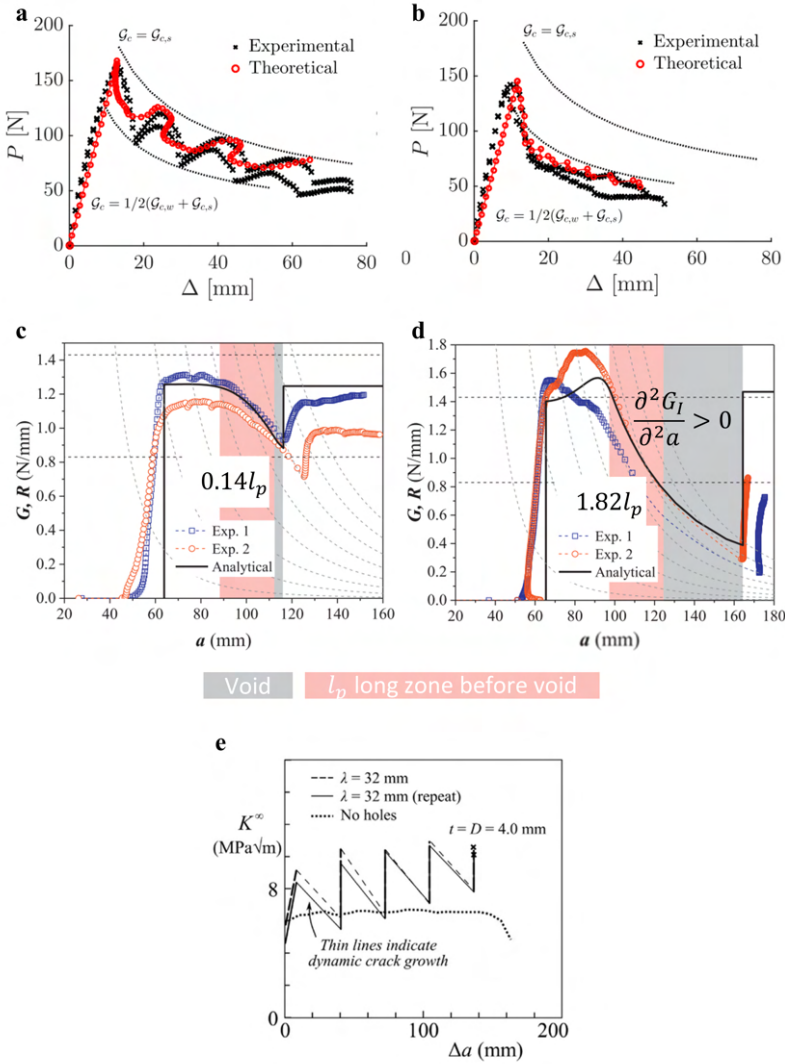


Figure 2.16 Load displacement curves for bondlines containing a) 20 mm long bonded zones followed by 10 mm long voids b) 3 mm long bonded and unbonded zones. R-curves in presence of c) 0.14 l_p long void and d) 1.82 l_p long void. e) R-curve of a 4 mm thick adhesive joint with holes of a diameter of 4 mm and spaced by 32 mm. Images taken from [121, 122] a) to d)) and [123] (e).

for larger void (length $\geq 0.45l_p$), the R-effect is more pronounced as the crack starts to propagate. Then, unstable crack propagation takes place at a distance about l_p from the void as for short voids. However, opposite to short voids, a critical state ($\partial^2 G_I / \partial^2 a > 0$) is attained at a distance ranging between $0.5l_p$ and l_p from the void indicating a snap-down behaviour due to the reduced load bearing capacity of the adhesive. The crack is then trapped by the stop hole and must be reinitiated. In this case, a toughening effect is generated compared to the classical joint. They concluded that a maximum toughening effect is obtained if the void has a length of at least $2l_p$.

Within the same idea, Maloney and Fleck [123] investigated the influence of the hole radius versus the adhesive thickness (r/t), the spacing between two consecutive holes and the influence of the adhesive thickness. They reported no influence of the stop hole radius and the adhesive thickness (1 and 4 mm) and no significant influence of the spacing between the stop holes.

Comparing both studies, the spacings investigated by Maloney and Fleck [123] ranged between 0.5 to $1.6l_p$ for the 4 mm thick bondline and between 0.8 and $2.3l_p$ for the 1 mm thick bondline. This agrees with the recommendations of Heide-Jørgensen and Budzik [121] regarding the bonded length. In addition, a similar failure mechanism is reported with a constant toughness away from the void that decreases when approaching it. Once trapped by the void, crack blunting results in a toughening effect but after reinitiation unstable crack propagation is directly reported until the crack tip reaches the next stop hole while Heide-Jørgensen and Budzik reported a more stable behaviour due to a less pronounced toughening effect, observed by comparing Figure 2.16a to Figure 2.16e. Heide-Jørgensen and Budzik used compliant PMMA adherends while Maloney and Fleck used stiff aluminium adherends. The modulus mismatch between the adherends and adhesive and the bending stiffness of the adherends could be at the origin of this difference, but this has not been investigated yet.

2.3.6 Crack bridging

Crack bridging is a well known toughening mechanism [11]. Upon crack propagation, bridging takes place behind the crack tip. To further propagate the crack, additional energy must be provided to the systems in order to deform and break the bridging material. Hence, the total fracture tough-

ness can be written as

$$G_{Ic}^{joint} = G_{Ic}^a + \Gamma^{Bridging}, \quad (2.5)$$

with G_{Ic}^a being the fracture toughness of the joint in the absence of bridging and $\Gamma^{Bridging}$ the contribution of the deformation of the bridging material. One considers a DCB with one bridging ligament behind the crack tip and at a distance L_b from the crack tip. The force carried by the bridging ligament F_b depends on the beam deflection at the position L_b and thus on the crack length and the applied force. The bending moment applied to the crack tip if $M_{bridg} = -Pa + F_b L_b$. If no bridging ligament is considered, the bending moment is equal to $M_a = -Pa$. The crack driving force is equal to $G_I = M^2 / WEI$. Crack propagation takes place when $G_I = G_{Ic}$, meaning that the bending moment must be equal to a critical value M_c . As for a given load P , $|M_{brid}| < |M_a|$, P leading to failure is higher in the presence of a bridging ligament. The total fracture toughness being given $G_{Ic}^{tot} = P^2 a^2 / WEI$, bridging results in a larger fracture toughness. Several strategies exist to trigger this phenomenon. A summary of the investigated strategies to induce bridging are shown in Figure 2.17.

In composites, crack bridging is triggered to improve the delamination resistance. For this purpose, fibres plies can be stitched together, thickness reinforcements called Z-pins are sometimes inserted or thermoplastic veils can be added at the mid-plane. Stitching leads to ply bridging behind the crack tip allowing for composite toughening. This toughening effect depends on the stitching pattern and the stitching density [125, 126]. Another alternative is to insert Z-pins which are reinforcements inserted through the thickness of the composite [127, 128]. Hoffmann and Scharr [128] investigated the influence of the amount of Z-pins and the shape of the Z-pins on the fracture toughness. By increasing the Z-pin volume content up to 2%, they improved the fracture toughness by a factor 42 for unidirectional laminates and a factor 13 for quasi-isotropic laminates for rectangular Z-pins. If circular pins are inserted, G_{Ic} is only increased by a factor 38 and 7. By changing the geometry, the contact area between the matrix and the Z-pins changes and is the highest with rectangular pins which can partly explain the higher fracture toughness as friction applies over a larger area. Cartié et al. [127] also reported a higher fracture toughness with increasing the frictional area between the matrix and the Z-pins as the main failure mechanisms are Z-pin debonding and pull-out under mode I loading. Un-

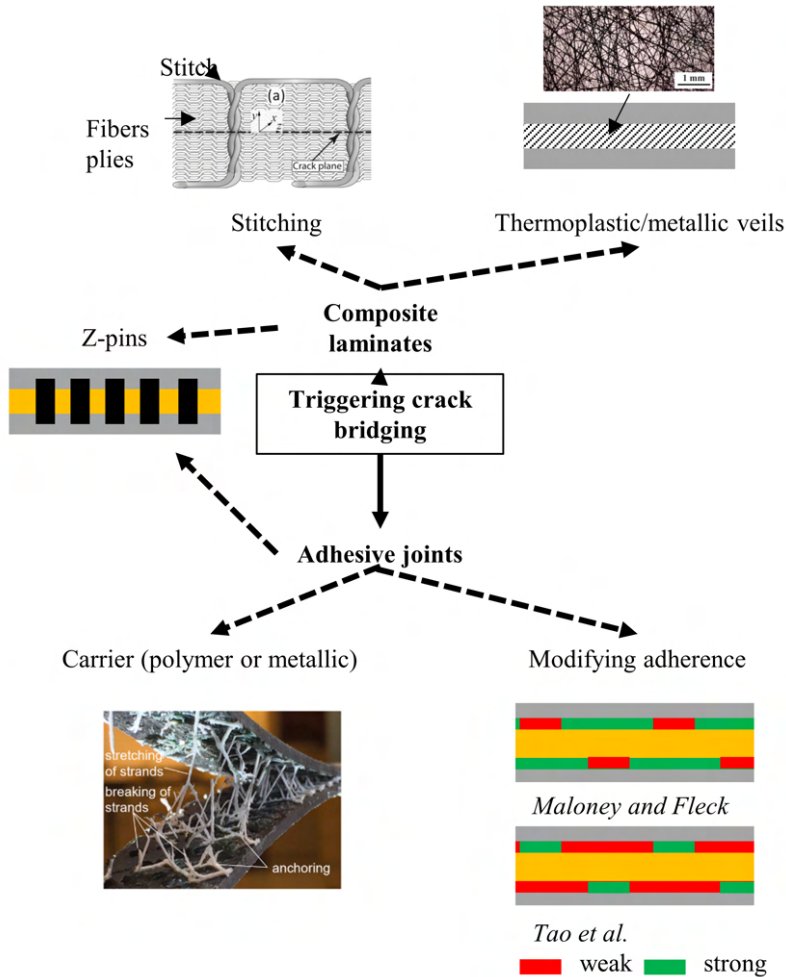


Figure 2.17 Strategies investigated in the literature to trigger bridging in composite laminates (stitching, thermoplastic/metallic veils and Z-pins) and adhesive joints (Z-pins, carriers, changes in adherence). Images taken from [81, 124, 125].

der mode II loading, transverse shear failure of the pins may occur. It is important to note that when the spacing between the Z-pins increases, the bending to shear ratio of the pins increases, modifying the failure modes. Finally, thermoplastic veils are inserted at the mid-plane of composite laminates to trigger crack bridging [124, 129–131]. Depending on the ther-

thermoplastic material, the areal weight and the composite architecture, the toughening may be more or less important. For example, Quan et al. [124] showed that the bridging effect is more important in unidirectional (UD) laminates than in non-crimp fibres and H5-satin laminates. In addition, they reported a higher toughness by adding PET and PPT veils than with PA veils in UD composites while in H5-satin laminates, the thermoplastic material did not influence the results. In addition, by increasing the thermoplastic areal weight, the fracture toughness is increased. The solubility of the thermoplastic material in the composite matrix has also an influence [129, 131]. Metallic fibres can also be added instead of thermoplastic ones [130].

Regarding adhesive joints, Z-pins can also be inserted, but other strategies have also been investigated as shown in Figure 2.17. A commercially used bridging strategy is the addition of a carrier [132]. They are mainly 2D polymer meshes. Following a similar idea, Yudhanto et al. [81], manufactured a 2D mesh but with some waviness through 3D printing. The waviness is introduced by some weft lines printed on both sides of the mesh. They did not observe any strong influence of the spacing between the weft lines. However, an influence of the amount of adhesive present in the bondline has been found. In saturated joints, the bridging strands are rather short and they are difficult to form. In unsaturated joints, bridging strands easily form and are longer, providing additional toughening. Regarding mode II fracture toughness, unsaturated joints are not characterized by a higher fracture toughness than saturated adhesive joints. Maloney and Fleck [123] also inserted a 3D copper mesh that proved to toughen their adhesive joint. Another strategy suggested by Maloney and Fleck [123] and Tao et al. [83, 133–135] is to locally vary the adherence between the adhesive and the adherends. The difference between the two works is shown in Figure 2.17. Maloney and Fleck introduced weak zones using PTFE films while Tao et al. introduced strong zones by controlling the laser blasting surface treatment. In both cases, the succession of strong and weak zones allows for the development of bridging ligaments. In the case of Maloney and Fleck, crack first propagates along one of the two interface. When sufficiently close from the weak zone at the opposite interface, a second crack is initiated at the weak point and propagates while the first one almost stops resulting in a bridging ligament that bridges both interfaces. In the case of Tao et al., the crack propagates along one interface and when it encounters a strong zone, a second crack starts to prop-

agates at the opposite weak interface. This also leads to a bridging ligament. However, the baseline fracture toughness corresponds to the one of the weak zones while in the work of Maloney and Fleck, it corresponds to the one of the strong zones. Tao et al. [83] numerically investigated the influence of the ratio between the cohesive strength and between the toughness of the strong and weak zones on the failure mechanisms. If the strength and toughness ratios are low, no bridging ligament is created. If the strength ratio slightly increases and the strong zones toughness remains limited, several bridging ligaments can be triggered. Finally, for high strength and toughness ratios, a bridging ligament is created but fails before creating the next one. Both groups also investigated the influence of the spacing between the strong and weak zones. Maloney and Fleck reported a higher toughness for short zero-adherence regions and small spacing between these regions. Tao et al. [134] also reported an improved toughness if the distance between the strong zones is decreased. Hence, the low adherence zones must be as short as possible to trigger a maximum number of bridging ligaments. Lubineau and co-workers [136, 137] also triggered crack bridging by introducing sacrificial cracks in adhesive joints

2.3.7 Elastic heterogeneities

Exploiting elastic heterogeneities along the crack path can also lead to some toughening effect. Kendall [138] and Xia et al. [139] investigated the peeling of strips of varying stiffness. Stiffness variation along the crack patch can be achieved either by locally changing the adherend Young's modulus or the adherend thickness. Both works reported an increase in peel force when the crack propagates from a compliant region to a stiffer region. As the crack propagates towards the stiffer material, its velocity decreases and the peeling force starts to increase resulting in a larger G_{Ic} . This arises from the fact that a significant proportion of the external work is transferred to the bending of the stiffer region, decreasing the crack tip driving force. As the crack further propagates, it is progressively accelerated and the crack reaches its initial velocity. If the crack propagates from the stiff to the compliant region, it is accelerated when reaching the interface before being decelerated. It is thus possible to create a peel strip with alternating stiff and compliant regions to create several arrest/accelerating events. According to Kendall, these regions must have a critical length which depends on the bending stiffness of the stiff and compliant regions. According to Xia et

al., the longer the heterogeneity with respect to the characteristic length scale of the bended film, the larger the toughening effect. This is true up to a certain length above which any increase in length does not further toughen the joint. This toughening effect does not completely vanish for small heterogeneities.

Following a similar idea, Luo and Turner [140] designed DCB specimens with varying bending stiffness along the DCB arms. There exists a minimum length for the stiff inserts to maximize the joint fracture toughness and this length increases with the beam thickness. Regarding the spacing between the inserts, there exists a critical spacing above which each stiff insert behaves independently of the other which corresponds to 1.5 times the beam thickness. Below this value, at least two consecutive stiff inserts interact together, further improving the joint stiffness. Following a similar idea, Morano et al. [82] manufactured channels in their adherends to create compliant regions. They investigated the influence of the shape of the channels, their spacing and their dimensions on the joint fracture toughness. Square channels result in a larger fracture toughness than ellipsoidal ones. For the squared channels, the fracture toughness is maximized if their height is equal to 0.65 times the beam thickness, their length to height ratio 1.25 and if the spacing between the channel is about 0.75 to 1 times their length. As observed previously, the toughness is higher when the crack propagates along the stiff regions than along the compliant regions.

2.3.8 Metamaterials as bondline material

The compliance of the bondline affects the stress distribution in the joint. A compliant bondline results in a broader stress distribution and lower stresses at the crack tip. Heide-Jørgensen et al. [141] changed the bondline compliance by designing pillar interfaces as shown in Figure 2.18a. They investigated the influence of the aspect ratio of the pillar (height over length) on the load at failure. The lowest failure load is reported for an aspect ratio of 1. Both, decreasing the aspect ratios below 1 and increasing it above 1 result in a higher failure loads. The bondline compliance follows a similar parabolic trend with first a decrease as the aspect ratio is increased up to 1 and then a slight increase of the compliance is reported if the aspect ratio increases. When the aspect ratio exceeds 1, an increase in aspect ratio results in a better stress redistribution with more pillars being affected and for a given load, the first pillars are subjected to lower stresses delaying failure as shown in Figure 2.18b-c. Pillar interfaces only toughen joints if

they are aligned parallel to the crack direction, but not perpendicular.

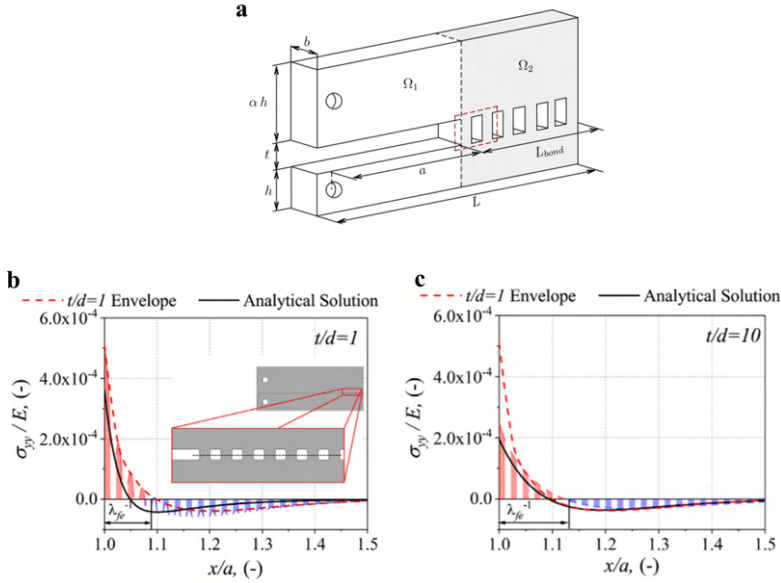


Figure 2.18 Interfaces architected with metamaterials: a) pillar unit cell, b) triangular unit cell (octet truss in 3D) and c) hexagonal unit cell (Kelvin unit cell in 3D). Images taken from [84].

Toughening must be activated in all possible crack propagation directions. Considering this, led to the design of interfaces with metamaterials. Different unit cells have been investigated. Athanasiadis et al. [84] and Hedvard et al. [142] inserted pillar, triangular and hexagonal unit cells in the bondline (Figure 2.19). Inserting metamaterials changes the process zone length depending on the dimensions of the unit cell resulting in a change in the stress distribution ahead of the crack tip. Depending on the unit cell pattern and its dimensions, different failure modes can be observed. The pillar and triangular unit cells can fail under buckling if the truss of the unit cells are too slender (low radius of truss over truss length radius) while for slenderness ratios approximately above 0.01, they fail under tension. The transition between the two failure modes also depends on the bondline thickness. Increasing the bondline thickness results in a larger slenderness ratio for which the transition between the two regimes takes place. In both cases, the load at failure increases as the slenderness ratio increases when

failure is dominated by buckling while it decreases in the tensile failure regime. Regarding the hexagonal unit, only a bending-dominated failure regime is reported with a decreasing load at failure as the slenderness ratio of the trusses increases. The advantage of introducing metamaterials is to cover a larger range of elastic process zones compared with classical adhesive joints by controlling their geometrical dimensions. In addition metamaterials as bondline materials result in lighter joints compared to plain adhesive joints.

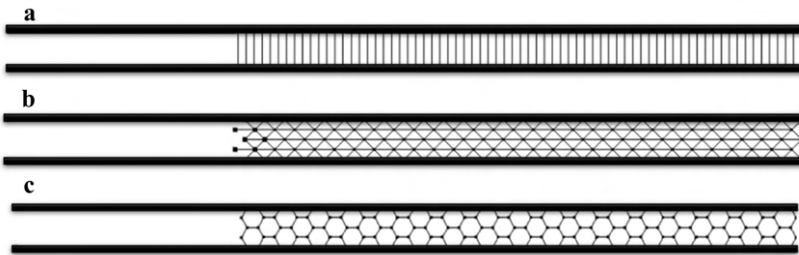


Figure 2.19 Interfaces architected with metamaterials: a) pillar unit cell, b) triangular unit cell (octet truss in 3D) and c) hexagonal unit cell (Kelvin unit cell in 3D). Images taken from [84].

2.3.9 Improving other joint mechanical properties

Fracture toughness is not the only important properties of adhesive joints. Indeed, adhesive joints are mainly loaded under shear in applications. Hence, the shear strength and stiffness are also important, but as the fracture toughness, they are influenced by the adhesive thickness. However, opposite to toughness, the shear strength and stiffness tends to decrease with the adhesive thickness [143–147]. The shear strength is an effective shear strength denoted τ^{f*} which depends on adhesive properties, geometrical parameters and adherend properties.

The most investigated configuration is the lap-shear joint in which an adhesive bonds two adherends over a certain length (overlap length). Analytical models predict an increase of the lap-shear strength with the adhesive thickness [148]. However, experimentally, a decrease is observed. Some argue that this arises from the fact that increasing t statistically leads to a larger number of larger microdefects (e.g. entrapped air) in the bondline [149]. In addition to increasing the number of defects, t has an influence

on the stress state in the bondline and stresses tend to localize close to the joint edges [143, 145, 150, 151] promoting adhesive instead of cohesive failure. This strength reduction is dependent upon the type of adhesive. Lap-shear tests have shown that this effect is less marked for ductile adhesives compared to brittle ones [147]. The stress distribution in lap-shear joint is shown in Figure 2.20 for a stiff/brittle and a compliant/ductile adhesive. Stress concentrations are more important for the stiff/brittle adhesive than for the compliant/ductile adhesive. The main issue is thus stress concentration at joint edges due to geometrical changes and material mismatch. Load eccentricity is also responsible for stress concentration by developing peel stresses at the edges. The decrease of strength with t is not only observed in lap-shear joint, but also in thick adherend shear tests (TAST) [151] or arcan tests [144]. Several strategies have been investigated to improve the shear strength of joint and are summarized in Figure 2.21 and explained here-below.

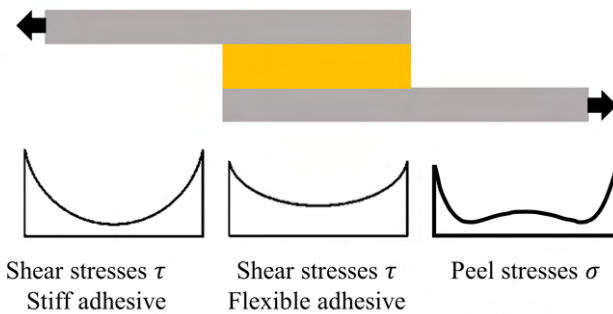


Figure 2.20 Lap-shear configuration with schematic representation of the shear stress distribution along the overlap for stiff and flexible adhesives and the general peel stress distribution along the overlap. Image inspired by [152].

The first strategy is reducing stress concentrations at joint edges [152]. It has been suggested to introduce spew fillets [153]. Adhesive fillets correspond to additional adhesive at the edges to reduce the geometrical discontinuity. The stresses act then over a larger region and are smoothed resulting in a lower peak stress at the ends of the overlap. The two most important parameters are the shape of the fillet and the adhesive properties. Rounded fillets may provide additional strength compared to tapered fillets. Stress concentrations can also be controlled by varying the shape of

the adherend corners [153–155]. Instead of having a sharp 90° corner, the corner is rounded. The important geometrical parameter in this case is the corner radius. Both strategies have a more beneficial effect on the strength of joints made of brittle adhesives than with ductile adhesives which are less sensitive to stress concentrations. A third strategy is to progressively decrease the adherend stiffness when approaching the joint edges by tapering the adherends [156]. Outside and inside tapers can be designed. The design of wavy-lap joints [157] has also been investigated to develop compressive stresses at the edges instead of strongly detrimental peel stresses. However, manufacturing wavy adherends is complex preventing further development of this strategy. As alternatives to tapered joints, scarf joints [152] have been designed. In this case, the tapered edges of the two joints are bonded to each other. The adherends being aligned, the load eccentricity is eliminated preventing the development of peel stresses at the edges of the overlap. The most important parameter is the scarf angle as it controls the overlap length and the amount of peel and shear stresses in the joint. Again, the manufacturing complexity limits their use. A last strategy is to design stepped joints which operate in a similar way as the scarf joints.

The second strategy category is the introduction of a gradient of properties in the bondline. The first strategy is to design bi-adhesive joints [152, 158]. The initial idea is to get advantage of the strength of stiff/brittle adhesives and of the development of lower stresses at the edges for ductile adhesives. Bi-adhesive joints are thus made of a ductile adhesive at their edges and a stiff/brittle adhesive at their centre. Such joints offer improved thermal resistance in addition to improved shear strength. However, their manufacturing is complex and results in many defects at the junctions between the two adhesives. Some strategies have been suggested to prevent the use of several adhesives for example, by introducing stiff particle at the centre or soft particles at the edges of the joint [159], by progressively changing the amount of particles added in the adhesive [160] or by changing the curing parameters by induction heating [161].

The last strategy is similar to the second one, but instead of grading the adhesive, the properties of the adherends are progressively changed [152]. The idea is to vary the adherend stiffness close to the edges to minimize the mismatch with the adhesive. For this purpose, a softer zone can be added close to the overlap edges. For composite adherends, this can be achieved by adding a polymer layer. Varying the stiffness can also be

achieved through additive manufacturing. This category also involves adherend tapering.

This is an exhaustive list of the strategies investigated in the literature. Other strategies have also been investigated such as the introduction of Z-pins [162] and have been proven to improve the joint shear strength in addition to the fracture toughness. In addition, adhesive joint can also be loaded under tension. In butt tensile joints, stress also tends to concentrate close to the edges at the interface with the adherends. The tensile strength improvement did not gain as much interest as the shear strength improvement. However, Maloney and Fleck [80] showed that their square-waved interfaces (see subsection 2.3.4) result in the one hand in toughness improvement and on the other other in a tensile strength improvement. Interface patterning is also known to improve the lap shear strength if the adherends interlock each other [115].

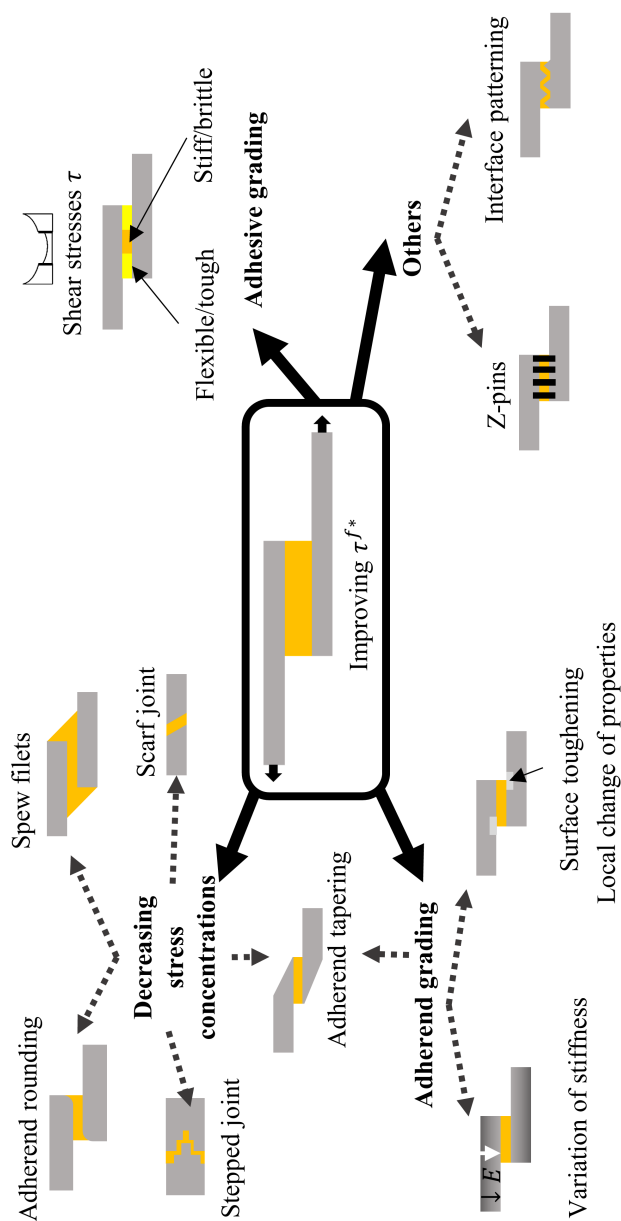


Figure 2.21 Summary of the strategies to improve the lap-shear strength of adhesive joints τ^{f*} .

PART I

Integrated manufacturing: thermoplastics for co-cured joints

PEI-epoxy interphases and co-cured composite joints

3.1 Introduction



This introduction covers literature results concerning PEI-epoxy blends, interphases and co-cured composites joints using a PEI film, as already presented in section 2.2 with more details.

Carbon fibre reinforced polymer composites (CFRPs) are broadly used in advanced structures such as in airplanes owing to their high strength and stiffness to weight ratios. Manufacturing these structures often requires assembling CFRPs together or with other materials. Primary load carrying structures are frequently joined through mechanical fasteners, altering the composites structural integrity [1]. Adhesive bonding offers a more uniform stress distribution and additional weight saving. However, limited resistance to crack propagation and long sequential bonding processes constitute two barriers for a more intensive use of this technology [1]. Regarding the manufacturing, composite bonding requires the curing of com-

posite parts prior to bonding with, possibly, some surface treatment steps. Regarding the toughness, even if adhesives used for high performance applications such as in aerospace have been intensively toughened by adding rubber particles, a thermoplastic (TP) phase, and/or short carbon fibres [9, 163] their fracture resistance remains a point of concern.

To tackle these two issues, it has been suggested to use other materials than epoxies such as adhesives, typically high-performance thermoplastics such as polyetherimide (PEI), polysulfone (PSU), polyethersulfone (PES), etc [15, 29–33]. These thermoplastics are inserted in the form of a film either in the middle of the composite lay-up [15, 29–33] or at the surface [35]. If it is added at the surface, the composite part can be joined to another composite part or to a metallic part via thermoplastic welding. Integrated manufacturing, also referred to as co-curing and co-bonding, can be achieved by inserting the TP film at the midplane of the lay-up prior to composite curing. The use of thermoplastics as bonding material can thus improve the fracture toughness of joints and reduce work- and time-intensity.

However, the integration of TP requires to create a tough interface between the composite and the thermoplastic film [5]. For this reason, thermoplastics exhibiting good interactions with epoxy have been used such as PEI, PES, PSU or polyetheretherketone (PEEK). These polymers can create a semi-interpenetrating network (semi-IPN) when in contact with the epoxy, leading to the development of a morphological gradient, and some functional groups can even react with the epoxide groups as found with polyamide. The formation of a PEI-epoxy interphase has been extensively investigated over the last years [44, 51, 53]. Hence, it is well-known that the curing cycle, the molecular weight of PEI or the resin viscosity can influence the development of the morphological gradient at the interphase. Recently, PEI has also been used as a surface layer material in epoxy CFRPs for ultrasonic welding [35], as a substrate material for the design of sensors for composites [164, 165] and also as an interlayer for co-curing [15, 29–32]. The addition of PEI results in an increase in fracture toughness compared to the composite in the absence of any TP film, and even compared to composites bonded with the FM3000 adhesive in some cases. However, the failure mechanisms taking place when a crack propagates in the interphase between PEI and epoxy has not been deeply investigated. The mechanics of PEI-epoxy blends has been more extensively explored.

An enhancement of the tensile strength, ductility and fracture toughness (G_{Ic}) is often reported as the PEI content increases. This is associated with a change of morphology and failure mechanism. Blends containing a low amount of PEI (typically <15 wt%) exhibit a sea-island morphology. The failure mechanism is governed by the brittle epoxy matrix and decohesion between the continuous epoxy phase and PEI domains takes place. Blends containing a larger amount of PEI (>20 wt%) undergo a phase inversion, with epoxy domains inside the PEI matrix. Failure occurs by yielding of the PEI and decohesion between the PEI and epoxy domains resulting in a significant improvement of the fracture toughness. A co-continuous morphology develops for intermediate concentrations of PEI. Decohesion of PEI and epoxy domains is often reported due to a difference in polarity and due to the lack of reacting functional groups [47]. PEI-epoxy blends have also been used as matrix for composites [166]. In that case, it has been found that the fracture toughness increases as the PEI content increases, but not linearly. If the PEI content exceeds 15 wt%, the increase in G_{Ic} of the composite is not proportional to the increase in G_{Ic} of the matrix. This arises from the fact that due to phase inversion, the fibres are in contact with the PEI phase instead of the epoxy phase. Indeed, the interfacial shear strength decreases when exceeding 15 wt% of PEI, indicating a weakening of the interface with the fibres when phase inversion takes place.

Regarding the epoxy-PEI interfaces, Kim and Kim [44], Vadugappatty Srinivasan and Vassilopoulos [54] and Farooq et al. [49] looked at crack propagation perpendicular to the interface. Kim and Kim [44] initiated the crack in a PEI-rich zone (nodular morphology) resulting in a high fracture toughness. Vadugappatty Srinivasan and Vassilopoulos [54] initiated the crack in the epoxy resin and observed that, when approaching the PEI film, the crack is deflected along the PEI film and does not propagate through it, significantly improving the fracture toughness. Farooq et al. [49] investigated the failure mechanisms of PEI-epoxy stacks cured following several curing cycles and with PEI films of different thickness. For low curing temperatures, short cracks were deflected along the interphase before propagating through the PEI film, while for high curing temperatures and/or long cracks, no deflection occurred, only propagation through the stacking. If the remaining PEI layer is thicker than the PEI plastic zone upon curing, macro toughening is reported. If it is thinner, only micro toughening is generated due to the presence of large epoxy particles in the PEI-rich part of the morphological gradient. Depending on the investigated system, these

authors measured fracture toughness values even larger than the one of PEI. Opposite to these studies, Voleppe et al. [12] propagated cracks parallel to the interface and reported a toughening effect associated with a crack trapping mechanism taking place in the part of the morphological gradient characterized by a nodular morphology. PEI-epoxy interphases are also observed in co-cured composites joints and lead to high fracture toughness competitive with classical aeronautic adhesives [31, 32]. In this case, cracks are forced to run parallel to the PEI film and fibres lie in the epoxy-rich zone of the morphological gradient and debond during delamination. Considerable plastic deformation, void growth and particle debonding take place when the crack propagates and even some cohesive failure in the PEI film. When performing lap-shear tests, failure occurs at the interface with the carbon fibres [30]. PEI particles are found on the fibres and act as mechanical lockers, ensuring a high shear strength. Opposite, Kim and Kim [166] reported a decrease of the interfacial shear strength between carbon fibres and epoxy matrix containing 15 wt% of PEI and more, preventing the full development of the toughening effect of PEI.

The goal of the present work is to further understand and mobilize this crack trapping mechanism, as observed by Voleppe et al. [12], and to unravel the failure mechanism taking place in PEI-epoxy interfaces. Indeed, the first experiments conducted by Voleppe et al. [12] suggested the existence of a soft region (lower yield strength) corresponding to the co-continuous part of the morphological gradient, which is opposite to common literature results as increasing the PEI content results in a larger strength [12, 44, 49, 54]. In order to investigate the failure mechanism, blends and interface samples are manufactured. The properties of the blends are determined through compression, nanoindentation and single edge notch bend (SENB) tests and nanoindentation tests are also performed along the PEI-epoxy interphase. For the samples containing an interface, small SENB samples are manufactured to perform tests under a microscope to follow the propagation of the crack along the interface and perpendicular to the interface. Finally, co-cured composite joints with PEI films are manufactured to investigate the failure mechanisms taking place and the fracture toughness of these joints to determine whether the crack trapping plays a role in PEI-composite joints or not.

3.2 Materials and Methods

3.2.1 Materials

The epoxy resin considered in this work is the aeronautic grade epoxy resin HexFlow RTM6 [167], provided by Hexcel. This highly-crosslinked epoxy resin is characterized by a glass transition temperature around 220°C upon curing. The second material is the polyetherimide (PEI) grade Ultem 1000 from Sabic Innovative Plastics, a high-performance thermoplastic. It is characterized by a molar mass about 50 000 g/mol and a glass transition temperature of 220°C. In order to manufacture homogeneous blends, PEI pellets are grinded using a pulverisette 14 machine from Fritsch. For the composite joints and the samples with an interphase, PEI films with thickness values equal to 50, 175 and 500 µm are used. For the composite samples, HexForce G0956 carbon fibres plies from Hexcel with an H5-satin architecture.

3.2.2 Sample processing

Three different types of samples are processed: (1) homogeneous PEI-epoxy blends, (2) samples exhibiting a PEI-epoxy interphase and (3) PEI co-cured composite joints. The processing method varies depending on the type of sample.

PEI-epoxy blends

Homogeneous PEI-epoxy blends containing 0, 5, 10, 15, 20 and 25 wt% of PEI are manufactured as follows. First, the PEI powder is dissolved into dichloromethane at room temperature. The RTM6 resin is added to the mixture which is stirred until obtaining an optically transparent solution. Then, the solvent is evaporated at 90°C during 30 minutes before pouring the mixture into the moulds. Glass tubes with a diameter of 13 mm are used to process the compression and nanoindentation samples and, rectangular aluminium moulds are used to manufacture the SENB samples. Once in the moulds, the mixture is degassed at 90°C for 30 minutes. The PEI-epoxy blends are then cured following the C180 curing cycle, shown in Figure 3.1, as the C160 led to the burning of the samples. The temperature increases up to 180°C at a rate of 0.5 mm/min and is kept constant for two hours before cooling down the samples at a rate of 5 mm/min. In practice, after removing the solvent, the blends containing 20 and 25 wt% of PEI

3 | PEI-epoxy interphases and co-cured composite joints

were too viscous to be poured into the moulds. Hence, only small samples could be produced and only nanoindentation tests were performed for these compositions. The compression samples are 12 mm high cylinders with a radius of 6 mm. The SENB samples are 52.8 mm long, 12 thick and 6 mm wide. The pre-crack length is about 6 mm.

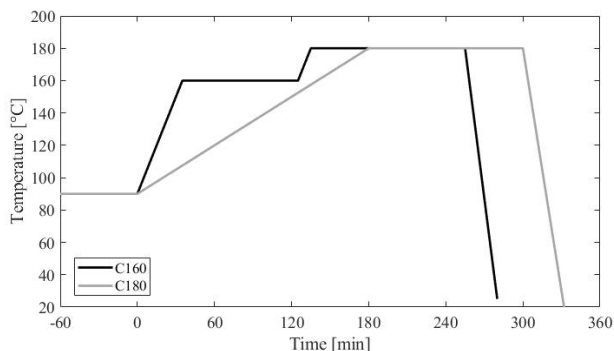


Figure 3.1 Curing cycles: the C160 cycle (black) is used to cure the samples containing a PEI film while the C180 cycle (gray) is used to cure the PEI-epoxy blends.

Samples with a PEI-epoxy interphase

The specimens with a PEI-epoxy interphase are processed using two-part aluminum moulds following the process of Voleppe et al. [12]. A 175 μm film and a 30 μm Kapton (Polyimide, PI) film from Dupont are inserted between the two parts. The Kapton film is inserted to create a pre-crack due to the lack of adherence between PI and PEI. The mould is then filled with RTM6 resin before degassing at 90°C for one hour. Curing takes place following the C160 curing cycle shown in Figure 3.1. The temperature is first increased at a rate of 2 mm/min up to 160°C, and maintained for 90 minutes in order to promote the swelling of the PEI film by the resin and the formation of the morphological gradient. The temperature is then increased to 180°C at a rate of 2 mm/min, and kept constant for two hours. The samples are finally cooled down at a rate of 5 mm/min. SENB samples are manufactured from these plates and the width W , the height H and the length L are set to 3, 6 and 30 mm. The ratio between the initial crack length and the width is set to 0.5 and to 0.7 for three samples. Two larger samples with dimensions $H = 14$ mm, $W = 7$ mm and $L = 61.6$ mm are also manufactured in order to compare the fracture surfaces at the small scale

and at the large scale. A pre-crack is introduced using a Kapton film with $a/W = 0.5$.

PEI co-cured composite joints

The PEI co-cured composite joints are manufactured through resin transfer moulding (RTM) as follows and shown in Figure 3.2. The first five G0926 carbon plies are laid down in the RTM mould. The PEI film is then inserted and two 70 mm long, 30 μm thick Kapton films (Dupont) are stacked on top of it to introduce a pre-crack. The last five plies are then deposited. The RTM mould is closed using a 8 bar press. The RTM6 resin is heated in a piston up to 90°C and injected at the same temperature into the mould until reaching a pressure of 6 bars. The composite is then cured following the C160 curing cycle shown in Figure 3.1.

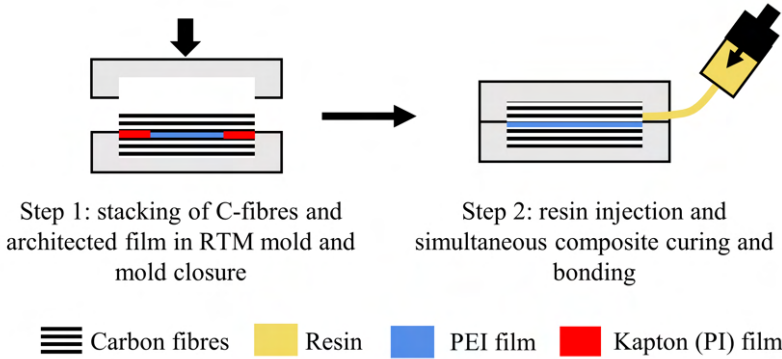


Figure 3.2 Manufacturing steps for PEI co-cured composite joints involving stacking of the plies and of the film and resin injection and curing.

3.2.3 Mechanical testing

Compression, nanoindentation and fracture toughness tests are performed in order to determine the mechanical properties of the homogeneous PEI-epoxy blends. Nanoindentation is also performed along the PEI-epoxy interphase to determine the local properties. In order to understand the failure mechanism of this interphase, SENB tests are carried out under optical microscope. Finally, the mode I fracture toughness of the PEI co-cured composite joints is determined through double cantilever beam (DCB) tests.

Compression

The uniaxial compression tests follow the ASTM D695 standard and the modifications made by Morelle et al. [168] in order to investigate the elastoplastic behaviour of the blends as epoxy then to be brittle under tensile loading. They are performed for homogeneous blends containing 0 to 15 wt% of PEI and for pure PEI samples. The tests are performed on cylindrical samples with a diameter and a height equal to 12 mm (Figure 3.3a) using a Zwick Z250 Roell screw driven machine equipped with a 250 kN cell force at a rate of 1 mm/min. A subpress supplied by Wyoming Instrument is added to the machine. In order to minimize friction and avoid barrelling, 60 μm Teflon films are inserted between the subpress platens and the samples. The tests are performed in a ventilated room at a temperature of 23°C and a relative humidity content of about 50%. In addition to the compression tests, a test is conducted on the subpress alone in order to consider the compliance of the mounting stage.

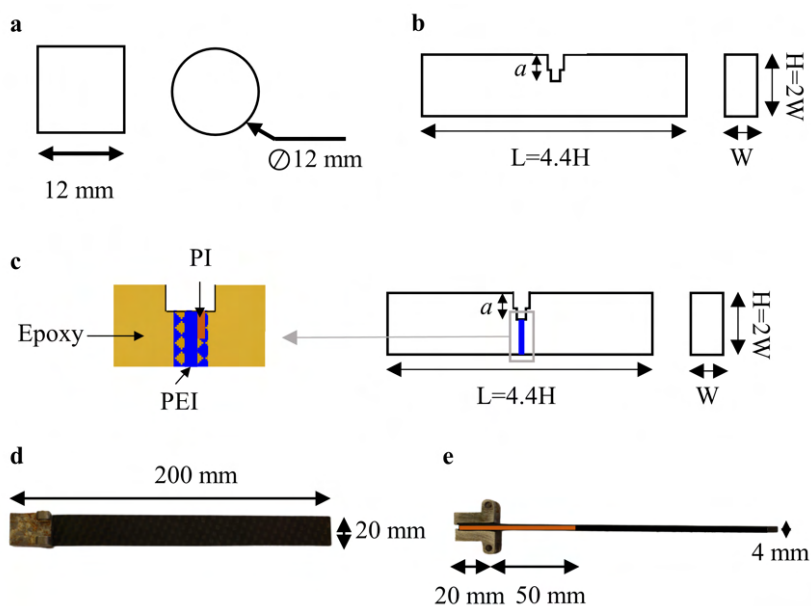


Figure 3.3 Schematic of a) compression and b-c) SENB test samples for b) blends and c) with an interphase. d-e) DCB samples from d) top view and e) lateral view.

Mode I fracture toughness of the blends

In order to determine the mode I fracture toughness of the homogeneous PEI-epoxy blends containing 5, 10 and 15 wt% of PEI, SENB specimens are tested following the ASTM D5045 standard [169]. The tests are performed on a Zwick Z50 Roell screw driven machine equipped with a 50 kN cell. A 3-point bending test bench is added with pins having a radius of 2.5 mm. The bottom pins are spaced by a distance of 48 mm. The SENB samples are 6 mm thick (W), 12 mm (H) high and 53 mm long (L) as shown in Figure 3.3b. Prior to testing, a 5.4 mm long notch is machined and a sharp crack is inserted using a dropping tower equipped with a razor blade. The tests are then performed at a rate of 1 mm/min.

Nanoindentation

Nanoindentation tests are performed on homogeneous blends containing 0 to 25 wt% of PEI, on pure PEI and along PEI-RTM6 interphases, using a G200 nanoindenter from Agilent Technologies with a pyramidal Berkovich diamond tip. The samples are polished prior to testing. 1200 and 4000 MD-Piano disks from Struers are used before finishing with a 0.3 μm alumina suspension (AP-D Suspension 0.3 μm , Struers Inc.). The Continuous Stiffness Measurement (CSM) method is applied and the Oliver and Pharr [170] method is used to extract the elastic modulus and hardness. Drift corrections are not considered as, for polymers, this correction is strongly affected by viscoelastic effects [171]. On homogeneous blends, 16 indents are performed on each sample. Regarding the samples exhibiting a PEI-epoxy interphase, indents are performed along the interphase and the measurement is repeated over 10 zones for each sample. In both cases, different penetration depths equal to 0.5, 1 and 2 μm are investigated.

SENB on samples with interphase

The SENB specimens shown in Figure 3.3b are mounted on a microtest 3-point bending stage equipped with a 2 kN cell (Deben UK Ltd., UK) and placed on the visualization stage of the VHX numerical optical microscope from Keyence. The tests are performed at a speed of 0.02 mm/s. Optical micrographs are recorded before, during (when possible) and after the test to observe the crack propagation path along the morphological gradient. Prior to testing the samples are polished following the same protocol as for nanoindentation.

DCB tests on co-cured composite joints

The DCB samples, shown in Figure 3.3c-d, are machined from the cured panels with a length of 200 mm and a width of 20 mm. The 70 mm long Kapton film inserted to create the pre-crack results in initial crack length of 50 mm. The total height of the samples is about 4 mm. The DCB tests are performed following the ISO 25217 standard [172] on a Zwick/Roell machine equipped with a 50 kN loadcell, at a constant cross-head speed of 1 mm/min. The machine is located in a ventilated room at constant temperature and relative humidity values close to 23°C and 50%, respectively. Prior to testing, the edges of the samples are graduated to measure the crack length. The evolution of the crack is monitored by taking a picture every 3 seconds with a Canon EOS1400D camera. G_{Ic} is then computed following the single beam theory [172].

3.2.4 Microscopy

The SENB tests on the samples exhibiting a PEI-epoxy interphase are performed under a VHX numerical optical microscope from Keyence. In addition, an Ultra 55 Field-Emission SEM (Zeiss, Germany) is used to characterize the fracture surfaces. The samples are coated with a 1 μ m thick gold layer and imaged at 5 keV. In order to determine the morphology associated to the PEI content of the homogeneous blends, fracture surfaces from compression samples are used, except for blends containing 20 and 25 wt% of PEI. For these two cases, fracture surfaces are produced by breaking the small processed samples with a hammer. The fracture surfaces of the SENB tests obtained from the blends and from the samples with an interphase are also characterized to understand the failure mechanisms taking place in these samples.

3.3 Results for epoxy-PEI blends

3.3.1 Influence of PEI concentration on blend morphologies

Depending on the PEI content, the morphology of the blend is modified. These morphologies, shown in Figure 3.4, have been identified by SEM characterization for the blends containing 5 to 25 wt% of PEI. Two different morphologies are identified. First, the blends containing 5 wt% and 10 wt% of PEI display a sea-island morphology (Figure 3.4a-b) with PEI nodules (white spheres) dispersed in the epoxy matrix. This morphology

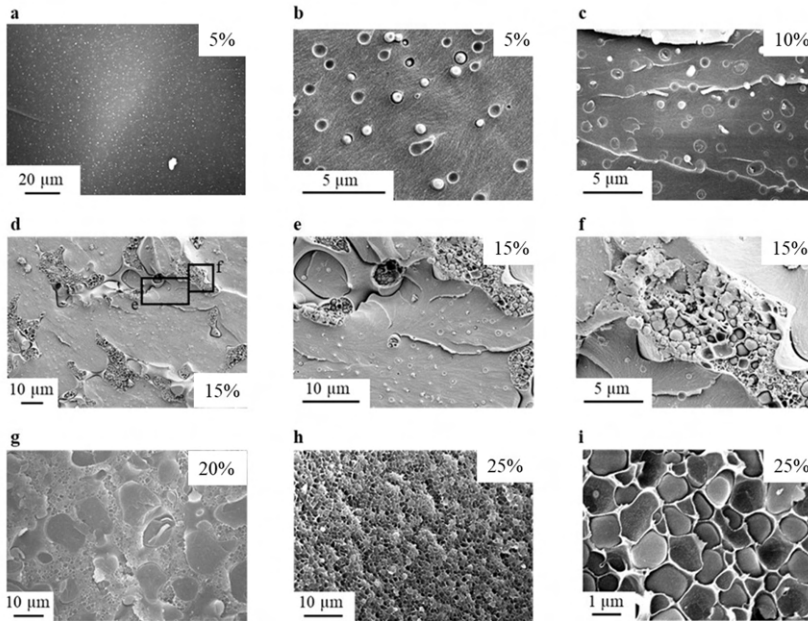


Figure 3.4 SEM micrographs of PEI-epoxy blends containing: a-b) 5wt% of PEI; c) 10 wt% of PEI; d-f) 15 wt% of PEI; g) 20 wt% of PEI and h-i) 25 wt% of PEI.

is formed via a nucleation and growth mechanism [44, 78]. Increasing the PEI content leads to an increase of the number of PEI domains dispersed in the epoxy matrix. Secondly, for the blend containing 25 wt% of PEI (Figure 3.4h-i), phase inversion takes place with nodular morphology formed via spinodal decomposition [44]. In this case, epoxy-rich domains are embedded in a PEI matrix. The blends containing 15 wt% (Figure 3.4d-f) and 20 wt% (Figure 3.4g) of PEI are made of a co-continuous morphology with some zones exhibiting a sea-island morphology (Figure 3.4f) and others exhibiting a nodular morphology (Figure 3.4e). The nodular morphology tends to be more present when increasing the PEI content from 15 to 20 wt%. In the literature, the same morphologies are often reported for similar PEI contents [44–48, 173]. It is important to note that the morphology of thermoplastic epoxy blends can be affected by the molecular weight of the thermoplastic [48], the presence of reactive end groups, the precure temperature or the curing agent [174]. In addition, the SEM micrographs shown in Figure 3.4 reveal decohesion between the PEI-rich and epoxy-

rich domains. Indeed, in Figure 3.4b, unbroken PEI domains can be observed or some holes corresponding to the imprint left by the decohesion between the epoxy and the PEI. Figure 3.4c shows that some PEI particles are broken indicating deformation of the PEI domains during facture and bridging of the crack as often reported [49]. Figure 3.4i indicates that the epoxy nodules are intact while the PEI matrix has been torn when breaking the samples. The epoxy domains have a diameter lower than $3\text{ }\mu\text{m}$, which is the characteristic length for PEI particles under which decohesion takes place instead of epoxy particles fracture, according to Farooq et al. [49]. A poor adhesion between the epoxy and the PEI is often reported as a consequence of the lack of reactive groups on the PEI chains and due to their difference in polarity [47].

3.3.2 Mechanical properties of PEI-epoxy blends

The elastoplastic response of PEI blends containing 5 to 15 wt% of PEI is determined by uniaxial compression tests, as shown in Figure 3.5a.

The Young's modulus decreases with increasing PEI contents from 0 to 15 wt%, even though PEI is stiffer than the RTM6 epoxy resin. According to Morelle et al. [168] a batch-to-batch variability can lead to variations of the Young's modulus of the RTM6 up to 10%. Hence, the variations of the Young's modulus values obtained for the blends are not significant. In the literature, the Young's modulus is reported to not be significantly influenced by the PEI content [44, 45, 47, 48].

Compression curves all exhibit a local maximum corresponding to the upper yield stress, followed by a local minimum referred to as the lower yield stress σ_y^l , indicating a true softening response as observed in a majority of polymer systems. The influence of the PEI content on these two properties is illustrated in Figure 3.5c. The upper yield stress σ_y^u tends to slightly increase from 127 MPa to 131 MPa when the PEI content is increased up to 15 wt%. The more significant increase is between the pure RTM6 sample and the blend containing 5 wt% of PEI. The PEI is characterized by a yield stress equal to about 150 MPa. Regarding the lower yield stress, it is almost not influenced by the PEI content and is about 110 MPa for all the blends. In the literature, tensile tests rather than compression tests have been performed on such blends with failure occurring before significant plasticity development, hence not enabling the determination of these quantities. For blends, the tensile strength increases by adding only 5 wt%

of PEI while further increasing the PEI content does not significantly lead to an increase of the tensile strength [45, 47] and can even lead to its decrease [12, 44, 48, 173]. This drop is associated to debonding at the interface between the PEI domains and the epoxy matrix for the sea-island morphology. If the PEI content increases above 15 to 20 wt%, a sudden surge in tensile strength is found associated to phase inversion [12, 44, 48] in agreement with the current results.

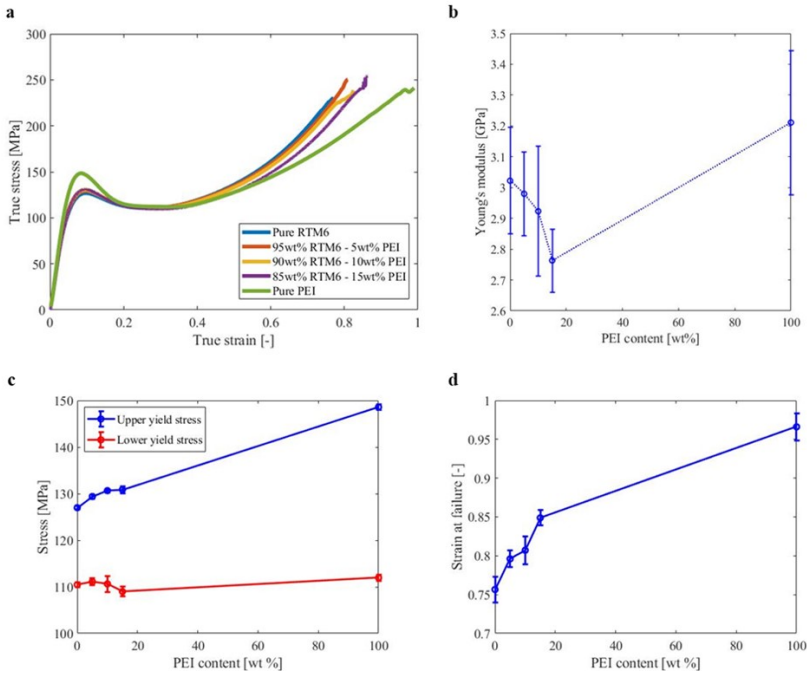


Figure 3.5 Influence of the PEI content on the mechanical response of PEI-epoxy blends under compression: a) uniaxial compression curves; b) variation of Young's modulus; c) variation of the upper and lower yield strength and d) variation of the strain at failure.

The PEI content also has an effect on the ductility (Figure 3.5d). Increasing the PEI content in the epoxy resin increases the strain at failure, in agreement with literature data [44, 48, 78, 173].

Figure 3.6 presents the hardness values measured by nanoindentation in the PEI-epoxy blends containing 5 to 15 wt% of PEI, but also in blends containing 20 and 25 wt% of PEI for which no compression samples could be

3 | PEI-epoxy interphases and co-cured composite joints

manufactured due to the high viscosity of the mixture before curing. Hardness is a measure of the resistance to plastic deformation and can therefore be related to the upper yield strength. For blends containing 5 to 15 wt% of PEI and the neat RTM6 and PEI samples, the ratio between the hardness and the upper yield stress is equal to 2.4. By increasing the PEI content from 0 to 20 wt%, the hardness only slightly increases by 2.6% as does the upper yield strength by increasing the content from 0 to 15 wt% of PEI. Further increase of the PEI content to 25 wt% results in a sudden increase of the hardness by 5%, and thus of the yield strength in agreement with literature data [44, 45]. Bulk PEI exhibits the highest value, consistently with the behavior found for σ_y^u .

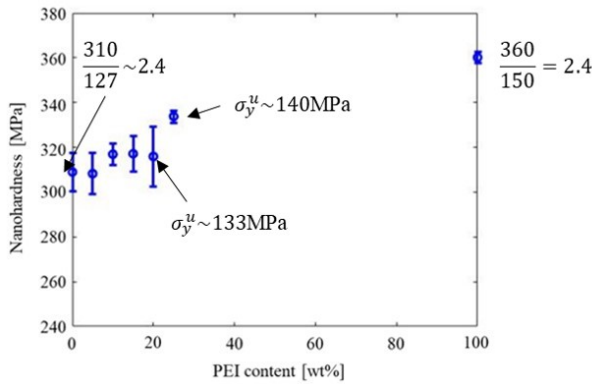


Figure 3.6 Variation of the hardness measured by nanoindentation as a function of the PEI content.

Figure 3.7 shows the variation of the toughness G_{Ic} for the blends and the corresponding fracture surfaces. The toughness slightly increases for 5 and 10 wt% PEI contents compared to the neat RTM6 epoxy. For the blend containing 5 wt%, riverlines, a mark of brittle failure, are observed on the fracture surface (Figure 3.7b-c). The riverlines extend from one PEI domain to another. Intact spherical domains or holes are found on the fracture surface, corresponding to the presence of PEI domains. The same conclusion can be drawn for the 10 wt% PEI blends (Figure 3.7d-e), except for the presence of riverlines. In addition to the debonded PEI particles, some broken PEI particles are observed on the fracture surfaces of blends with 10 wt% of PEI indicating the deformation of PEI and the bridging of the

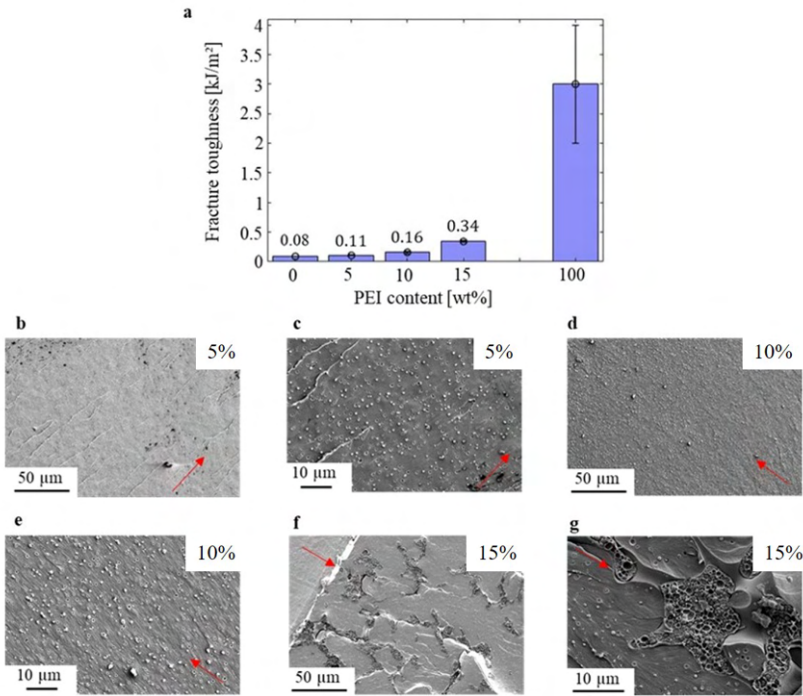


Figure 3.7 a) Variation of the fracture toughness with the PEI content of the blends and SEM micrographs of the corresponding fracture surfaces: b)-c) 5wt% of PEI; d)-e) 10 wt% of PEI; f)-g) 15 wt% of PEI. Red arrows indicate the crack propagation direction.

crack leading to toughening, as previously reported [49]. The toughness of these blends is thus mainly governed by the epoxy resin, but the presence of the PEI particles results in a slight toughening effect. G_{Ic} is more than doubled when increasing the PEI content from 10 to 15 wt%, associated to the emergence of a co-continuous morphology (Figure 3.7e-f). In the zones characterized by a sea-island morphology, the same features as those presented for 5-10 wt% PEI are observed. In the zones exhibiting a nodular morphology (Figure 3.7f), the crack propagates through the tough PEI phase and decohesion between the epoxy nodules and PEI occurs, as well as PEI tearing. No test has been performed for blends containing 20 or 25 wt%, but from the literature [44, 78], it can be assumed that G_{Ic} will further increase due to the phase inversion. In this case, yielding and fracture of the PEI matrix are reported to be the toughening mechanisms. The

increase of G_{Ic} in the blend containing 15 wt% of PEI is due to the presence of zones exhibiting the nodular morphology, in which the crack is forced to propagate through the tough PEI.

Voleppe et al. [12] reported upper and lower yield strength values equal to 115 and 95 MPa for the 15 wt% PEI blend, respectively. These values are lower than those found in this work which are equal to 127 and 110 MPa, although the same materials were used. In the current study, it has been checked that all compression samples exhibited a co-continuous morphology through imaging of the fracture surfaces. This information is not confirmed for the work of Voleppe et al. [12] However, the transition from sea-island morphology to co-continuous morphology takes place at around 15 wt% of PEI. It is thus possible that the blends of Voleppe et al. [12] exhibited a sea-island morphology for the batch tested under compression, if the exact composition was slightly below the transition value. In this case, a lower yield stress could be measured as many weak interfaces between the PEI domains and the epoxy matrix would be present. This would be in agreement with some results of the literature [44]. It has been checked through finite elements simulations that this difference does not affect the interface fracture toughness as explained in Appendix A.

3.3.3 Linking morphology and mechanical properties

Figure 3.8 summarizes in a schematic way the influence of the PEI content on the microstructure, yield strength, hardness, ductility and fracture toughness. Blends with less than 15 wt% PEI exhibit a sea-island morphology characterized by spherical PEI domains dispersed in the epoxy matrix. These blends only demonstrate a slight improvement of the mechanical properties. The properties are governed by the epoxy matrix which is softer and more brittle than PEI. Therefore, the presence of PEI contributes to a modest improvement in performance. For instance, the cracks are bridged by the PEI particles, which contributes to some shielding and reduction of the local stress intensity factor. Increasing the PEI content above 20 wt% leads to a phase inversion and, to a sharp increase of all the investigated properties. As the epoxy particles are now embedded in a PEI matrix, the tough and strong PEI governs the properties. With a co-continuous morphology (blends containing 15-20 wt% of PEI), the elastoplastic response is not drastically modified, but the fracture toughness is significantly improved by the presence of zones exhibiting a nodular morphology in which the crack is forced to penetrate.

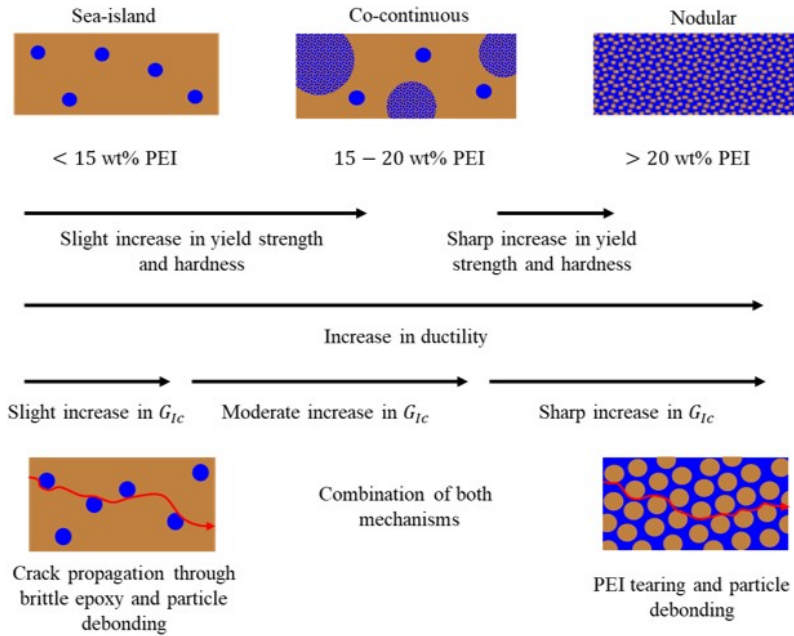


Figure 3.8 Summary of the influence of the PEI content on the microstructure, mechanical properties and failure mechanisms in PEI-epoxy blends.

3.4 Results for PEI-epoxy interphase

Although the mechanical properties and failure mechanisms of PEI-epoxy blends have already been investigated in the literature [44, 45, 47, 48], the response of PEI-epoxy interphases has been much less studied. Hereafter, the morphological gradient is characterized and nanoindentation tests are performed along the interphase. SENB experiments are carried out in order to further investigate the crack trapping mechanism described by Voleppe et al. [12] in PEI-RTM6 systems.

3.4.1 Morphological and chemical gradients

The morphological gradient, which is shown in Figure 3.9, involves five different zones. The first two zones correspond to regions where only one of the two polymers is present. The first one is the pure PEI zone, corre-

3 | PEI-epoxy interphases and co-cured composite joints

sponding to the inserted film, while the second one is the pure RTM6 zone located 100 μm away from the PEI film. Close to the PEI film, Figure 3.9e shows a region exhibiting a nodular morphology referred to as the PEI-rich zone. Close to the pure RTM6 zone, a sea-island morphology is found, see Figure 3.9c, referred to as the epoxy-rich zone. Between the epoxy- and PEI-rich zones, a more tortuous interlayer can be observed, referred to as the transition zone, see Figure 3.9d. At the boundary between the transition and the epoxy-rich zones, a thick PEI layer can be observed as highlighted by the yellow arrows in Figure 3.9d. The decohesion between the PEI boundary layer and the epoxy resin takes place when the crack propagates perpendicular to the interphase, in agreement with the low adhesion between PEI and epoxy already reported before.

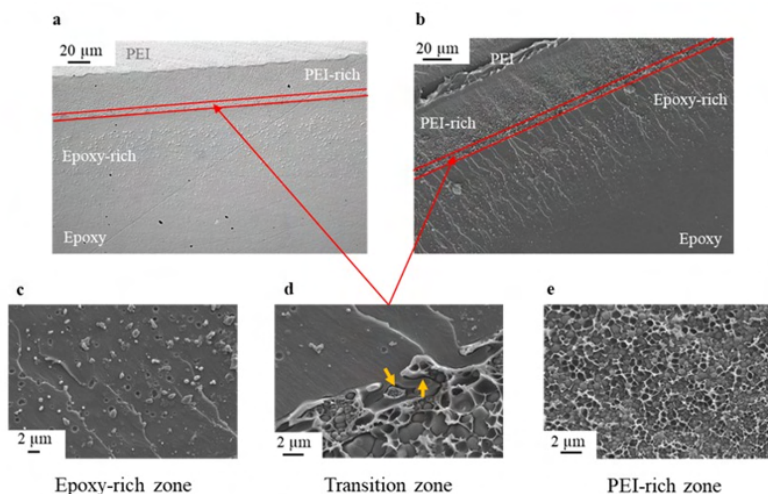


Figure 3.9 a) Optical micrograph of the interphase; SEM micrographs of b) the interphase after fracture perpendicular to the interphase, c) the epoxy-rich zone, d) the transition zone and e) the PEI-rich zone. The yellow arrow in d) indicates the decohesion between the PEI boundary layer and the epoxy.

As previously mentioned, the morphologies are dependent on the PEI concentration. From the micrographs like the one shown in Figure 3.10b, the chemical gradient can be determined by drawing computation lines. Over each line, the amount of black phase (PEI) and gray phase (epoxy) is computed. The resulting chemical gradient is given in Figure 3.10a. The PEI

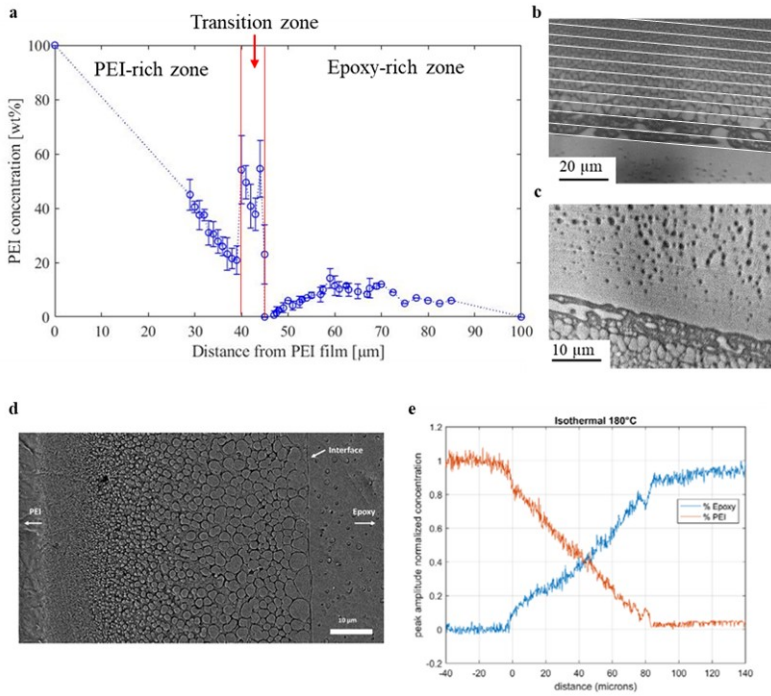


Figure 3.10 a) Chemical gradient of the RTM6-PEI interphase; b) High concentration PEI zone corresponding to the transition zone with computation lines; c) PEI depletion zone after the transition zone; d) common morphological gradient reported in the literature and e) the corresponding chemical gradient (taken from [52]).

concentration linearly decreases away from the PEI film. However, in the transition zone, a sudden increase of the PEI concentration is found, increasing from 20 to 40-60 wt%. The transition zone in the optical micrograph (Figure 3.10b) is darker than the rest of the micrograph, indicating a high PEI content. This jump had already been observed by Voleppe [69]. In addition, just after the transition zone, where the epoxy-rich zone starts, the PEI content is about 0 wt% and then increases, over a distance of 15 μm, to 12 wt% before decreasing down to 0 wt% over the next 40 μm. Figure 3.10c shows this depletion in PEI right beyond the transition zone. This chemical gradient is different from what is commonly reported (see Figure 3.10e), see e.g. the profile suggested by Lestriez et al. [175], but the morphological gradient formed at the RTM6-PEI interphase is also dif-

ferent from the one often reported in the literature. Indeed, the tortuous structure observed in the transition zone in Figure 3.9 does not show up in Figure 3.10d. The chemical gradient corresponding to the interphase of Figure 3.10d is also found in Figure 3.10e. The PEI content almost linearly decreases away from the PEI film, and no concentration peak is observed at the transition from the PEI-rich zone to the epoxy-rich zone [44, 52]. It is known that the curing cycle, the curing agent, the viscosity of the resin or the molecular weight of the TP can all influence the morphological gradient, providing a series of explanation for the observed differences [44, 51, 53]. The formation of this morphological gradient is left for further investigations.

3.4.2 Mechanical properties

The variation of the hardness along the interphase is evaluated through nanoindentation. In addition, a comparison can be made with the blends through the chemical gradient. Figure 3.11 shows a good match when combining the blends hardness and the chemical gradient to the hardness measured along the interphase. The hardness decreases when moving away from the PEI film in the PEI-rich zone. This decrease is almost linear until the end of the transition zone. At the interface between the transition zone and the epoxy-rich zone, the hardness drops, but remains higher than in bulk RTM6 before slightly increasing over 10 μm due to the increase in PEI content in this region, see Figure 3.11b. Finally, the hardness decreases and tends towards the value of bulk RTM6 epoxy resin. In the transition zone, a higher hardness was expected based on the chemical gradient, but not found. Note that the indentation response is affected by the surrounding environment depending on indentation depth normalized by the distance to the regions exhibiting a different behavior. As the two zones framing the transition zone contain less PEI, they are softer and the extracted hardness of the transition zone can therefore appear lower than the true value.

Through nanoindentation, it is also possible to measure the elastic modulus, although the value is different from the modulus measured under compression for blends. For blends, the modulus measured by nanoindentation was found to increase with PEI content with no minimum for the 15 wt% PEI blend unlike what was obtained from the compression tests. A decrease of the modulus is also found along the interphase as the PEI content decreases.

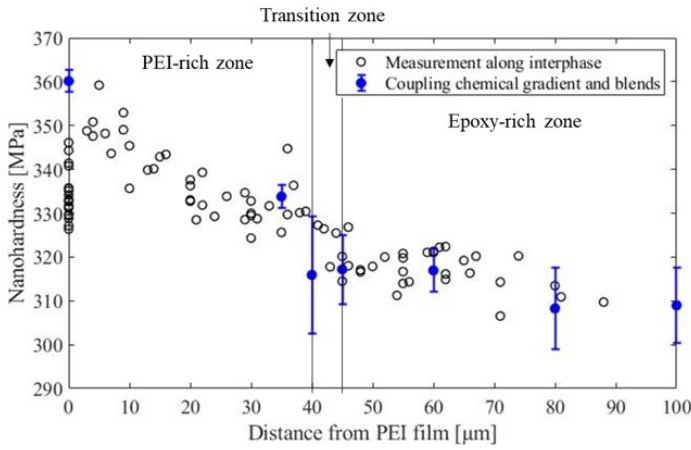


Figure 3.11 Variation of the hardness along the interphase: measurement along the interphase and coupling between the chemical gradient and the results on PEI-epoxy blends.

3.4.3 Investigation of the failure mechanisms

One of the goals of the work was to further investigate the trapping mechanism proposed by Voleppe et al. [12] for a system similar to the one addressed in this work. In the work of Voleppe et al. [12], it was argued that the trapping of the crack between the PEI-film and the epoxy-rich zone was due to the presence of a transition zone softer than the PEI- and epoxy-rich zones. In the present study, the transition zone is characterized by a tortuous structure exhibiting a nodular morphology similar to the one observed by Voleppe et al. Nanoindentation and compression results show that this region is not, in our case, softer than the neighboring regions.

In order to understand the crack trapping mechanism, SENB tests similar to those of Voleppe et al. [12] are performed. At first, smaller SENB samples are tested under optical microscope. The interphase observed by optical microscopy prior to testing and after failure and the fracture surfaces are characterized by SEM. The same crack trapping mechanism as that found by Voleppe et al. [12] is identified, as shown in Figure 3.12 when the crack is initiated in the tough PEI-rich zone (Figure 3.12a and Figure 3.12c-e). Indeed, the crack first propagates in a zone exhibiting a nodular morphology (Figure 3.12c-e) before propagating along a flat featureless surface (Figure 3.12e-f). Figures 3.12d-e focus on the transition from crack

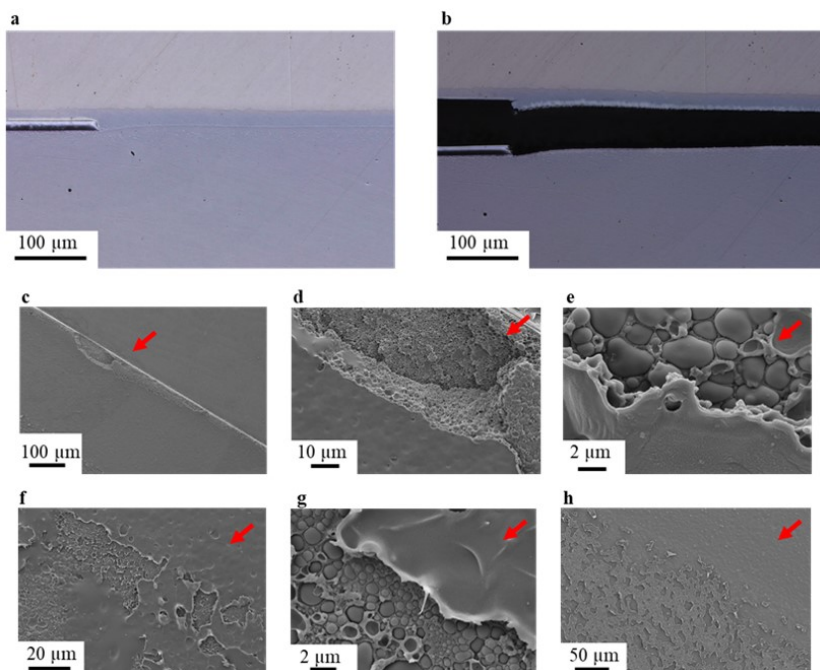


Figure 3.12 Crack initiation and propagation loci when the pre-crack lies in the PEI-rich zone. Optical micrographs a) before loading and b) upon fracture. SEM micrographs of fracture surface c) initiation locus in the PEI-rich zone with d) a zoom in the initiation zone and e) at the location of the deflection along the interface between the PEI boundary layer and the epoxy resin; f) slight deflection in the PEI-rich zone during propagation, g) zoom highlighting the cutting of the PEI boundary layer to propagate in this zone, h) opposite surface.

propagation in the PEI-rich zone to propagation along the featureless zone. Figure 3.12e shows that the crack propagates through the PEI layer before being deflected and propagating along the following featureless surface. This PEI layer corresponds to the PEI boundary layer at the interface between the transition zone and the epoxy-rich zone. In addition, during the propagation along this featureless surface, the crack sometimes propagates into the transition zones as shown in Figure 3.12f to h. Some areas exhibiting a nodular morphology are detected. Again, at the boundary of these zones (Figure 3.12g), the crack is found to propagate through the PEI boundary layer forming the interface between the transition zone and

the epoxy-rich zone. This short propagation through zones exhibiting a nodular morphology is believed to originate from the tortuous structure of the transition zone. This mechanism probably requires less energy to the crack to propagate straight into this region instead of being deflected along the weak PEI-epoxy interface, which would introduce some local mode II components. A propagation close to the transition zone agrees with the optical micrographs shown in Figure 3.12a and b taken prior and upon failure. Comparing these two micrographs confirms that the crack propagates close to this interface. The privileged scenario is crack propagation along this interface due to low adhesion between PEI and epoxy resin, the weakest point of the interphase. For the other samples, the PI film used to insert a pre-crack moved during the curing and did not lay in the PEI-rich zone anymore. In this case, the crack is initiated in the epoxy-rich zone. Indeed, Figure 3.13 shows that the crack propagates away from the PEI, and a sea-island morphology is observed on the fracture surface indicating crack propagation in the epoxy-rich zone. In this case, no propagation is detected along the interface between the transition zone and the epoxy-rich zone.

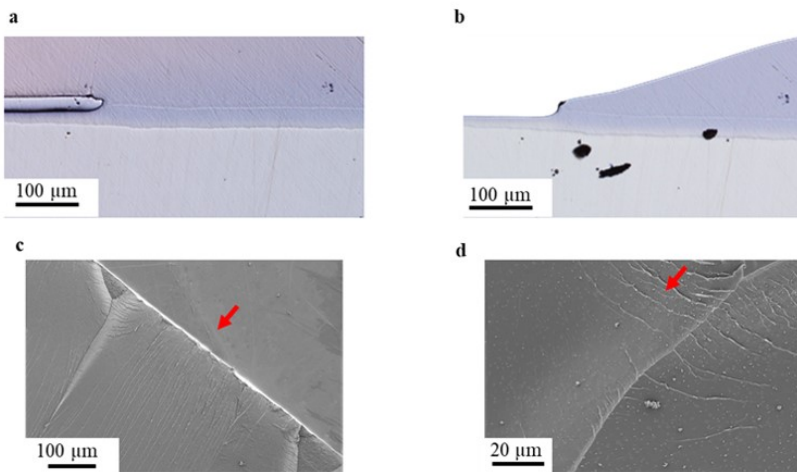


Figure 3.13 Crack initiation and propagation loci when the pre-crack lies in the epoxy-rich zone. Optical micrographs acquired a) before loading and b) upon fracture. SEM micrographs of one of the fracture surfaces: c) initiation locus in the PEI-rich zone, d) zoom on the fracture surface highlighting propagation in the epoxy-rich zone exhibiting a sea-island morphology.

To verify that the smaller size of our SENB specimens compared to those made by Voleppe et al. [12] did not alter the failure mechanisms, SENB specimens of the size selected by Voleppe et al. [12] are also tested but not under the optical microscope. Figure 3.14 shows one fracture surface of a crack initiated in the PEI-rich zone. The crack first propagates in the PEI-rich zone, characterized by a nodular morphology in Figure 3.13b, and a leaves-like pattern is observed in this region. Then, at some point, the crack starts to propagate along a flat featureless surface as mentioned earlier. At the transition between these two loci, the crack propagates through the PEI boundary layer of the transition zone before being deflected and propagating along the featureless interface. The same failure mechanism is thus observed as in the small SENB specimens.

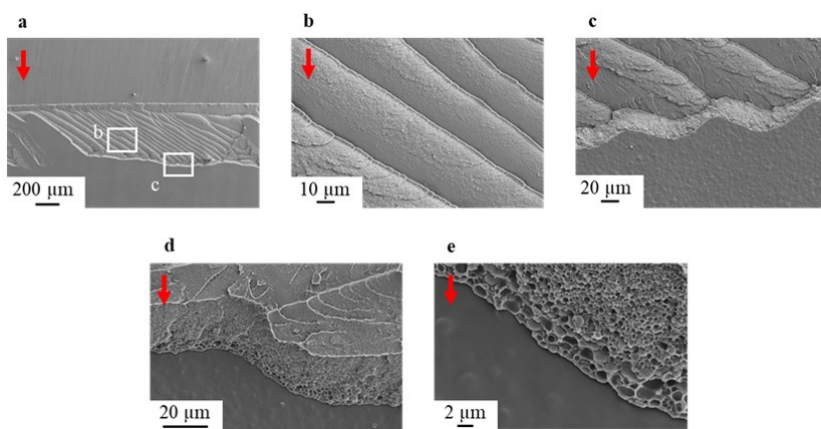


Figure 3.14 SEM micrographs of fracture surfaces of large SENB samples: a) initiation in the PEI-rich zone with b) a leaves-like pattern, c-e) deflection at the interface between the PEI boundary layer of the PEI-rich zone and the epoxy-rich zone. c-d) opposite images and e) zoom in d) at the deflection locus.

The failure mechanisms are summarized in Figure 3.15. Two cases can be distinguished, depending on the crack initiation locus: (i) in the PEI-rich zone or (ii) in the epoxy-rich zone. In the first case, the crack first propagates through the tough PEI-rich zone before being deflected at the interface between the transition and epoxy-rich zones where the crack propagates between the epoxy matrix of the epoxy-rich zone, and the PEI boundary layer of the transition zone, due to the lack of adhesion between PEI

and epoxy resin. In the second scenario (ii), the crack is initiated in the epoxy-rich zone and propagates through it without being deflected towards the interface with the transition zone.

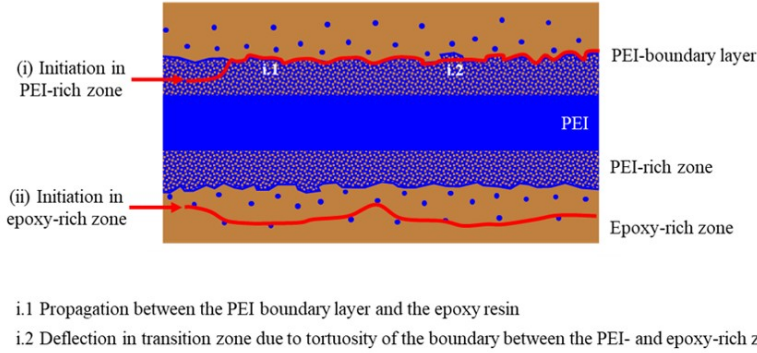


Figure 3.15 Schematic summary of the failure scenarios with (i) a pre-crack in the PEI-rich zone and (ii) in the epoxy-rich zone. In case (i), the crack propagates along the PEI boundary layer and the epoxy resin (i.1) but can be deflected in the PEI-rich zone due to tortuosity of the interface (i.2).

Finally, we discuss now the fracture toughness values measured by Voleppe et al. [12]. For SENB specimens tested in the present work, no crack propagation is observed before complete failure of the samples. This means that an initiation value is measured and, not a propagation one. Hence, the measured value of 880 J/m^2 in the case of the crack starting in the PEI-rich zone corresponds to the initiation fracture toughness. A high value is expected in this zone as the nodular morphology resulting from the phase inversion is known to be beneficial. The fracture toughness of the interface between the PEI boundary layer of the transition zone and the epoxy-rich zone cannot be measured through SENB testing. This highlights the fact that when testing a thermoplastic-epoxy interphase, or any other one, not only initiation is important but also propagation because the pre-crack does not always stay in the weakest part of the interphase, resulting in an overestimation of its fracture toughness. This requires to design a test set-up to properly test interfaces as suggested in Appendix B.

In summary, this study confirms that if the crack initiates in the PEI-rich region, G_{Ic} is high. However, the crack does not necessarily stay in the tough region and propagates in a more brittle way with much less dissi-

pation. In other words, the scenario of Voleppe et al. [12] of the crack being trapped in a softer zone is only very partly recovered. There are two reasons for this. The first is that the generation of a softer region in the interphase region is not guaranteed and requires conditions that are delicate to reproduce. The second reason is that Voleppe's work did not look at the propagation beyond the first stage of cracking and maybe had similar effects. This cannot be verified as the specimens are no longer available. This does not mean that the idea of crack trapping in a soft/tough region/layer is not a rich idea to keep pushing in further studies, just that the PEI/RTM6 is not necessarily the best system to activate this mechanism partly due to the difficulty to control in a reproducible way the local graded microstructure. Finally, note that the transverse toughening when the crack penetrates transversely the interphase has been investigated in the literature [49, 54] and is efficient.

3.5 Results for PEI co-cured composite joints

3.5.1 Fracture toughness and failure mechanisms

Figure 3.16 shows the R-curves resulting from the DCB testing of the composite parts and of the PEI co-cured joints. Inserting a 50 μm thick PEI film results in a fracture toughness equal to $284 \pm 89 \text{ J/m}^2$ while a 175 μm thick film results in a lower G_{Ic} equal to $146 \pm 28 \text{ J/m}^2$. The highest fracture toughness is achieved with a 500 μm film and is equal to $370 \pm 47 \text{ J/m}^2$. The G_{Ic} associated to interply delamination is equal to $359 \pm 109 \text{ J/m}^2$. The insertion of a 500 μm film is thus competitive with the delamination energy, but not with classical structural adhesives which are characterized by G_{Ic} values around 800 to 1000 J/m^2 . Inserting a 50 μm thick PEI film results on average to a lower G_{Ic} compared to composite delamination, although some results are competitive with composite delamination. The 175 μm film always leads to a lower fracture toughness, compared to the reference composite.

The adhesive thickness is known to influence the joint fracture toughness by affecting the degree of constraint in the bondline, and thus the stress triaxiality and the constraint on the development of the plastic zone. As the adhesive thickness increases, the degree of constraint decreases and the plastic zone can develop resulting in an increase in fracture toughness up to a thickness equal to twice the plastic zone size of the adhesive, if

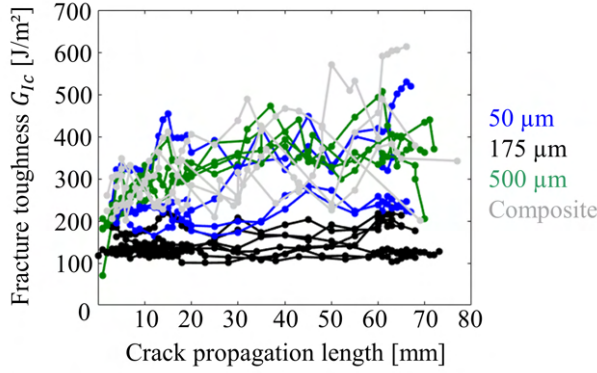


Figure 3.16 R-curves for the co-cured composite joints with a PEI a 50 μm (blue), a 175 μm (black) and a 500 μm (green) thick film. The grey lines correspond to the reference composite.

the crack propagates in the middle of the joint. The thickness has also an effect for cracks propagating along an interface. If the interface cohesive strength is about 2 to 4 times the adhesive yield strength, the maximum toughness is reached if the adhesive thickness is equal to twice the plastic zone size, which depends on the elastic mismatch between the adhesives and the adherends and is given by

$$R_0 = \frac{2}{3\pi(1-\beta^2)} \left[\frac{1-\nu_s^2}{E_s} - \frac{1-\nu_a^2}{E_a} \right]^{-1} \frac{G_0}{\sigma_{0a}^2}, \quad (3.1)$$

with ν_s and ν_a the Poisson ratios of the substrate and the adhesive, respectively, E_s and E_a the Young's modulus of the adherends and the adhesive and σ_{0a} , the adhesive yield strength. β is the second Dundur's parameter expressed as

$$\beta = 0.5 \frac{\mu_a(1-2\nu_a) - \mu_s(1-2\nu_s)}{\mu_a(1-\nu_a) + \mu_s(1-\nu_s)}, \quad (3.2)$$

where μ_i is the shear modulus of material i , given by $E_i/2(1+\nu_i)$. In the present case, this results in a plastic zone size close to 350 μm . Hence, the maximum toughness is achieved with the 500 μm film. Quan et al. [31] reported a maximum fracture toughness for PEI layers of about 175 μm thickness.

Figure 3.17 shows the failure mechanisms taking place in co-cured com-

posite joints with a 50 μm thick PEI film. Due to the architecture of the carbon fibres plies, the crack can propagate in resin-rich pockets or along carbon fibres. Upon curing, a morphological gradient like that reported by Voleppe et al. [12] develops. First, in the resin-rich pockets (Figure 3.17b-e), this gradient can fully develop and the crack propagates along the interface between the resin and PEI-rich zones (Figure 3.17d). Sometimes, it is also slightly deflected into the PEI-rich zone (Figure 3.17e). In the second case, the crack propagates away of a resin pocket (Figure 3.17f-i). The crack can propagate through the PEI film (Figure 3.17f and g), generating some short bridging PEI ligaments and some local mode-mixity due to the deflection. If it does not propagate through the film, it propagates along the carbon fibres located in the PEI-rich zone owing to the development of the morphological gradient (Figure 3.17h and i).

Figure 3.18 shows the failure mechanisms taking place in co-cured composite joints with a 175 or 500 μm thick PEI film. The crack is no longer able to propagate through the PEI film as shown in Figure 3.18a. As with the 50 μm thick PEI film, in resin-rich pockets, the crack propagates along the interface between the PEI-rich and epoxy-rich zones of the morphological gradient (Figure 3.18d-e). Away from the resin pockets, crack propagation takes place along the fibres in the PEI-rich zone (Figure 3.18f-g). Opposite to the present results, in adhesive joints, an increase in G_{Ic} is reported with the adhesive thickness and, up to now, the same behaviour was observed by increasing the PEI film thickness between 90 and 250 μm . Hence, it is believed that the higher toughness resulting from the insertion of a 50 μm PEI film originates from the crack deflection through the PEI film, that results in some mode-mixity at the crack tip and in the formation of small PEI bridging ligaments. Crack deflection [119] and bridging [124] are two well-known toughening mechanisms in adhesive joints.

An additional feature that can be observed on all fracture surfaces is the presence of ellipsoidal circles as shown in Figure 3.18b. Zooming on these features do not reveal any of the previously mentioned morphologies. These spots seem to correspond to zones without interdiffusion between PEI and epoxy. This lack of interdiffusion could result in decohesion at the boundary between the PEI film and the epoxy matrix. The origin of these ellipsoidal circles is unknown but as these are not observed in PEI-epoxy interphase, they could arise from the manufacturing process and resin flows during injection. In addition, such features are not reported by Quan and co-workers who used pre-pregs without resin injection, preventing resin

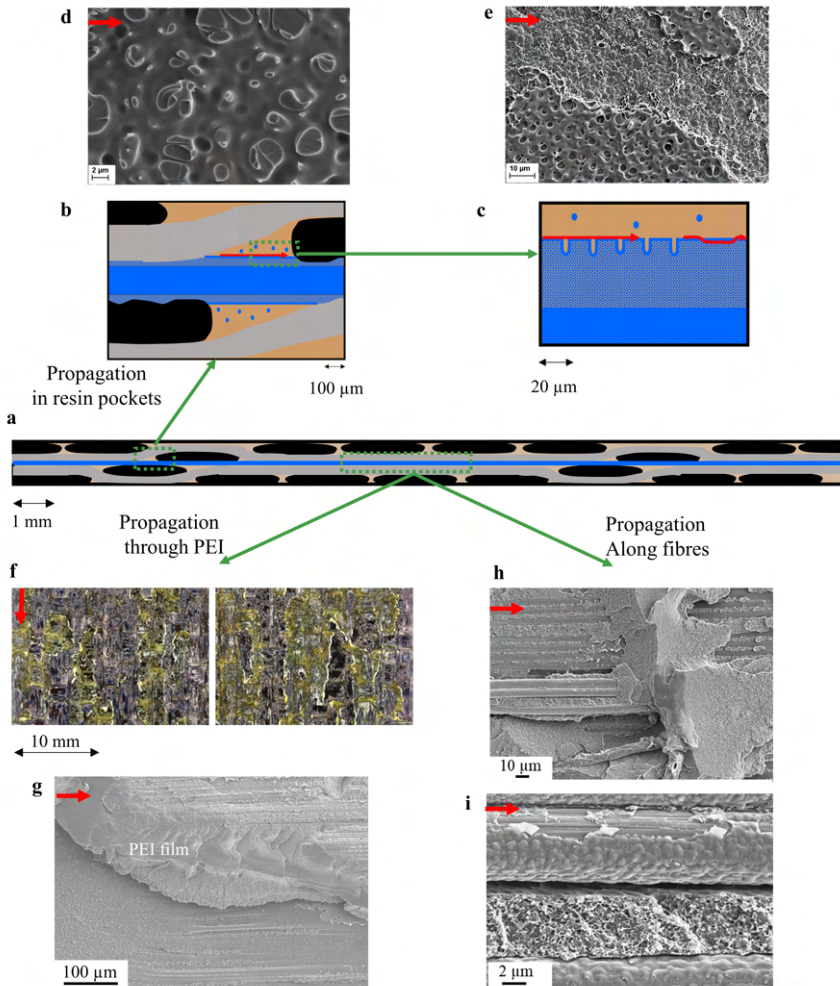


Figure 3.17 Failure mechanisms taking place in co-cured composite joints with a 50 µm thick PEI film: a) scheme of the joint, b) morphological gradient in a resin pocket and c) zoom on the PEI-rich/epoxy-rich interface where the crack propagates d) along the PEI-rich/epoxy-rich interface or e) locally in the PEI-rich zone, f) fracture surface with PEI on both sides indicating g) propagation through the PEI film. h-i) Propagation along the fibres present in the PEI-rich zone away of the resin pockets. The red arrows indicate the crack propagation direction.

flow.

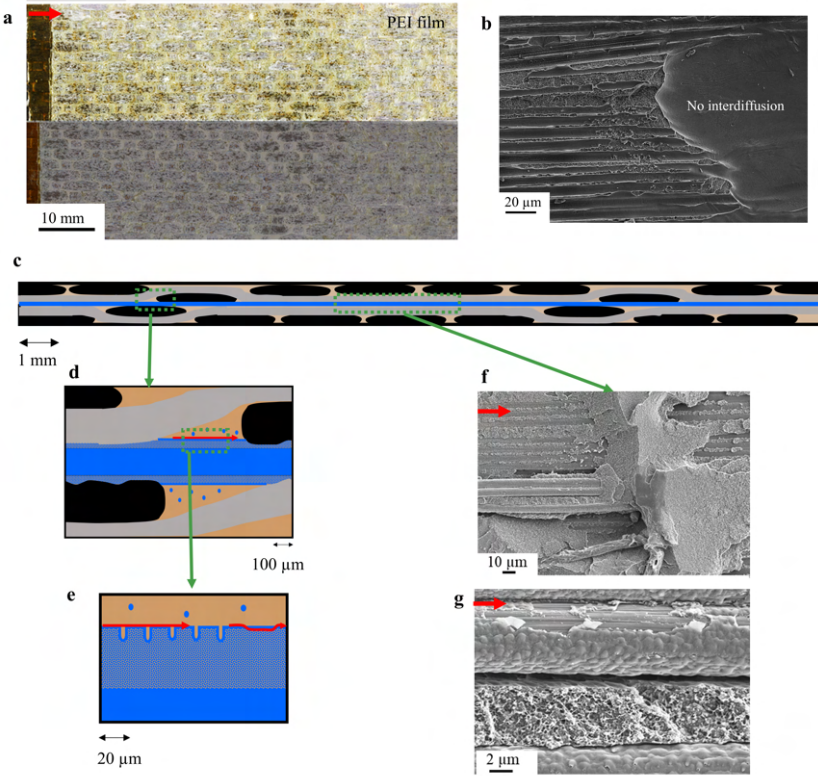


Figure 3.18 Failure mechanisms taking place in co-cured composite joints with a 175 or 500 μm thick PEI film: a) whole fracture surfaces with the PEI film remaining only on one face, b) SEM micrograph of a zone without interdiffusion, c) scheme of the composite (PEI in blue), d-e) propagation in resin rich pockets where f) the morphological gradient develops and g) the crack propagates along the PEI-rich/epoxy-rich interface or slightly in the PEI-rich zone, f) propagation along the fibres in g) the PEI-rich zone. The red arrows indicate the crack propagation direction.

Opposite to the results of Quan and co-workers [30, 31], inserting a PEI film results in a lower G_{Ic} compared to that of the reference composite. Along the fibres, delamination in the PEI-rich zone was also reported. However, crack propagation along the PEI-rich/epoxy-rich interface and through the PEI film were not observed by Quan et al. [30, 31]. Propagation through

the film cannot explain the lower G_{Ic} , while the propagation along the PEI-rich/epoxy-rich interface could but this mechanism only takes place very locally and cannot explain such a difference. One of the explanations can be related to the nature of the materials that are used. Quan et al. [30, 31] used the 8552 epoxy resin, which is harder than RTM6 and PEI. Hence, a decrease of the hardness and strength is expected from the epoxy-rich zone to the PEI film while in the present case, the RTM6 is softer than PEI resulting in an increase in hardness. In the work of Quan et al., the PEI can potentially act as a dissipative layer [79, 97] but does not for the investigated PEI-RTM6 system. In addition, the 8552 resin is toughened by a thermoplastic and the sizing of the fibres could be adapted to this particularity while not for the present used carbon fibres resulting in a weaker interface with the carbon fibres, decreasing the fracture toughness. Finally, the process can also be at the origin of the lower fracture toughness as Quan et al. [30, 31] used pre-pregs combined with autoclave curing without subsequent resin injection while resin is injected during the RTM process in the present work. However, this must be further investigated. The comparison of these two works show that it is important to understand what happens during the manufacturing process but it is also important to consider the material properties. It could be interesting to test systems made of other epoxy resins to more deeply understand the failure mechanism taking place in the PEI co-cured joints. In addition, manufacturing processes such as pre-pregs with autoclave curing or RTM could be compared to determine whether the process has an influence on the co-cured joint performances.

3.5.2 Trapping mechanism and its implication on composite-PEI joints

Figures 3.17 and 3.18 show that due to the composite architecture, resin pockets form at the locations where the carbon fibre orientation changes. In these pockets, the morphological gradient can completely develop, resulting in the formation of a sharp boundary between the PEI-rich and epoxy-rich zones similar to that observed in PEI-RTM6 interphases (Figure 3.9). The crack propagates along this boundary as in SENB specimens, indicating that the interface between the two zones is the weakest point in these pockets. This confirms that the weak interface between these two zones is at the origin of the crack trapping mechanism observed by Voleppe et al. [12]. The development of this weak interface must be prevented and consequently, the full development of the morphological gradient too. This

requires to force the carbon fibres to be located in the PEI-rich zone by depositing the PEI film directly into contact with the carbon fibres. Hence, the distance between the PEI film and the carbon fibres must be lower than the length of the PEI-rich zone. If not, the crack will propagate along the weak interface. Understanding the failure mechanism is therefore important for the design of tough co-cured PEI-composite joints and in general for TP-epoxy interphases, requiring the design of an adequate test set-up (Appendix B).

3.6 Conclusion and perspectives

In order to reduce the processing intensity of adhesive bonding, co-curing is an interesting solution, though it requires the adhesive and composite resin to have matching curing cycles and it is limited to process without any flow of resin. An alternative to the use of an adhesive is the insertion of a thermoplastic film such as PEI. Upon curing, the interdiffusion of PEI and epoxy results in a morphological gradient with two main zones: a PEI-rich and an epoxy-rich zone separated by a thin transition zone. The first goal of the current study was to understand the failure mechanisms taking place in PEI-epoxy interphases when a crack propagates parallel to the PEI film as Voleppe et al. [12] reported a fracture toughness up to 880 J/m^2 for such interfaces. The second goal was to manufacture co-cured PEI-composite joints through RTM processing in order to assess the fracture toughness of such joints.

Investigating the properties of PEI-RTM6 interphases and blends revealed that the softening found in the transition zone by Voleppe et al. [12] could not be reproduced. Indeed, the yield strength/hardness and the Young's modulus tend to increase with the PEI content resulting in a decrease of these properties along the interphase when moving away from the PEI film. By performing SENB tests, two failure mechanisms have been observed. The first one is observed when the crack is initiated in the epoxy-rich zone and does not go back to the PEI film. The second one corresponds to the crack trapping mechanism observed by Voleppe et al. [12], and takes place when the crack is initiated in the PEI-rich zone. Once initiated the crack tends to propagate towards the epoxy-rich zone without reaching it. The crack then propagates along the interface between the PEI-rich and epoxy-rich zones, due to the lack of adhesion between PEI and epoxy. Hence, the toughness value computed by Voleppe et al. [12]

corresponds to an initiation toughness in the PEI-rich zone, and not to the fracture toughness of the weak interface. To determine the fracture toughness of the weak interface, another geometry must be used than the SENB one. We suggest to perform wedge-peel tests, but this is left for further investigation.

Regarding PEI-composite co-cured joints, the toughness is found to depend on the PEI thickness, which controls the failure mechanism and the plastic dissipation. At low thickness ($50\mu\text{m}$), the crack can propagate through the PEI film resulting in a fracture toughness higher than for a film of $175\mu\text{m}$. For thick films, the degree of constraint is lower and the fracture toughness increases if we compare the $175\mu\text{m}$ and $500\mu\text{m}$ thick films. However, compared to composite delamination, the toughness is only slightly improved, limiting the interest for such joints. Opposite, Quan et al. [31] reported an improved toughness. Hence, there could be an influence of the epoxy resin properties with respect to those of PEI and also an influence of the process. However, this must be further investigated. Finally, the presence of a weak interface must be considered in the design of such joints. Indeed, if the spacing between the PEI film and the fibres exceeds the length of the PEI-rich zone, the weak interface can develop and this could be detrimental to the joint fracture toughness as well.

As a perspective, in order to confirm the presence of the weak interface and compute its fracture toughness, crack propagation tests along PEI-epoxy interphases must be performed. For this purpose, we suggest to perform wedge-peel tests following the method described in Appendix B. This could be applied to any type of interface to guarantee a measure of the interface fracture toughness, and not only of an initiation fracture toughness at a location that does not correspond to the weakest point of the interface. In addition, failure mechanisms for different PEI/epoxy systems could be investigated (with the epoxy being harder or softer than PEI) as well as for joints manufactured through different processes.

3.7 Appendices

3.7.1 Appendix A: Influence of transition zone properties

It has been shown that the crack propagates along the interface between the transition zone and the epoxy-rich zone. In addition, different properties were obtained for this transition zone in the work of Voleppe et al. [12]

and in the current study. This raised the question “Does the elastoplastic response of the transition zone influence the fracture toughness measured when a crack propagates along the interphase?” To answer this question, finite element analyzes are performed using cohesive elements with Abaqus. For the cohesive elements, a UEL using the Gauss integration scheme and a trapezoidal cohesive law has been implemented. The geometry is the one of a single edge notch tensile (SENT) specimen as shown in Figure 3A.1a and is meshed using 4 noded plane strain elements with reduced integration (CPE4R). The interphase is modelled by dividing it into 5 zones: the PEI film, the PEI-rich zone, the transition zone, the epoxy-rich zone and the pure epoxy zone as shown in Figure 3A.1b-c. The width W of the SENT is equal to 500 times the plastic zone size of RTM6 R_0^{RTM6} and the total height of the SENT is equal to $5.5W$. The initial crack length is equal to $0.5W$.

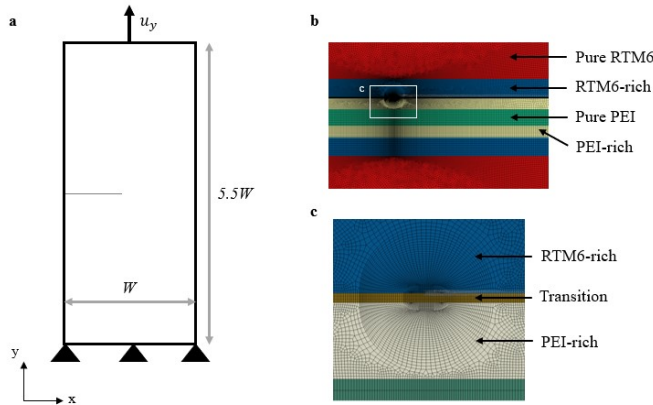


Figure 3A.1 a) Geometry of the SENT sample. b-c) Partitions to model the interphase with the 5 zones: pure RTM6, RTM6-rich zone, transition zone, PEI-rich zone and pure PEI.

Constitutive laws

Mechanical properties have been assigned for each zone of the interphase:

- Pure RTM6: compression curve for bulk RTM6 obtained in the present study, but which is similar to the one of Voleppe et al. [12].
- Epoxy-rich zone: compression curve for PEI-RTM6 blend containing 5 wt% of PEI obtained in this study, similar to the one of Voleppe et

al. [12].

- Transition zone: compression curve for the blend containing 15 wt% of PEI. The properties assigned in this zone differ in the two investigated cases.
- PEI-rich zone: extrapolated compression curves based on nanoindentation data for the blend containing 25 wt% of PEI.
- Pure PEI: compression curve for bulk PEI obtained in the present study, similar to the one of Voleppe et al. [12].

What differs between the two cases is the elastoplastic response of the transition zone. In the first case, the response corresponds to the one shown in Figure 3.5a for the 15 wt% blend. For the second case, the curve has been adapted to match the upper and lower yield stress obtained by Voleppe et al. The stress-strain curves used as constitutive laws are shown in Figure 3A.2. In what follows the upper yields stress will be referred to σ_y^u .

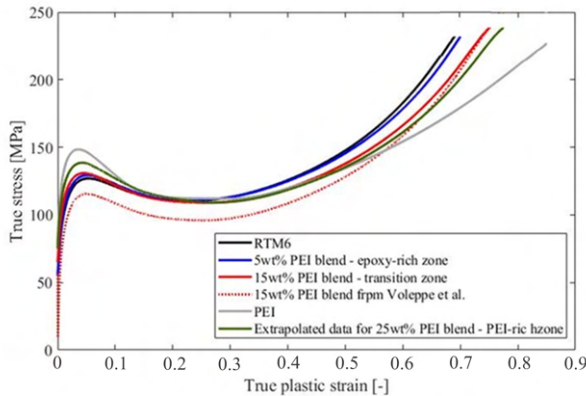


Figure 3A.2 True stress - true plastic strain response for the different materials used in the simulations based on compression curves obtained at a constant cross-head speed of 1 mm/min.

The PEI-RTM6 blends exhibit strain-softening which is known to lead to the development of shear bands that can lead to mesh-dependent results. To circumvent this issue, strain-rate sensitivity has been introduced by considering a strain-rate sensitivity exponent of 0.03 [168]. Although, this did

not completely remove the mesh-dependency. Hence, the two cases are compared using similar structured meshes.

Cohesive zone elements

Cohesive elements are zero-thickness interface elements used to model crack propagation as shown in Figure 3A.3a. Their behaviour is governed by a cohesive law relating the traction σ to the nodal separation (relative displacement) δ . In the present case, a trapezoidal cohesive law is used as the one shown in Figure 3A.3b. It is characterized by an elastic part for $\lambda_1 = \delta/\delta_c \leq 0.15$ with δ_c being the critical opening displacement. This part is followed by a constant stress part over which the traction is equal to the critical stress σ_c and extends from $\lambda_1 = 0.15$ to $\lambda_2 = 0.5$. Finally, traction decreases when $\lambda \geq 0.5$ and vanishes when $\lambda = 1$ corresponding of the breaking of the element. The cohesive energy is given by

$$\Gamma_c = \frac{\sigma_c \delta_c}{2} (1 - \lambda_1 + \lambda_2). \quad (3A.1)$$

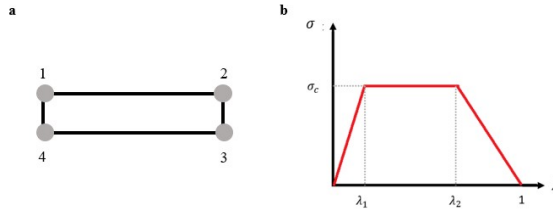


Figure 3A.3 a) Schematic representation of a cohesive element and its node numbering. b) Trapezoïdal cohesive traction: traction σ versus adimensional separation λ .

Cohesive elements are already implemented in Abaqus. However, in preliminary studies, it has been found that with excessive plasticity ($\sigma_c/\sigma_y^u > 3$), the COH2D4 elements do not properly open. Abaqus cohesive elements use the Newton-Cotes integration scheme which has already been reported to fail in presence of plastic deformations around the cohesive zone [176, 177]. Consequently, a user element subroutine (UEL) has been implemented in Fortran and in which the Gauss integration scheme is used.

□ UEL implementation

One considers of 4 node elements as shown in Figure 3A.3 and the nodal displacement vector is given by $\mathbf{u} = (u_x^1 \ u_y^1 \ u_x^2 \ u_y^2 \ \cdots \ u_x^4 \ u_y^4)^T$ with u_i^j being the nodal displacement of node j in direction i . The separation vector is then

$$\Delta = \begin{bmatrix} u_x^1 - u_x^4 \\ u_y^1 - u_y^4 \\ u_x^2 - u_x^3 \\ u_y^2 - u_y^3 \end{bmatrix}. \quad (3A.2)$$

Based on the nodal separation vector Δ , the separation at the integration points δ can be obtained using the interpolation matrix $\mathbf{S}$. δ has two components: the normal separation δ^N and the tangential separation δ^T and is given by

$$\delta = \begin{bmatrix} \delta^T \\ \delta^N \end{bmatrix} = \mathbf{S}\Delta = \begin{bmatrix} S_1 & 0 & S_2 & 0 \\ 0 & S_1 & 0 & S_2 \end{bmatrix} \Delta, \quad (3A.3)$$

with S_1 and S_2 begin the two interpolation functions given by $S_1 = (1 - \eta)/2$ and $S_2 = (1 + \eta)/2$ with η being the position of the integration point. η is set to 0 for the Newton-Cotes integration scheme while it is equal to 0.57735 for the Gauss integration scheme. δ is the separation vector in the global coordinates while in the local coordinates, the separation vector is denoted by $\mathbf{v}$ and is given by

$$\mathbf{v} = \Theta\delta = \Theta\mathbf{S}\mathbf{H}\mathbf{u} = \begin{bmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{bmatrix} \mathbf{S}\mathbf{H}\mathbf{u} = \mathbf{N}\mathbf{u}. \quad (3A.4)$$

The cohesive elements contribute to the virtual work that postulates that the internal virtual work (right term in Equation 3A.5) is equal to the external virtual work (left term Equation 3A.5). The internal virtual work is the sum of the virtual strain energy and the cohesive fracture energy on the fracture surface.

$$\int_{S_{ext}} \delta \mathbf{d}^T \cdot \mathbf{F} dS = \int_V \delta \boldsymbol{\varepsilon}^T \boldsymbol{\sigma} dV + \int_{\Gamma_c} \delta \mathbf{v} \cdot \mathbf{T} dS, \quad (3A.5)$$

$\delta \mathbf{d}$ represent the virtual displacement vector, $\delta \boldsymbol{\varepsilon}$ the corresponding variations of the small strain tensor, $\mathbf{v}$ the separation between the faces of the cohesive element and $\mathbf{T}$ the traction acting on the cohesive surface. The contribution of the cohesive elements is equal to the second right term and

3 | PEI-epoxy interphases and co-cured composite joints

can be rewritten as

$$\int_{\Gamma_c} \delta \mathbf{v} \cdot \mathbf{T} dS = \int_{\Gamma_c} \delta \mathbf{v}^T t dS = \int_{\Gamma_c} (\mathbf{N} d\mathbf{u})^T t dS = d\mathbf{u}^T \int_{\Gamma_c} \mathbf{N}^T \mathbf{T} dS = \mathbf{f} \delta \mathbf{u}, \quad (3A.6)$$

where $\mathbf{f}$ represents the internal forces vector and is given by

$$\mathbf{f} = \int_{\Gamma_c} \mathbf{N}^T \mathbf{T} dS = \int_{-1}^1 \int_1^1 \mathbf{N}^T \mathbf{J} d\eta d\zeta = B \int_{-1}^1 \mathbf{N}^T \mathbf{T} |\mathbf{J}| d\eta, \quad (3A.7)$$

where B is the out-of-plane thickness of the cohesive element and $|\mathbf{J}|$ is the determinant of the Jacobian and is equal to $l_{coh}/2$ with l_{coh} being the length of the cohesive element. The Right-hand side matrix can be then computed and is given by

$$\mathbf{RHS} = -\mathbf{f} = -B \int_{-1}^1 \mathbf{N}^T \mathbf{T} |\mathbf{J}| d\eta. \quad (3A.8)$$

The stiffness matrix, denoted by AMATRX in Abaqus, is given by

$$\mathbf{AMATRX} = \mathbf{K} = \frac{\partial \mathbf{f}}{\partial \mathbf{u}} = B \int_{-1}^1 \mathbf{N}^T \frac{\partial \mathbf{T}}{\partial \mathbf{v}} \frac{\partial \mathbf{v}}{\partial \mathbf{u}} |\mathbf{J}| d\eta = B \int_{-1}^1 \mathbf{N}^T \frac{\partial \mathbf{T}}{\partial \mathbf{v}} \mathbf{N} d\eta. \quad (3A.9)$$

The link between the traction and the separation is introduced through the cohesive law. A trapezoidal law is used in the present case. Figure 3A.3 shows the evolution of the traction σ depending on the adimensional separation parameter λ given by

$$\lambda = \sqrt{\left(\frac{\delta^N}{\delta_c^N}\right)^2 + \left(\frac{\delta^T}{\delta_c^T}\right)^2}, \quad (3A.10)$$

with δ^N and δ^T being the current normal and tangential separations while δ_c^N and δ_c^T are their critical values above which no traction is exerted anymore and the cohesive element breaks.

□ Issues with cohesive elements

Some preliminary simulations have been run following those performed by Hutchinson and Tvergaard [94] by considering material properties of polymers. The yields stress σ_0 is equal to 56 MPa and the σ_0/E ratio is set to 0.019. The cohesive energy G_c is set to 100 J/m². The cohesive strength is varied. These simulations showed that with extensive plastic deformation a compromise must be found between:

1. The critical opening displacement must be lower than the half of the length of the cohesive element otherwise the first element infinitely opens at the side of the crack tip and does not open at its other edge.
2. A sufficient number of elements (3 to 5) must be found in the fracture process zone that is given by $l_s = \Gamma_c E / 3\pi(1 - \nu^2)\sigma_c^2$ if there is plasticity
3. The plastic zone must be sufficiently discretized

The first rule imposes a maximum length to the cohesive length l_{coh} while the others impose a minimum length. The evolution of the maximum cohesive element length following these three rules is shown in Figure 3A.4. Ideally, the cohesive element length must be lower than indicated by the blue and red curves corresponding the sufficient discretization of the plastic zone and the process zone. It can be observed that if $l_{coh} = 5\delta_c$ or $l_{coh} = 10\delta_c$, for σ_c/σ_0 , the second and third rules are not satisfied.

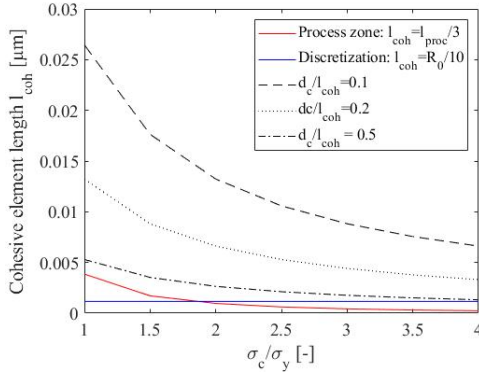


Figure 3A.4 Variation of the cohesive element length with the cohesive strength for the three rules allowing to determine l_{coh} .

To circumvent this issue, we investigated a scaling of the material properties by keeping the plastic zone size R_0 constant. Hence, the Young's modulus is increased by a factor X and the Γ_c is divided by X . Simulations with values of $X=5, 7$ and 10 have been run. It is observed in Figure 3A.5 that X has no influence on the results. This agrees with the results Hutchinson and Tvergaard [94] who showed that the ratio σ_0/E has only a limited influence on the results. Hence, this strategy has been kept for the simula-

tions of the interphase.

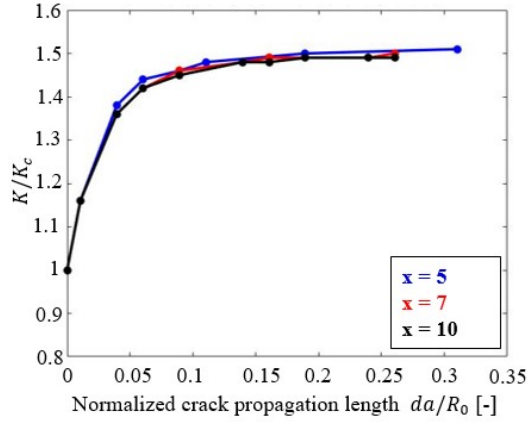


Figure 3A.5 Influence of the scaling factor x for G_c and E on the fracture toughness in terms of stress intensity factor. The scaling factor is set to 5, 7 and 10 and $l_{coh} = R_0/20$.

Results

Before modelling SENT samples with the interphase, SENT with homogeneous materials have been modelled using the transition properties determined in the current work and in the work of Voleppe et al. [12]. Owing to long computation times (> 400 hours), the steady-state fracture toughness G^{SS} is taken when the crack has propagated over a distance equal to $0.1R_0$, R_0 being the plastic zone size of the investigated material. It can be observed that as reported by Depinoy et al. [178], the evolution of the fracture toughness with the cohesive strength exhibits a sigmoidal shape (Figure 3A.6a). For a given range of σ_c , G^{SS} increases (toughening regime) while for high cohesive strength, it is almost constant (saturation regime). For a given σ_c/σ_y^u ratio, G^{SS} tends to be slightly higher with the properties for the 15 wt% PEI blend of the present work compared to the one of the work of Voleppe et al. [12]. However, this difference is not significant. The results are in agreement with those of Depinoy et al. [178].

As first case for SENT with an interphase, a crack is inserted in the RTM6-rich zone at a distance of $65 \mu\text{m}$ from the transition zone. Figure 3A.6b shows that the properties of the transition zone do not influence the steady-

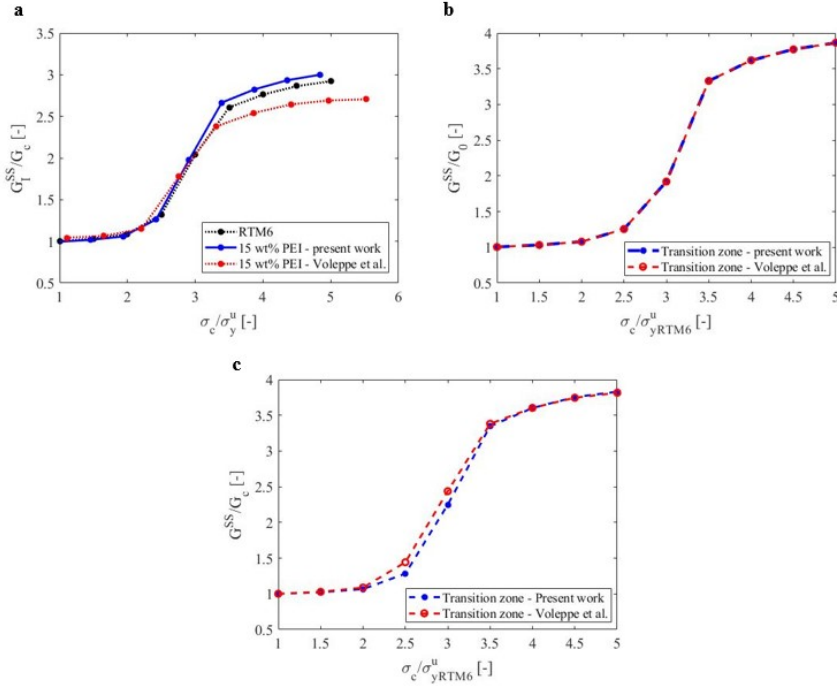


Figure 3A.6 Evolution of steady-state fracture toughness with cohesive strength for a) homogenous SENT made of RTM6 and 15 wt% PEI blends from the present work and the work of Voleppe et al. [12], b) for a crack propagating in the RTM6-rich zone and c) a crack propagating at the interface of the transition zone.

state fracture toughness for a given σ_c . G^{SS} is computed after the crack propagated over a distance equal to $0.25R_{0RTM6}$. Indeed, the plastic zone size of the RTM6 being about $12\ \mu\text{m}$, the plastic zone size developing ahead of the crack tip does not interact with the transition zone that is only elastically deformed. As for homogeneous blends, a sigmoidal shape is observed for the evolution of the steady-state fracture toughness.

Finally, the cohesive zone is inserted at the interface between the transition zone and the RTM6-rich zone. The evolution of G^{SS} evaluated after a propagation length of $0.25R_{0RTM6}$ with the cohesive strength is shown in (Figure 3A.6c). It can be observed that for a given σ_c , G^{SS} is not affected by the transition zone properties. Indeed, main of the plastic dissipation

takes place in the RTM6-rich zone and in the PEI-rich zone as the transition zone is thin compared to the plastic zone size. In addition, in homogeneous SENT, the difference between the two cases was already marginal. To conclude, the presence of a soft zone in the transition zone of Voleppe et al. [12] does not affect the interface fracture toughness compared to the current study in which no softening is observed.

3.7.2 Appendix B: Testing method for TP-epoxy interphases

In PEI-RTM6 interphases, it has been shown that when the crack initiates in the PEI-rich zone, it is deflected towards the epoxy-rich zone. However, it does not reach this zone due to the presence of a weak interface between the PEI-rich and epoxy-rich regions. By performing SENB tests, the initiation fracture toughness is measured. Hence, the G_{Ic} value measured by Voleppe et al. [12] corresponds to the one of the PEI-rich zone and not the one of the weak interface. It is thus important to design a test set-up allowing to measure the fracture toughness during crack propagation. We suggest to manufacture wedge-peel samples made of epoxy blocks with a PEI film inserted at the middle. The first step is thus to manufacture 4 mm thick samples with a width of 10 mm and a length of 150 mm. This requires to design a mould in which the PEI film can be inserted. For this purpose, a mould with two parts has been designed and these parts join each other with screws. The mould is shown in Figure 3B.1.

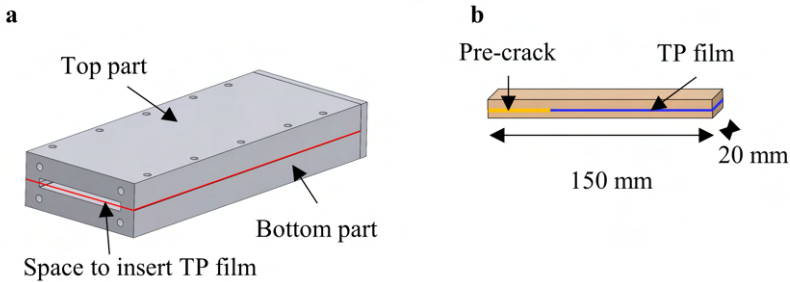


Figure 3B.1 a) Mould to manufacture wedge-peel samples and b) wedge-peel samples and its dimensions.

Once the samples are manufactured, wedge-peel tests can be performed by inserting a razor-blade between the Kapton and the PEI films. The tests are performed under a 3D microscope allowing to determine the beam deflections to determine the crack tip position. The distance between the crack

tip and the razor blade is referred to as the crack length a and the fracture toughness is given by

$$G_{Ic} = \frac{3Eh^3D^2}{16a^4(1-\nu^2)} \quad (3B.1)$$

with E being the beam Young's modulus, D the razor blade thickness, h the epoxy beam thickness and ν the Poisson's ratio of the beam material.

The idea is thus to measure the fracture toughness for different crack tip locations and then determine by the analysis of the fracture surfaces, the corresponding crack propagation locus. This combination will allow to determine the toughness of the interface if the crack propagates along it.

Such tests have not been performed as the manufacturing of the samples has been found to be challenging with leaks of resin taking place during curing. This probably requires to optimize the mould design.

4

Co-cured composite joints: selection of potential TP interlayers

Chapter 3 indicates that PEI co-cured composite joints manufactured by the RTM do not result in joints with superior toughness as expected. As other thermoplastics (TP) than PEI can interact with epoxy, this chapter investigates the potential candidates for thermoplastic interlayer material in co-cured composite joints. Hence, the focus is on the mode I fracture toughness of the joints and the main failure mechanisms as a first step towards a more in depth analysis of TP layers for co-curing. We do not cover the chemistry beyond the formation of the interphases and the failure mechanisms along the interphases as covered in Sections 3.3 and 3.4 for the PEI-epoxy interphases.

4.1 Introduction



This introduction covers literature results concerning co-cured composite joints and TP-epoxy interphases already presented in Section 2.2 but with more details.

Carbon fibre reinforced polymer composites (CFRPs) are broadly used in advanced structures such as in airplanes owing to their high strength and stiffness to weight ratios. Manufacturing these structures often requires assembling CFRPs together or with other materials. Primary load carrying structures are frequently joined through mechanical fasteners, altering the composites structural integrity [1]. Adhesive bonding offers more uniform stress distribution and additional weight saving. However, limited resistance to crack propagation and long sequential bonding processes constitute two barriers for a more intensive use of this technology [1].

To tackle these two issues, it has been suggested to use other materials than epoxies as adhesives, typically high-performance thermoplastics such as polyetherimide (PEI), polysulfone (PSU), polyethersulfone (PES) ... [15, 29–33]. These thermoplastics are inserted in the form of a film either at the middle of the composite lay-up [15, 29–33] or at the surface [35]. If added to the surface, the composite part can be joined to another composite part or to a metallic part via thermoplastic welding. Integrated manufacturing, also referred as co-curing and co-bonding, can be achieved by inserting the TP film at the mid-plane of the lay-up prior to composite curing. The use of thermoplastics as bonding material can thus improve the fracture toughness of joints and reduce work and time-intensity.

However, the integration of TP requires the creation of a tough interface between the composite and the thermoplastic film [5]. For this reason, thermoplastics exhibiting good interactions with epoxy have been used such as PEI, PES, PSU or PEEK. These polymers can create a semi-interpenetrating network when in contact with the epoxy leading to the development of a morphological gradient and some functional groups can even react with the epoxide groups as observed with the use of polyamide. One of the most investigated material is PEI for the manufacturing of co-cured composite joints [15, 29–32, 35]. Although, Quan and co-workers [15, 29–32] reported improved G_{Ic} compared to composite delamination when using autoclave

curing, in Chapter 3, it has been shown that PEI co-cured composite joints manufactured through resin transfer moulding suffer from limited fracture toughness ($< 500 \text{ J/m}^2$).

The goal of the present chapter is to investigate the manufacturing of co-cured composite joints with other thermoplastics than PEI. For this purpose, polyethersulfone (PES), polysulfone (PSU), polyamide 6 (PA6), polycarbonate (PC) and polymethylmethacrylate (PMMA) films are inserted in a composite joint. The mode I fracture toughness of these joints is investigated as well as the morphological gradient developing upon curing and the local properties through nanoindentation. This is a preliminary study which does not consider the chemical effects and the details of the failure mechanisms have not been deeply investigated. The idea is to identify potential candidates for composite joints co-curing through a TP layer.

4.2 Materials and methods

4.2.1 Materials

For the composite adherends, the HexFlow RTM6 resin and the G0926 Hex-Force carbon fibres fabrics are used. Then, six different thermoplastics are inserted: polyetherimide (PEI), polyethersulfone (PES), polysulfone (PSU), polycarbonate (PC), polyamide 6 (PA6) and polymethylmethacrylate (PMMA). They are inserted under the form of $500 \text{ }\mu\text{m}$ films supplied by GoodFellow. The PSU film is initially $900 \text{ }\mu\text{m}$ thick but the thickness is reduced using a hot static press and a $500 \text{ }\mu\text{m}$ thick spacer. The properties of these materials are given in Table 4.1.

4.2.2 Samples manufacturing

Epoxy-TP interphases

The specimens with a TP-epoxy interphase are processed by inserting the TP films in glass tubes. The tubes are filled with RTM6 resin before degassing it at 90°C for one hour. Curing takes place following the C160 curing cycle (Figure 4.1). The temperature is first increased at a rate of 2°C mm/min up to 160°C before maintaining it for 90 minutes in order to promote the formation of the morphological gradient [12, 68]. The temperature is then increased to 180°C at a rate of 2°C mm/min before keeping it constant for two hours.

4 | Co-cured composite joints: selection of potential TP interlayers

Table 4.1 Properties of the thermoplastic materials used for composite joints co-curing. A stands for amorphous while S-C means semi-crystalline. Values under brackets are the values obtained in Chapter 3 while other values come from EduPack [179].

Material	T _g [°C]	E [GPa]	σ_y [MPa]	G _{Ic} [kJ/m ²]	Amorphous ?
RTM6	220	2.8	100 (127)	0.08	Thermoset
PEI	215	3 (3.4)	80 (140)	1.5-5	A
PES	220	2.5	50-75	2	A
PSU	190	2.7	75	2-7	A
PMMA	100	2.4-3.3	54-72	0.2-0.9	A
PC	150	2.4	60	2	S-C
PA6	50-60	1.5-2	50	6-8	S-C

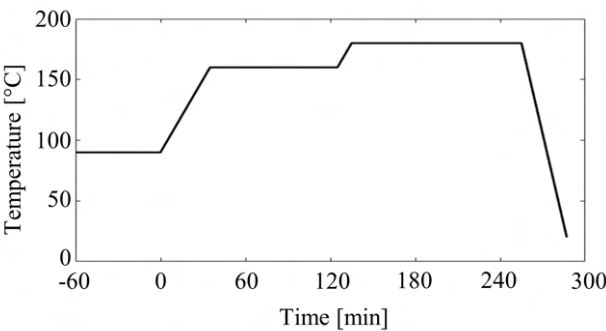


Figure 4.1 C160 curing cycle used for processing of the TP-epoxy inter-phases and the co-cured composite joints.

Co-cured composite joints

The TP co-cured composite joints are manufactured through resin transfer moulding (RTM) following the process illustrated in Figure 3.2. The first five G0926 carbon plies are first laid down in the RTM mould. The TP film is then inserted and two 70 mm long, and 30 μ m thick Kapton films (Dupont) are stacked on the top of it to introduce a pre-crack. The five last plies are added. The RTM mould is then closed using a 8 bars press. The RTM6 resin is then heated in a piston up to 90°C before being injected at the same temperature in the mould until reaching a pressure of 6 bars. The composite is finally cured following the C160 curing cycle shown in Figure 4.1.

4.2.3 Mechanical testing

Nanoindentation along interphase

Nanoindentation tests are performed along TP-epoxy interphases using a G200 nanoindenter from Agilent Technologies with a pyramidal Berkovich diamond tip. The samples are polished prior to testing. 1200 and 4000 MD-Piano disks from Struers are used before finishing with a 0.3 μm alumina suspension (AP-D Suspension 0.3 μm , Struers Inc.). The Continuous Stiffness Measurement (CSM) method is used relying on the Oliver and Pharr [170] method to extract the elastic modulus and the hardness. Drift corrections are not considered as, for polymers, this correction is strongly affected by viscoelastic effects [171]. Indents are performed along the interphase with repetitions over 3 different zones for each sample. The penetration depth is set to 1 μm .

DCB testing

The DCB samples have been machined from the cured panels with a length of 200 mm and a width of 20 mm. The 70 mm long Kapton film inserted to create the pre-crack induces an initial crack length of 50 mm. The total height of the DCB is about 4.4 mm. The DCB tests are performed following the ISO 25217 standard [172] on a Zwick/Roell machine equipped with a 50 kN loadcell, at constant cross-head speed of 1 mm/min. The machine is located in a ventilated room at constant temperature and relative humidity content about 23°C and 50%, respectively. Prior to testing, the edges of the samples are graduated to measure the crack length, allowing to track the crack by taking one picture every 3 seconds with a Canon EOS1400D camera. G_{Ic} is then computed following the single beam theory [172]. For each condition, 4 samples are tested.

4.2.4 Microscopy

A VHX optical microscope from Keyence and an Ultra 55 Field-Emission SEM (Zeiss, Germany) are used to characterize the fracture surfaces of the DCB samples and to characterize the morphologies of the TP-epoxy interphases. For SEM imaging, the samples are coated with a 1 μm thick gold layer and imaged at 5 keV. For samples containing an interphase without fibres, they have been broken perpendicular to the film using a razor blade.

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4.2.5 Infrared spectroscopy

The chemical gradient of PMMA-epoxy and PC-epoxy interphases is qualitatively investigated with infrared (IR) spectroscopy. Spectra are acquired every 5 μm along a line extending over the interphase. The experimental Spectra are measured using a Nicolet iN10 MX infrared imaging microscopy, configured with the following detectors: a room temperature DTGS, a single point MCT-A and a linear array MCT-A detector. Each sample is placed on the sample post of an imaging ATR microscope accessory and raised to make contact with the germanium ATR crystal for measurement (pressure of 25 bar). 64 scans are taken over 22 seconds and the spectral resolution is set to 4 cm^{-1} . The qualitative profile is determined based on peak amplitude normalization. For the PMMA interphase, the peaks at 1725 (ester function of PMMA) and 1551 mm^{-1} (aromatic group of epoxy) wavelength are used. For the PC-interphase, the peaks at 1771 (carbonate function of PC) and 1613 mm^{-1} (aromatic group of epoxy) wavelength are used.

4.3 Results and discussion

4.3.1 Epoxy-polyetherimide interphase

The results regarding the epoxy-PEI interphase have already been presented in Chapter 3. Upon curing a morphological gradient develops with a PEI-rich zone and an epoxy-rich zone. The G_{Ic} of the joints is shown again in Figure 4.2. The PEI film thickness influences the results with the higher toughness being attained with the 500 μm thick film and which is equal to $370 \pm 47 \text{ J/m}^2$. In PEI co-cured composite joints, the crack propagates along the carbon fibre located in the PEI-rich zone. However, due to the architecture of the carbon fibres fabrics, resin pockets are locally found where the full morphological gradient develops. At these locations, the crack propagates along the weak interface between the PEI-rich and epoxy-rich zones. Finally, for the 50 μm thick film, the crack is also able to propagate through the tough PEI film.

4.3.2 Epoxy-polyethersulfone interphase

Upon curing, a morphological gradient develops in PES-epoxy systems with a PES-rich zone and an epoxy-rich zone as shown in Figure 4.3a. The 10 μm long PES-rich zone is characterized by a nodular morphology while

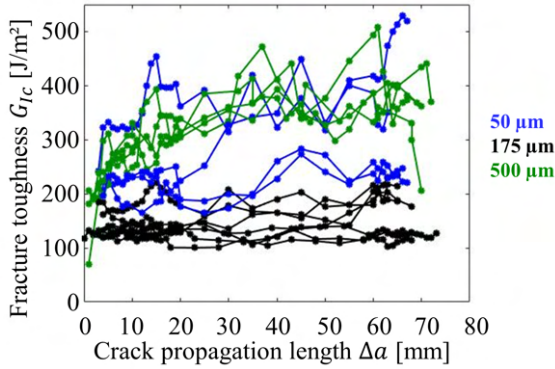


Figure 4.2 R-curves for the co-cured composite joints with 50 μm (blue), a 175 μm (black) and a 500 μm thick PEI films.

a sea-island morphology is observed in the epoxy-rich zone. This morphological gradient is identical to the one reported by Voleppe et al. [68] for the same system. A sharp transition is observed between the two zones and according to Voleppe et al. [68], it is at the origin of the low toughness of this system. Indeed, as for PEI and epoxy, PES and epoxy functional groups do not react with each other to create strong bonding.

Figure 4.3b shows the evolution of the hardness along the interphase. As the PES-rich zone is quite thin, a progressive increase of the hardness from the PES film to the epoxy-rich zone is not found as the indents in the PES-rich zone are affected by their surrounding environments. If a tough interface could be formed, PES could potentially be an interesting material to be used as dissipative layer [79, 97] owing to its lower hardness with respect to RTM6.

Figure 4.4a shows the R-curves extracted from the DCB tests performed on the PES co-cured composite joints. These joints are characterized by a G_{Ic} equal to $70 \pm 11 \text{ J/m}^2$. This is a low value. Figure 4.4b shows the fracture surface which is completely featureless. The crack propagates adhesively at the interface between the PES film and the composite. Figure 4.4c shows that interdiffusion takes place during co-curing. Based on the results of Voleppe [69], the main hypothesis is that the crack propagates at the interface between the PES- and epoxy-rich zones. This again highlights the importance of (1) being able to correctly determine the interface G_{Ic} and (2) to force the fibres to be in the TP-rich zone as already explained in Chapter

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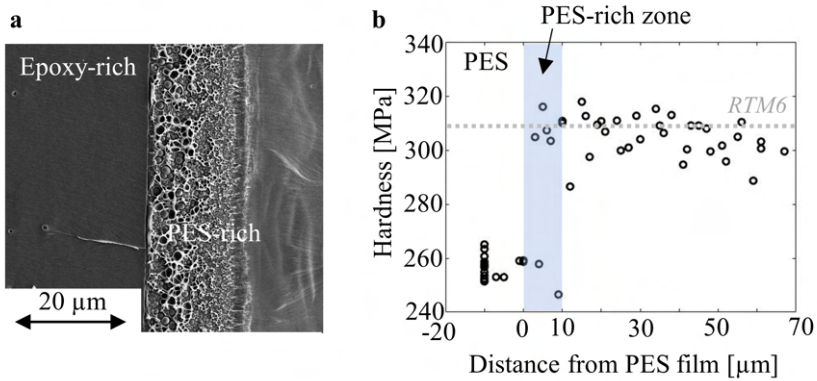


Figure 4.3 a) PES-epoxy interphase and b) evolution of hardness along the interphase.

3 for PEI-epoxy interphases to avoid the development of the PES-epoxy weak interface between the PES film and the carbon fibres.

4.3.3 Epoxy-polysulfone interphase

The PSU-epoxy interphase is shown in Figure 4.5. A PSU-rich zone exhibiting a nodular morphology is observed close to the PSU film (Figure 4.5c) while away from the film, the epoxy-rich zone is characterized by a sea-island morphology (Figure 4.5d), as reported by Min and Kim [60]. Between these two zones, a transition zone develops with a gradual change in morphology as shown in Figure 4.5b which differs from the transition zone reported by Min and Kim [60]. This zone exhibits a co-continuous morphology combining the nodular and the sea-island morphologies. Opposite to PEI-epoxy and PES-epoxy interphases, the transition between the TP-rich and the epoxy-rich zone is smooth. In addition, the interphase is thicker than the one observed in PEI- and PES-epoxy systems as it has thickness about 100 μm.

Figure 4.6 shows the evolution of the hardness along the interphase. An increase of the hardness is found along the interphase when approaching the epoxy-rich zone. A similar behaviour is expected for the PES-epoxy interphase although it is difficult to observe it due to the thin TP-rich zone while for the PEI-epoxy interphases, the opposite evolution is found. As PES, it is a good candidate to create a dissipative layer.

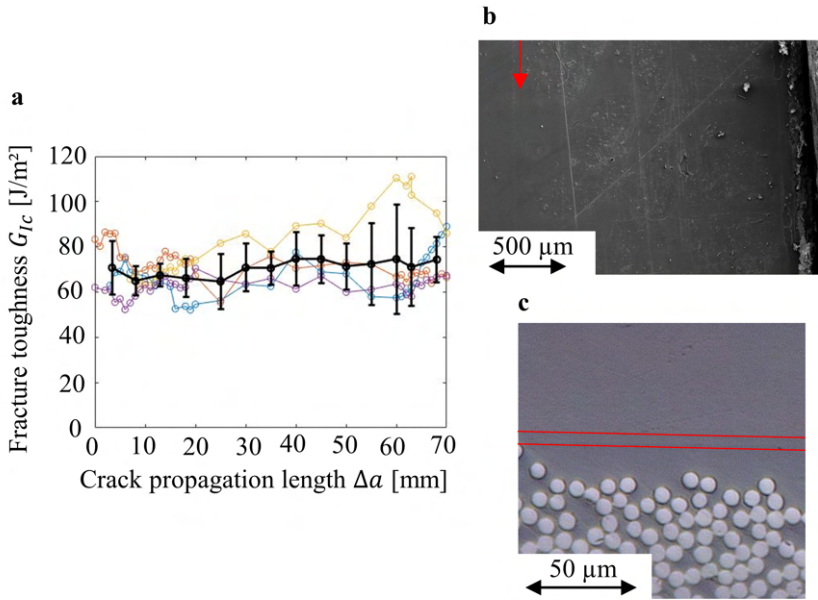


Figure 4.4 a) R-curve for the PES co-cured composite joints (black curve = average). b) SEM micrograph of the fracture surface and c) optical micrograph of the joint cross-section highlighting the interdiffusion zone.

The R-curves for the PSU co-cured joints are shown in Figure 4.7a. The joints are characterized by a toughness equal to $203 \pm 60 J/m^2$. Looking at the fracture surfaces reveal crack propagation between the PSU film and the composite. On the SEM micrographs, delamination along the carbon fibres laying in the PSU-rich zone is found (Figure 4.7b). However, many featureless spots similar to those appearing in PEI co-cured composite joints are observed as shown in Figure 4.7b-c. These spots presumably correspond to zones without any interdiffusion. In these zones, the crack propagates along the interface between the film and the resin. This interface is believed to be brittle due to the lack of interdiffusion, resulting in joints characterized by a low G_{Ic} .

4.3.4 Epoxy-polyamide interphase

As shown in Figure 4.8a, no morphological gradient develops upon curing in PA-epoxy based systems. When breaking the samples perpendicular

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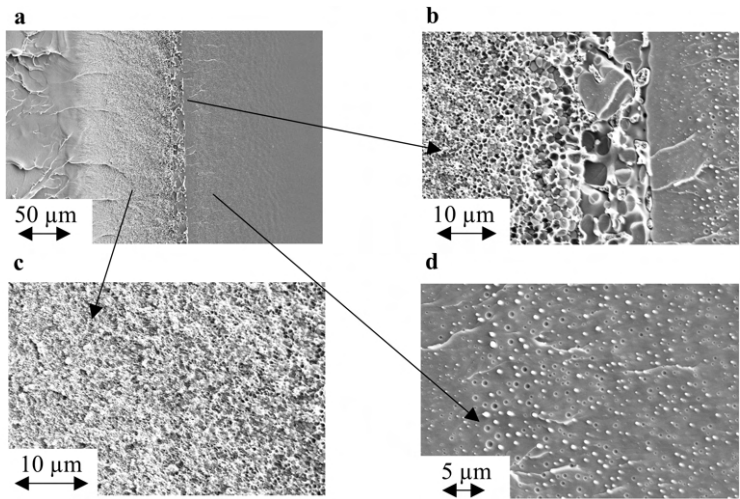


Figure 4.5 SEM micrographs of a) the PSU-epoxy interphase with b) the transition zone, c) the PSU-rich zone and d) the epoxy-rich zone.

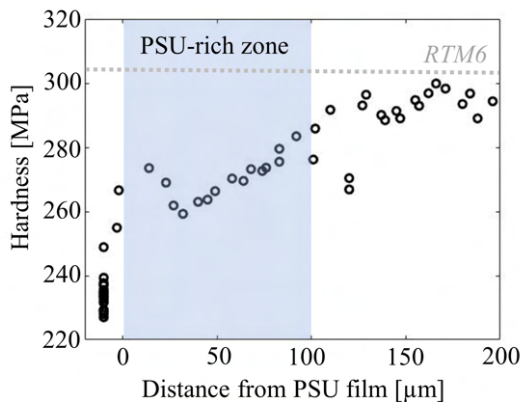


Figure 4.6 Evolution of the hardness along the PSU-epoxy interphase.

to the film, the crack is directly deflected along the PA-epoxy interface. This indicates the presence of a brittle interface. The presence of this weak interface is confirmed by the DCB tests as G_{Ic} is found to be equal to the very low value of $26 \pm 6 \text{ J/m}^2$. In addition, adhesive failure can be observed between PA and the composite. Two perfectly smooth, featureless fracture

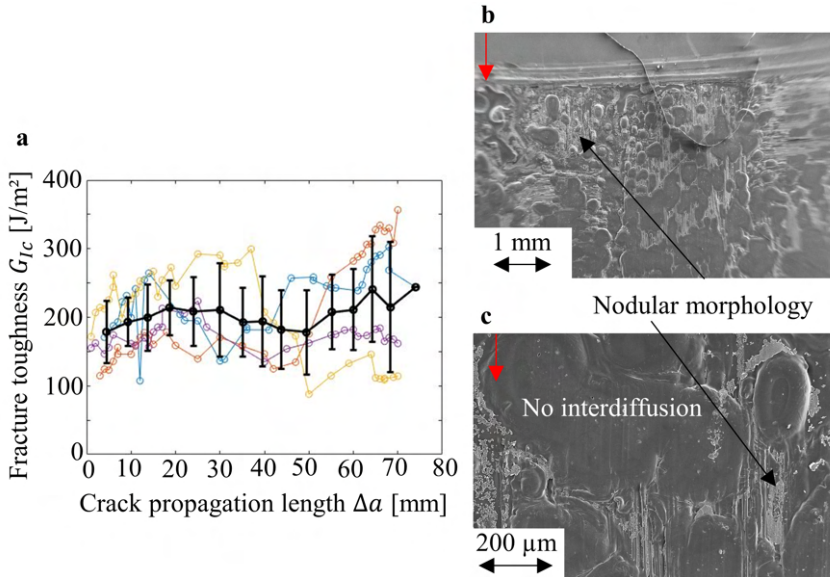


Figure 4.7 a) R-curve for the PSU co-cured composite joints (black curve = average). b) SEM micrograph of the fracture surface and c) zoom in a zone without interdiffusion.

surfaces are obtained as shown in Figure 4.8b and SEM micrographs are completely featureless.

Additional tests have been performed with sandblasted PA films in order to improve the toughness of PA co-cured composite joints. Because of the flexibility of the film, it is difficult to obtain an homogeneous roughness over the whole surface film. The toughness of these joints is equal to $120 \pm 34 \text{ J/m}^2$. This represents an increase of the toughness by a factor 5 compared to the untreated PA film. Adhesive failure still takes place but opposite to the untreated film, the fracture surface exhibits some roughness. The roughness induced by sandblasting can thus be responsible for the improvement of the toughness by inducing some mechanical interlocking.

PA-epoxy blends are obtained by dissolving the two species in an alcohol-based solvent [55, 56]. Dissolution favours the interactions between PA and epoxy. Opposite, Kim et al. [57] mixed Nylon powder with epoxy and found that the mixing temperature must be at least above 215°C to

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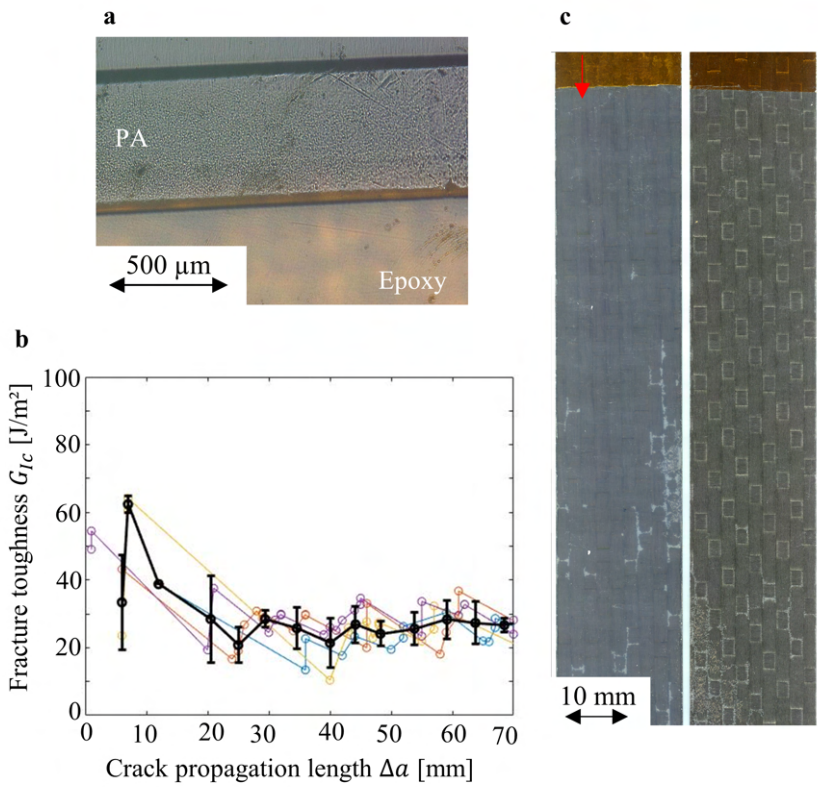


Figure 4.8 a) Optical micrograph of the PA-epoxy interface. b) R-curve for the PA co-cured composite joints (black curve = average). c) Optical micrograph of the fracture surface of one joint.

induce a reaction between the two polymers. In the present case, the curing temperature (160°C) is thus too low to favour chemical interactions and, consequently, to achieve bonding.

4.3.5 Epoxy-polycarbonate interphase

Upon curing, epoxy and polycarbonate interact with each other resulting in the formation of a chemical gradient with a very fine morphology similar to the one observed by Lee et al. [72] as shown in Figure 4.10. This corresponds to the phase-inverted morphology found for the PEI, PSU and PES-epoxy interphases. Interdiffusion is confirmed by the FTIR analysis

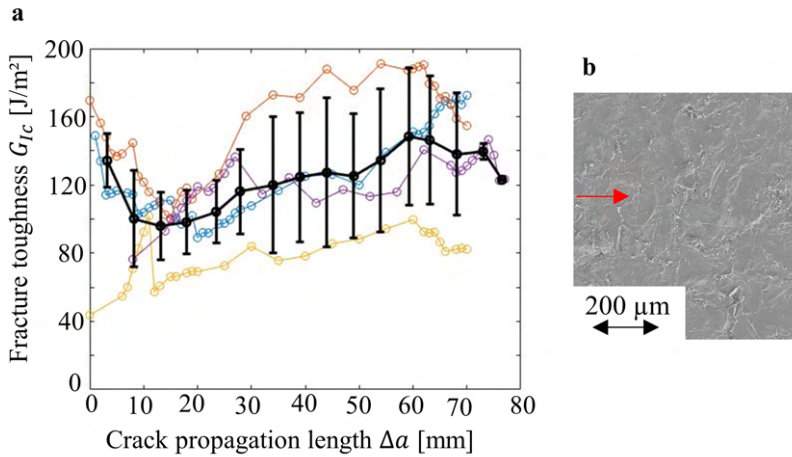


Figure 4.9 a) R-curve for sandblasted PA co-cured composite joints (black curve = average). b) SEM micrograph of the fracture surface.

as shown in Figure 4.10d, confirming that interdiffusion took place during curing. However, simultaneously, large voids form at the interface due to the reaction between epoxy and PC.

The evolution of the hardness along the interphase could not be determined as the indents could not be found over the specimen surface upon testing. Although, the PC hardness is around 200 MPa, lower than the one of the RTM6 which is about 310 MPa. Owing to this mismatch, PC could act as a dissipative layer.

The G_{Ic} of PC co-cured composite joints (Figure 4.11a) is equal to 804 ± 511 J/m². A stick-slip behaviour takes place during crack propagation with toughness values up to 2000 J/m². As shown in Figure 4.11b, the fracture surfaces reveal crack propagation through the PC film. The local maxima found in Figure 4.11a corresponds to the locations of crack deflection towards the opposite interface. During curing, voids nucleate at both interfaces with the composite. It is believed that these voids are at the origin of crack deflection. No fibres are observed on the fracture surfaces when the cracks propagate along the PC-composite interface. The crack seems to be propagating in the PC-epoxy interphase. Indeed, in the absence of an interphase, the crack would propagate along the fibres as in composites produced without TP film.

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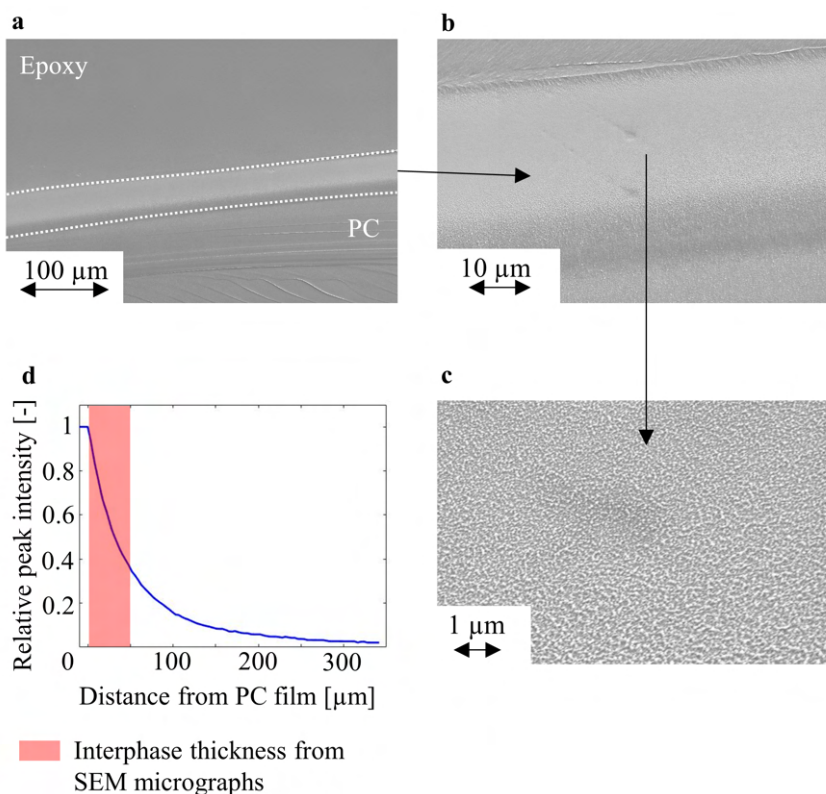


Figure 4.10 SEM micrographs of the PC-epoxy interphase at different magnifications showing a) the interphase region, b) the PC-rich zone of the interphase, c) the morphology of the PC-rich zone and d) FTIR profile for the carbonate group.

4.3.6 Epoxy-polymethylmethacrylate

Regarding the PMMA-epoxy system, epoxy and PMMA interact with each other during curing, as already reported by Zweifel et al. [37]. The morphological gradient cannot be accurately distinguished on the micrographs. Nevertheless, changes on the fracture surface features are found on the fracture surface indicating the potential presence of a morphological gradient. As highlighted in Figure 4.12a-b, a rougher zone is observed over 40 μm. By zooming in this rough zone (Figure 4.12c), some small features can be distinguished. It is impossible to accurately separate the two phases (PMMA and epoxy). However, as this zone is close to the PMMA film

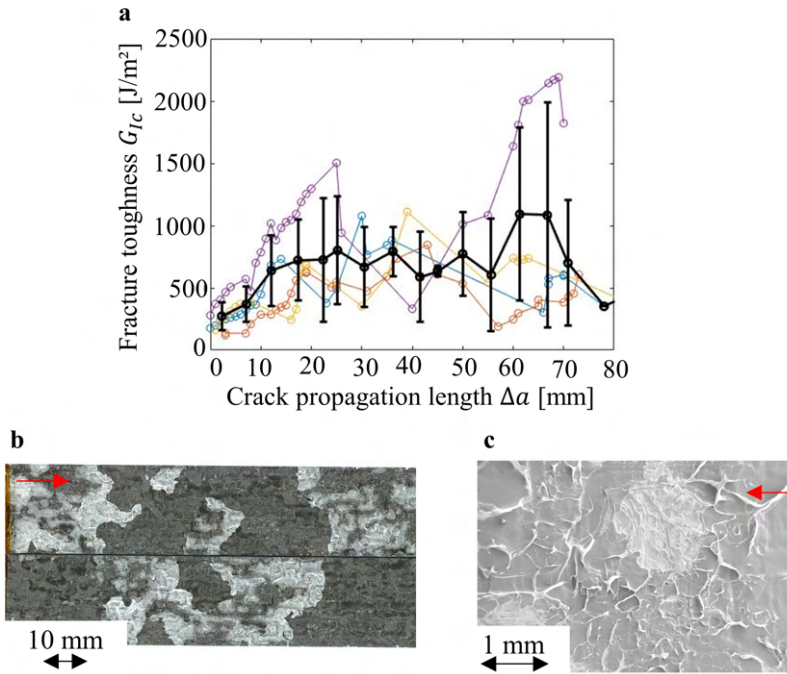


Figure 4.11 a) R-curve for the PC co-cured composite joints. b) optical micrograph of the fracture surface and c) SEM micrograph of the fracture surface

and that the fracture surface features change away from it, it probably corresponds to the PMMA-rich region of the morphological gradient which should be tougher than the epoxy-rich region. Interdiffusion is confirmed by the FTIR analysis as shown in Figure 4.12d and an interphase thickness of 40 μm is found, in agreement with the SEM micrographs.

Figure 4.13 shows that the hardness increases over 40 μm away from the PMMA film, in agreement with the changes in morphology. This confirms the formation of an interphase.

PMMA co-cured composite joints are characterized by a toughness equal to $670 \pm 138 \text{ J/m}^2$ (Figure 4.14a). The crack only propagates between the PMMA film and the composite, as shown in Figure 4.14b. However, almost no imprints of fibres appear on the fracture surfaces as shown in

4 | Co-cured composite joints: selection of potential TP interlayers

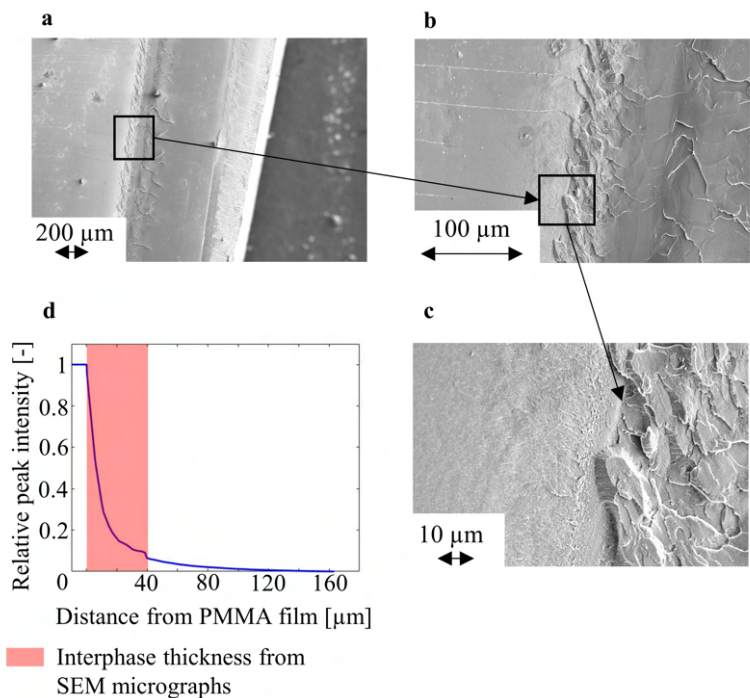


Figure 4.12 a)-c) SEM micrographs of the PMMA-epoxy interphase with different magnifications such that b)-c) show the rough zone corresponding to the interphase. d) FTIR profile for PMMA along the interphase.

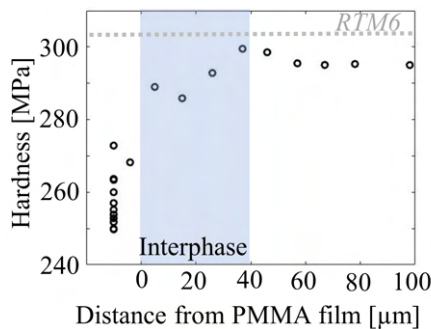


Figure 4.13 Evolution of the hardness along the PMMA-epoxy interphase.

Figure 4.14d-e. The cracks seems thus to propagate either in the PMMA film close to the interface with the composite or in the interdiffusion zone. However, microscopic analyses could not accurately reveal the crack propagation locus. At crack initiation (Figure 4.14c), different features resembling to fingerprints are observed but not during further crack propagation. These features have not been observed in PMMA-epoxy interphases and are only found near the initiation site related to the PI insert.

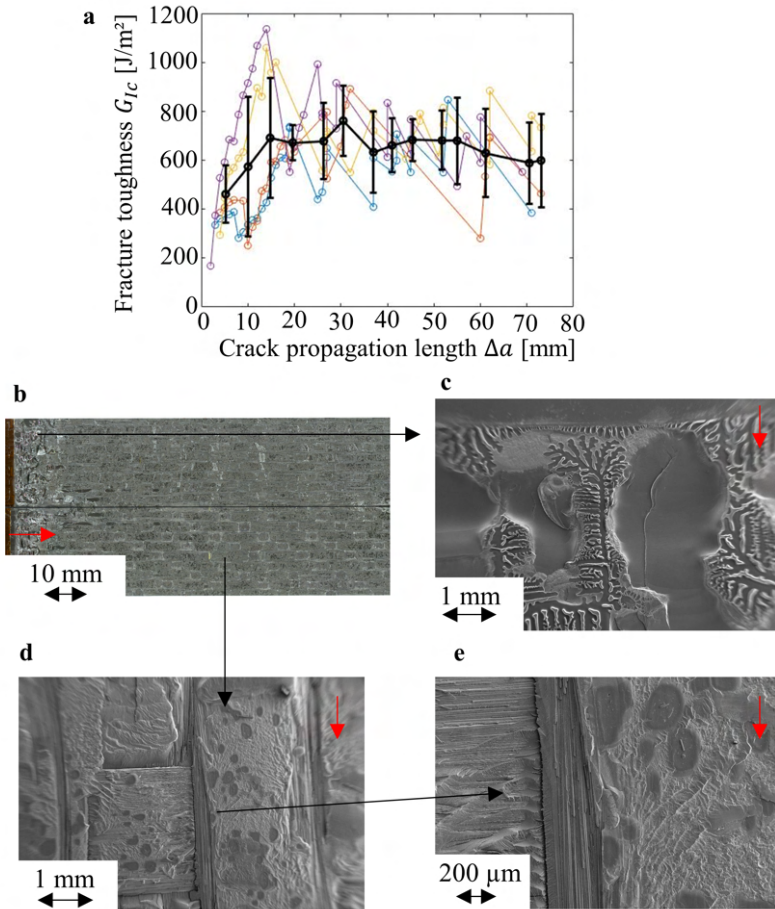


Figure 4.14 a) R-curve for the PMMA co-cured composite joints. b) optical micrograph of the fracture surface with the SEM micrographs of c) the initiation zone and d-e) steady-state propagation region.

4.4 Conclusion and perspectives

Figure 4.15a compares the toughness of the different co-cured joints with the associated failure mechanisms (Figure 4.15b) while Figure 4.15c shows the variation of the hardness along each interphases. Some conclusions can be drawn from these results:

1. Interdiffusion favours adhesion. PA joints with no interdiffusion show very poor toughness.
2. TP film roughening improves the joint toughness (improved G_{Ic} of PA joints by sandblasting).
3. If there is interdiffusion, the presence of a weak interface between the TP-rich and the epoxy-rich zones between the TP film and the fibres must be avoided (low G_{Ic} of PES joints). The interphase thickness must be at least such that the TP-rich zone reaches the fibres, but it does not mean that it must be as thick as possible. Indeed, depending on the thickness of the interphase, the morphological features can have different size resulting in different toughening effects as observed by Farooq et al. [49]. The optimum thickness interphase is thus a parameter to be investigated that depends for example on the curing cycle.
4. Crack deflection through the TP film is favourable, but is random and results in large scatter.
5. If there is interdiffusion in the region including fibres and the crack does not deflect, a softer TP layer may be favorable to improve G_{Ic} by increasing the plastic dissipation (PSU and PMMA joints).
6. PMMA joints result in the highest G_{Ic} for the tested conditions.

Based on these conclusions, the two best systems are the PEI (500 μm) joints and the PMMA joints which compete with composite delamination. However, PMMA is characterized by a lower glass transition temperature and has thus a lower thermal resistance preventing its use in high temperature applications compared to PEI. However, PMMA co-cured joints could find some applications such as in windmills as Arkema already manufactures PMMA windturbine blades [180].

In this work, only an exhaustive list of TP materials has been investigated

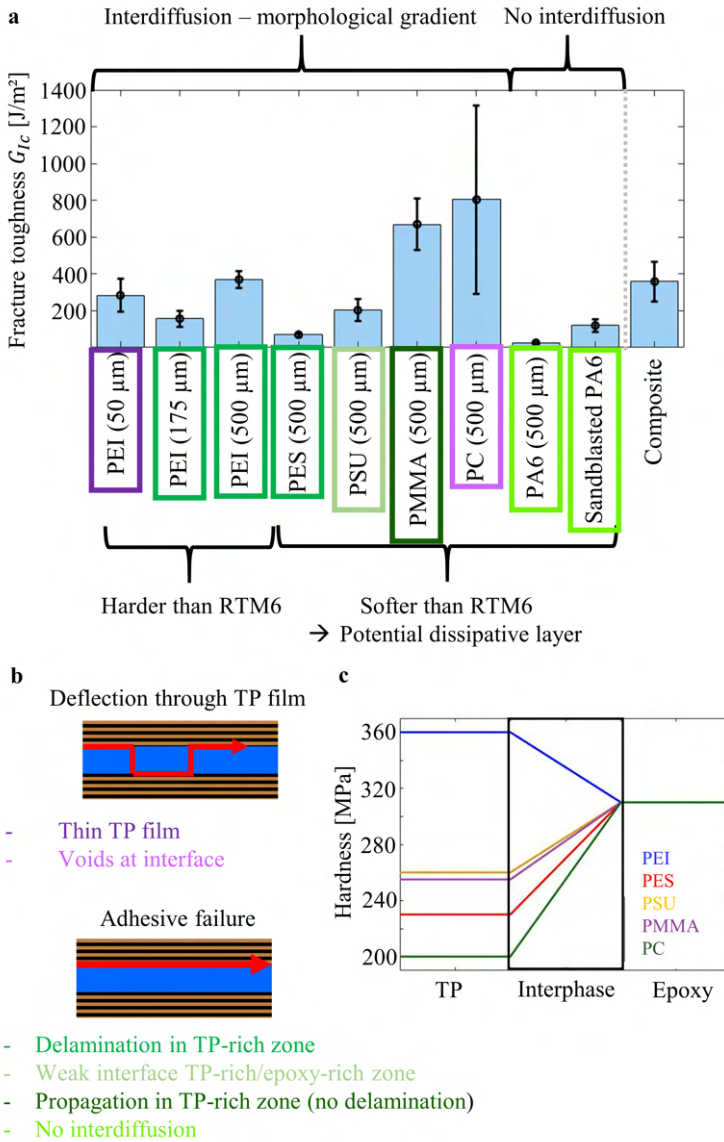


Figure 4.15 Summary of the results for the different TP co-cured composite joints: a) fracture toughness, b) failure mechanisms and c) schematic evolution of the hardness along the interphase.

4 | Co-cured composite joints: selection of potential TP interlayers

as interlayer for composite joints co-curing. This already gives some indications such as the toughest joints will be obtained if interdiffusion between the TP and the composite resin occurs, the morphological gradient must be thick enough to develop within the composite and that G_{Ic} can be improved by roughening the TP film. However, to determine the best system, additional research is required.

1. Influence of the curing cycle: The development of a morphological gradient depends on many parameters such as the curing cycle, the resin viscosity or the TP molecular weight [44, 51, 53, 69]. Further work should thus focus on these aspects to determine the processing conditions resulting in the toughest joints.
2. Influence of the degree of crystallinity for semi-crystalline TP: PC and PA6 are both semi-crystalline polymers. Depending on the curing cycle, the degree of crystallinity might change. For both bulk thermoplastics and thermoplastic composites, a decrease of the fracture toughness is reported as the degree of crystallinity increases [181–183].
3. Influence on other joint properties: Not only mode I fracture toughness is important in joints but also lap shear strength and stiffness as joints are mainly loaded under shear. Hence, performing lap-shear tests is also important to compare the joints and evaluate their mechanical performances. In addition, thermal resistance, resistance to solvents and other conditions (UV, humidity ...) must be investigated.
4. Influence of film thickness: The joint properties are also influenced by the remaining film thickness (depending on initial film thickness for given processing conditions) as it is for classical adhesive joints. The thicker the joint up to a thickness about twice the plastic zone size, the higher the toughness if plastic dissipation takes place in the joint. Hence, by varying the film thickness, another system than the PMMA one could be the best.
5. Influence of the manufacturing process: Locally, no interdiffusion takes place (with PSU and PEI). Consequently, the presence of these defects should be understood to manufacture reliable joints. For the RTM process, the influence of some parameters could be investigated such as the injection speed or the pressure in the mould. In addition,

several manufacturing processes could be envisaged such as autoclave curing using pre-pregs or infusion. Changing of manufacturing process could lead to a change in materials such as the epoxy resin or the initial degree of resin curing and these parameters can influence the formation of an interphase and thus the joint properties.

To conclude, a large amount of work is still needed to optimize co-cured composite joints using a TP interlayer which is a topic that started to gain interest during the last five years [15, 29–32, 35–39].

PART II

**Toughening strategies for
co-cured joints**

5

Crack bridging in co-cured joints



This chapter is based on an article that will be submitted soon: Charline van Innis, Michal K. Budzik and Thomas Pardoën, *Crack bridging in co-cured joint*.

5.1 Introduction



This introduction covers literature results concerning co-cured composite joints and extrinsic toughening strategies, as already presented in Sections 2.2 and 2.3 with more details.

Carbon fibre reinforced polymer composites (CFRPs) are broadly used in advanced structures such as airplanes, wind turbine blades or marine structures, owing to a high strength and stiffness to weight ratios. The man-

ufacturing of these structures requires the joining of CFRP components. Mechanical fastening is one option but drilling holes in the adherends lowers the structural integrity due to the generation of stress concentrations [1]. Adhesive bonding offers more homogeneous stress transfer and lightweight. However, adhesives often suffer from limited mode I toughness G_{Ic} and cost and time intensive bonding process [1], preventing a generalized use of this joining technique in advanced applications such as in airplanes.

Co-curing is an alternative to secondary bonding, with a decrease of the manufacturing intensity. Co-curing involves simultaneous curing and bonding of the composite parts. In secondary bonding, the adherends must first be cured apart, the surfaces must be treated and the CFRPs adherends can finally be bonded together. Co-curing is only possible when no resin has to be injected during the process to prevent the adhesive to flow out. Hence, co-curing is possible using pre-pregs, but not with resin transfer moulding process (RTM). On the one hand, co-cured composite joints are known to be characterized by a lower mode I fracture toughness compared to secondary bonded joints due to the development of defects at the adhesive/-composite interface [23–25]. On the other hand, co-curing results in improved geometrical accuracy [22] of the components and are less affected by hygrothermal ageing [26].

In order to solve the issue of lower G_{Ic} , Quan and co-workers [30, 31] suggested to replace the adhesive by a thermoplastic film compatible with the composite resin [5]. This led Quan and co-workers to insert polyetherimide (PEI) and UV-treated polyetheretherketone (PEEK) films between pre-pregs made of the 8552 epoxy resin. Regarding the PEI co-cured joints, at room temperature, an improved mode I toughness was reported compared to the composite toughness during delamination, but lower than the G_{Ic} of the co-cured adhesive joint. The PEI film results in higher mode II fracture toughness (G_{IIc}) and lap-shear strength compared to composite delamination with an adhesive-based co-cured joint. In addition, increasing the PEI film thickness improves the mode I, mode II fracture toughness and lap-shear strength with an optimum thickness around 200 μm . The good adherence between the composites and the PEI results from the interdiffusion of PEI and epoxy during curing. Other candidates for co-curing include among others phenoxy [33], polyethersulfone (PES) [33, 34], and polysulfone (PSU) [60]. Recently, we co-cured composite joints using PEI films of varying thicknesses (see Supplementary Material 1). However,

these co-cured joints also suffer from low mode I fracture toughness when compared to the composite delamination toughness, requiring the exploration of effective toughening strategies.

Several toughening strategies have been investigated for adhesives. The first consists in enhancing the plastic energy dissipation in the joint by increasing the adhesive thickness [13, 93, 99, 102]. Plastic energy dissipation can also be improved by inserting a softer layer close to the crack propagation plane in the assembly [79, 97]. Crack deflection by architecturing the adherends has been proven effective to raise the mode I fracture toughness through forcing local mode-mixity at the crack tip [80, 106, 107, 119]. A crack can also be deflected from the propagation plane by adding stiff particles or cavities. If a cavity is added, it can trap the crack and if large enough, it blunts the crack, delaying its reinitiation and increasing the effective G_{Ic} [80, 122]. It has even been suggested to create arrays of cavities referred as pillar interfaces [141]. Following this idea, more generally, the bondline can be structured using metamaterials concepts [84, 142] to trigger complex failure modes. Elastic heterogeneities [138, 139] are also known to influence the effective G_{Ic} and can be achieved by architecturing the adherends [82]. Finally, one mechanism that has proven very efficient is crack bridging. Bridging is a phenomenon commonly taking place in composites due to fibres attached to both crack faces behind the crack tip. It can also be triggered by adding Z-pins [127, 128], by inserting a carrier in the adhesive [80, 81, 132] or by changing the local adherence at both interfaces [80, 135]. In composite parts, thermoplastic veils [124, 130] are also often introduced at interplies to trigger bridging and improve the delamination toughness. The areal weight of such veils is known to have an influence on G_{Ic} with an existing optimum areal weight. The resulting toughness varies depending on the thermoplastic from which the veil is made and on the architecture of the carbon fibre fabrics (e.g. unidirectional UD, H5-satin or non-crimp fibre NCF). For example, Quan et al. [124] reported a higher improvement in UD composites compared to NCF and to H5-satin laminates. Bridging is the toughening principle addressed in the present study.

Several models have been developed to predict the mechanical response of structures involving fibre bridging. In the context of finite element analysis, cohesive zones model are often used with multilinear traction-separation laws [184]. Analytical or closed-form models have also been proposed such as the one developed by Cox and Marshall [185, 186] or by Sørensen and Jacobsen [187] who considered the continuous effect of bridging and non-

discrete bridging events opposite to Spearing and Evans [188], Szekrényes and Uj [189] and Li et al. [190]. In these models, a bridging law relating the bridging stresses to the opening displacements is required. The bridging law shape depends on the bridging phenomenon. For instance, if pull-out takes place, bridging stresses decrease with increasing opening displacement. One of the main issues in modelling bridging is the occurrence of snap-back instability upon failure of a ligament resulting in a sudden decrease of the applied displacement and of the force as the energy released by the advancing crack exceeds the energy dissipated to form the new fracture surfaces [190]. Sequence linear analysis [191] and crack length control schemes [192] are more adequate than a classical Newton-Raphson scheme to solve such problem.

The present work aims at triggering crack bridging in co-cured PEI composite joints using polyethylene ligaments provided as under the form of fishing lines deposited perpendicular to the crack propagation direction. The influence of the bridging ligament strength and spacing is investigated experimentally and modelled analytically. Increasing the spacing between the fishing lines and the tensile strength of the lines results in a significant improvement of the fracture toughness. An important parameter is also the tensile elongation of the bridging ligaments, which has not been tested experimentally but addressed based on the analytical model. Guidelines are given for the optimization of joints toughened through the addition of bridging ligaments.

5.2 Materials and Methods

5.2.1 Materials

The composite adherends are made of the Hexflow RTM6 epoxy resin [167] and HexForce G0926 H5-satin carbon fabrics. Each adherend consists of five plies stacked parallel to one another. The two composite adherends are joined by co-curing with a PEI film in a RTM press. The PEI film (Ultem 1000, Sabic, sold by GoodFellow) is 50 μm thick (see Supplementary Material 1 for the choice of the thickness) and is degreased using acetone prior to co-curing. Fishing lines made of Ultra-high-molecular-weight polyethylene (UHMPE) from the Decathlon brand *Caperlan* are inserted between the carbon fabrics and the PEI film. Different diameters and strength of fishing lines were tested, listed in Table 5.1. The strength is given in kg but in

terms of stress, all the lines should have a similar tensile strength as they are made of similar material.

Table 5.1 Properties of the fishing lines used to investigate the influence of tensile strength and spacing

Fishing line	Description	Tensile strength [kg]	Diameter of the line (diameter of one strand) [mm]	Investigated spacing [mm]
Caperlan TX4 60 µm	4 strands braid	3.9 (38 N)	0.26 (0.06)	1, 2, 5, 10
Caperlan TX8 60 µm	8 strands braid	5.7 (56 N)	0.19 (0.06)	1, 2
Caperlan TX8 100 µm	8 strands braid	8.3 (81 N)	0.25 (0.1)	1, 2
Caperlan TX8 200 µm	8 strands braid	17.2 (167 N)	0.35 (0.2)	1, 2

5.2.2 Joints manufacturing

Prior to joint manufacturing, the fishing lines are manually positioned with fixed spacing at both sides of the PEI film. We followed different procedures to introduce the fishing lines. For the joints made of the *Caperlan TX4 60µm* lines, the PEI film has been first stacked on the top of a paper sheet with graduation. Afterwards, each line has been positioned at a time and attached at its edges on the PEI film. This process was repeated at the other side of the PEI film. As an alternative, shown in Figure 5.1, a board has also been designed with spikes spaced by a given distance. This prevents cutting of each strand deposited on the film. When, all the fishing lines are deposited, the lines are attached along their edges to prevent any motion.

The joints are manufactured by resin transfer moulding. The first five G0926 carbon plies are first laid down in the RTM mould. The PEI film with fishing lines on both sides is then inserted and two 70 mm long, 30µm thick Kapton films (Dupont) are stacked on top of PEI in order to act as pre-crack for the fracture tests. The five last plies are then deposited. The RTM mould is then closed under 8 bar pressure. The RTM6 resin is heated in a piston up to 90°C before being injected at the same temperature in the

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mould until reaching a pressure of 6 bar. The composite is cured following the optimized curing cycle for PEI-RTM6 interphase developed by Voleppe et al. [12]. The temperature is increased at a rate of $2^{\circ}\text{C}/\text{min}$ up to 160°C for 90 minutes. Finally, the temperature is increased up to 180°C at a rate of $2^{\circ}\text{C}/\text{min}$ and the temperature is maintained constant for two hours. From the panels, double cantilever beam (DCB) samples are machined as shown in Figure 5.2.

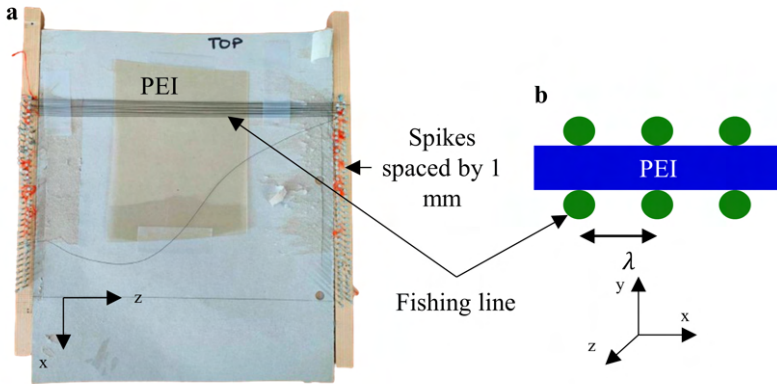


Figure 5.1 a) Board with spikes spaced by 1 mm to deposit the fishing line on the PEI film every 1 mm. Fishing lines are inserted at both sides of the PEI film. b) Scheme of the fishing line distribution at both sides of the film.

5.2.3 Mechanical testing

DCB samples, shown in Figure 5.2a and b, have been machined from the cured panels with a length of 200 mm and a width of 20 mm in a direction such that the fishing lines are aligned perpendicular to the crack propagation direction (direction z in Figure 5.2c). The 70 mm long Kapton film inserted to create the pre-crack corresponds to an initial crack length of 50 mm. The total height of the samples is about 4 mm. The DCB tests are performed following the ISO 25217 standard [172] on a Zwick/Roell machine equipped with a 50 kN loadcell, at constant cross-head speed of 1 mm/min. The machine is located in a ventilated room at constant temperature and relative humidity content about 23°C and 50%, respectively. Prior to testing, the edge of the samples are graduated to measure the crack length by taking one picture every 3 seconds with a Canon EOS1400D cam-

era. The mode I toughness G_{Ic} is then computed following the single beam theory [172]. Four replicate samples are tested for each condition, except for a spacing of 7 mm for which only one sample was available.

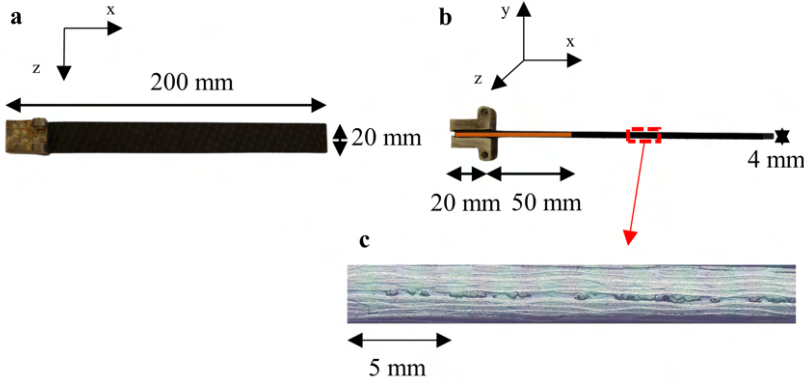


Figure 5.2 a-b) DCB sample and dimensions from a) the top and b) the side. c) Optical micrograph of the bondline with fishing lines spaced by 1 mm.

5.2.4 Microscopy

Scanning electron microscopy (SEM) with Ultra 55 Field-Emission SEM (Zeiss, Germany) under 5 keV and optical microscopy with a VHX microscope from Keyence has been used for fractography analyses. This required cutting joints along the crack propagation direction and polishing the cross-section prior characterization.

5.3 Analytical model

5.3.1 Governing equations

An analytical model, similar to the one of Li et al. [190], has been developed in order to predict the joint toughness. The model considers the joint toughness in the absence of fishing lines G_0 , line spacing λ , Young's modulus E^f , tensile strength F^f and radius r of the fishing lines. The materials properties and dimensions of the composite adherends enter the problem. The DCB is of width B , arm thickness h and made of a material of Young's modulus E . The crack has a length a . In this DCB, n fishing lines are posi-

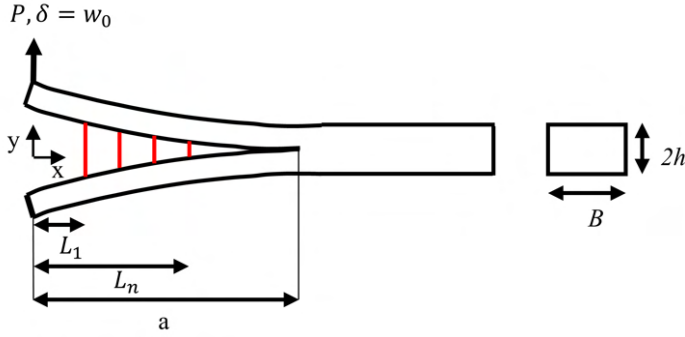


Figure 5.3 Double cantilever beam configuration with a width B , a height $2h$. A crack of length a is bridged by n ligaments in the wake, located at the positions L_i .

tioned with spacing λ and are thus located at the positions L_i ($i=1, 2, \dots, n$) as shown in Figure 5.3. The deflection at the position L_i is given by

$$w_i(L_i) = \frac{PL_i^3}{6EI} - \sum_{j=1}^i \frac{F_j}{EI} \left(\frac{L_i^3}{6} - \frac{L_j L_i^2}{2} \right) + C_i L_i + D_i \quad (5.1)$$

where P is the applied force, F_j is the force carried by the bridging ligament at position L_j while C_i and D_i are two constants (see Supplementary Material 2). If i is set to 0, one gets the deflection at the loading point. Solving these equations requires to derive a bridging law relating the applied force to the bridging ligament located at L_i to the beam deflection at the same point (see subsection 5.3.2). In addition to these equations, a crack propagation criterion is required. The crack propagates when the crack tip driving force G_{tip} is equal to the toughness of the joint in the absence of bridging ligament G_0

$$G_{tip} = \frac{1}{BEI} \left(Pa - \sum_{j=1}^n F_j (a - L_j) \right)^2 = G_0. \quad (5.2)$$

The unknowns of the problem are thus the deflections at the bridging ligament positions, w_i ($i=1, 2, \dots, n$) and the macroscopic force P while the inputs are B, h, E, a, G_0 and the bridging law. The $(n+1)$ equations correspond to the n equations coming from Equation 5.1 and one last equation

coming from Equation 5.2. A Newton-Raphson method is used with as guess for the k th iteration $X_k = (W_{1k}, W_{2k}, \dots, W_{nk}, P_k)$ and the function to solve is

$$H(X_k) = \begin{pmatrix} W_{1k} - w_1(L_1) \\ W_{2k} - w_2(L_2) \\ \vdots \\ W_{kn} - w_n(L_n) \\ G_{tip} - G_0 \end{pmatrix} = 0. \quad (5.3)$$

This is repeated for each crack length a in order to predict the corresponding the macroscopic load $P(a)$ and total fracture toughness $G_{Ic}^{tot}(a)$.

5.3.2 Bridging law

The bridging law relates the bridging force F_i to the deflection at the position of the bridging ligament $w_i(L_i)$. The distance between the two DCB arms at the position L_i is equal to $2w_i$. Hence, the bridging law gives $F(2w_i)$. Although the bridging law influences the results, Spearing and Evans [188] showed for two different laws, but characterized by the same fracture energy (surface area under the curve), that G_{Ic} is similar. In the present case, the four laws shown in Figure 5.4 have been addressed each of them being selected as partly representative of the phenomenology of the bridging specific to our configuration (see later with the results). Law # 1 considers that the fishing lines behave as springs while for Law # 2, the fishing lines must first be unfolded before being loaded under tension. Law # 3 considers that the ligament is attached at the edges at opposite interfaces while Law # 4 considers a ligament attached to the bottom interface, but debonded over a distance d and which is bonded to the opposite interface at the middle of the debonded length. The two last laws strongly depend upon the fishing line Young's modulus E^f and radius r . In all cases, the fishing lines break when the force exerted along the loading axis is equal to the tensile strength F^f . More details are given in Supplementary Material 2. An influence of the bridging law is found on the results. However, as long as the surface area under the bridging law is constant, a similar toughness is reached while if it is increased, G_{Ic} increases. However, depending on the law, the tensile elongation of the fishing line differs. The higher the tensile elongation, the less steep the R-curve and the maximum toughness is reached after a longer crack propagation length, in agreement with the results of Li et al. [190].

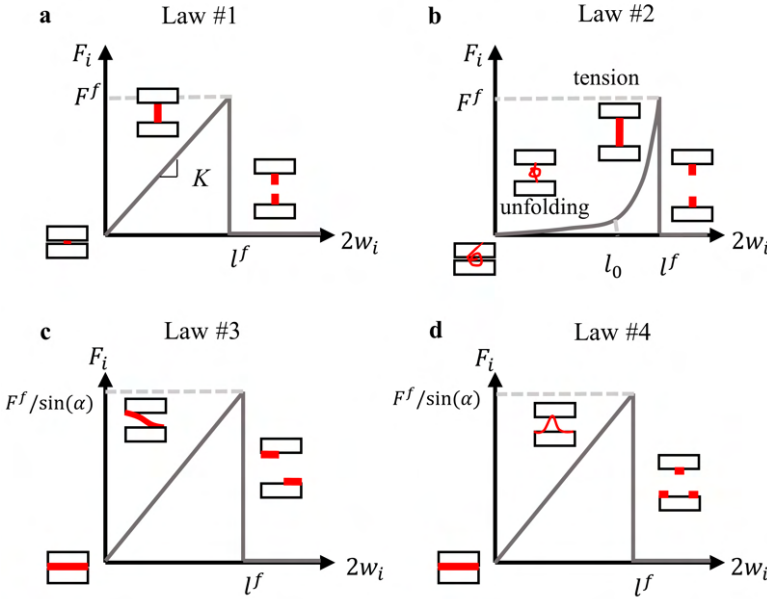


Figure 5.4 Bridging laws: a) Law # 1 the ligament behaves as a spring, b) Law # 2 the ligament first unfold before being loaded under tension, c) Law # 3 the ligament is attached at the edges of the DCB, d) Law # 4 decohesion takes place over a short distance d while the debonded part adheres to the opposite interface.

5.4 Results

5.4.1 Influence of spacing from experimental DCB tests

Figure 5.5a shows the experimental R-curves extracted from the DCB tests performed on co-cured joints with a $50\text{ }\mu\text{m}$ thick PEI film and fishing lines with different λ . No matter the spacing, the toughness first increases over a certain crack propagation distance (Δa^{ss}) that varies with λ before reaching a plateau corresponding to the steady-state region. For a spacing of 1 mm, the steady-state toughness is equal to $763 \pm 102\text{ J/m}^2$ and the R-effect takes place over a crack propagation length $\Delta a^{ss} = 10\text{ mm}$ (defined in Figure 5.5b). If the spacing is increased up to 2 mm, the steady state G_{Ic}^{ss} is about $718 \pm 89\text{ J/m}^2$ and is reached after a crack propagation length about 10 to 15 mm. For spacings equal to 5, 7 and 10 mm, the steady-state is

reached after 25 mm of crack propagation and the steady-state toughness is equal to 433 ± 66 , 614 ± 34 , 563 ± 99 for each spacing, respectively. One can also define G_{Ic}^{init} for $\Delta a \rightarrow 0$ by linear extrapolating the results as shown in Figure 5.5b. The plain joint fracture toughness G_0 (without fishing lines) is equal to 284 ± 89 J/m² (see Chapter 3). The key results are gathered in Figure 5.5c. Bridging by the fishing lines leads to a maximum G_{Ic}^{ss}/G_0 ratio of about 2.7. The toughening effect decreases as the spacing increases.

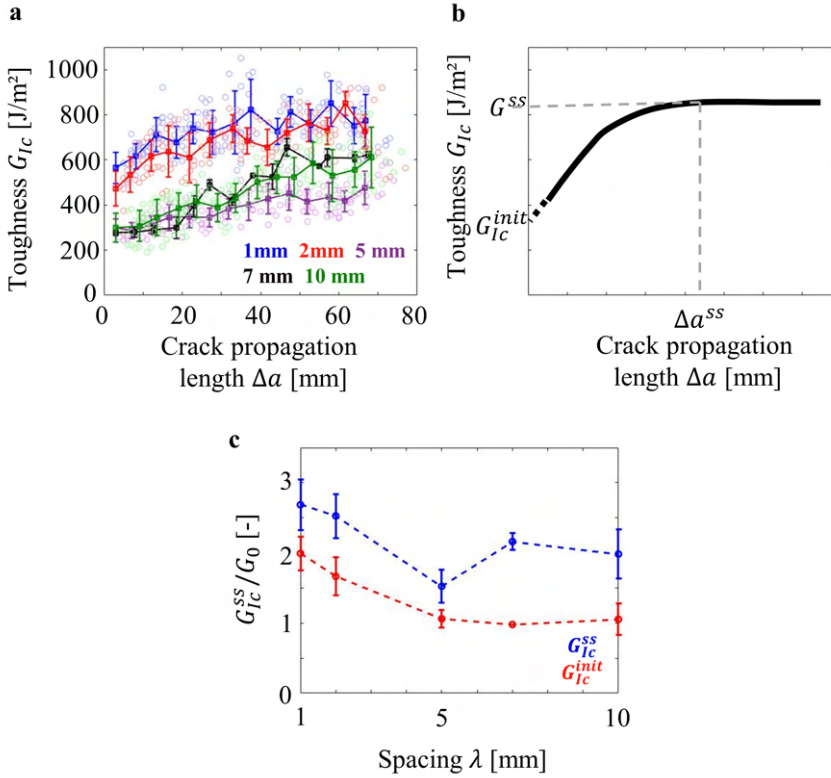


Figure 5.5 a) R-curves for PEI co-cured composite joints with fishing lines perpendicular to the crack propagation direction spaced by a distance λ equal to 1 (blue), 2 (red), 5 (purple), 7 (black) and 10 (green) mm. b) Scheme of R-curve defining G_{Ic}^{init} , G_{Ic}^{ss} and Δa^{ss} . c) Evolution of steady-state G_{Ic}^{ss} and initiation toughness G_{Ic}^{init} with spacing.

5.4.2 Influence of strength from experimental DCB tests

The R-curves resulting from the crack propagation in PEI co-cured composite joints in which fishing lines with different tensile strengths are inserted are shown in Figure 5.6. Figure 5.6a corresponds to joints in which the fishing lines are spaced by 1 mm while in Figure 5.6b, they are spaced by 2 mm. For joints containing fishing lines spaced by 1 mm and characterized by a tensile strength of 5.7, 8.3 and 17.2 kg, the steady-state toughness G_{Ic}^{ss} is equal to $559 \pm 141 \text{ J/m}^2$, $502 \pm 135 \text{ J/m}^2$ and $931 \pm 237 \text{ J/m}^2$, respectively. If the spacing is increased up to 2 mm, the 5.7 kg fishing lines result in a toughness equal to $537 \pm 95 \text{ J/m}^2$, the 8.3 kg to a G_{Ic}^{ss} equal to $432 \pm 83 \text{ J/m}^2$ and the 17.2 kg to a toughness equal to $494 \pm 187 \text{ J/m}^2$. This is summarized in Figure 5.6c. In the case of the 17.2 kg fishing lines spaced by 2 mm, a first peak is reached for a crack propagation length of 20 mm with an associated toughness of about 690 J/m^2 before a sudden decrease. For the joints made with 1 mm spaced fishing lines, the toughness tends to increase with the tensile strength of the fishing lines while when spaced by 2 mm, G_{Ic} is almost constant although a slight increase is observed at the beginning of the R-curve with the tensile strength. To conclude, increasing the fishing line strength tends to be beneficial for the joint fracture toughness. In addition, the tensile strength does not influence the distance over which the R-effect is taking place and ranges from 10 to 20 mm in all cases.

5.4.3 Fractography

Figure 5.7 shows the fracture surfaces giving interesting indications on the failure mechanisms taking place during crack propagation in samples with fishing lines spaced by 1 mm (results are similar for 2 mm spacing). Figure 5.7a schematizes decohesion between a fishing line and the composite (Figure 5.7b) or between a fishing line and the PEI film (Figure 5.7c) sometimes takes place. In addition, the crack can propagate along the composite outside of the morphological gradient that forms upon co-curing as fibre imprints can be observed in the micrograph of Figure 5.7b. As for joints without fishing lines, the crack propagates through the tough PEI film (Chapter 3 and Figure 5.7b). In addition, fibre bridging takes place as shown in Figure 5.7d, originating from a change in crack propagation plane. At a given crack length, along the DCB width, the crack propagates above and below a given fishing line as shown in Figure 5.7e and upon loading, the fishing line breaks as observed on the fracture surfaces in Figure 5.7f and g. In addition, inserting fishing lines results in a wavy PEI

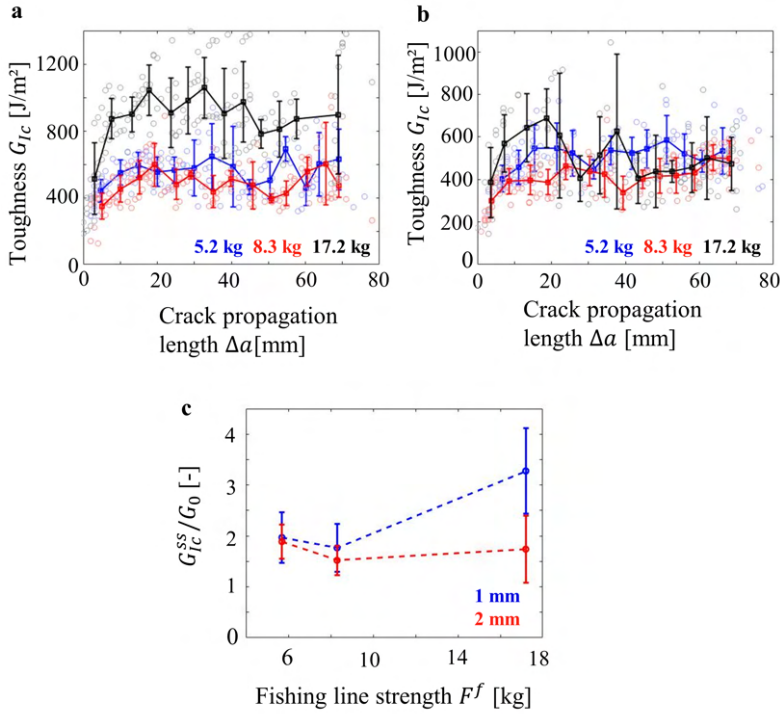


Figure 5.6 R-curves for PEI co-cured joints containing fishing lines of varying tensile strength: 5.2 kg (blue), 8.3 kg (red) and 17.2 kg (black) spaced by a) 1 mm and b) 2 mm. c) Evolution of G_{Ic}^{ss}/G_0 with the fishing line strength.

film and the fishing lines cause some degree of crack deflection. Crack deflection and bridging induce toughening, but it is believed that the major contribution arises from crack bridging by the fishing lines. Figure 5.7h schematically represents a cross-section of a joint containing fishing lines spaced by at least 5 mm, showing that the PEI film is less wavy. Close to the fishing lines, similar mechanisms as those shown in Figure 5.7a are found while between two consecutive fishing lines, the fibres are close to the PEI film and lay thus in the morphological gradient. In these regions, failure mechanisms similar to joints without fishing lines are reported in Chapter 3: delamination in the PEI-rich zone, propagation along the PEI-rich/epoxy-rich interphase in resin pockets and propagation through the PEI film.

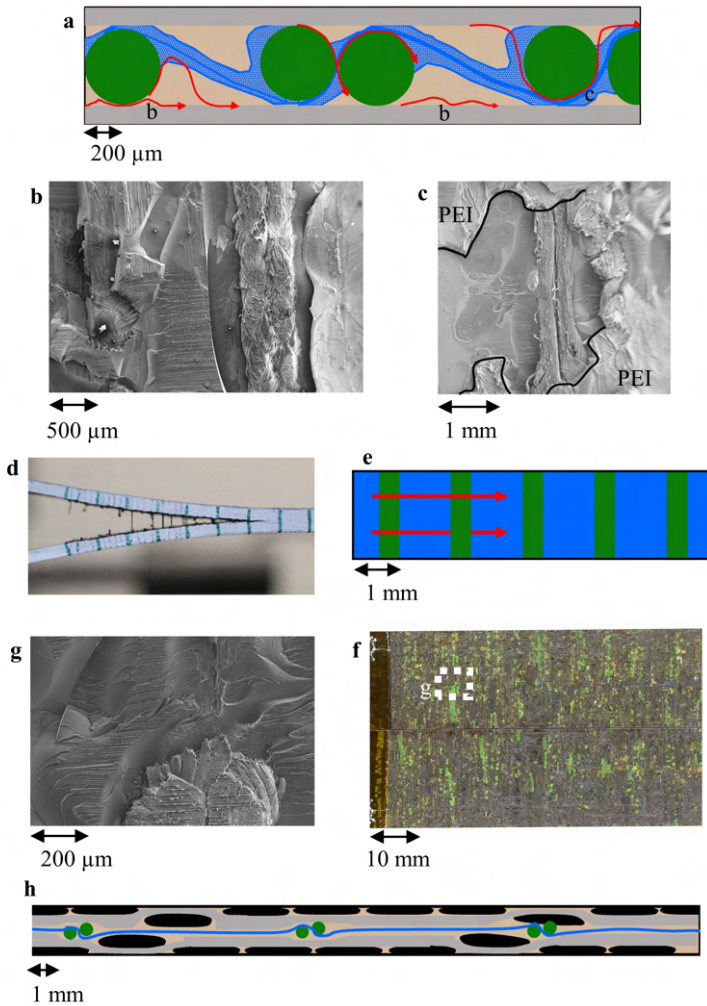


Figure 5.7 Failure features in PEI co-cured joints with fishing lines spaced by 1 or 2 mm: a) schematic representation with decohesion of the fishing line along b) the composite fibres or c) the PEI film with crack propagation through the PEI film. d) DCB picture taking during a test with bridging by the fishing lines due to e) a crack propagation at both sides of a fishing line that f-g) breaks upon loading f) optical micrograph and g) SEM micrograph. h) Schematic representation along the DCB length for a joint containing fishing lines spaced by 5 mm.

Figure 5.8 shows the fracture surface of a joint made of 17.2 kg strength fishing lines spaced by 2 mm. The bottom part exhibits zones with limited number of fishing lines segments (highlighted in gray), revealing the surface of the composite. In these regions, a limited number of fishing lines act as bridging ligament reducing the potential toughening. The same is observed on all fracture surfaces for this condition, probably explaining the sudden drop in toughness for a crack advance above 20 mm in this case (Figure 5.6b, black line).



Figure 5.8 Optical micrograph of the fracture surface of a co-cured composite joint with 17.2 kg strong fishing lines spaced by 2 mm (orange strands). The grey regions correspond to zones with a limited number of fishing lines acting as bridging ligaments.

Figure 5.9 schematizes the bridging phenomenon. As shown in Figure 5.9b, at a given x position, the crack propagates either between the fishing line and the PEI film or between the fishing line and the composite depending on the z position. As shown in Figure 5.9c-d, this results in the development of bridging ligaments. This change of crack path location can take place several time along the joint width, resulting in many bridging ligaments as shown in Figure 5.9e which upon deformation break (Figure 5.9f), resulting in fracture surfaces with small fishing line parts as shown in Figure 5.9g and Figures 5.7f and Figure 5.8.

5.5 Discussion

The experimental results confirm that the introduction of fishing lines in PEI co-cured composite joints results in significant toughening owing to

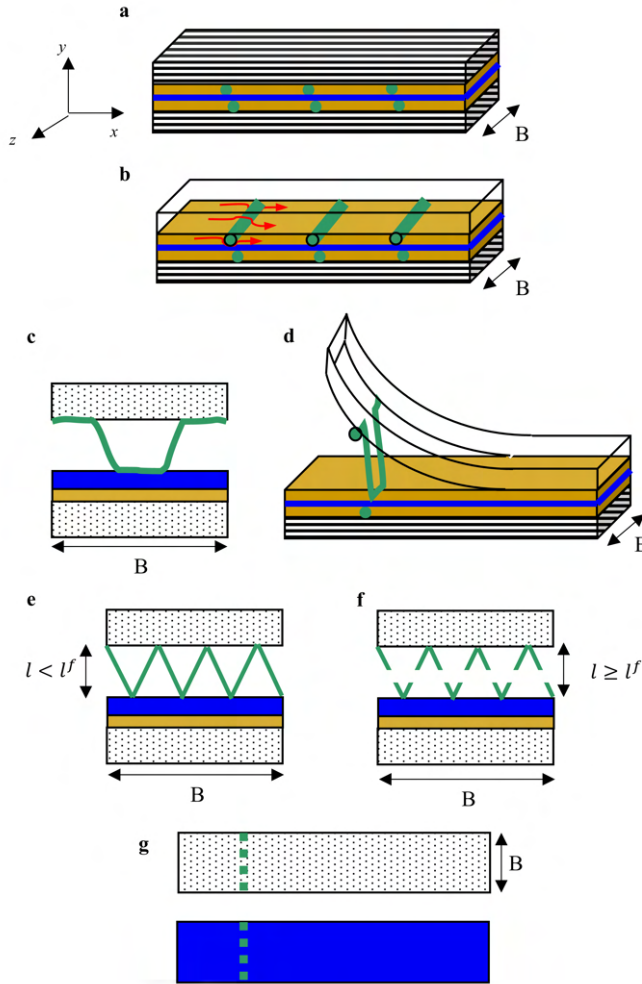


Figure 5.9 a) Scheme of a joint with fishing lines. b-g) Schematic representation of the bridging phenomenon with b) a crack propagating above and under the fishing line for a given $x = L_i$ position resulting in the configuration shown in c) for the view from the opened side and d) for the 3D view upon loading. e-f) Generalization to the actual joints where one bridging ligament can result in several bridging ligaments e) before ligament failure and f) upon failure. g) Scheme of the fracture surface at the composite and PEI film side with bridging ligament parts at both sides.

crack bridging. The toughness is increased by a factor of minimum 1.5 up to 3.3 depending on the nature and spacing of the fishing lines. In H5-satin composites, fibre bridging is known to have only a moderate influence on G_{Ic} when inserting thermoplastic veils leading to toughening factors (G_{Ic}^{ss}/G_0) lower than 2 [131]. The areal weight is comparable to the one of thermoplastic veils ($\approx 10 \text{ g/m}^2$) for a joint with lines spaced by 1 mm ($\approx 20 \text{ g/m}^2$). Hence, extensive toughening was not expected. This also shows that there is space for potentially improving thermoplastic veils by controlling the orientation of the fibres.

Experimentally, the strength of the fishing lines and spacing significantly impact the toughness. Upon loading, one part of the bridging ligament is attached to the top composite part and the other one to the bottom part. Many segments can be observed on the fracture surface as shown in Figure 5.7f and Figure 5.8, indicating the bridging mechanism can be better assimilated to the behaviour of Law # 4 described in subsection 5.3.2 (Figure 5.4d).

5.5.1 Identification of the model parameters

The only unknown in the model relying on Law # 4 (Figure 5.4d) is the debonding distance d which is used as fitting parameter. The identification of d is shown in Figure 5.10 for the 3.9 kg strong fishing lines spaced by 1 and 2 mm. In order to reproduce the experimental results, d must range between 0.25 and 0.5 mm. If now, the 5.7 kg strong fishing lines are considered, d must be set to 0.1 mm, a value similar to the radius of the fishing line. Hence, in this case, bending dominates over tension (Supplementary Material 2). If this bending is considered, the results cannot be properly fitted anymore. This may be explained by the low Young's modulus of the fishing lines resulting in large deformations, not captured by the analytical model. The model also neglects potential defects that can be present in joints.

The distance d is thus an important fitting parameter but it cannot be controlled by inserting the fishing lines. In addition, the model requires to consider large deformations and tension and bending dominated regimes, including the transition, which is beyond the scope of the present analysis. Consequently, in order to discuss the influence of the spacing and strength on the joint fracture toughness, Law # 1 will be used for what follows.

With Law # 1, the fitting parameter is no longer d but the tensile elonga-

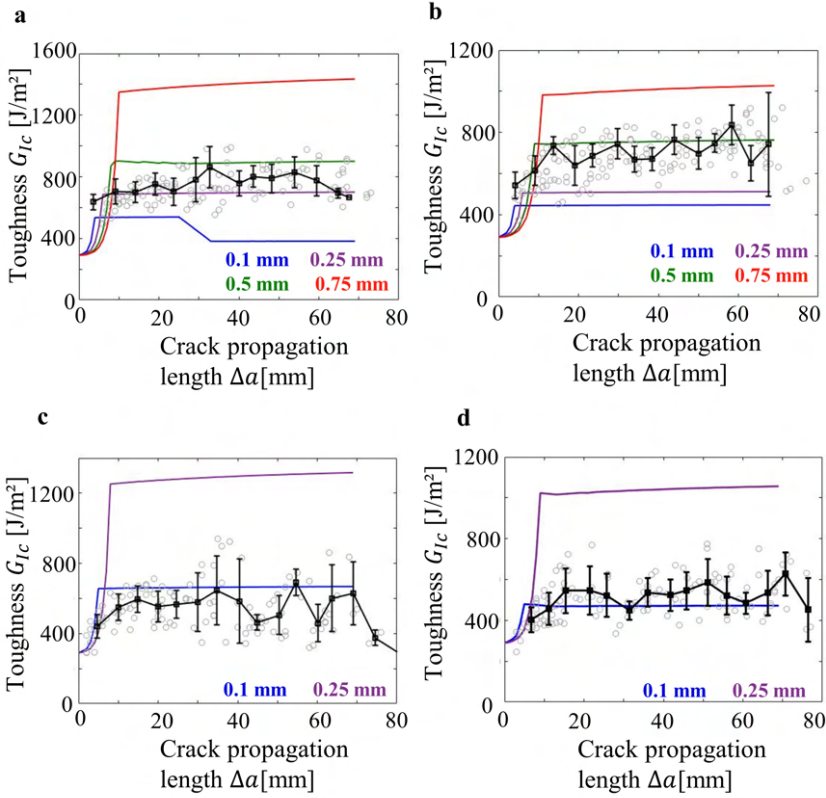


Figure 5.10 Fitting of experimental results (grey dots and dark line) using the 4th bridging law with different debonding distances d for a) 3.9 kg – 1 mm, b) 3.9 kg – 2 mm, c) 5.7 kg – 1 mm and d) 5.7 kg – 2 mm.

tion of the fishing line l^f . Although this law does not accurately represent the bridging mechanism, it is possible for each condition to determine l^f values so that the predicted joint toughness matches the experimental G_{Ic}^{ss} . The tensile elongation l^f is determined for the joints with 3.9 kg strong fishing lines spaced by 1, 2, 5, 7 and 10 mm as shown in Figure 5.11. Ideally, l^f , as d with Law # 4, should be equal to the same value for all conditions. However, for the 3.9 kg strong fishing lines, values of l^f ranging between 0.4 and 0.6 mm are found to fit the results for $\lambda=1$ mm while for $\lambda=2$ mm, l^f must be set with a value between 0.7 mm and 1 mm. Using values ranging from 0.5 to 1.5 mm allows capturing the G_{Ic} of joints with fishing lines

spaced by 5, 7 and 10 mm, but, as with Law # 4, the distance over which the R-effect (Δa^{ss}) takes place is not well predicted. This may arise from a distribution in the tensile elongation of the fishing lines as they can debond over different length. For the fishing lines characterized by a strength of 5.7, 8.3 and 17.2 kg and λ set to 1 and 2 mm, l^f values ranging between 0.1 et 0.5 mm reproduces the results, except if the strength is about 5.7 kg and λ is set to 2 mm, a condition for which l^f must range between 0.7 and 1 mm (see Supplementary Material 3).

Once using Law # 1, l^f can strongly influence the results and by analogy the same occurs if d in Law # 4 is used. By fitting the experimental results, it is found that for the joints for which the fishing lines have been placed manually, l^f and d are larger compared to the joints manufactured by inserting the fishing lines using the board with spikes. Placing the fishing lines using the board results in a higher tension in the fishing lines prior to bonding which could have an influence on residual stresses. The initial tension in the fishing lines may thus have an influence, but this parameter is hard to control. Now, we must recognize that the present model involves a lot of assumptions and can only be considered as semi-quantitative, although sufficient to discuss next some key trends.

In what follows, the influence of the spacing and of the strength will be discussed by only considering Law # 1 and keeping l^f constant but using different values within the identified spectrum. The goal at this point is thus to use this model to unravel the influence of the spacing (subsection 5.5.2) and the fishing line strength (subsection 5.5.3) in order to optimize the joint fracture toughness (subsection 5.5.6).

5.5.2 Influence of spacing

Figure 5.12a shows that the maximum toughness G_{Ic}^{max} decreases as the spacing between the fishing lines decreases in agreement with experimental results. Above a spacing λ_{c1} , a stick-slip behaviour (Figure 5.12c, $\lambda=4$ mm) is observed because when one fishing line breaks, the crack advances but without additional fishing line introduced behind the crack tip. This results in a saw-tooth R-curve as shown in Figure 5.12c (dashed lines). Above a spacing λ_{c2} , the spacing does no longer influence the maximum and minimum G_{Ic} attained during crack propagation and the minimum G_{Ic} (G_{Ic}^{min}) is equal to G_0 . For the fishing lines characterized by a tensile elongation of 0.25 mm and 1 mm, this happens when the spacing between the fish-

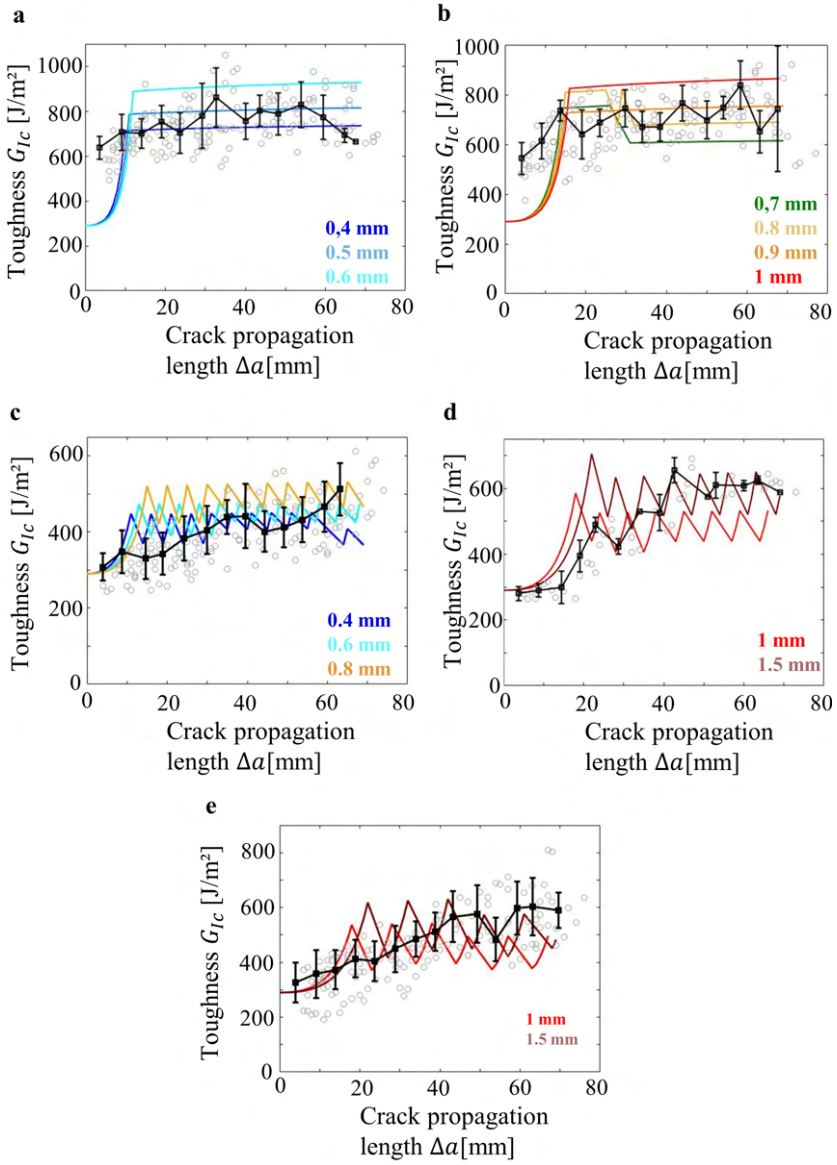


Figure 5.11 Identification of l^f on experimental results (grey dots and dark line) using the bridging law # 1 for different spacings by adjusting the tensile elongation: a) 1mm, b) 2mm, c) 5 mm, d) 7 mm, e) 10 mm.

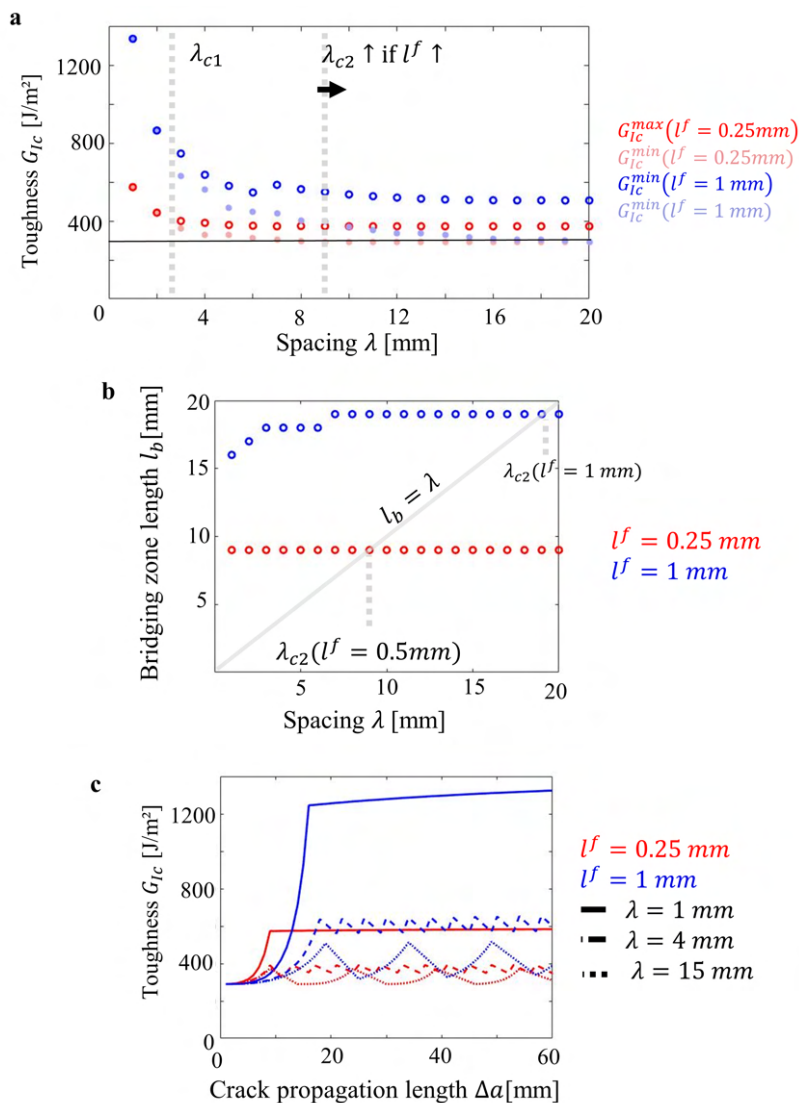


Figure 5.12 Influence of the line spacing on the joint toughness and response of the joint for two tensile elongations of the lines equal to 0.5 and 1 mm: a) evolution of toughness with spacing, b) evolution of the bridging zone length with the spacing and c) R-curves for spacings of 1, 4 and 15 mm.

ing lines is equal to 9 and 19 mm, respectively. As shown in Figure 5.12b, this occurs when the bridging zone length l_b is equal to λ . The bridging zone length corresponds to the zone comprised between the first intact fishing line behind the crack tip and the crack tip. If λ is lower than l_b , there is always at least one fishing line acting as bridging ligament so that $G_{Ic}^{min} > G_0$. If $\lambda > \lambda_{c2}$, a stick-slip behaviour is also found as shown in Figure 5.12c with the maximum toughness corresponding to the toughness in the presence of one fishing line and the minimum one to the joint toughness without fishing lines ($G_{Ic}^{min} = G_0$). In this case, any increase of spacing does not affect the toughness but as λ increases, the frequency at which a peak toughness is reached decreases, reducing the number of toughening events. For values of l^f up to 5 mm, λ_{c1} is not affected by l^f and is equal to 2 while λ_{c2} increases as l^f increases as it increases l_b .

5.5.3 Influence of the strength of the fishing lines

Increasing the strength can be achieved by changing the ligament material, by triggering bridging ligaments acting in parallel [190] or by increasing the diameter of the ligament. For given spacing and tensile elongation of the lines, increasing the strength results in an improved toughness, but not in all cases as shown in Figure 5.13a. This arises from the fact that as the strength increases, the bridging zone length progressively decreases. For two ligaments of similar strengths such that $F^{f1} < F^{f2}$, it is possible to have $l_b^1 > l_b^2$ resulting in a more pronounced toughening effect for fibres of strength F^{f1} . This is shown in Figure 5.13c. In addition, the toughening effect resulting from an increase in strength also depends on the tensile elongation of the fishing lines. The toughening effect is enhanced when increasing l^f . Figure 5.13a shows that although the spacing between the lines is set to 1 mm, some conditions result in a maximum and minimum toughness. A R-curve corresponding to such case is shown in Figure 5.13d. As the crack advances, the deflection at a distance l_b slightly increases and may exceed the tensile elongation of the fishing line reducing the bridging zone length and the fracture toughness. For a given tensile elongation of 0.5 mm, the combined influence of tensile strength and spacing is shown in Figure 5.13b. Increasing the strength results in an increase of λ_{c2} .

5.5.4 Influence of tensile elongation of the lines

The influence of the tensile elongation has been previously discussed with the tensile strength and the spacing. An increase in tensile elongation re-

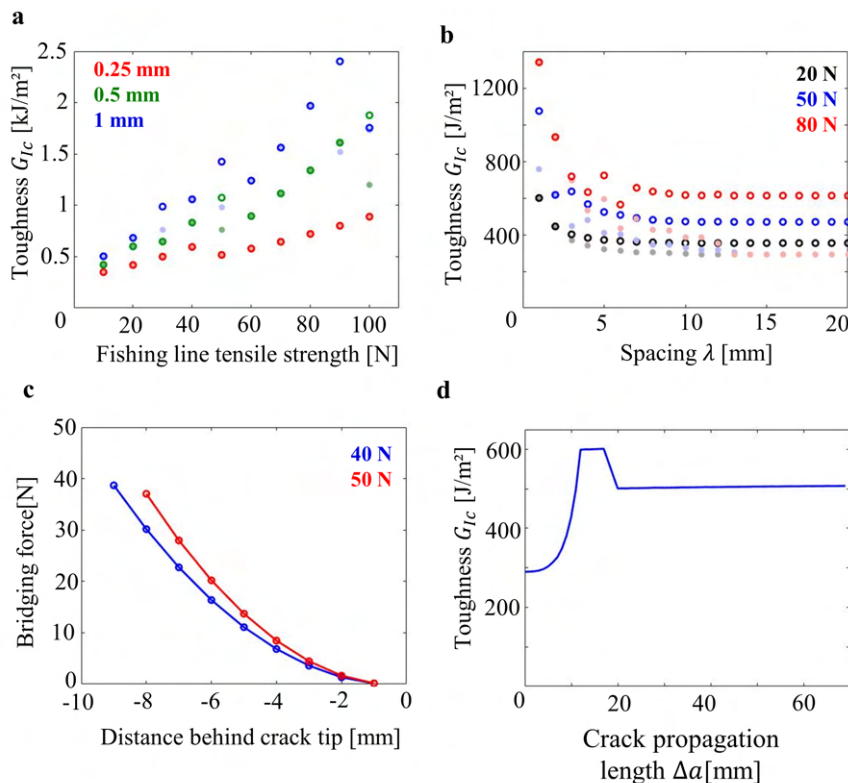


Figure 5.13 Influence of the tensile strength of the fishing lines on the toughness: a) spacing of 1 mm and different elongations (0.25, 0.5 and 1 mm), b) combined influence of strength and spacing, c) Increase in tensile strength with a decrease in bridging zone length reducing the fracture toughness ($\lambda=1$ mm, $l^f=0.5$ mm), d) R-curve exhibiting a local stick-slip behaviour as the bridging zone length is suddenly reduced.

sults in an increase in toughness. However, as the tensile elongation is increased, the distance over which the R-effect takes place increases, delaying the attainment of the maximum toughness. In addition, by decreasing the tensile elongation, the toughening effect resulting from the decrease in spacing and the increase in strength is reduced. For the 5.7, 8.3 and 17.2 kg strong fishing lines, using bridging Law # 1, the tensile elongation is assumed to be around 0.1 and 0.2 mm (Supplementary Material 3), explaining why the toughening effect resulting from an increase in tensile strength

is not properly captured experimentally.

5.5.5 Influence of the joint toughness in the absence of fishing lines

Depending on the initial joint fracture toughness, the toughening effect from the use of fishing lines can be more or less important. In any case, for a given set of (λ, l^f, F^f) , the toughness G_{Ic}^{ss} increases with G_0 . However, the toughening effect is reduced as shown in Figure 5.14a for different F^f and l^f and $\lambda=1$ mm. For a bare joint without fishing lines, if G_0 increases, the beam deflection at a distance x from the crack tip increases. Hence, when the fishing lines are inserted, for low values of G_0 , the distance between the two DCB arms at a position L_i does not exceed l^f while this value is well exceeded for larger values of G_0 . Hence, the bridging zone length decreases as G_0 increases as shown in Figure 5.14b, reducing the energy dissipated in the bridging zone. Hence, to obtain a similar toughening effect, the strength of the bridging ligament must be increased if the initial joint toughness is larger. Figure 5.14c shows that the toughening effect is governed by the ratio between the energy required to break the ligament $U_{bridge} = F^f l^f / 2$ and G_0 . Finally, it is important to note that G_0 only slightly influences the distance over which the R-effect takes place.

This explains why the toughening effect is slightly larger in joints with a 175 μm PEI film than in joints with a 50 μm thick film. The 50 μm thick film results in a higher toughness with and without bridging compared to the 175 μm film. However, if the toughening ratios (toughness with ligaments over toughness without) are compared, it is higher for the 175 μm thick film.

5.5.6 Optimizing joints with architected bridging features

The influence of the bridging ligament properties, distribution and the initial joint toughness have been investigated in the previous sections. Increasing the tensile strength and the tensile elongation of the fishing lines and decreasing the spacing between the ligaments results in an improved toughening effect. The tensile strength can be increased by (i) changing the ligament materials for a stronger material, (ii) increasing the radius of the ligament as the maximum tensile force matters and not the maximum tensile stress or (iii) by triggering bridging ligaments acting in parallel. In contrary, the tensile elongation cannot be easily controlled. The toughening effect is also dependent on G_0 .

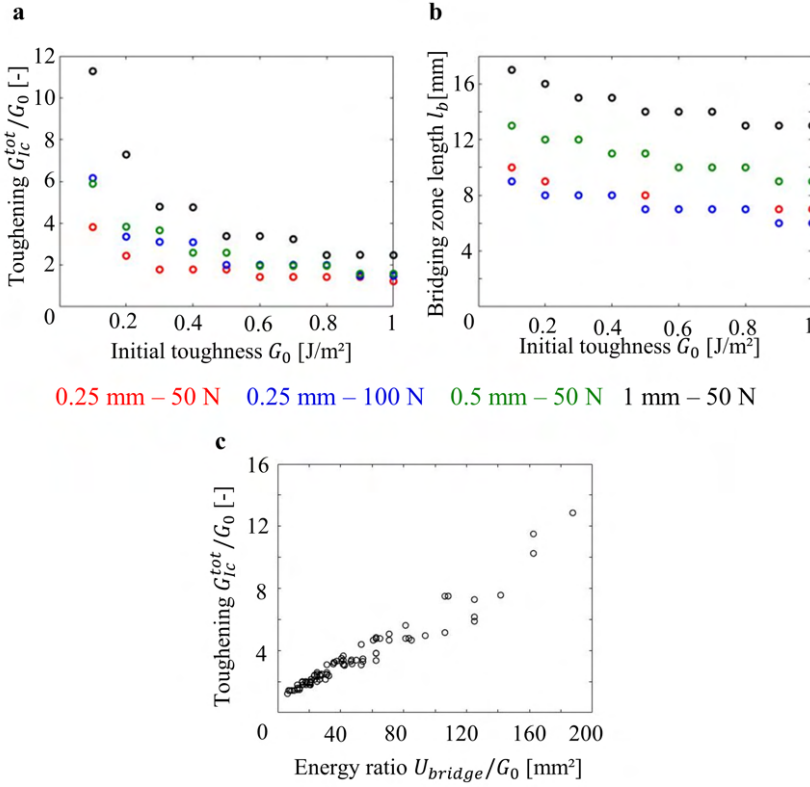


Figure 5.14 Influence of G_0 on the total joint toughness G_{Lc}^{tot} for $\lambda = 1$ mm and different l^f and F^f : a) toughening effect depending on G_0 and b) influence of G_0 on the bridging zone length l_b . c) Evolution of toughening ratio with the ratio between the energy required to break a ligament and G_0 .

Regarding the tensile elongation, it increases the joint fracture toughness but also the distance over which the R-effect takes place. Hence, the joint toughness increases more progressively as the crack propagates if it is enhanced. Hence, in some case, for short crack propagation distance, if one considers two joints made of the same bridging ligaments but with different elongation, the joint with the less ductile ligaments can be tougher than the one with the ductile ligaments. For example, this is shown in Figure 5.11 and in Figure 5.12c. However, for long crack propagation distances, the joint with the ductile ligaments is the toughest. Consequently,

the design of the joint is dependent upon the maximum allowable length of the crack present in the joint.

Adhesive joint selection analysis [101] can be qualitatively applied to the present case. However, in the present case, the shear behaviour of the joints with fishing lines is unknown. Although, it can be argued that if the radius of the fishing lines increases, the bondline thickness will increase resulting in a decrease of the shear stiffness and shear strength. In addition, it will result in a slight mass increase of the joint. If the objective is to minimize the joint mass or if the constraints on shear stiffness and strength are quite severe, joints with thin bridging ligaments must be preferred. If one considers joints with thin ligaments and that the objective is to maximize the effective tensile strength of the joint, the selection depends on the crack length. Consequently, a trade-off must be found between the ligament strength and elongation for this crack length. If the objective is to maximize the joint fracture toughness, the selection is crack length independent and joints containing strong and long ligaments will be preferred. The answer is more nuanced when minimizing the joint weight. If the ligaments are thin, it can be assumed that the mass is negligible compared to the rest of the joint components. Hence, a spacing of 1 mm will be an optimum although the joint weight is slightly higher than for joints with larger spacings. In addition, this gain in weight will be counterbalanced by the improved toughness. However, this is only true for short spacings as for long spacings, the joint toughness is no longer dependent on the spacing. Hence, the design of joints containing bridging ligaments depends on the initial toughness G_0 of the joint, the minimum toughness that must be attained and the maximum distance over which the crack can propagate before attaining G_{Ic}^{ss} . Depending on this and on the choice of λ , F^f and l^f can be tuned to meet the requirements. This is true if one considers the bridging ligaments acting as springs (bridging law # 1 – Figure 4a) while if bridging Laws # 3 and # 4 (Figure 5.4c-d) accounting for bending are considered, the bridging ligament Young's modulus must also be considered in the design.

5.6 Conclusion

Co-curing of composites with an adhesive is an alternative method to secondary bonding, but it results in a lower joint toughness. Hence, it has been suggested to replace the adhesive by a thermoplastic film such as PEI.

Using pre-pregs, this method showed improved joint toughness compared to composite delamination. However, when the co-curing between the composites and the PEI film is achieved through the Resin Transfer Moulding process, a lower toughness than composite delamination is achieved, requiring the development of mitigating toughening strategies. In the present work, crack bridging has been triggered by inserting fishing lines at both sides of the PEI film resulting in an improvement of the toughness by a factor +100% to +200%. Fishing lines joints are characterized by a toughness significantly larger than the delamination toughness of the composite and can compete, depending on the design parameters, with some classical adhesive joints. Triggering crack bridging by inserting fishing lines in PEI co-cured joints proved thus to be efficient.

The toughness of the joint can be optimized by controlling the spacing between the ligaments, the tensile strength and the tensile elongation of the strands. Low spacing and high tensile strength allow to improve the joint fracture toughness. Increasing the tensile elongation also results in a larger fracture toughness, but it delays the attainment of the maximum fracture toughness. Hence, for small crack propagation length, joints with less ductile ligaments can be tougher than joints with ductile ligaments. Consequently, the joint design must consider the maximum allowable crack extension and the level of toughness required upon a certain crack propagation distance in order to fulfill the mechanical requirements. If the crack is allowed to only extend over a small distance, it must be preferred to increase the bridging ligament strength by changing of material, increasing the ligament radius or have ligaments acting in parallel than increasing the tensile elongation. If the crack extends without restriction, increasing the tensile elongation of the ligament is an option, however, difficult to control experimentally.

5.7 Supplementary materials

5.7.1 Supplementary material 1: Preliminary study



The original version of this supplementary involves results presented in Chapter 3 and have been removed here to not be redundant.

Owing to the lower mode I fracture toughness of adhesive co-cured composite joints with respect to secondary bonded joints, Quan and co-workers [30, 31] suggested to replace the adhesive by a thermoplastic film such as polyetherimide (PEI). They inserted PEI films of varying thicknesses between composite pre-pregs made of the 8552 epoxy resin. They reported a larger mode I fracture toughness compared to composite delamination but lower than compared to secondary bonded joints. In addition, they reported an increase in G_{Ic} with the PEI film thickness with an optimum thickness around 200 μm . To our knowledge, co-curing of composite joints using a PEI film has never been performed through the resin transfer moulding (RTM) process. In the case of pre-pregs, the carbon fabric plies are already impregnated by the resin leading to a resin layer between the PEI film and the carbon fibres before curing. Opposite, in RTM, the carbon fibres are in contact with the PEI film and the resin is then injected. Hence, the fibres can remain in contact with the PEI films.

Chapter 3 investigates the toughness of co-cured composite joints through RTM and the influence of the PEI film thickness on G_{Ic} and the failure mechanisms taking place. The fracture toughness was found to be lower than 400 J/m^2 for all film thickness (50, 175 and 500 μm). The goal is to determine whether adding fishing lines or not between the PEI film and the carbon fibres could be beneficial for the joint toughness.

For this purpose, fishing lines spaced by 5 mm have been inserted at both sides of 50 and 175 μm thick PEI films prior to co-curing. The fishing lines were deposited one by one and when there coated with resin to prevent their motion. The resulting R-curves are shown in Figure 5S2.1. Inserting fishing lines in the joint joints with the 50 μm film results in a steady-state fracture toughness about $837 \pm 50 \text{ J}/\text{m}^2$ which represent an increase by a factor 2.9. If fishing lines are inserted with the 175 μm thick film, G_{Ic} is about $464 \pm 55 \text{ J}/\text{m}^2$, an increase by a factor 3.1 compared to the joint without fishing lines. This toughening originates from the fishing lines acting as bridging ligaments behind the crack tip.

Inserting fishing lines into a co-cured composite joint with a PEI film results in toughening effect originating from the bridging induced by the fishing lines. No matter the thickness of the PEI film, the mode I fracture toughness is increased by a factor 3 by inserting fishing lines at both sides of the PEI film every 5 mm. For the following experiments, a 50 μm thick film has only be used as it leads to the highest fracture toughness. In addi-

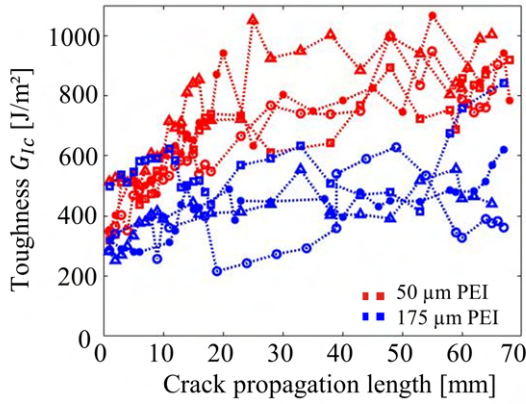


Figure 5S1.1 R-curves the co-cured composite joints with a PEI a 50 μm (red) and a 175 μm (blue) thick film in which fishing lines are inserted, spaced by 5 mm.

tion, this could also lead to the strongest joint as the tensile and lap-shear strength of joints are known to decrease with the adhesive thickness. The joint containing a 50 μm thick film thus probably offers the trade-off in term of toughness, stiffness and strength.

5.7.2 Supplementary material 2: Analytical bridging model

Governing equations

The considered configuration is the one of a double cantilever beam (DCB) of a thickness $2h$, width B and in which a crack of length a is present. In addition, toughening occurs by bridging due to the presence of n bridging ligaments spaced by a distance λ and that are placed at the L_i positions ($i = 1, 2, \dots, n$) as shown in Figure 5S2.1.

When the DCB opens, the bridging ligaments are deformed and carry each one a force F_i that depends on the beam deflection at the position $w_i(L_i)$ such that $F_i = F_i(w_i(L_i))$. This requires to define a bridging law. The equations describing the beam deflection are first derived based on the Euler-Bernoulli beam theory. The relationship between the beam deflection and the load for a simple DCB is given by

$$\frac{\partial w^2}{\partial x^2} = -\frac{M}{EI} = \frac{Px}{EI}, \quad (5S2.1)$$

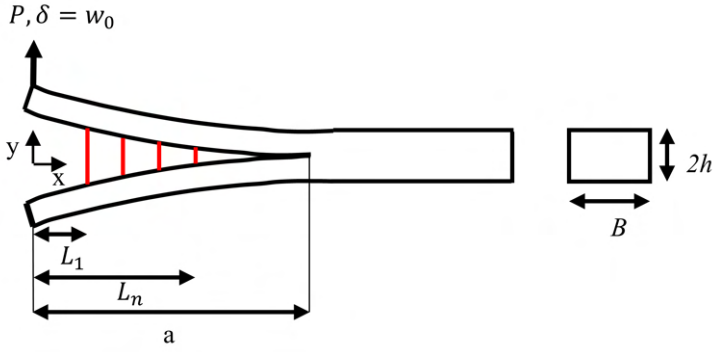


Figure 5S2.1 Double cantilever beam configuration with a width B , a height $2h$. A crack of length a is present with n bridging ligaments present in its wake, placed at the positions L_i .

with x being the position along the DCB with the origin corresponding to the loading point, M the bending moment resulting from the applied load P . E is the beam Young's modulus and I is the second moment of inertia equal to $Bh^3/12$. If n bridging ligaments are now considered, the deflection of the beam between two consecutive ligaments located in L_i and L_{i+1} ($i = 1, 2, \dots, n$) is given by

$$\frac{\partial w^2}{\partial x^2} = -\frac{M}{EI} = \frac{Px - \sum_{j=1}^i F_j(x - L_j)}{EI} \text{ for } L_i \leq x \leq L_{i+1}. \quad (5S2.2)$$

This leads to an expression for $w_i(x)$, the beam deflection equation for $L_i \leq x \leq L_{i+1}$:

$$w_i(x) = \frac{Px^3}{6EI} - \sum_{j=1}^i \frac{F_j}{EI} \left(\frac{x^3}{6} - \frac{L_j x^2}{2} \right) + C_i x + D_i. \quad (5S2.3)$$

If i is set to 0, the second term is equal to 0 and the deflection and the loading point $\delta = w_0$ is determined. The boundary equations needed to determine the C_i and D_i constants are

$$\begin{cases} w_n(a) = 0 \\ \frac{\partial w_n(a)}{\partial x} = 0 \\ w_i(L_{i+1}) = w_{i+1}(L_{i+1}) \\ \frac{\partial w_i(L_{i+1})}{\partial x} = \frac{\partial w_{i+1}(L_{i+1})}{\partial x} \end{cases}. \quad (5S2.4)$$

The two first boundary conditions allow to determine the C_n and D_n coefficients of w_n such that $L_n \leq x \leq a$

$$C_n = -\frac{Pa^2}{2EI} + \sum_{j=1}^n \frac{F_j}{EI} \left(\frac{a^2}{2} - l_j a \right), \quad (5S2.5)$$

$$D_n = -\frac{Pa^3}{6EI} - C_n a + \sum_{j=1}^n \frac{F_j}{EI} \left(\frac{a^3}{6} - \frac{L_j a^2}{2} \right). \quad (5S2.6)$$

The two last boundary conditions provide a generic formula for the other C_i and D_i coefficients for $i = 0, 1, 2, \dots, n-1$. $i = 0$ corresponding to the deflection equation for $x \leq L_1$.

$$C_i = C_{i+1} - \frac{F_{i+1} L_{i+1}^2}{2EI}, \quad (5S2.7)$$

$$D_i = D_{i+1} + (C_{i+1} - C_i) L_{i+1} + \frac{F_{i+1} L_{i+1}^3}{3EI}. \quad (5S2.8)$$

Equations 5S2.3, 5S2.5, 5S2.6, 5S2.7 and 5S2.8 are used to compute the beam deflection at the position of each bridging ligament. In order to determine the toughness by considering the action of the bridging ligament, one needs a crack propagation criterion. The total toughness is given by

$$G_{total} = G_0 + G_{bridging} \quad (5S2.9)$$

with G_0 being the fracture toughness in the absence of the bridging ligaments and $G_{bridging}$ the contribution of the bridging ligaments behind the crack tip. The crack propagates when the crack driving force at the crack tip G_{tip} (Equation 5S2.10) is equal to G_0 .

$$G_{tip} = \frac{1}{BEI} \left(Pa - \sum_{j=1}^n F_j (a - L_j) \right)^2 = G_0. \quad (5S2.10)$$

In order to determine the G_{tip} , P and the bridging forces F_j must be determined. Determining the F_j forces requires to determine the deflection of the DCB at the positions L_j , $w_j(L_j)$ as the bridging force and the deflection are linked together through the bridging law. A system of $n+1$ equations is obtained that can be solved using the Newton-Raphson method. The guess of iteration k is denoted $X_k = (W_{1k}, W_{2k}, \dots, W_{nk}, P_k)$ with W_{ik} being

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the guess of iteration k for the deflection in L_i while P_k is the guess for the applied load. The guess for the next iteration $k + 1$ is given by

$$X_{k+1} = X_k + \frac{G(X_{k+1})}{H'(X_{k+1})}, \quad (5S2.11)$$

where H is a real-value function of X_k and H' is the jacobian of H . H is given by

$$H(X_k) = \begin{pmatrix} W_{1k} - w_1(L_1) \\ W_{2k} - w_2(L_2) \\ \vdots \\ W_{kn} - w_n(L_n) \\ G_{tip} - G_0 \end{pmatrix}. \quad (5S2.12)$$

Convergence is achieved when the residuals $|X_{k+1} - X_k|$ are lower than a certain value ϵ . In the present case $|W_{ik+1} - W_{ik}| < 0.001$ and $|P_{k+1} - P_k| < 0.1$.

Bridging law

The bridging law relates the bridging force F_i to the deflection at the position of the bridging ligament $w_i(L_i)$. The distance between the two DCB arms at the position L_i is equal to $2w_i$. Hence, the bridging law gives $F(2w_i)$. It is well known that the bridging law influences the predicted DCB response. For example, Spearing and Evans [188] showed that if the area under the different bridging laws and the bridging ligament strength are kept constant, the final fracture toughness is also constant. They considered bridging laws corresponding to a perfectly plastic ligament, a linear elastic ligament and fibre pullout (softening). The increasing part of the R-curve is modified with the steady-state fracture toughness being reached first for the perfectly plastic law and then for the pull-out case and finally, the elastic ligament. In addition, the perfectly elastic and plastic laws result in an exponential increase of the toughness as the crack propagates while the increase is smoother when considering fibre pull-out. Li et al. [190] used a bilinear trapezoidal law and the showed that the ligament ductility can influence the response by impacting the snap-back instability and the resulting total fracture toughness. Four types of behavior have been considered here and are shown in Figure 5S2.2.

- Law # 1: The bridging ligaments behave as springs (Figure 5S2.2a). Each one has a stiffness K and breaks when the applied force reaches

the tensile strength F^f .

- Law # 2: The bridging ligament is first unfolded and starts to be loaded under tension when it reaches a length l_0 and breaks when the tensile force is equal to F^f (Figure 5S2.2b).
- Law # 3: The bridging ligament is attached to the DCB arms at the opposite interfaces close to the DCB edge. Depending on the radius of the bridging ligament and the width of the DCB, tensile or bending conditions dominate (Figure 5S2.2c and e).
- Law # 4: The bridging ligament is bonded to one arm except in one point where it is attached at the opposite interface and decohesion between the ligament and the two adherends take place over a distance d . Depending on this distance and the ligament radius, bending or tension dominates (Figure 5S2.2d and f).

□ Bridging law # 1: springs (Figure 5S2.2a)

The bridging ligaments act as springs. The force linearly increases with the applied displacement and they break when the applied force is equal to the bridging ligament tensile strength F^f and when their final extension is equal to l^f . The stiffness of the ligament is given by $K = F^f/l^f$. The bridging law is thus written as

$$F_i(2w_i) = \begin{cases} 2Kw_i & 0 \leq 2w_i \leq l^f \\ 0 & l^f \leq 2w_i \end{cases} \quad (5S2.13)$$

□ Bridging law # 2: elongation and tension (Figure 5S2.2b)

This bridging law considered that the bridging ligament is loaded under tension when it has a length l_0 meaning that a bridging force is applied on the ligament when $w_i \geq l_0$. For $w_i < l_0$, the ligament is unfolded. The ligament breaks when the applied force is equal to the bridging ligament tensile strength F^f and when their final extension is equal to l^f . If the ligament is perfectly elastic, l^f depends on the ligament Young's modulus E^f and is given by $l^f = \left(1 + \left(F^f/AE^f\right)\right) l_0$ with A being the fishing line cross-section area. This would require to have a curve similar to the dashed one shown in Figure 5S2.2, but convergence issues appear due to the sharp increase in stiffness when $w_i = l_0$. Consequently, it is assumed

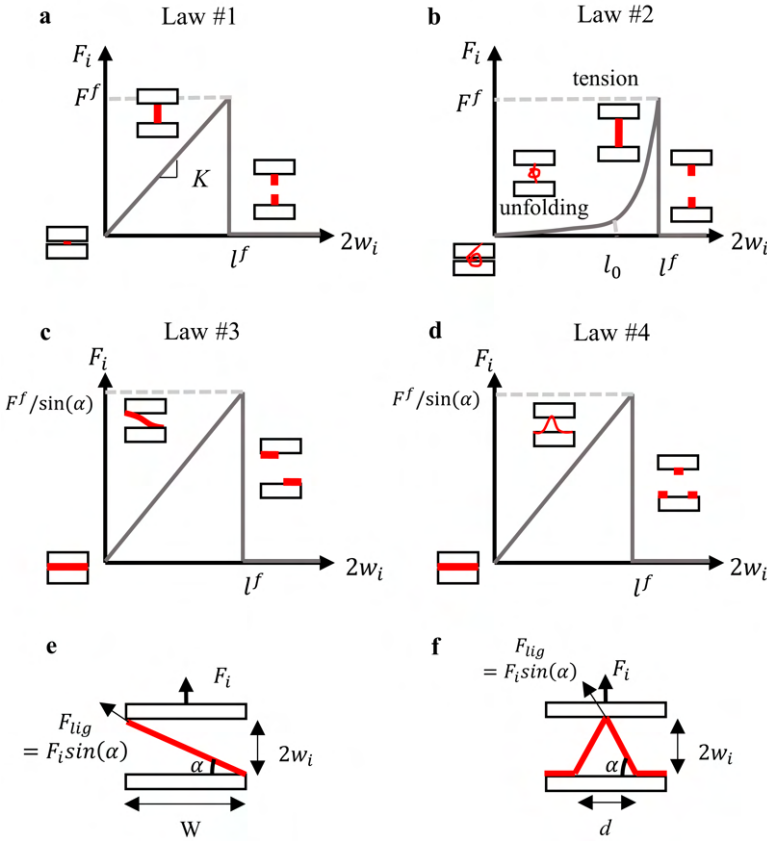


Figure 5S2.2 Bridging laws: a) the ligament behaves as a spring, b) the ligaments first unfold before being loaded under tension (dashed-curve: ideal case; plain curve: modelling), c) the ligament is attached at the edges of the DCB, d) decohesion takes place over a short distance d while the debonded part adheres to the opposite interface. e-f) Bridging ligament configuration for e) the law in c) and f) the law in d).

that it slightly increases during unfolding and the ideal bridging law is approximated by an exponential function such that $F_i(w_i = l_0) < 0.1F^f$. The bridging law is thus given by

$$F_i(w_i) = \begin{cases} b_1 \exp(2b_2 w_i) & 0 \leq 2w_i \leq l^f \\ 0 & l^f \leq 2w_i \end{cases}, \quad (5S2.14)$$

with b_1 and b_2 being two fitting coefficients.

- Bridging law # 3: ligament attached at opposite interface close to the edges (Figure 5S2.2c)

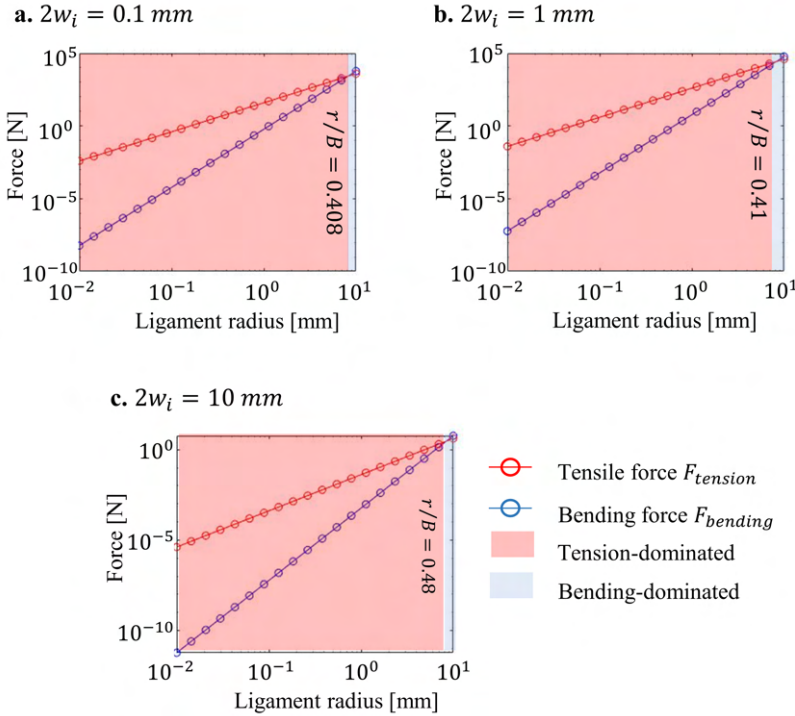


Figure 5S2.3 Variation of $F_{bending}$ and $F_{tension}$ for the bridging Law # 3 with the bridging ligament radius for different displacements a) 0.1 mm, b) 1 mm and c) 10 mm. The width B is set to 20 mm and the ligament Young's modulus to 5 GPa. The r/W ratio at which the transition from the tension-dominated regime to the bending dominated regime takes place is given in each graph.

The bridging ligament is considered to be attached to the DCB beams only at the edges of the DCB. Along the width of the DCB, decohesion takes place. Depending on the radius of the bridging ligament and the width, bending or tension dominates. Under small deformation, the force re-

quired to bend the ligament is given by

$$F_{bending}(L_i) = \frac{3E\pi r^4(2w_i)}{B^3}, \quad (5S2.15)$$

while the tensile force applied on the ligament is given by

$$F_{tension}(L_i) = \frac{1 - \cos(\alpha)}{\cos(\alpha) \sin(\alpha)} E\pi r^2, \quad (5S2.16)$$

with α being the angle between the bridging ligament and the DCB arm given by $\alpha = \arctan(2w_i/B)$. Figure 5S2.3 shows the evaluated $F_{bending}$ and $F_{tension}$ for several values of radius r and displacement values $2w_i$. The transition from the “tension dominated” mode to the “bending dominated” mode takes place when r/B is about 0.4 for small deflections and increases as the deflection increases. This transition ratio is independent on E^f which only affects the magnitude of $F_{bending}$ and $F_{tension}$.

Assuming that tension dominates (thin bridging ligaments), it is possible to determine the force required to cause the ligament failure so that $F_{tension}(w_i) \sin(\alpha) = F^f$ which also provides the beam deflection at failure l^f .

□ Bridging law # 4: local decohesion (Figure 5S2.2d)

The fourth bridging law assumes that the bridging fibre debonds from the bottom arm over a distance d and is attached at its middle to the upper arm as shown in Figure 5S2.2d and f. Under small deformation, the force required to bend the bridging ligament is given by

$$F_{bending}(L_i) = \frac{48E\pi r^4(2w_i)}{d^3}. \quad (5S2.17)$$

The force applied on the DCB if tension in the bridging ligament dominates is given by

$$F_{tension}(L_i) = 2 \frac{1 - \cos(\alpha)}{\cos(\alpha) \sin(\alpha)} E\pi r^2, \quad (5S2.18)$$

with $\alpha = \arctan(4w_i/d)$, the angle between the bridging ligament and the bottom DCB arm. Figure 5S2.4 shows the evolution of $F_{bending}$ and $F_{tension}$ with the fibre radius and the displacement. It can be observed that the transition from a tensile-dominated regime to a bending-dominated regime always occurs for a r/d ratio about 0.2. If tension is assumed, the

force required to apply failure can be determined based on $F_{tension}(w_i) = 2 \sin(\alpha) F^f$ from which the deflection at failure l^f can be determined.

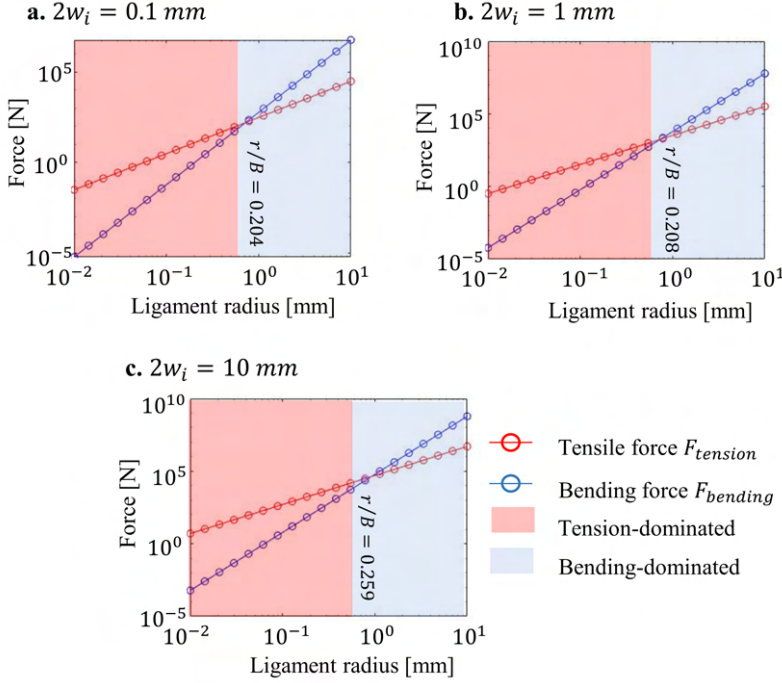


Figure 5S2.4 Variation of $F_{bending}$ and $F_{tension}$ for the bridging Law # 4 with the bridging ligament radius for different displacements a) 0.1 mm, b) 1 mm and c) 10 mm. The distance d is set to 5 mm and the ligament Young's modulus to 5 GPa. The r/W ratio at which the transition from the tension-dominated regime to the bending dominated regime takes place is given in each graph.

Numerical algorithm

The algorithm is schematized in . The inputs of the models are the DCB beam Young's modulus, thickness and width, the spacing between the bridging ligaments λ , the initial crack length a_0 and the final crack length for computations a_{max} , the position of the first bridging ligament L_1 and the joint toughness in the absence of bridging G_0 . In addition, depending on the selected bridging law, several inputs can be required such as the ligament Young's modulus, radius, tensile strength, tensile elongation or ini-

tial unfolded elongation. Then for each crack length a_m , the position of all potential bridging ligaments L_i are determined based on L_1 . The Newton-Raphson algorithm is then used to determine the macroscopic load P_m and the $w_i(L_i)$ deflections. When it has converged, the algorithm checks if all ligaments are still unbroken or not. If not, L_1 is modified such that it corresponds to the position of first intact ligament. The same computations are performed for each a_m ranging from a_0 to a_{max} . Finally, knowing the macroscopic load P_m for each value of a_m , the toughness is computed based on the Euler-Bernoulli beam theory:

$$G_{Ic} = \frac{12P_m^2 a_m^2}{EB^2h^3} . \quad (5S2.19)$$

The equation used to compute the toughness has an influence on the R-curve. Li et al. [190] compared the compliance method and the single beam theory. Using the compliance method, G_{Ic} is constant until the ligament starts to be damaged and G_{Ic} increases until the ligament breaks. Using the single beam theory, as soon as the ligament is active, G_{Ic} increases until the ligament fails.

Influence of the bridging law on the results

The four bridging laws introduced in Section 5.7.2 are compared in term of the DCB response. However, one must determine how the different parameters will be adapted depending on the bridging law to obtain some equivalents. For this purpose, 5 cases have been analyzed and are given in Table 5.2. For every bridging law one considers a DCB having an initial crack length of 50 mm and the only considered bridging ligament is located at a position $L_1=60$ mm.

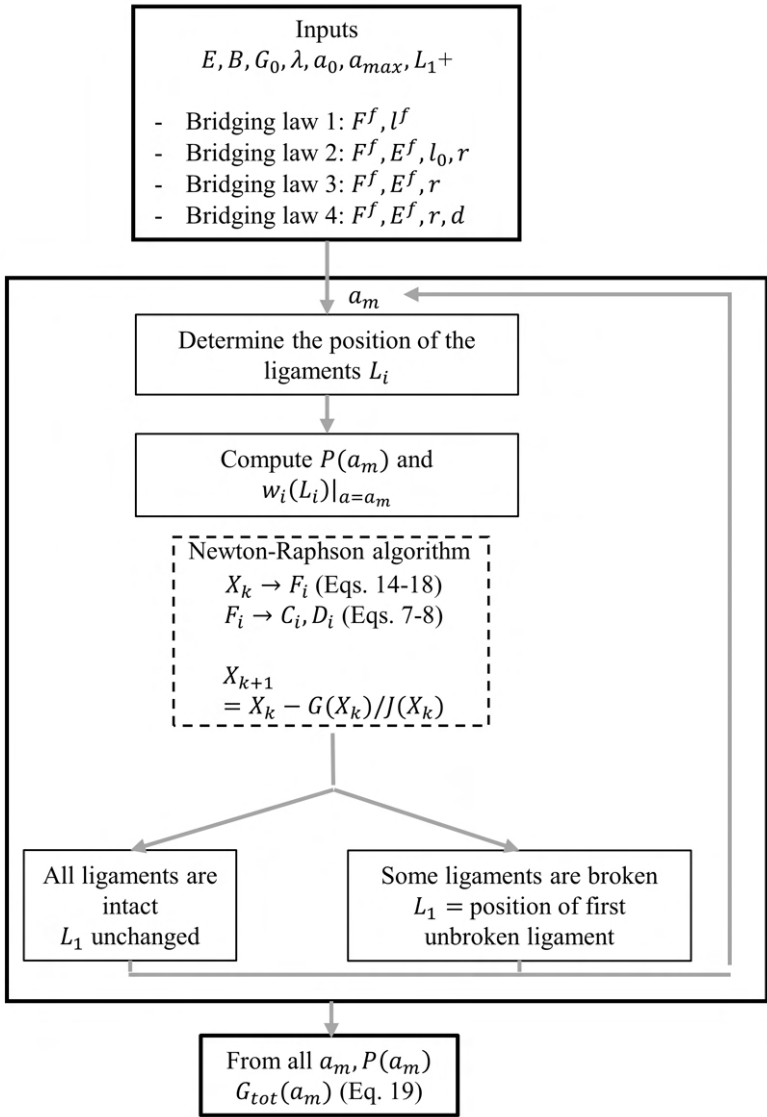


Figure 5S2.5 Algorithm for computing the joint fracture toughness considering the presence of bridging ligaments.

Table 5.2 Parameters considered for the different cases to determine the influence of the bridging law and the resulting fracture toughness.

Case number	Law	Parameters	Energy to break one ligament [mJ]	Maximum total fracture toughness
1	Bridging law #1 - same extension as for law #3	$F^f = 30 \text{ N}$ $l^f = 12.93 \text{ mm}$	193.95	1100 J/m ²
2	Bridging law #1 - same energy as law #2 (case 3)	$F^f = 30 \text{ N}$ $l^f = 3.09 \text{ mm}$	46.35	625 J/m ²
3	Bridging law #2 - same extension as law #3 (case 4)	$F^f = 30 \text{ N}$ $E^f = 5 \text{ GPa}$ $r = 0.1 \text{ mm}$ $l^f = 12.93 \text{ mm}$	46.35	625 J/m ²
4	Bridging law #3	$F^f = 30 \text{ N}$ $E^f = 5 \text{ GPa}$ $r = 0.1 \text{ mm}$	355.58	1800 J/m ²
5	Bridging law #4	$F^f = 30 \text{ N}$ $E^f = 5 \text{ GPa}$ $r = 0.1 \text{ mm}$ $d = 5 \text{ mm}$	169.95	1600 J/m ²

These cases can be compared to one another. Comparing case 1 and case 2, the energy dissipated by the deformation and breaking of the ligament is larger in case 1 than in case 2 due to the higher tensile elongation in case 1, resulting in a higher toughness. The bridging ligaments of cases 1 and 3 are characterized by similar properties. However, case 3 consider unfolding of the bridging ligament resulting in lower dissipated energy and lower G_{Ic} as consequence and a smoother R-effect. For cases 2 and 3, the resulting toughness is similar as the same energy is dissipated by the breaking of the bridging ligaments. However, the ligaments are not characterized by the same tensile elongation. In case 2, the tensile elongation is lower and the toughness raises faster than in case 3. The tensile elongation influences thus the distance over which the R-effect is taking place. Case 4 can be compared to case 1 as the fibres are characterized by similar properties. However, in case 1, the force applied on the fibre only acts parallel to it while in case 4, it acts at angle α with respect to the horizontal. Hence, the tensile force acting on the fibre is lower than the macroscopic load. Consequently, the macroscopic load can be increased up to 55 N resulting in more energy dissipated by the bridging ligament and a higher fracture toughness. However, in both cases, the maximum fracture toughness is reached for a similar crack length which governs the beam deflection at the ligament position. In cases 4 and 5, a lower amount of energy is dissipated by the ligament in case 4 and its tensile elongation is much lower resulting in a lower G_{Ic} and a steeper R-effect, in agreement with the previous comparisons. For case 5, the decohesion length d influences the results as the shorter d , the lower the tensile elongation but the macroscopic load at failure is not affected. Increasing d results thus in a higher fracture toughness. These examples showed that to be able to extrapolate or fit experimental results using a bridging model, three main parameters are important:

- The energy dissipated by one fibre: for a given F^f , similar G_{Ic} values are attained for a given amount of energy dissipated by bridging.
- The tensile elongation of the fibre controls the crack propagation length over which G_{Ic} increases, the shorter, the steeper the increase.
- The bridging event: depending on the bridging phenomenon (law), the two above mentioned parameters are influenced.

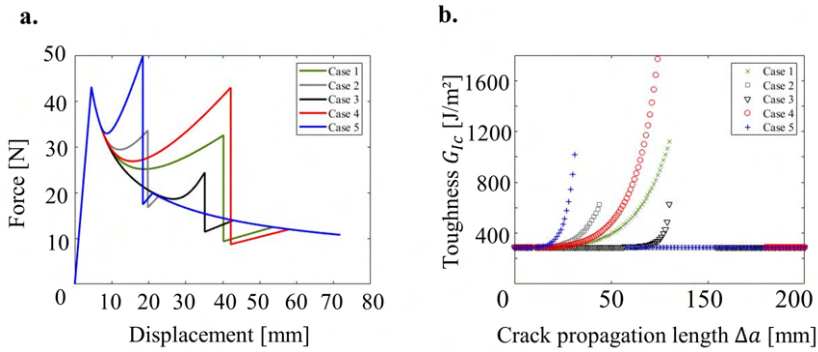


Figure 5S2.6 a) Force-displacement curves and b) R-curves for the five cases of Table 5.2.

5.7.3 Supplementary material 3: Identification of fitting parameters of experimental results

The experimental results for the joints made of *Caperlan TX4 60 μ m* fishing lines spaced by 1 to 10 mm are already shown in Figure 10. Figures 5S3.1 and 5S3.3 show the fitting of the experimental results for the other fishing lines spaced by 1 and 2 mm, respectively.

The parameter l^f is also used to fit the experimental fracture toughness. Although, a fit of the R-behaviour is not really observed. For all conditions, the tensile elongation is almost the same and ranges between 0.1 and 0.3 mm, indicating that the deposition method using the spikes board leads to reproducible results in this case while for the hand lay-up technique, a larger scattering is observed as observed in Figure 5.11 when comparing the l^f ranges. In addition, hand lay-up deposition results in longer bridging ligaments. Hence, the deposition method could have an influence on the l^f parameter, but this parameter remains difficult to control.

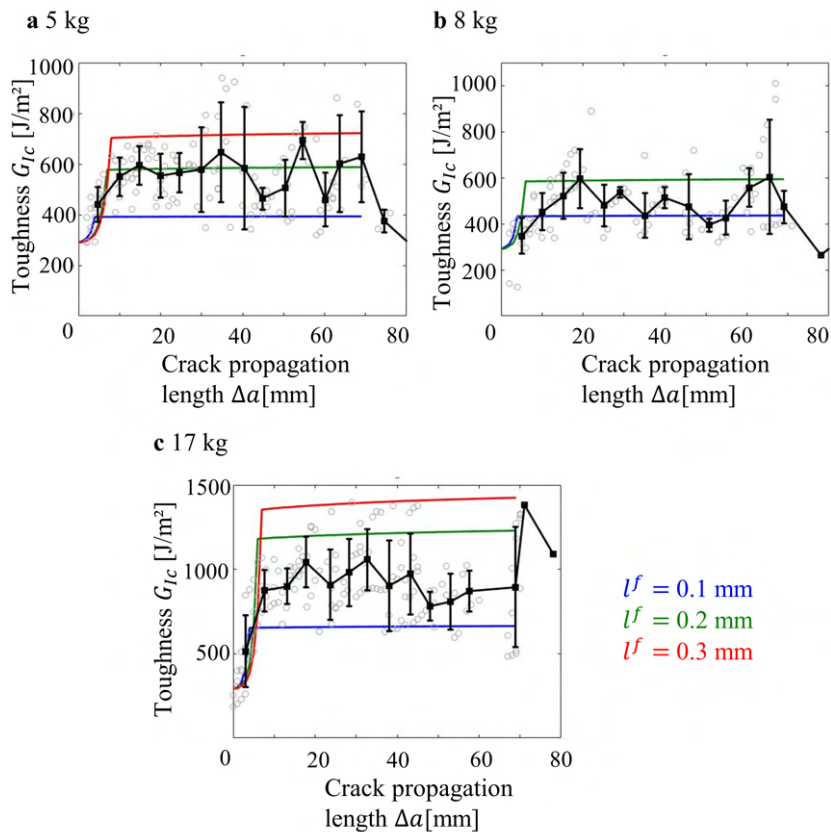


Figure 5S3.1 Fitting of experimental results (grey dots and dark line) using the 1st bridging law for different a spacing of 1 mm and tensile strengths of a) 5.7 kg, b) 8.3 kg and c) 17.2 kg.

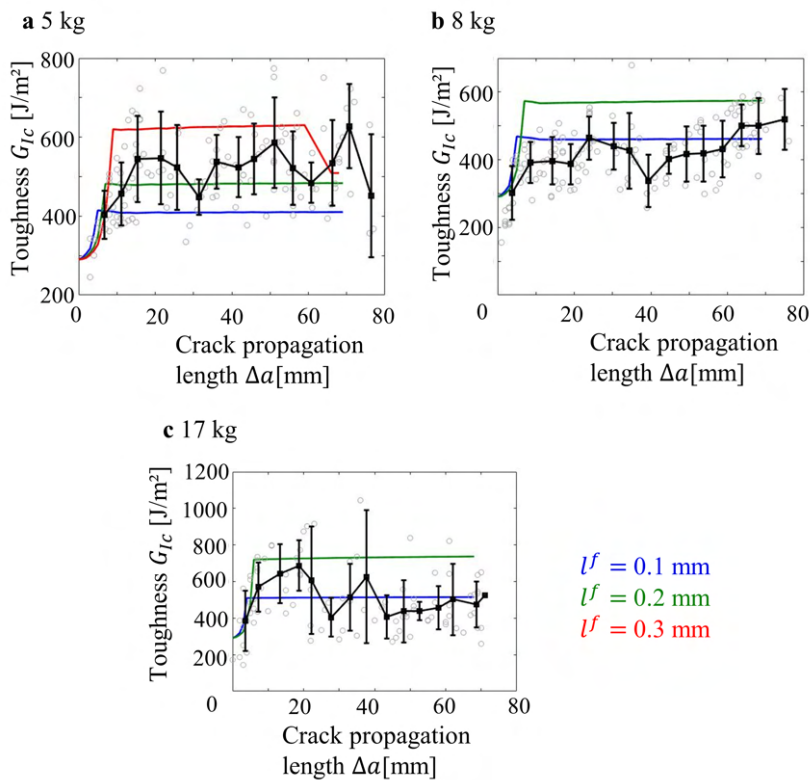


Figure 5S3.3 Fitting of experimental results (grey dots and dark line) using the 1st bridging law for different a spacing of 2 mm and tensile strengths of a) 5.7 kg, b) 8.3 kg and c) 17.2 kg.

6

Thermoplastic film architected with channels



This chapter is based on the article published in *Composites part A: applied manufacturing and sciences*: Charline van Innis, Michal K. Budzik and Thomas Pardoën, *Ultra-tough architected adhesive joints for integrated composite processing and bonding*, Comp. A, February, 2024.



This chapter is also linked to the patent application number EP23161330.8. (C. van Innis, T. Pardoën, *Film for joining parts*, EP23161330.8)

6.1 Introduction

Carbon fibre reinforced polymer composites (CFRPs) are broadly used in advanced structures such as in airplanes, wind turbine blades or marine structures, owing to a high strength and stiffness to weight ratio. Manufacturing these structures often requires assembling CFRPs together or with other materials. Depending on the application, the adhesive thickness can vary from 100 μm (e.g. aeronautic) to several centimeters (e.g. windmills or marine structures) [7]. Primary load carrying structures are frequently joined through mechanical fasteners, altering the composites structural integrity [1]. Adhesive bonding offers more uniform stress distribution and additional weight saving. However, limited resistance to crack propagation and long sequential bonding processes remain two barriers for more intensive use of this technology [1].

Adhesive bonding is a time and labour-intensive process involving careful surface treatment to reach acceptable mechanical performances [133]. Adherends join upon curing of the adhesive under clean and controlled environment. Co-curing is an attractive alternative enabling the simultaneous bonding and composite curing, without an adhesive, hence reducing the number of steps and removing the need for surface treatments. Integrated manufacturing of joints can be achieved through classical composite processes such as resin transfer moulding (RTM), i.e. the interface between the two adherends can be seen as the bondline, made of a thin resin matrix layer. However, even high performance epoxy resins such as RTM6 [167, 193] are significantly more brittle than the best adhesives, characterized by a fracture toughness typically lower by one order of magnitude. This sets a major challenge for the development of tough joints.

Several strategies have been proposed to enhance the crack propagation resistance of adhesive joints. Figure 6.1a shows a double cantilever beam (DCB) joint made of two composite adherends (gray), a “bondline” (blue) and two interlayers (yellow) corresponding to a thin surface layer of polymer matrix on the composite adherends. Many failure scenarios can be anticipated based on which class of toughening mechanism is activated. Enhanced plastic dissipation is the primary known mechanism to toughen adhesive joints [94]. Plastic dissipation (Figure 6.1-i) is dependent upon adhesive thickness [13, 93, 99, 102]. In the presence of a soft layer (Figure 6.1-ii) adjacent to the layer where crack propagates, plastic dissipation

can take place, enhancing the fracture toughness [79, 97].

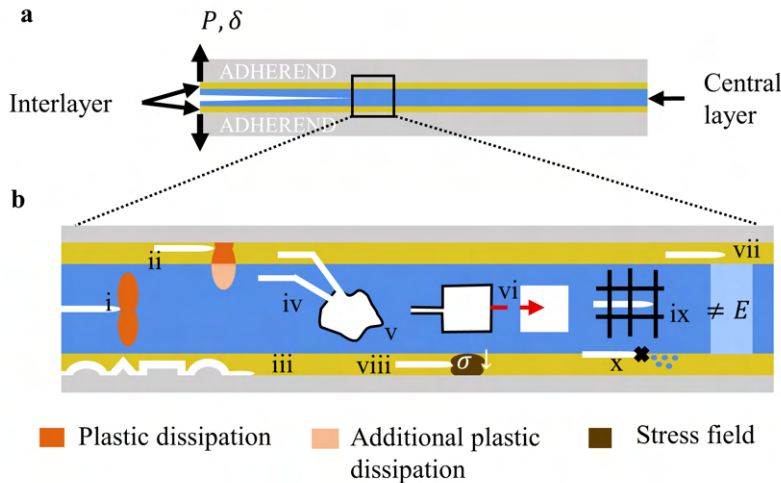


Figure 6.1 Schematic illustration of (a) a DCB joint with the composite adherends (gray), central bondline (blue) and two interlayers (yellow) on both sides, and (b) of the toughening mechanisms that can take place in the joint depending on the cracking mechanism – failure scenario. (i) Plastic dissipation, (ii) additional plastic dissipation due to soft layer, (iii) structured interface, (iv) crack deflection by cavity and (v) subsequent trapping and blunting, (vi) pillar interfaces, (vii) local stiffness variation, (viii) shielding by a cavity, (ix) crack bridging by a mesh and (x) tough interface due to polymer chemical interactions.

Crack deflection and zigzagging requires more energy than straight mode I cracks as a result of the introduction of a mode II component (Figure 6.1-iii). Sinusoidal interfaces [106, 107] and squared interfaces [80] have been designed to increase the crack surface area. Adding surface features [194, 195] and modifying surface roughness [22, 110] also improves mechanical interlocking. In homogeneous materials, crack deflection can also be triggered through stiff particles [196] or cavities [197] (Figure 6.1-iv). The influence of the relative position of pores with respect to the crack leads to crack trapping and anti-shielding (i.e. an effective G_{Ic} lower than the local material G_{Ic}) if located ahead of the crack tip.

Crack trapping by cavities leads to crack tip blunting [121–123] (Figure 6.1-v) with a positive effect on fracture toughness as the crack must reinitiate

to further propagate. If the cavities are large with respect to the layer thickness and close to each other, the joint may behave as pillar interfaces [141] (Figure 6.1-vi). More generally, metamaterials can be used as bondline to improve the fracture toughness [84].

Varying the local properties of the adherends or of the adhesive is one additional toughening strategy (Figure 6.1-vii). Morano et al. [82] designed adherends with channels (Figure 6.1-viii) to improve crack propagation resistance, involving parameter optimization. The adherend-adhesive interface strength through surface treatment can also be locally varied to force crack meandering from one weaker zone to another [138, 198, 199]. This further leads to bridging ligaments by alternating the strong interface zones [123, 135]. Crack bridging proved to be a very efficient toughening mechanism. Thermoplastics veils [130, 200] are, for these same reasons, inserted in composites to improve the interlaminar fracture toughness. Metallic [123] and polymeric [81] meshes in adhesive joints can be inserted to emulate this mechanism (Figure 6.1-ix).

The first major challenge addressed in this study is to simultaneously activate most of the toughening principles to anticipate all possible failure scenarios and avoid a weak link option for crack propagation, and to reach joint toughness values unattainable with homogeneous joints. A key element to implement this vision is to consider the strength of the different interfaces which are: (i) the composite resin – carbon fibres interface for the interlayer-composite interface, (ii) the interface between the interlayer and the central bondline, i.e. between two polymers. For the latter, chemical interactions between compatible polymers can lead to tough interfaces [5]. Polyamide (PA), polyetherimide (PEI), polyethersulfone (PES), polycarbonate (PC) and polyetheretherketone (PEEK) are good candidates to promote a tough interface (Figure 6.1-x). The second objective is to integrate the bonding step into a seamless manufacturing process. This implies insertion of the architected bondline at the midplane of the composite before resin injection. The architecture is designed to trigger most of the toughening mechanisms addressed above for all possible scenarios of crack propagation. To embody this vision, the core novelty has been the design of a bondline made of 3D printed Nylon (PA) thin bands with regularly spaced channels, which consists of a tough polymer but sufficiently soft (owing to the presence of cavities). The 3D printed film is shown in Figure 6.3. Upon injection, two extreme joint configurations can be produced with either (i) all channels filled by resin or (ii) all left empty depend-

ing whether the channels are left open or not at the edges of the film. In addition, some intermediate configurations are obtained with (iii) the filling of only a fraction of the channels. In case (i), crack deflection is prevented, but local variation of properties can lead to crack trapping. Case (ii) can potentially lead to the behaviour observed for pillar interfaces when the crack propagates in the central layer [141]. Case (iii) permits a combination of the two modes. As the mechanical resistance under shear loading is also important in applications, it will be addressed by finite element modelling to determine the influence of architecture. Eventually, the Ashby's material selection approach [201] is used to analyze the trade-off between toughness and shear resistance.

6.2 Materials and methods

6.2.1 Joints manufacturing

Figure 6.2 schematically outlines the bonding process from the architected film printing to final joint (see Appendix B for a description of the full process). The Nylon structures, layer height of 100 μm , are 3D printed using NylonWhite material (supplied by Markforged) on a MarkTwo machine with the default/optimized manufacturer settings. The design rules for adherends with channels set by Morano et al. [82] based on finite element simulations are used to select the height (H), the width (W) and the spacing (S) between the cavities. The total bondline thickness is referred as t . These authors found that G_{Ic} is maximized if H/t has the largest possible value (0.65 in their study), if W/H is equal to 1.25 and if S is equal to W . In addition to these rules, the structure thickness is set by the available RTM mould. The dimensions are: thickness of 1.4 mm, cavity height and width of, respectively, 1 mm and 1.2 mm and spacing between cavities is equal to 1 mm. The 3D printed structure, shown in Figure 6.3a, is inserted at the midplane of the composite in the mould after visual inspection. The two composite adherends are made of four plies of H5 satin fabric made of carbon fibres each (HexForce G0926 supplied by Hexcel). At the midplane, in addition to the architected Nylon tape, a non-adhesive polyimide (Kapton) film is inserted to create a pre-crack required for DCB fracture testing. Once the RTM mould is closed by a pressure of 8 bar, the epoxy resin RTM6 from Hexcel is injected at a temperature of 90°C. Then, the temperature is increased up to 160°C at a rate of 2°C/min and this temperature is held for 90 minutes before increasing it up to 180°C at the same rate, with a

post-curing duration of two hours. During injection, the pressure inside the mould is increased and maintained to 6 bar. Upon curing, the channels have a height of 0.8 mm and a length of 1.1 mm, with 1 mm spacing (Figure 6.3d).

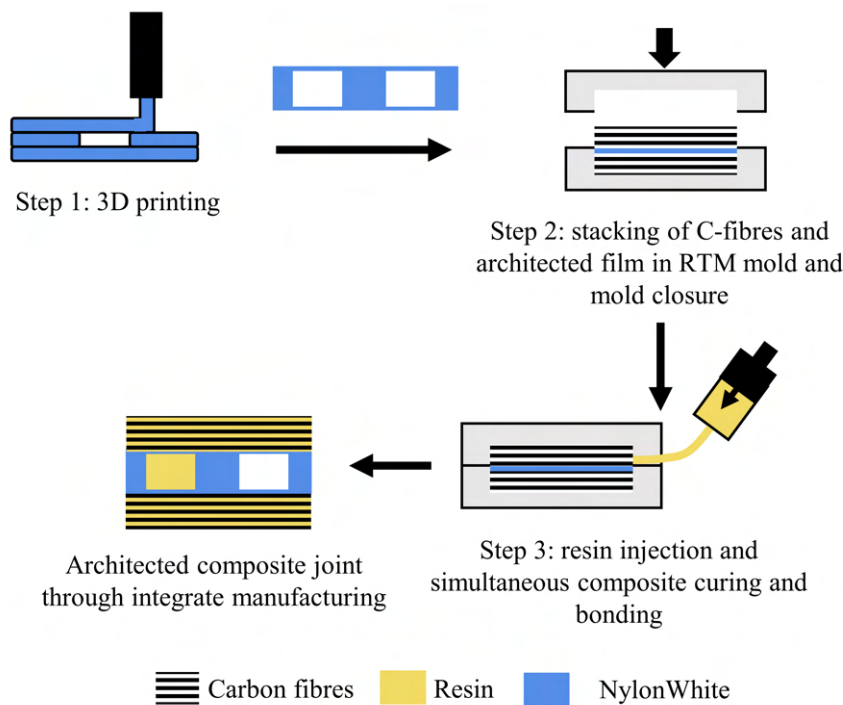


Figure 6.2 Manufacturing steps for architected joints from 3D printing of the architected Nylon part to the unmoulding.

6.2.2 Mechanical testing

The DCB samples, shown in Figure 6.3a, have been machined from the cured panels with a length of 200 mm and a width of 20 mm. The 70 mm long Kapton film inserted to create the precrack results in initial crack length of 50 mm. The total height of the samples is about 4.4 mm including the structured insert of 1 mm. The DCB tests are performed following the ISO 25217 standard on a Zwick/Roell machine equipped with a 50 kN loadcell, at constant cross-head speed of 1 mm/min. The crack tip position is tracked using Digital Image Correlation (DIC) using two cameras and

the Istra4D software. For the DIC analysis, the facet size and the grid spacing are set to 21 and 7 pixels, respectively. G_{Ic} is then computed following the single beam theory. As large deflections take place, the corrected beam theory and the experimental compliance method should be preferred but cannot be applied in the present case due to the stick-slip behaviour [172] which introduces some uncertainty on the exact value of G_{Ic} .

Tensile tests are performed, using the same mechanical testing machine and settings as before, on flat dogbone NylonWhite specimens to determine the tensile properties. The specimens shown in Figure 6.3e have a thickness of 2 mm and width of 5 mm along the 30 mm long gauge section. Extensometers with an initial spacing of 20 mm are used to determine the strain.

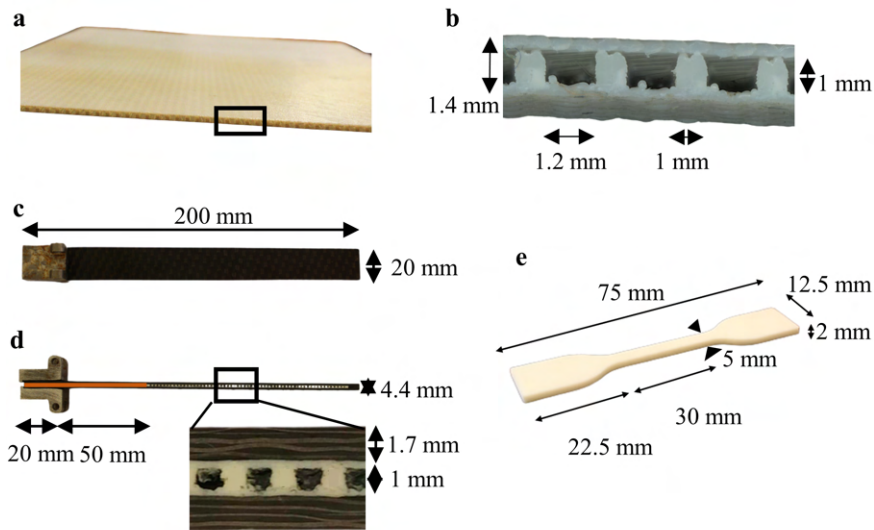


Figure 6.3 Nylon film and specimens with their dimensions: (a) Nylon film after 3D printing, (b) zoom on the edge of the Nylon film with the dimensions of the cavities after printing, (c) DCB specimen from the top, (d) lateral view of the DCB specimen and zoom on the film after bonding, (e) tensile specimen.

6.2.3 Fractography

For fractography analyses, scanning electron microscope (SEM) and optical microscope have been used. An Ultra 55 Field-Emission SEM (Zeiss,

Germany) has been used to analyze the fracture surfaces with a voltage of 5 keV. An Olympus Provis AX70 optical microscope has been used to analyze fractured joints. This required to cut the joints along the crack propagation direction and to polish the cross-section prior to observation.

6.3 Finite element modelling

Two finite element (FE) models are developed. The first one is for the DCB configuration and is used to investigate the failure mechanisms taking place in the architected joints and observed experimentally. The second one addresses the lap-shear joint configuration to investigate the influence of the architected film and of the position and fraction of filled /empty cavities in the joint on the shear properties.

6.3.1 Double Cantilever Beam configuration

The DCB specimen with 100 mm length, 50 mm pre-crack and 20 mm width is modelled using the commercial FE code Abaqus (Dassault Systemes, France). A 2D model is created with four-nodes plane-strain elements with reduced integration (CPE4R) to perform computations under static conditions. Different configurations are modelled by varying the channels left empty or filled by resin to determine how the fracture toughness is affected when varying some key parameters based on a failure criterion of the type “maximum principal stress at a critical distance”. Six cavities are considered, but only the state of the third, fourth and fifth cavities in Figure 6.4a is changed. If a joint made of only empty cavities is modeled, 12 channels are considered in the model. The initial crack is introduced as a seam and the mesh is refined around the crack tip with the smallest elements having a length of 5 μm . Different meshes have been tested to check the convergence of the results in terms of mesh refinement.

Regarding the material properties, the Nylon tensile test data of Figure 6.4c and data from previous experimental campaigns of our laboratory are used for the epoxy resin and the composite (Figure 6.4c-d). The composite is characterized by an in-plane Young’s modulus of 70 GPa, an out-of-plane modulus equal to 3 GPa and a Poisson ratio equal to 0.3 [167]. Nylon is assumed to be isotropic although the properties depend on the printing direction. The epoxy resin behaviour is modelled with isotropic linear elasticity and with the Drucker-Prager yield criterion for the plastic response,

see Morelle et al.[168].

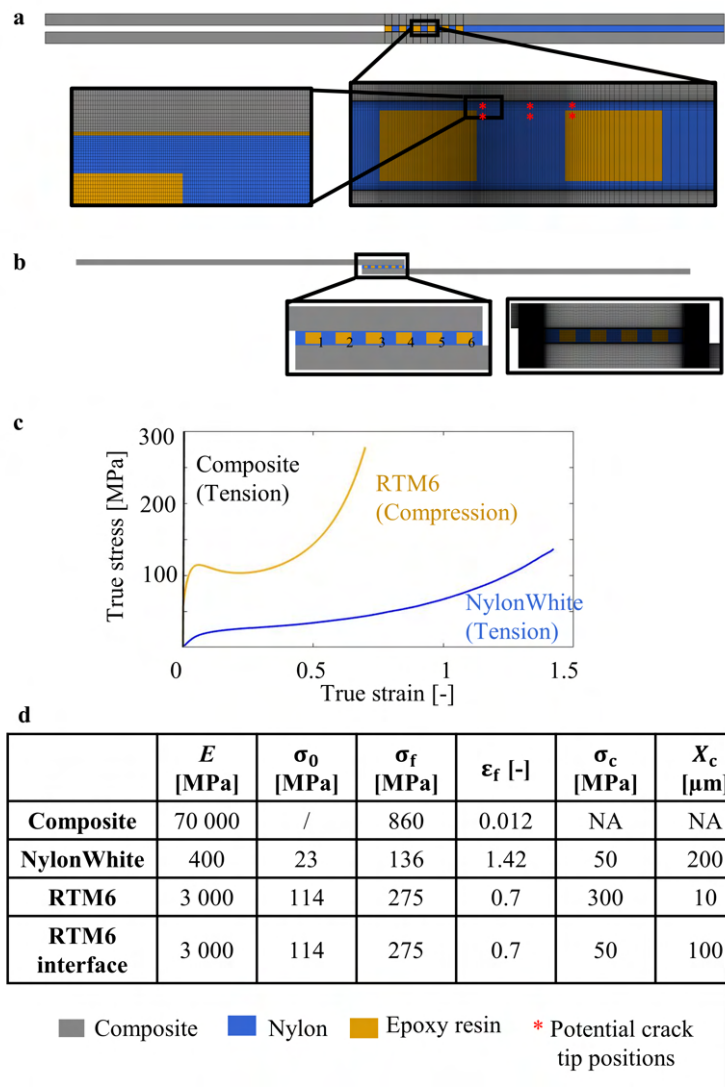


Figure 6.4 Geometries of the finite element model and corresponding mesh: (a) Double Cantilever Beam configuration and (b) Lap-shear configuration. (c) Engineering stress-strain curve data for NylonWhite, RTM6 and the composite used to model the Nylon film. (d) Properties of the materials and parameters of the failure criterion.

The failure criterion requires the determination of a critical stress σ_c associated to a critical distance X_c for each material and interface. Such a two-parameter criterion has been shown to better capture thickness effects than cohesive zone models in adhesive joints [92]. The crack is assumed to propagate in the channels filled by RTM6 resin when the maximum principal stress is equal to 300 MPa over a distance of 10 μm , as identified in a former study [193]. Although the tensile strength of the Nylon is equal to 136 MPa (when tested in the longitudinal direction, see Figure 6.4c), a value of 50 MPa is used for the critical stress in this case. This low value is justified by the significant anisotropy in the fracture resistance associated to the 3D printing process which potentially leads to much smaller cracking resistance in the direction orthogonal to the printed layers. Furthermore, a larger critical stress leads to extreme deformation close to the crack tip and to convergence issues before fulfilling the criterion. Hence, the choice of the 50 MPa value at a distance of 200 μm for the printed Nylon is much more phenomenological (and based on extrapolating data from Kinloch and Williams [120]) than the values chosen for RTM6 which were based on a systematic analysis of the fracture mechanisms [193]. The interface with the composite is assumed to fail when the maximum principal stress is equal to 50 MPa over a distance of 100 μm . When one of the failure criteria is met, the corresponding applied load P is extracted in order to determine G_{Ic} using the ISO 25217 standard [172]:

$$G_{Ic} = \frac{4P^2}{EW^2} \left(\frac{3a^2}{h^3} + \frac{1}{h} \right) \quad (6.1)$$

and the orientation of the maximum principal stress is also extracted in order to determine the crack propagation direction. The algorithm is shown in Figure 6.5.

6.3.2 Single lap-shear joint

A lap joint shown in Figure 6.4b made of the architected film comprising six cavities that can be either filled or left empty is modelled. The adherends length, width and height are set to 100 mm, 20 mm and 1.7 mm, respectively. The channels are symmetrically distributed. A 10 μm thick resin layer is modelled between the composite adherends and the architected film. As for the DCB simulations, CPE4R elements are used as well as the same material constitutive laws as described earlier. In addition, the fraction and position of the filled cavities are varied. The shear strength is

Step 1: Definition of the model (see Figure 6.4)

Step 2: Model running (Abaqus)

Step 3: Post-processing

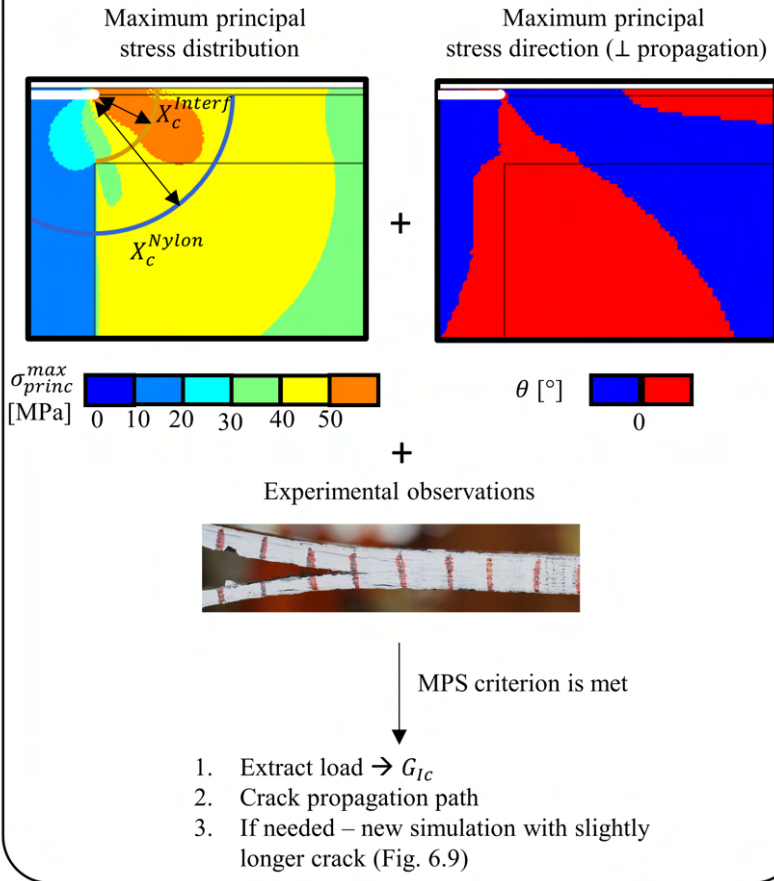


Figure 6.5 Finite element procedure implemented to determine the fracture toughness corresponding to given failure scenario and the crack path.

computed when one of the failure criteria described above is met.

6.4 Results and discussion

6.4.1 Mode I fracture toughness

The experimental crack resistance curves are given in Figure 6.6a while Figure 6.6b shows a typical crack resistance curve giving the variation of mode I critical energy release rate, G_{Ic} , as a function of crack growth Δa . Three regimes of G_{Ic} are distinguished. The configurations associated to these regimes is shown in Figure 6.6c, as well as the G_{Ic} values predicted by the FE model. Regime (1) is characterized by G_{Ic} between 600 and 1200 J/m² (precisely 828 ± 157 J/m²). Regime (2) corresponds to 1500 to 2500 J/m² (precisely 1704 ± 422 J/m²) while the largest G_{Ic} are above 3000 J/m² (precisely 4267 ± 846 J/m²) up to exceptionally high values larger than 5000 J/m² in regime (3). For comparison, the interlaminar toughness of composite made of G0926 carbon fibres fabrics and RTM6 is ≈ 350 J/m². Adhesives used in aerospace such as FM300 or EA9695K05 have G_{Ic} in the range 800 to 1000 J/m² [27]. The two best regimes exhibit a resistance 1.5 to 5.5 times of classical adhesives while being produced in a single co-curing step. Even the lowest Regime (1) results in resistances higher than the interlaminar toughness. Independent or not of the presence of a weaker regime, ultimately, crucial for crack propagation resistance is the presence of a strong crack arrestor mechanism as triggered in the present joints.

In Regime (1), Figure 6.7a-b shows that the crack propagates along the interface between the composite and Nylon structure. The variations originate from the coexistence of three sub-mechanisms. First, broken fibre bundles along the Nylon side indicate that fibre bridging is activated (Figure 6.7b) with, we assume, only a minor toughening role - the H5 satin fabrics are indeed known for involving limited contribution of fibre bridging to G_{Ic} [202, 203]. Subsequently, the crack propagates between two adjacent 3D printed layers resulting in small bridging ligaments. The crack deflection between two printed layers has not been fully elucidated, but probably originates from poor local adhesion between layers. Finally, the main path is along the composite-Nylon interface where the crack is not deflected into the composite or into the Nylon structure. Opposite to the current results, Tao et al. [135] inserted a PA film in the middle of an adhesive layer, but with a detrimental effect on G_{Ic} . This configuration in-

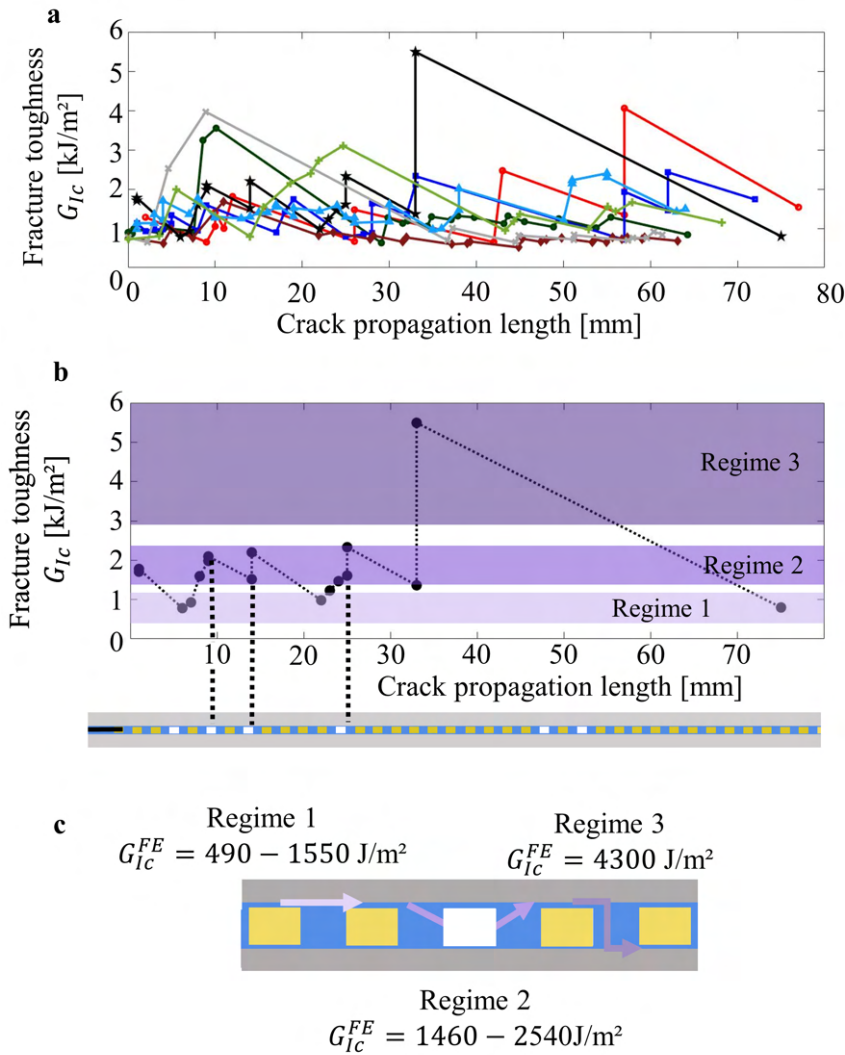


Figure 6.6 (a) Experimental crack resistance curves for architected joints. (b) One of the R-curves with the associated joint configuration. (c) Microstructure associated to each regime with the G_{Ic} computed by FE modelling.

cluding only filled cavities is modelled by finite elements. As shown in Figure 6.7c, the computed G_{Ic} varies from 490 to 1550 J/m² depending on

the crack tip location. This comes from the difference in energy dissipated by plastic deformation. Indeed, when the crack is located above a filled channel, the plastic zone is constrained by the stronger epoxy resin in the channel. An increase in fracture toughness above the channels due to the higher local stiffness as in Morano et al. [82] was expected opposite to the present results. This indicates a competition between the contribution of plastic dissipation and the effect of stiffness variations. In the present case, plastic dissipation dominates. In addition, when the crack tip is located between two adjacent cavities, stresses first concentrate at the interface above the filled channel ahead of the crack tip, leading to the initiation of a secondary crack that interacts then with the main crack. Plastic dissipation in the Nylon film thus contributes to the toughness associated to Regime (1) as it acts as a dissipative layer [79, 97]. In addition, 3D printing generates some roughness on the film surface that can lead to mechanical interlocking also contributing to the increase in fracture toughness.

The second regime corresponds to crack trapping by a channel (Figure 6.6 and Figure 6.8) left empty upon resin injection, while the others are filled as shown in Figure 6.8a-b. The finite element model shows that when the crack is located at the interface above the last filled channel before the void, stresses tend to concentrate between the crack tip and the cavity, see Figure 6.8c. This leads to crack deflection towards the cavity when the failure criterion is met, and it becomes trapped by the empty channel. Although the crack propagates through Nylon, the Nylon fracture toughness is not attained as the void leads to an anti-shielding effect [204] resulting in the apparent fracture toughness equal to about 1500 J/m^2 in the simulation. The micrographs show that the crack propagates through the tough Nylon film from the cavity towards the same interface as previously. In this case, the finite element model indicates, as observed in Figure 6.8d, that stress concentrates at the interface close to the filled channel ahead of the void resulting into the initiation of a small crack that progressively propagates. At some point, the secondary crack and the main crack interact and join each other, see Figure 6.8e, leading to the main crack propagation. This mechanism is summarized in Figure 6.8f and the computed fracture toughness is about 2500 J/m^2 . In addition, the main crack is not sharp and, as explained by Maloney and Fleck [123] and Heide-Jørgensen et al. [121, 122], the crack tip gets extensively blunted and must then re-initiate, improving the joint toughness. Propagation of the crack through the tough Nylon contributes to the G_{Ic} improvement, although the bulk G_{Ic} of Nylon is never attained.

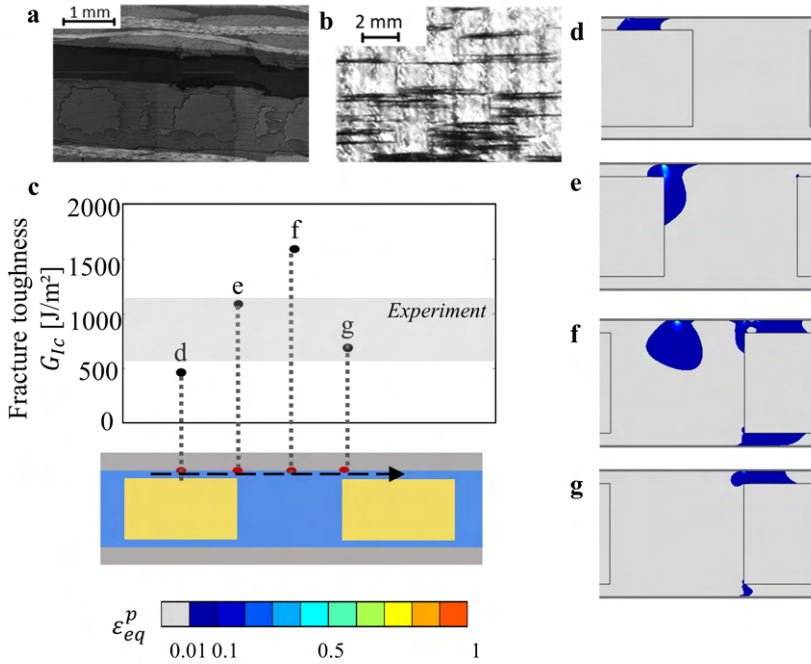


Figure 6.7 Optical micrographs corresponding to the fracture surface in Regime (1) from (a) the side and (b) the top of the Nylon film. (c) Variation of G_{Ic} , through FE modelling, with the crack tip position and the corresponding plastic zones: crack located (d) above the filled cavity, in the middle of it; (e) above the last corner of the filled cavity; (f) between two consecutive filled cavities and (g) above the first corner of the filled cavity.

The experimental scatter on G_{Ic} is explained by the partial filling of some of the channels (Figure 6.8b). Deflection is observed when the channel is empty at least over one third of the joint width. In this case, only a small part of the crack front is trapped by the empty cavity while the rest of the crack still propagates along the weak composite-Nylon interface. To conclude, the toughness of Regime (2) is due to the crack propagation inside the tough Nylon film originating from its deflection by the channel and its deflection back to the interface when reinitiated due to the initiation of microcracks along the interface.

Regime 3, observed in Figure 6.9, is triggered when the crack propagates

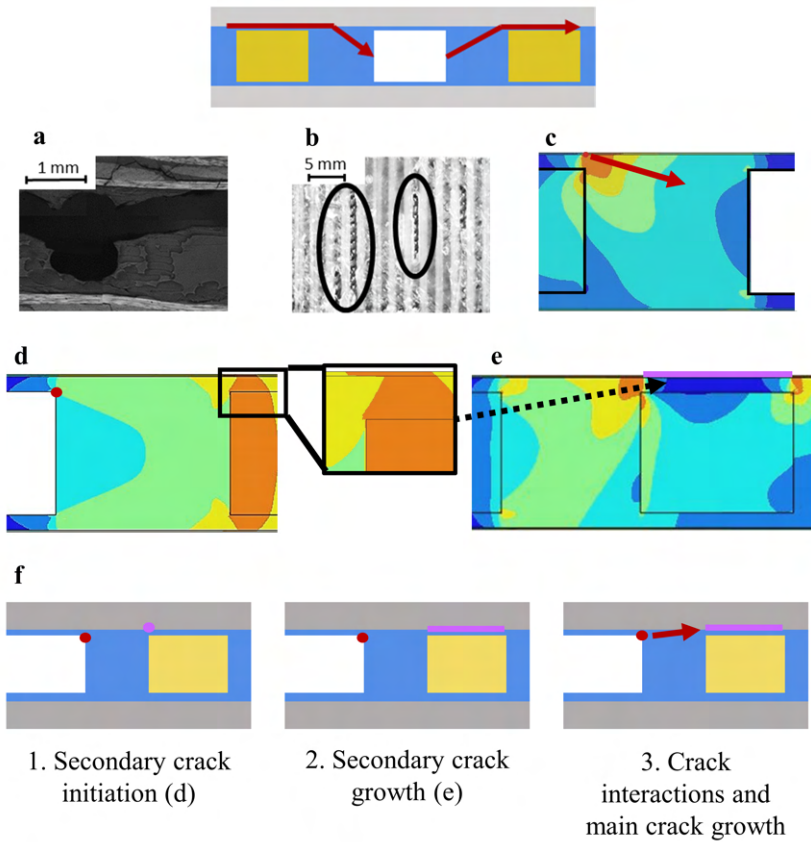


Figure 6.8 Failure mechanism associated to the presence of an empty channel (Regime 2): (a)-(b) Optical micrographs from (a) the side of the DCB and (b) the top surface of the PA film. (c)-(e) Stress distributions when (c) the crack is located behind the cavity and (d)-(e) the crack is trapped by the cavity (d) before and (e) after secondary crack initiation. (f) Summary of the failure mechanism when the crack has been trapped by the cavity.

through the thickness of the film, from one interface to the other. This crack path is observed for all the points, obtained on different specimens, characterized by a fracture toughness larger than 3 kJ/m^2 in Figure 6.6a. The fractography could not reveal the root cause of this deflection. Although the crack is arrested for the cases for which the crack is trapped by an empty channel or when it propagates through the entire thickness of the structure,

an unstable crack propagation jump is observed along the weak composite-Nylon interface when reaching G_{Ic} . It is deemed that it originates from a sudden change in the adherence at one of the interfaces. The top interface in Figure 6.9c is stronger than the bottom one, leading to the crack deflection at the weakest interface (Figure 6.9c-e) as it has been reported by Tao and co-workers [83]. This is modeled by inserting a secondary crack at the bottom interface. It can be observed in Figure 6.9f-g that the two cracks interact with each other and that the primary crack propagates through the Nylon film. A maximum fracture toughness of about 4300 J/m^2 is computed using the FE model for this Regime 3, in very good agreement with the experimental results.

In addition to these three experimental observations, FE simulations have been performed for other configurations in order to guide towards even larger theoretical joint design. First, a joint made only of empty channels is modelled. In this case G_{Ic} reaches 40 kJ/m^2 with the Nylon structure acting as bridging ligaments. This value is extremely high and, although most probably unrealistically high, but provides a motivation for further optimization. However, when one wall fails, a large amount of energy is released which is not considered by the model and could be detrimental to structural integrity and could be associated to a snap-back instability. However, under the displacement-controlled conditions, when one of the walls fails, the energy will be released following unloading paths. Thus, once the next wall is attained, the energy stored in the loaded composite beams will be much lower than the fracture energy and the propagation is altered [122]. In addition, integrated manufacturing of such joint is complicated, asking for perfect control of the printing process preventing the resin from entering the channels. Note that other failure scenarios could take place such as composite delamination and, though not considered by the model.

As a further step, the influence of the second channel ahead of the cavity on the failure process has been considered (subsection 6.7.1). The process zone length is evaluated (Figure 6A.1a) to be around 3 to 4 mm. Given the channels dimension, the second cavity ahead of the crack tip could also have an influence on G_{Ic} . An empty channel has a shielding effect (Figure 6A.1b-e), leading to higher G_{Ic} . Thus, an empty channel can result in two antagonistic effects depending on its position with respect to the crack tip, i.e. anti-shielding if located ahead and shielding if located far enough.

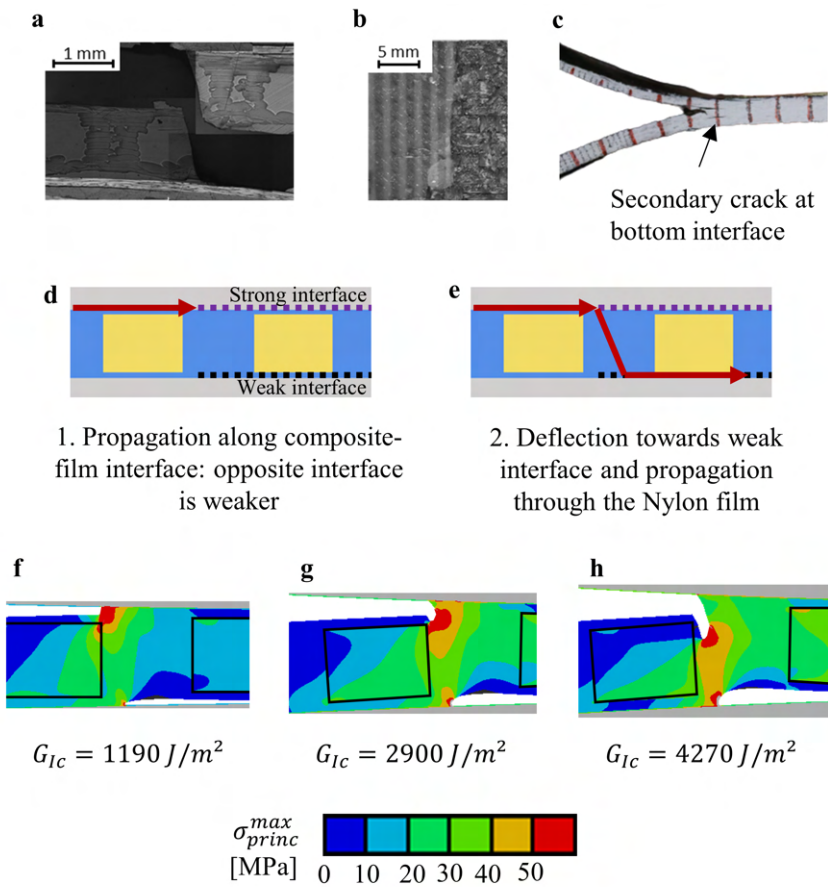


Figure 6.9 Optical micrographs corresponding to the fracture surface in Regime (2) from (a) the side and (b) the top of the Nylon film. Stress distributions when the crack is located (c) before the void, (d) is trapped by the void and (e) when the two cracks interact with each other. (f)-(g) Progressive propagation through the film thickness due to main crack interactions with a secondary crack at the opposite interface.

6.4.2 Lap-shear joints

The fracture toughness of the joint is not the only important property to consider when designing bonded joints under shear dominated loads. In aerostructures, the adhesive thickness must remain below a certain threshold to limit the decrease of shear stiffness and strength. This has been ad-

dressed, using as an example, the lap shear joint configuration with the architected bondline. For this purpose, the number of empty and filled channels varies as well as the position. Table 6.1 shows that increasing the number of filled cavities improves the joint strength and stiffness. However, it can also be observed that, for a given amount of filled channels, the position of the filled channels does influence the shear strength.

Table 6.1 Joint stiffness, strength and fracture toughness of architected joints depending on the configuration, determined by finite element modelling.

Amount of filled cavity	Configuration	Apparent shear modulus [MPa]	Strength [MPa]	Maximum G_{Ic} [kJ/m ²]
0%	EEE	52	6	40
33%	FEE	138	9	16
	EFE	138	11	16
	EEF	137	12	16
66%	EFF	221	14	2.2
	FFE	222	12	2.2
	FEF	223	13	2.2
100%	FFF	306	14	1.5
Nylon	/	143	12	8.5 – 11.5 [179]
Resin	/	1070	20	0.09[167]

First, the two extreme cases, EEE and FFF models, are compared to one another. Figure 6.10a-d shows that, for a given applied load, shear stresses are smaller in the joint with all channels being filled. This arises from the fact that by filling the channels, the effective cross-section area over which the applied shear force act is increased, reducing the magnitude of the local shear stresses. In addition, filling all the cavities also results in lower peel stresses near the edges. Regarding failure, the criterion is first met at the joint corner for EEE while it is first met in the interface above the first filled cavity for FFF. The shear strength is thus improved by increasing the amount of filled channels, but also the shear stiffness, as when filling all the cavities, a stiffer material is present in the bondline.

When joints containing 33% and 66% of filled channels are considered, an influence of the location of the empty and filled channels with respect to the

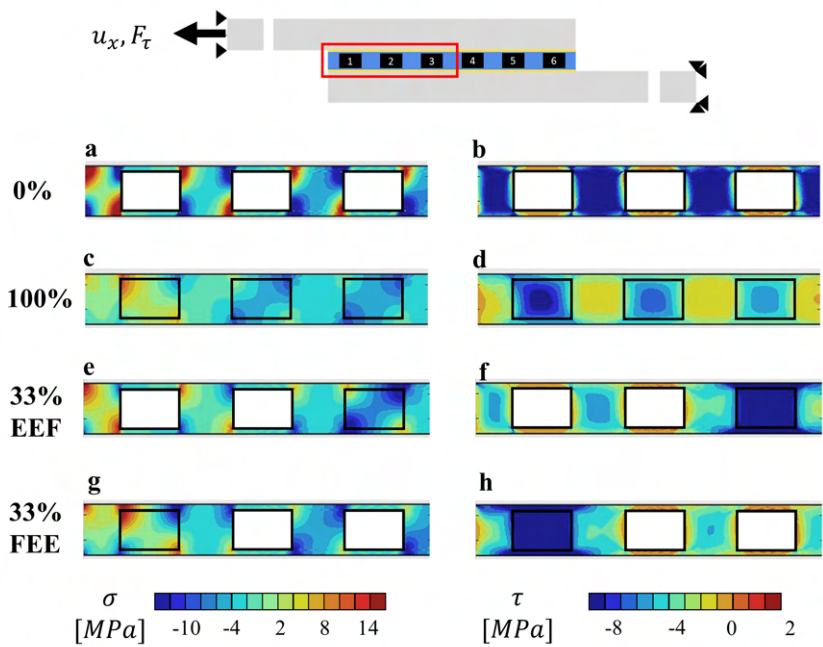


Figure 6.10 Peel (left) and shear (right) stress distributions in lap-shear joints containing (a)-(b) 0%, (c)-(d) 100%, (e)-(h) 33% of filled channels. (e)-(f) The filled channel is located at the joint centre or (g)-(h) at the edge of the lap joint.

joint edges is found (Table 6.1). Indeed, if the channel close to the edge is left empty, the shear strength is improved with respect to the case for which the edge channels are filled with resin. The stress distributions are shown in Figure 6.10e-h for the FEE and EEF joints, both containing 33% of filled channels. The shear stresses (Figure 6.10e, g), in both situations, are the highest right above the filled cavity. Regarding the peel stresses, they tend to be the highest near the joint edges in both cases, but their magnitude is slightly larger when the first channel is left empty which could negatively affect the shear strength. However, due to the combination of the large peel stresses and the shear stresses near the filled channel when located near the edge, the failure criterion is first met for the joint with the filled channel near the edges, resulting in a lower shear strength. The introduction of an empty channel near the joint edges thus improves the stress distribution and the shear strength. The same accounts for a joint containing 66% of

filled channels.

These findings agree with common results found in the literature. Indeed, Kumar et al. [159] tailored the stiffness of adhesive bonds. They determined an improved strength when the centre of the joint is stiffer than the edges. Here, a larger shear strength has been found when for a given amount of filled cavities, the empty cavities are located at the edges of the joint. When the edge activity is left empty, peel stresses near the joint corners tend to be larger. However, shear stresses near the edge are lower and the cavity allows for better shear stress distribution in the joint, resulting in higher shear strength. Indeed, when the first channel is filled, high shear stresses localize near the corner, in the interface right above the filled cavity leading to premature failure. As a further perspective, it would be interesting to investigate the influence of the amount of filled channels and their position in the joint for industrial assembly by using a more complex model.

6.5 Materials selection for joints with tunable properties

In the previous section, the influence of the position of the filled and empty channels on the joint properties have been investigated. Contrary to the fracture toughness, stiffness and strength are improved by increasing the amount of filled channels (Table 1). Hence, depending on the joint configuration, the properties of the joint can be modified. To determine the best joint configuration, a rational materials selection procedure [201] has been followed as further developed elsewhere [101]. The objective of the selection chosen here is to minimize the mass of the structure, $m = \rho L W t$ with ρ the density of the architected film, L and W the length and width of the joint, and t the film thickness. The joint is subjected to two forces: a tensile force F_σ and a shear force F_τ (Figure 6.11a) resulting in a tensile stress $\sigma_\perp$ and a shear stress τ . This leads to two constraints: (1) F_σ cannot lead to crack propagation in the presence of a crack of length $2a$ implying that $K < K_{IC}$ and (2) F_τ cannot result in a displacement larger than $u_\tau|_{max}$. Through FE modelling, the joint stiffness and fracture toughness are computed for different amount of filled cavities (0, 33, 66, 100 %) and architected film thicknesses ranging from 0.05 to 1 mm. It is important to recognize that small film thicknesses cannot be used with the current manufacturing procedure but could be produced with other techniques such as selective laser sintering, stereolithography or selective thermoplastic elec-

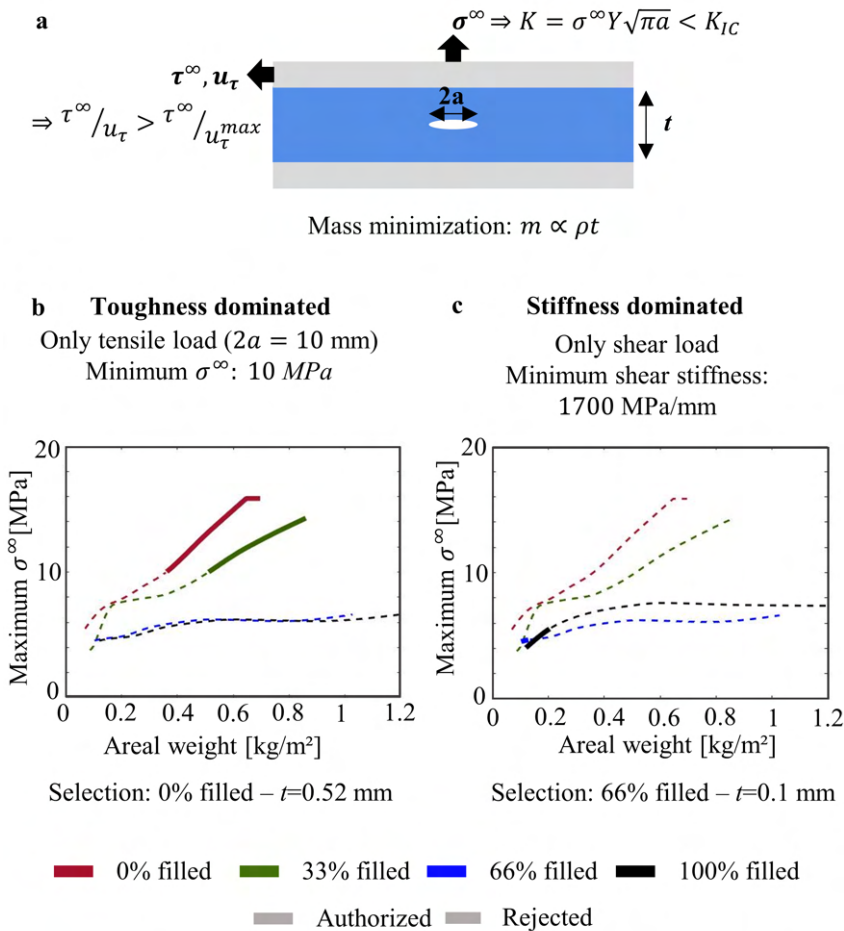


Figure 6.11 Materials selection approach for architected adhesive joints (a) problem description, (b) case where the toughness dominates the problem due to high peel stress and (c) the problem is stiffness-dominated.

trographic process. The lowest attainable film thickness is thus a matter of access and progress in 3D printing technology. If the joint is mainly subjected to tensile stresses (Figure 6.11b), the selection is dominated by G_{IC} and the best joint is made of empty cavities. On the other hand, if only shear is applied to the joint, the shear stiffness constraint dominates (Figure 6.11c), the best joint contains 66% or 100% of filled cavities. These

two examples show that depending on the loading, the best configuration changes. The degree of freedom in the architecture allows tuning performances depending on the application to get an optimum response. Further work will focus on extending this selection analysis to a wider range of objectives, constraints and load configurations as attempted elsewhere for mainly classical adhesives [101].

6.6 Conclusion

Intensive use of adhesive bonding in composite applications is prevented by the moderate fracture toughness of available adhesives and long manufacturing times. To simultaneously unlock these two barriers, we printed architected Nylon films with channel holes to bond composites by co-curing. This leads to several major findings:

- First, exceptionally high fracture toughness larger than 5000 J/m^2 is attained during crack propagation with intermediate regimes also of high fracture toughness. Several toughening mechanisms are triggered involving: plastic dissipation, crack deflection, crack tip blunting and crack shielding.
- Second, single step co-curing is enforced by simultaneously bonding and processing of the composite panels, saving time and energy, and avoiding surface treatments.
- Third, the shear stiffness and strength can be modulated by playing with the architecture depending on the structural requirements.
- Aside from proving excellent toughening potential, this study made use of the simple stress-at-a distance failure criterion. This failure criterion proved very effective to predict the magnitude of G_{Ic} for the different cracking mechanisms occurring along the crack path after identification from independent data on the bulk materials under consideration.
- An original materials selection approach is adapted to adhesive joints to guide the design approach of the best joint configuration depending on the application.

As a perspective, controlling the filling of the cavities and the film thickness can lead to a wide variety of ultra-tough joints with controlled strength

and stiffness based on a single architected film. However, controlling the film thickness requires to change manufacturing technique which is left for further investigation.

6.7 Appendices

6.7.1 Appendix A: Influence of the second cavity ahead of the crack tip

An important parameter to consider when designing architected joints is the length of the process zone. Heide-Jørgensen and Budzik [122] define it as the inverse of the elastic foundation wavelength λ_w for adhesive joints given by

$$\lambda_w = \sqrt[4]{\frac{m E_a W}{4 t E_s I}} \quad (6A.1)$$

where E_a and t are, respectively, the Young's modulus and the thickness of the adhesive which has a Poisson ratio ν_a , the composite modulus E_s , the DCB width W , the second inertia moment I while m defines the stress state at the crack tip in the following way:

- For plane-strain conditions and plane stress conditions in the transverse direction: $m = 1/(1 - \nu_a^2)$;
- For plane strain conditions in in-plane and out-of-plane directions: $m = (1 - \nu_a)/[(1 - 2\nu_a)(1 + \nu_a)]$;
- For plane stress conditions in all directions: $m = 1$.

Assuming that the central layer is made of NylonWhite only, this equation leads to values of process zone lengths ranging from 3.12 mm to 3.95 mm. As the length of one channel is 1.1 mm and the spacing between two adjacent channels is 1 mm, when the crack is located above the right (crack A) or the left (crack B) corner of a cavity, the process zone spans over cavity III as shown in Figure 6A.1a. Consequently, channel III must be considered.

Leaving cavity III in Figure 6A.1a empty has a beneficial effect as the computed fracture toughness corresponding to when the first failure criterion is met is higher. Depending on the case, the fracture toughness is increased by 10 to 25% when the cavity III is left empty. When the two first cavities (I and II) are filled and if cavity III is empty, the fracture toughness is improved by 7 to 15% when the crack propagates along the composite interface between the two first channels. The improvement is only 7% when it

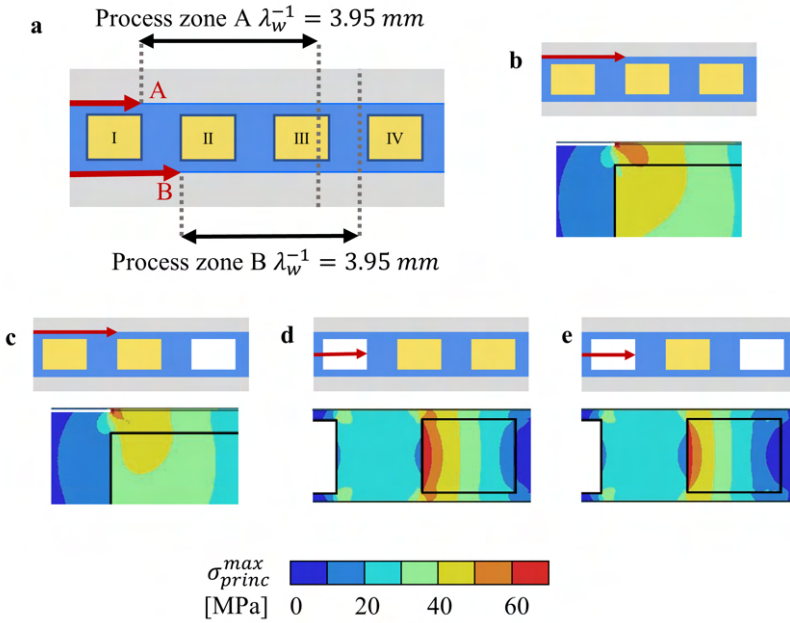


Figure 6A.1 (a) Schematic view of the process zone length and the part of the joint covered by this zone if the crack is located at the left (crack B) or at the right corner (crack A) of a channel. (b)-(e) Maximum principal stress distribution when (b) and (c) the crack propagates along the interface and (d)-(e) when the crack has been trapped by a cavity and the first cavity ahead of the crack tip is full of resin. In (b) and (d), the last considered cavity is filled while in (c) and (e) it is left empty resulting in an anti-shielding effect.

is close to the right corner of cavity I while the increase is of 15% when the crack is close to cavity II. The stress distributions in this last case are shown in Figure 6A.1b-c for an arbitrary load applied on the DCB. The stresses near the crack tip are lower when the last cavity is left empty. If channel I is empty while channel II is filled, the fracture toughness increases by 25% if cavity III remains empty. The maximum principal stress distributions are shown in Figure 6A.1d-e, with lower stresses near the interface when cavity III is empty. To conclude, if the empty cavity is located far enough from the crack tip and is located in the process zone, it has a shielding effect while it has an anti-shielding effect when it is close from the crack tip (this is the case where the crack is deflected towards the cavity).

6.7.2 Appendix B: Detailed description of the manufacturing steps of the joints

The different steps of the integrated manufacturing process needed to obtain the architected joints are presented by showing pictures (while only schematics are given in the paper). It is made of four steps: 3D printing of the film, stacking of the film and the carbon fibres plies in the mould, curing of the lay-up and unmoulding of the panel.

- Step 1 (Figure 6B.1) 3D printing of the Nylon film. The Nylon film upon 3D printing is shown in Figure 6.3b.

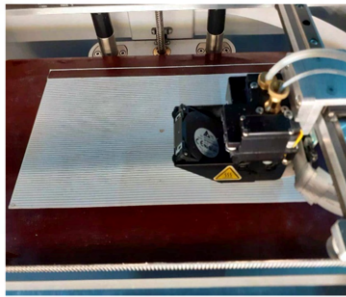


Figure 6B.1 Step 1 - 3D printing of the Nylon film made of NylonWhite with a MarkTwo printed from Markforged.

- Step 2 (Figure 6B.2): Stacking of the Carbon fibres plies and the films and mould closure
- Step 3 (Figure 6B.3): Resin injection and curing of the panel
- Step 4 (Figure 6B.4) – Unmoulding. After unmoulding the panel is machined to obtained DCB samples, shown in Figure 6.3c-d.

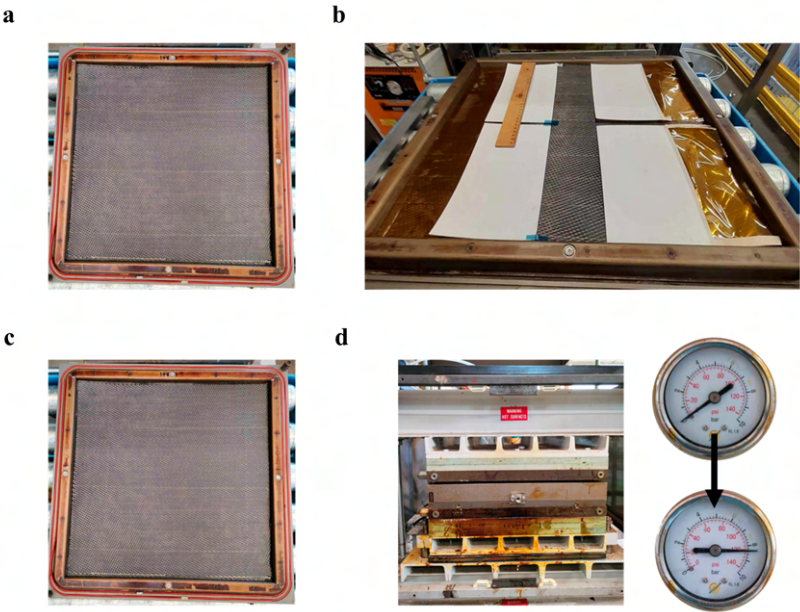


Figure 6B.2 Step 2 - a) Stacking of the first four Carbon plies, b) stacking of the Nylon film and the Kapton film to insert the precracks, c) stacking of the last four plies of Carbon fibres and d) mould closure by inserting it in the press and increasing the pressure to 8 bar.

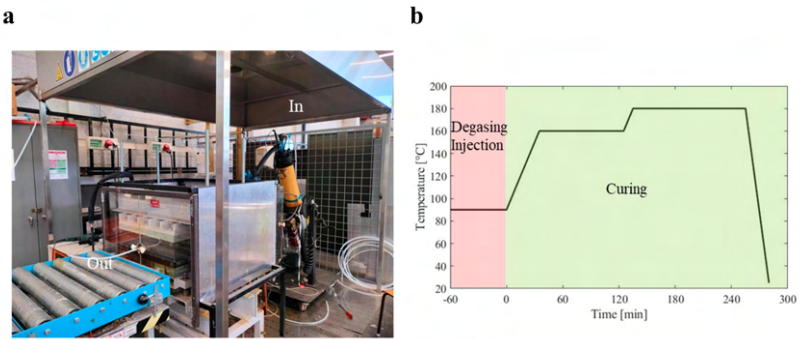


Figure 6B.3 Step 3 – a) RTM processing lab during resin injection and curing; the resin is injected by the piston at the right and goes out of the mould at the left side. b) Curing cycle of the joints.



Figure 6B.4 Step 4 – Composite panel after opening the mould.

7

Toughening through a dissipative layer

7.1 Introduction

Several potential toughening mechanisms have been described in Section 2.3 including plastic dissipation, crack deflection, crack bridging, crack blunting, toughening through elastic heterogeneities or the use of metamaterials in the bondline. Crack bridging and bondline architecturing have been investigated for co-cured composite joints in Chapters 5 and 6, respectively. The current chapter investigates the toughening that can be potentially generated by adding a dissipative layer in the joint.

This work has several motivations. First, Voleppe et al. [12] claimed that the trapping of the crack in the PEI-epoxy interphases was the result of the presence of a soft evidenced transition zone towards which the crack is deflected and in which it gets stuck, quite similarly to a crack propagating in a laminated glass that initiates in the glass and then propagates into the adhesive film. The second motivation comes from the high toughness of the PA film-composite interface evidenced in Chapter 6 which is assumed to partly originate from the plastic dissipation taking place in the soft PA

film. Finally, this concept of a dissipative layer has already been applied in the molecular bonding context [97, 105] and proved to be very efficient. The goal is to apply this concept to adhesive joints.

To meet the objective, the configuration of Figure 7.1 is investigated. It consists of a multi-layer made of two elastoplastic layers stacked between two elastic rigid adherends. The two elastoplastic layers have different Young's modulus, yield strength and toughness. In addition, their thickness h_1 and h_2 is varied. The goal is to determine the magnitude of the overall joint toughness (G_c) depending on these parameters. One key difficulty is that G_c depends on the position of the crack in the stack. Hence, the crack propagation locus must be determined. This leads to the two main questions of this chapter:

1. Depending on the material properties and the layer thickness, where does the crack propagate? The selected crack path will be referred to as Y_{crack} .
2. Depending on the material properties, h_1 , h_2 , what is the joint fracture toughness knowing Y_{crack} ?

This is investigated numerically using the steady-state FE code developed by Martiny et al. [13, 92] on the basis of a first version of the code written by Landis et al. [205]. We intend to derive guidelines regarding the design of tough adhesive joints including a dissipative layer.

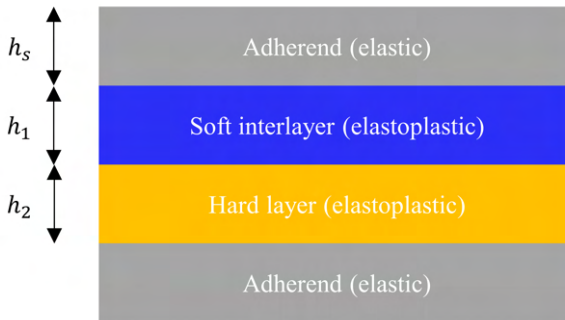


Figure 7.1 Multi-layer configuration to investigate the toughening effect of a dissipative layer in the joint.

7.1.1 Crack propagation in multilayers

Crack propagation in multi-layers is a topic that has already been addressed by several authors [206–208]. The focus was on elastic materials or more generally to cases for which Linear Elastic Fracture Mechanics (LEFM) can be applied. Crack path selection and stability can be addressed in terms of the asymptotic stress field ahead of the crack tip. The stress field at a distance r from the crack tip is expressed as

$$\begin{bmatrix} \sigma_{xx} & \tau_{xy} \\ \tau_{xy} & \sigma_{yy} \end{bmatrix} = \frac{K_I}{\sqrt{2\pi r}} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + \frac{K_{II}}{\sqrt{2\pi r}} \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} + \begin{bmatrix} T & 0 \\ 0 & 0 \end{bmatrix} + O(\sqrt{r}) \quad (7.1)$$

with K_I and K_{II} being the mode I (opening) and mode II (shearing) stress intensity factors. It is often assumed that the crack chooses the trajectory for which $K_{II} = 0$. T is constant and denotes the T-stress acting parallel to the crack plane. The T-stress has several origins such as resulting from internal stresses or from the geometric constraint. It governs the crack stability. Cotterell and Rice [209] demonstrated that a crack is stable (in terms of remaining straight) if $T < 0$ and unstable if $T > 0$.

Based on LEFM, Fleck et al. [206] investigated crack propagation and stability in brittle adhesive joints assuming infinite rigid adherends. The mode-mixity at the crack tip is given by the phase angle ψ defined as $\arctan(K_{II}/K_I)$. ψ depends on the elastic mismatch between the adherends and the adhesive through the first and second Dundur's parameters α and β . ψ also depends on the position of the crack across the adhesive thickness. The T-stress depends on the elastic mismatch through α and β , on the adhesive thickness, on the adherend thickness and on the internal stress [206, 210]. For an adhesive joint loaded under mode I, four scenarios can take place as shown in Figure 7.2a. The only case in which the crack propagates in a stable way in the bondline is when $\partial K_{II}/\partial y > 0$ and $T < 0$ and as shown in Figure 7.2b, it depends on α (Equation 7.2) and β (Equation 7.3). Fleck et al. [206] made the assumption of infinite adherends. Chen et al. [210] showed that the T-stress increases if the adherend thickness h_s decreases or if the adhesive thickness t increases, favouring unstable crack propagation.

$$\alpha = \frac{(1 - \nu_a)/\mu_a - (1 - \nu_s)/\mu_s}{(1 - \nu_a)/\mu_a + (1 - \nu_s)/\mu_s} \quad (7.2)$$

$$\beta = 0.5 \frac{(1 - 2\nu_a)/\mu_a - (1 - 2\nu_s)/\mu_s}{(1 - \nu_a)/\mu_a + (1 - \nu_s)/\mu_s} \quad (7.3)$$

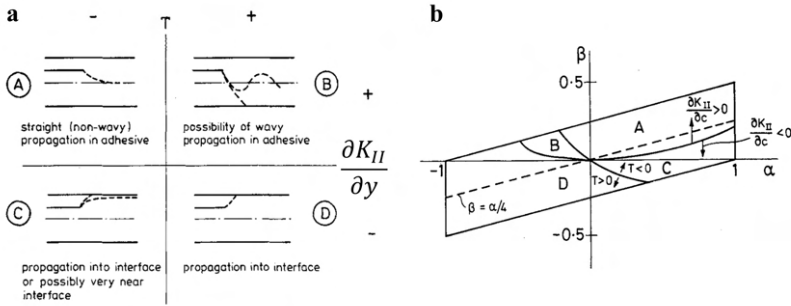


Figure 7.2 a) Crack path selection scenarios in adhesive joints depending on $\partial K_{II}/\partial y$ and T and b) regimes of crack path selection depending on α and β . Taken from [206].

The crack path location predicted by Fleck et al. [206] has been verified in adhesive joints, but also in other systems such as glass/copper/glass [211, 212], alumina/aluminium/alumina [211]. However, McNaney et al. [211] also showed that the distance between the interlayer and the crack is an important parameter. If the interlayer is located too far away, the crack propagation is not affected by the presence of the interlayer. These studies only focused on elastic materials. In addition, the presence of a weak interface can also have an influence on the crack path selection [212]. For example, in glass/copper systems, the crack should propagate away from the copper layer but if the glass/copper interface is weak, the crack propagates towards the interface.

If the interlayer is elastoplastic and a crack propagates parallel to the interface, there is a competition between (i) the stress concentration induced by the lower compliance of the soft layer attracting the crack and (ii) the enhanced plastic dissipation resulting in a higher G_c if the crack propagates towards the soft layer [213, 214]. If the amount of plasticity remains limited, the crack deflection is governed by the elastic mismatch while if it is important, the deflection will be dominated by the plasticity [212, 213]. In this last case, a crack can be deflected away from the interface while the theoretical analysis by Fleck et al. [206] predicts the crack to deflect towards the interface.

7.1.2 Fracture toughness of multilayers

Bertholet et al. [97] and Dépinoy et al. [79] investigated the influence of h_1 , h_2 and of the material properties of the layers on G_c for interfacial cracks between one adherend and the hard layer. Both studies demonstrated that h_2 (hard layer) must be minimized in order to maximize the plastic dissipation in the soft interlayer. Increasing h_1 (soft layer) results in an improved G_c . However, above a certain value, the improvement is no longer significant. More details about these findings are given in subsection 2.3.2.

7.1.3 Problem statement

The precise configuration analyzed here is a four-layered joint shown in Figure 7.3. The two external layers are the adherends characterized by a Young's modulus E_s and a Poisson ratio ν_s with a thickness h_s . The two adherends surround two elastoplastic layers referred to as the "adhesive stack". The top layer (subscript 1) is softer than the bottom layer (subscript 2). Both are characterized by a Young's modulus E_i , a Poisson ratio ν_i , a yield stress σ_{0i} , a hardening exponent n_i . They are modelled using a power-law given by Equation 7.4 and using the von Mises theory of plasticity considering isotropic hardening. In addition, each layer is characterized by an intrinsic fracture toughness G_{0i} .

$$\sigma = \begin{cases} E_i \varepsilon & \text{if } E_i \varepsilon \leq \sigma_{0i} \\ \sigma_{0i} \left(\frac{E_i \varepsilon}{\sigma_{0i}} \right)^n & \text{otherwise} \end{cases} \quad (7.4)$$

In addition, the crack propagation path and the joint toughness are predicted based on a cohesive zone model through a traction separation law. The cracking resistance of each elastoplastic interlayer is characterized by a cohesive strength σ_{ci} and a cohesive energy G_{0i} . A trapezoidal cohesive law is used, defined by the cohesive strength σ_{ci} , the critical opening δ_{ci} and the cohesive energy $\Gamma_c = G_0$. The traction depends on the parameter λ defined by $\sqrt{(\delta^n / \delta_c^n)^2 + (\delta^t / \delta_c^t)^2}$ with δ^k being the normal ($k = n$) or tangential ($k = t$) opening displacement while δ_c^k is the critical opening displacement in mode k . It is assumed here that $\delta_c^t = \delta_c^n = \delta_c$ and is given by Equation 7.5. Consequently, the mode I and II cohesive energies are equal. Traction increases until reaching $\lambda = \lambda_1 = 0.15$ and remains constant until $\lambda = \lambda_2 = 0.5$ before decreasing and vanishing when $\lambda = 1$. The cohesive

7 | Toughening through a dissipative layer

strength, energy and critical opening displacement are related by

$$G_{0i} = \frac{\sigma_{ci}\delta_c}{2}(1 - \lambda_1 + \lambda_2). \quad (7.5)$$

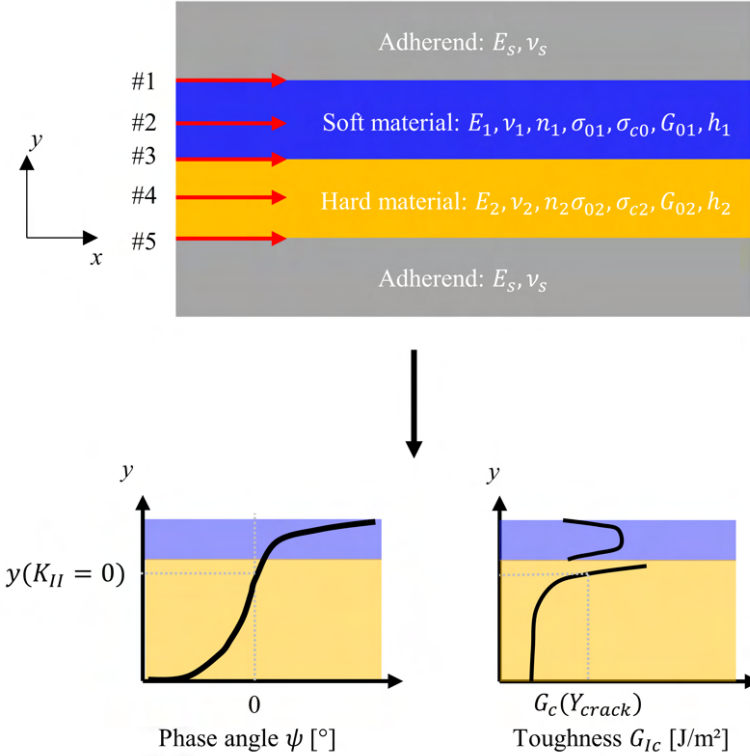


Figure 7.3 Modelling strategy: a joint made of two layers in which the crack position is change. Then, ψ and G_c are computed for each position y to determine $Y_{crack} = y(\psi = 0)$ and $G_c Y_{crack}$.

To determine the crack propagation locus in the joint, several simulations are run for one set of parameters by changing the position of the cohesive elements in the joints as shown in Figure 7.3. For each simulation, ψ and the joint toughness G_c are computed giving their evolution with the crack position in the joint y . The crack path ($y = Y_{crack}$) is the one such that $\psi(y) = 0$ and the joint toughness is then taken as $G_c(Y_{crack})$. Some assumptions are made:

- The two elastoplastic materials are characterized by the same intrinsic toughness (cohesive energy) $G_{01} = G_{02} = G_0$.
- Perfect interfaces are assumed to neglect crack deflection towards a weak interface.

These assumptions lead to the following dimensional analysis for the joint fracture toughness G_c , depending on the material properties and some geometric parameters.

$$\frac{G_c}{G_0} = F \left(\frac{E_1}{E_s}, \frac{E_2}{E_1}, \nu_1, \nu_2, \nu_s, n_1, n_2, \frac{\sigma_{01}}{E_1}, \frac{\sigma_{02}}{\sigma_{01}}, \frac{\sigma_{c1}}{\sigma_{01}}, \frac{\sigma_{c2}}{\sigma_{c1}}, \frac{h_1}{R_{01}}, \frac{h_2}{R_{02}}, \frac{h_1}{h_2}, y \right) \quad (7.6)$$

where R_{0i} is the plastic zone size of material i given by

$$R_{0i} = \frac{G_{0i} E_i}{3\pi(1 - \nu_i^2) \sigma_{0i}^2}. \quad (7.7)$$

As a first step, we assume that the joint is only made of the soft layer (1). A first analysis will be conducted for linear elastic material response to compare with the results from Fleck et al. [206] but here using a cohesive zone model. Then, plasticity is introduced to determine the influence of σ_{c1}/σ_{01} on the crack propagation path. Then, the second interlayer is introduced. First, an elastic analysis is performed again to determine the crack path depending on E_1/E_2 and h_1/h_2 , before introducing plasticity in both layers.

7.2 Finite element model

The steady-state code developed by Martiny et al. [13, 92] based on the work of Landis et al. [205] is used, to determine the steady-state fracture toughness G_c of a joint.

The model assumes that deformation and damage take place in a restricted zone ahead of the crack tip. This zone is referred to as the fracture process zone denoted as l_s . In the present model, it is represented by the traction versus separation condition across the crack plane described earlier. Cohesive elements are thus inserted along the joint at a position y . The surrounding materials are considered as continuum. The possible toughening mechanism is plastic dissipation in the elastoplastic layers. Hence,

G_c is equal to $G_c = G_0 + \Gamma_1^p + \Gamma_2^p$ with Γ_i^p being the contribution of plastic dissipation in layer i .

The model is implemented within a quasi-static plane-strain, large rotations, steady-state finite element formulation developed by Martiny et al. [13, 92]. More details about the formulation can be found in [13]. The adherends and the two interlayers are modelled with quadrangular elements. As a wedge-peel configuration is assumed, the wedge is considered with the help of displacement-like boundary conditions.

The phase angle ψ is estimated by determining the energy dissipated in the cohesive element located at a distance d from the crack tip corresponding to $0.5l_s$ with the process zone length of material i (1 or 2) given by

$$l_{si} = \frac{G_0 E_i}{\sigma_{ci}} M, \quad (7.8)$$

where M is a parameter depending the cohesive law model and on the surrounding materials [215]. If the surrounding material is only elastically deformed, it is equal to 1 while it is set to $1/3\pi(1 - \nu_i^2)$ if it is plastically deformed. Knowing the shear and normal opening displacements of the cohesive elements in d , it is possible to determine the amount of energy dissipated in mode I (opening) and mode II (shearing), referred to as Γ_I and Γ_{II} , respectively. ψ is then obtained using the relationship $\psi = \arctan(K_{II}/K_I) = \arctan(\sqrt{\Gamma_{II}/\Gamma_I})$.

7.3 Monolayer adhesive joint

This section focuses on a joint with a single layer between the two adherends referred to as the adhesive. First, we consider a perfectly elastic material to recover the results from Fleck et al. [206] and we investigate the influence of σ_{c1}/σ_{01} and h_1 on the mode-mixity evolution along the adhesive thickness. Then, we consider an elastoplastic material to investigate the influence of σ_{c1}/σ_{01} and h_1/R_{01} on the crack path selection. In both cases, $h_s = 5$ mm, $\nu_s = \nu_1 = 0.3$ and E_s and E_1 are such that $\alpha = 0.89$ and $\beta = 0.26$. Note that G_c is not influenced by the crack position as it is assumed that the mode I and II cohesive energies are equal.

7.3.1 Elastic adhesive

The objective of this section is to re-assess with the steady-state FE code and using a cohesive model the results of Fleck et al. [206] which are based on a LEFM approach. Here, the CZM introduces a length scale. This length scale is the cohesive zone length l_s which is the distance between the crack tip and the point where the cohesive strength is first attained. According to Parmigiani and Thouless [208], l_s influences the measured phase angle in an asymmetric DCB if the ratio between l_s and the thickness of the thinnest beam is larger than 10. For larger values of l_s , ψ decreases. If l_s is small enough, the LEFM should be recovered.

First, Figures 7.4 to 7.6 show that the predicted crack propagation locus is at the centre of the adhesive bondline, no matter the values of σ_{c1} and h_1 , in agreement with the results of Fleck et al. [206]. Indeed, for α and β values equal to 0.89 and 0.26, regime A of Figure 7.2 is expected.

Influence of the distance at which mode-mixity angle is determined

The distance ahead of the crack tip d at which the mode-mixity angle ψ is evaluated has an impact on the value of ψ due to the oscillatory nature of the stress field [209]. More precisely, LEFM predicts a variation of ψ ahead of the crack tip if $\beta \neq 0$. For this reason, in asymmetric DCBs with the thinnest beam having a height h , ψ is commonly evaluated at a distance $d = h$ from the crack tip. The variation of ψ with d is given by $\psi(d) = \psi(h) + \varepsilon \ln(d/h)$ [207]. Figure 7.4a shows the evolution of ψ with y/h_1 if $h_1 = 1$ mm and $d = 0.5h_1$ and $d = 0$ mm (i.e. in the first element in front of the crack tip). For $\sigma_{c1} > 100$ MPa, satisfying the condition $l_s > h_1$, the ψ profile is strongly affected by the choice of d at which it is determined. ψ tends towards 0° if d is large, predicting values far from those of Fleck et al. [206]. The cohesive elements located at such a distance from the crack tip are not loaded showing that this choice is not relevant. The mode-mixity must be estimated at a position which is representative of the failure process.

This raises the question of choosing an arbitrary value for d which is quite small and which is lower than the value of l_s for all values of σ_{c1} . In Figure 7.4b, it is fixed to $0.5l_s$ ($\sigma_{c1} = 400$ MPa). In that case, there is no overlap between the ψ profiles, opposite to what is found for $d=0$ mm. Hence, using cohesive zone modelling, d cannot be set to a constant value independent of σ_{c1} . Indeed, the evolution of the stress field ahead of the crack

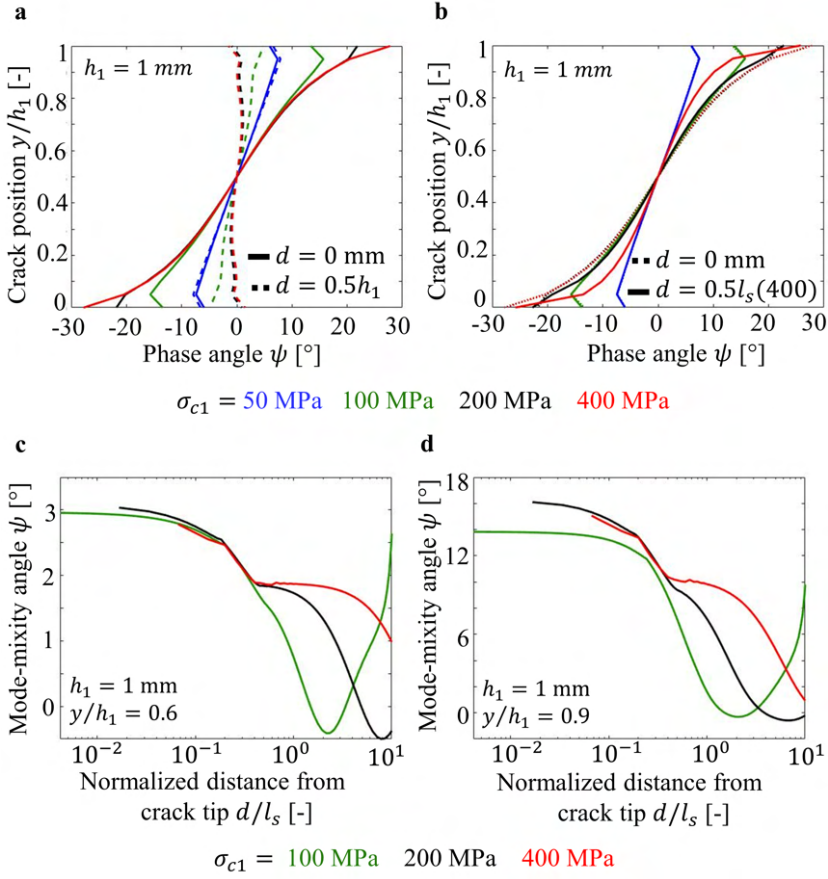


Figure 7.4 a-b) Variation of the phase angle ψ along the adhesive thickness for different values of σ_{c1} and d set to a) 0 mm and b) $d = 0.5h_1 = 0.5 \text{ mm}$. c-d) Evolution of the phase angle ψ ahead of the crack tip for $h_1 = 1 \text{ mm}$, different values of σ_{c1} and c) $y/h_1 = 0.6$ and d) $y/h_1 = 0.9$.

tip depends on the process zone length. If the distance ahead of the crack tip is normalized by the process zone length and if $l_s < \Delta h_1$ (distance to the closest adherend), for a given d/l_s ratio, ψ reaches a similar value for all values of σ_c as shown in Figure 7.4c while if $l_s > \Delta h_1$, the magnitude of ψ changes as shown in Figure 7.4d for $\sigma_{c1} = 100 \text{ MPa}$. An idea could be to take d as being a fraction of l_s , for example $d = 0.5l_s$. In that case, as for $d = 0 \text{ mm}$, the profiles overlap as shown in Figure 7.5a and b. However,

it is shown in Figure 7.5c, that d influences the value of $\psi(y/h_1)$ but crack propagation is still predicted to take at the centre of the joint. d is set to 0 mm for what follows.

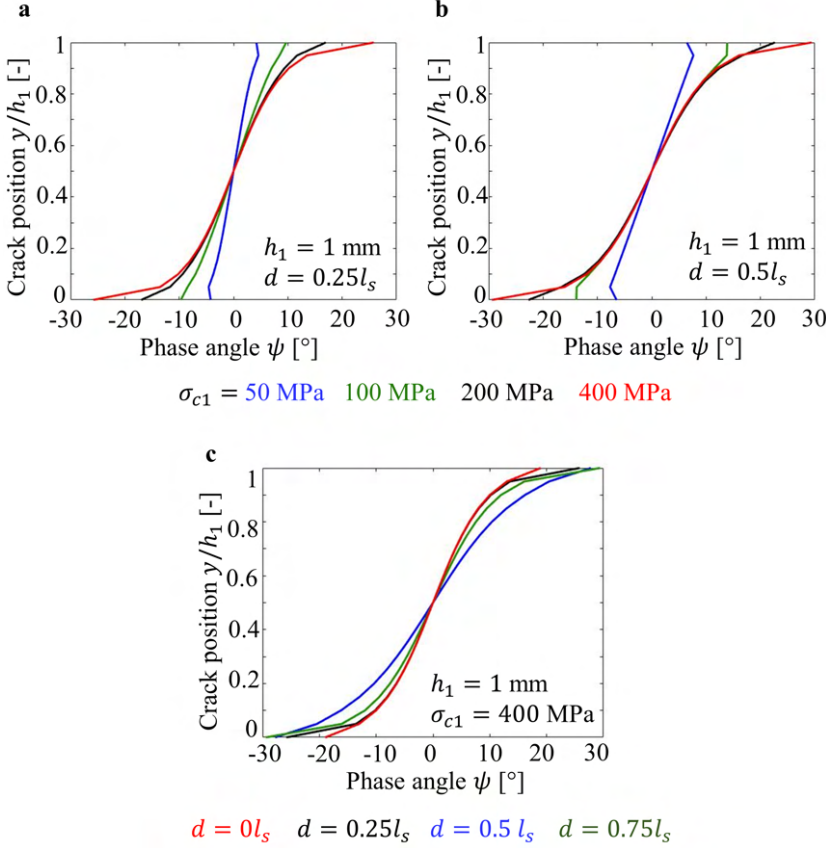


Figure 7.5 a-b) Evolution of the phase angle ψ along the adhesive thickness for four values of σ_{c1} (50, 100, 200 and 400 MPa) and an adhesive thickness h_1 equal to 1 mm for d set to a) $0.25l_s$ and b) $0.5l_s$. Evolution of ψ with y/h_1 for $\sigma_{c1} = 400$ MPa and different values of d .

Influence of σ_{c1} and l_s

The variation of ψ along the adhesive thickness is shown in Figure 7.6 for four different σ_{c1} ($l_s \propto 1/\sigma_c^2$) resulting in l_s values equal to 1.2, 0.3, 0.075 and 0.019 mm. The adhesive thickness is set to 1, 0.5 and 0.1 mm in Figures 7.6a, b and c, respectively. It is found that for σ_c values ranging from 100

to 400 MPa, the evolution of ψ along the joint thickness is quite similar if $h_1=1$ mm, except close to the interface with the adherends. If h_1 is set to 0.5 mm, σ_{c1} equal to 200 and 400 MPa predict similar values of $\psi(y/h_1)$ while $\sigma_{c1}=100$ MPa predicts lower values of ψ . If h_1 is set to 0.1 mm, σ_{c1} influences the values ψ . In agreement with Parmigiani and Thouless [208], for low values of l_s/h_1 , the ψ profiles given obtained for different values σ_{c1} overlap.

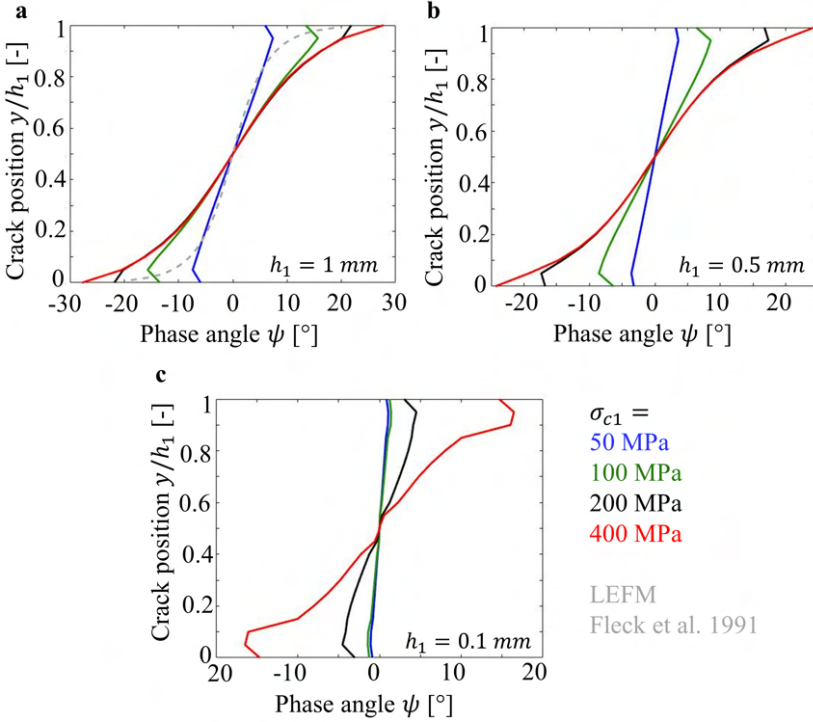


Figure 7.6 Variation of the phase angle ψ for crack located at different positions across the adhesive thickness for four values of σ_{c1} (50, 100, 200 and 400 MPa) for adhesive thickness h_1 equal to a) 1 mm, b) 0.5 mm and c) 0.1 mm. The grey dashed line in a) corresponds to the solution from Fleck et al. [206] with $\alpha = 0.8$ and $\beta = 0.2$.

Although an overlap is observed between the profiles obtained for the different values of σ_{c1} for the 1 mm thick adhesive, the predicted ψ profiles do not overlap close to the interface and ψ then depends on σ_{c1} . For σ_{c1} equal to 100 and 400 MPa, the profiles overlap if $0.4 \leq y/h_1 \leq 0.6$. Let us denote

Δh_1 as being the distance between the crack tip and the closest adherend. If y/h_1 is equal to 0.4 or 0.6, Δh_1 is equal to 0.3 mm which is the length of the cohesive zone. If the ψ profiles for σ_{c1} equal to 200 and 400 MPa are compared, an overlap is found for $0.1 \leq y/h_1 \leq 0.9$ if $h_1=1$ mm and for $0.3 \leq y/h_1 \leq 0.7$ if $h_1=0.5$ mm. In both cases, outside this range Δh_1 is lower than l_s .

To conclude, in agreement with Parmigiani and Thouless [208], there is thus an influence of the cohesive zone length on ψ when $l_s > \Delta h_1$. Although, even if $\Delta h_1 < l_s$ or $l_s > h_1$, the shape of ψ profile is well predicted, accurately predicting the crack propagation locus. It only has an effect on the predicted value for ψ . In our problem, what matters is not the accurate value of ψ , but well the shape of the profile to determine the crack propagation locus.

Influence of the adhesive thickness

Figure 7.7 shows the evolution of ψ across the adhesive thickness for different values of h_1 when σ_{c1} is set to 400 MPa. A clear influence of h_1 on the value of ψ is found while not predicted by Fleck et al. [206]. The ψ profiles exhibit similar shapes and predict crack propagation at the centre of the adhesive. Note that for very thin bondlines the shape of the profile close to the interface is slightly modified and suggests potential crack deflection towards the adherend. However, this could be an artefact coming from the fact that $l_s < \Delta h_1$ close to the interface for these cases.

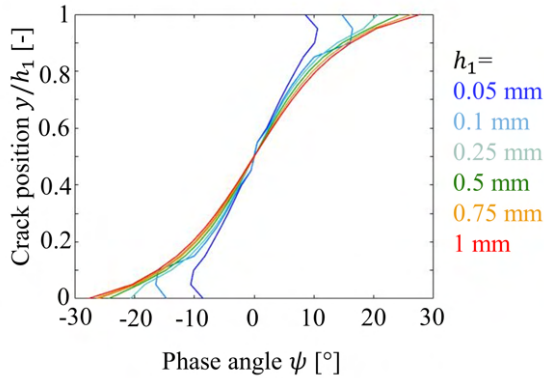


Figure 7.7 Variation of the phase angle ψ along the adhesive thickness for different values of adhesive thickness h_1 when $\sigma_{c1} = 400$ MPa.

One considers an asymmetric DCB with beams of thickness t_1 and $t_2 > t_1$. In this case, Chen et al. [210] showed that if t_1 increases, the phase angle ψ tends to decrease. In the present case, if a crack propagates at a given position y/h_1 in the joint, the bottom beam has a thickness $h_{bot} = h_s + (y/h_1)h_1$ while the top beam has a thickness $h_{top} = h_s + (1 - (y/h_1))h_1$. Figure 7.8 shows the evolution of h_{top}/h_{bot} with h_1 for y/h_1 equal to 0.6, 0.7, 0.8 and 0.9. The thicker the adhesive, the lower the h_{top}/h_{bot} . Based on the results of Chen et al. [210], it means that the thicker the adhesive, the larger the mode-mixity at the crack tip. Hence, it should be expected that $\psi(y/h_1)$ increases if h_1 increases in agreement with the results shown in Figure 7.7. Remember that Fleck et al. [206] considered infinite adherends.

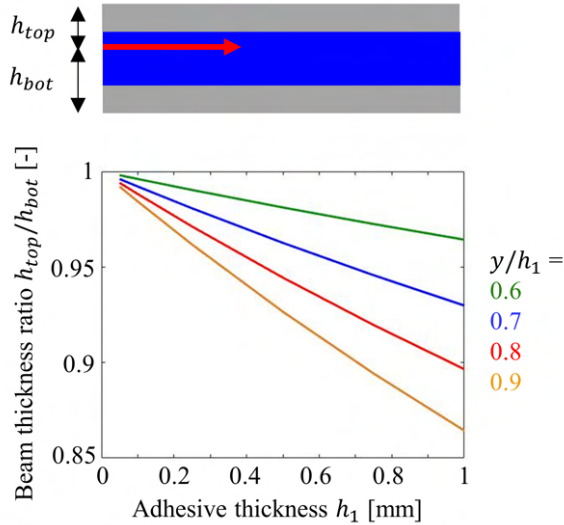


Figure 7.8 Variation of the beam thickness ratio h_{top}/h_{bot} with the adhesive thickness for y/h_1 equal to 0.6, 0.7, 0.8 and 0.9.

Finally, the adhesive thickness is known to influence the magnitude of the T-stress [206, 210]. Increasing h_1 tends to lead to positive values of the T-stress instead of negative values. In the present case the effect of the T-stress is not analysed. Hence, crack propagation in the centre of the bond-line is predicted even for thick joints although it could not be the case experimentally [216].

7.3.2 Comparison to LEFM

Simulation have been run using Abaqus as explained in Appendix A in order to compare the results obtained with the steady-state code and cohesive elements to those predicted by LEFM. The results are shown in Figure 7.9 for ψ evaluated at a distance of 10 μm from the crack tip. It is observed that for a thick adhesive layer, the LEFM and the steady-state CZ solutions are similar. Discrepancies appear for thin adhesive joints. LEFM also predict an influence of the adhesive thickness on the mode-mixity angle for a given crack position in the joint.

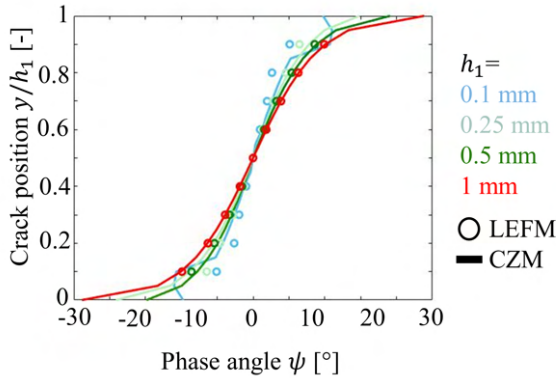


Figure 7.9 Comparison between the predicted mode-mixity angle given by LEFM and the steady-state cohesive solution for different adhesive thicknesses.

To conclude, the results of Fleck et al. [206] are reproduced with a cohesive zone modelling approach but some extra findings emerge from the analysis. First, the value of the phase angle ψ measuring the mode-mixity depends on (1) the distance from the crack tip d at which it is evaluated and (2) on the adhesive thickness. It is also influenced by σ_{c1} if the $l_s < \Delta h_1$. In addition, d must be lower than l_s to generate meaningful results. For what follows, it has been decided to take d equal to 0.

7.3.3 Elastoplastic adhesive

The focus is now on an adhesive characterized by an elastoplastic response with a yield strength σ_{01} and a strain hardening exponent $n_1 = 0.1$. The influence of σ_{c1}/σ_{01} and h_1/R_{01} on the joint toughness and on the crack propagation locus is investigated.

Influence of σ_{c1}/σ_{01}

The influence of σ_{c1}/σ_{01} is investigated first. In bulk materials, plastic dissipation takes place if σ_{c1}/σ_{01} is typically larger than 3 [207], depending on the strain-hardening capacity, while at an interface between an elastoplastic layer and an elastic adherend, the threshold ratio significantly depends on the elastic mismatch [95]. The influence of σ_{c1}/σ_{01} on $\psi(y/h_1)$ is investigated for adhesive thickness h_1 equal to 1.1 and 2.1 R_{01} in Figure 7.10 while the evolution of G_c with y/h_1 is shown in Figure 7.11.

First, for σ_{c1}/σ_{01} values lower than 3 (Figure 7.10a-b), the joint toughness is independent of y/h_1 and is only slightly enhanced. Plastic dissipation remains limited. The ψ profiles are similar to those reported for an elastic adhesive in Section 7.3.1, predicting crack propagation in the centre of the bondline (Figure 7.10a-b). If plasticity remains limited, the crack path selection is thus governed by the elastic mismatch, in agreement with Tilbrook et al. [213].

For σ_{c1}/σ_{01} equal to 3.5 and 3.75 (Figure 7.10d-e), for both thickness, G_c increases by more than 40%, indicating enhanced plastic dissipation with a dependence on y/h_1 . G_c is the lowest if interfacial failure takes place. In addition, the evolution of ψ with y/h_1 indicates crack deflection towards the adherends. Indeed, ψ decreases across the bondline thickness, opposite to the elastic case. The development of plasticity thus modifies the crack path selection, in agreement with Tilbrook et al. [213].

Figure 7.10c shows that ψ is influenced by h_1/R_{01} for σ_{c1}/σ_{01} equal to 3.25. For $h_1 = 1.1R_{01}$, G_c is almost independent of y/h_1 . In addition, plastic dissipation remains limited. The analysis of $\psi(y/h_1)$ shows that the crack propagates at the centre of the bondline (blue dot). If $h_1 = 2.1R_{01}$, plastic dissipation is enhanced and G_c is improved by a factor 1.5 at the centre of the bondline. The ψ profile is modified in that case, predicting crack propagation close to the interface (red dots).

To conclude, plasticity influences the crack path selection. In the absence of plastic dissipation and if the plasticity remains limited, the elastic mismatch governs the crack path selection. If the plastic dissipation taking place in the adhesive is important, it is no longer the case and the crack tends to be deflected towards one of the interfaces, in agreement with the results of Tilbrook et al. [213].

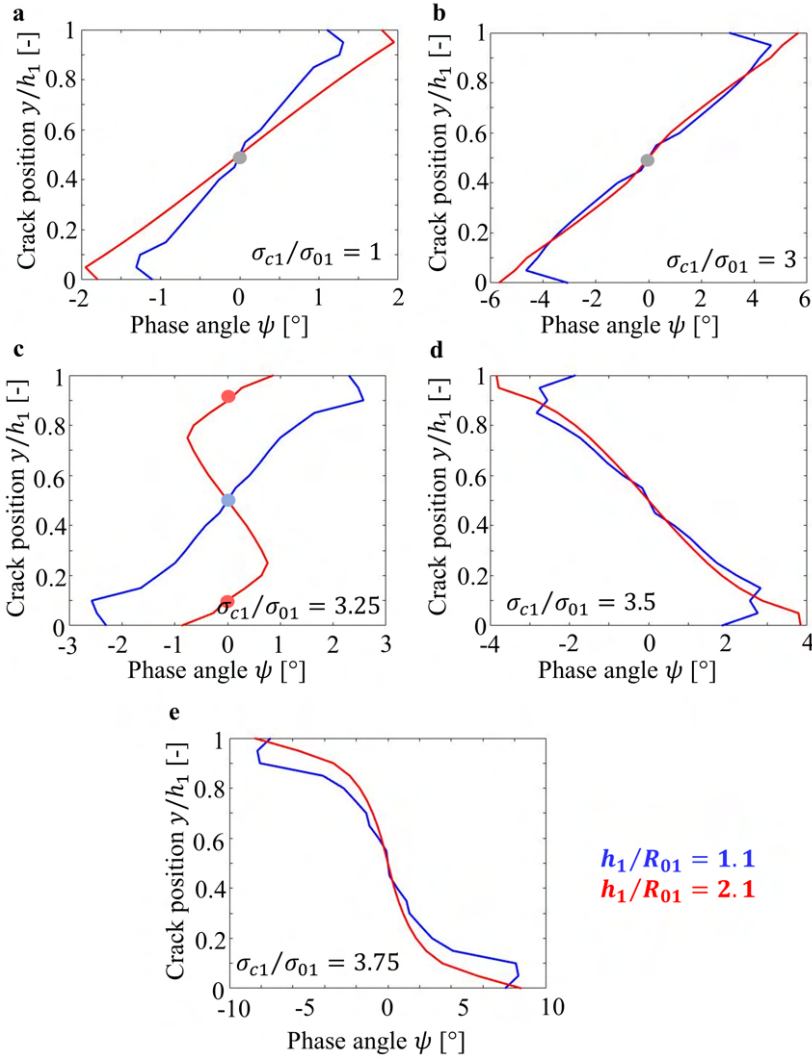


Figure 7.10 Variation of ψ along the adhesive thickness for h_1/R_{01} equal to 1.1 (blue) and 2.1 (red) for σ_{c1}/σ_{01} equal to a) 1, b) 3, c) 3.25, d) 3.5 and e) 3.75.

Influence of the adhesive thickness h_1/R_{01}

In adhesive joints, the adhesive thickness influences the fracture toughness by influencing the plastic dissipation taking place in the joint.

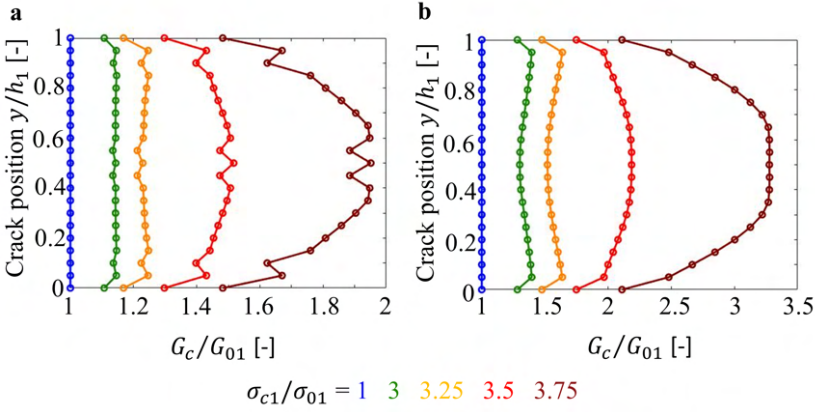


Figure 7.11 Variation of G_c depending on the crack location and σ_{c1}/σ_{01} when h_1/R_{01} is equal to a) 1.1 and 2.1.

For $\sigma_{c1}/\sigma_{01} \leq 3$, h_1 does not influence the selection of the crack path if the evolution of the T-stress with h_1 is neglected. The joint toughness is improved by increasing the adhesive thickness if $\sigma_{c1}/\sigma_{01} \geq 2.5$, indicating some plastic dissipation. For $2.5 \leq \sigma_{c1} \leq 3$, $G_c(y/h_1)$ exhibits a concave parabolic shape with an enhanced toughening effect when the crack runs close to the interface as shown in Figure 7.12 for thick joints. At the centre of the bondline, for thick adhesives, one can assume a situation similar to bulk materials and the enhancement in G_c remains limited in agreement with Hutchinson and Tvergaard [94]. Close to the adherends, the crack is shielded by the adherends and for a given σ_{c1}/σ_{01} ratio, the toughening effect is larger than in the bondline, in agreement with Tvergaard and Hutchinson [95]. Figure 7.17b indicates that the zone over which plastic dissipation is larger for a crack close to the adherend than for a crack at the centre of the bondline. For $\sigma_{c1}/\sigma_{01} < 3$, the crack tends thus to propagate in the centre of the bondline because (i) plastic dissipation is limited and elastic mismatch governs the selection of the crack path and (ii) if plastic dissipation takes place, G_c is the lowest at the centre.

For σ_{c1}/σ_{01} values larger than 3, Figures 7.13 and 7.14 show that the adhesive thickness influences the shape of the ψ profile. For low values of h_1/R_{01} , plasticity in the joint remains limited and the elastic mismatch governs the crack path selection. For thicker bondlines, the ψ profile tends to be inverted. As shown in Figure 7.13a, for $\sigma_{c1}/\sigma_{01} = 3.25$, crack propagation

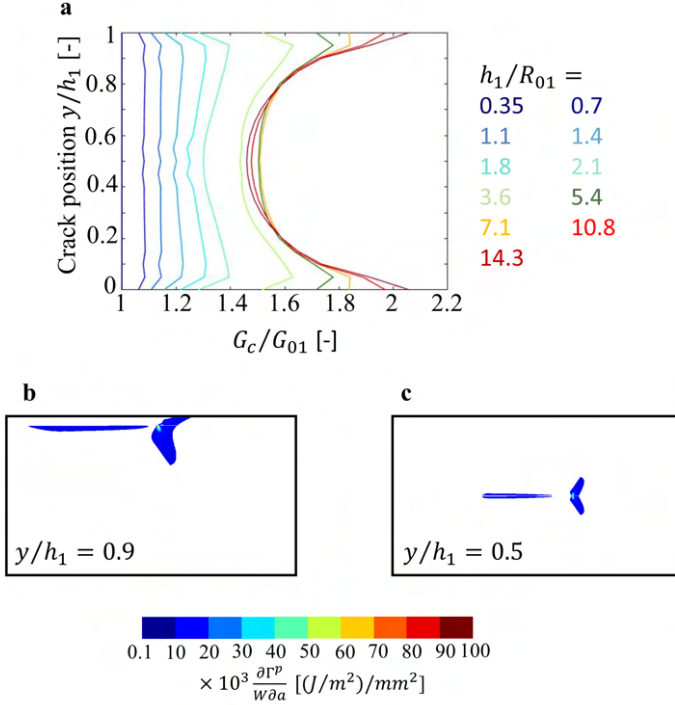


Figure 7.12 a) Variation of G_c depending on the crack location and h_1/R_{01} for σ_{c1}/σ_{01} equal to 3. b-c) Plastic dissipation zones for $h_1/R_{01} = 10.8$, $\sigma_{c1}/\sigma_{01} = 3$ for b) $y/h_1 = 0.9$ and c) $y/h_1 = 0.5$.

is predicted to be away from the centre of the bondline for h_1/R_{01} values ranging from 1.4 to 5.4. In this case, plasticity develops in contact with the adherends (Figure 7.13c). For thicker joints, the ψ profile is again inverted and is similar to what is reported for an elastic adhesive, predicting crack propagation in the centre of the joint. This occurs when the plastic dissipation zone is not in contact with the adherends as shown in Figure 7.13c. In addition, for $h_1/R_{01} \geq 7.1$, G_c starts to be the lowest at the centre of the bondline as shown in Figure 7.13b.

For $\sigma_{c1}/\sigma_{01} = 3.5$, the ψ profile (Figure 7.14a) is also inverted for low values of h_1/R_{01} , for which the plastic dissipation zone expands over the whole adhesive thickness. In addition, the crack tends to propagate at the interface, characterized by the lowest G_c as shown in Figure 7.14b. As h_1 increases, the shape of the G_c profile changes and exhibits a concave

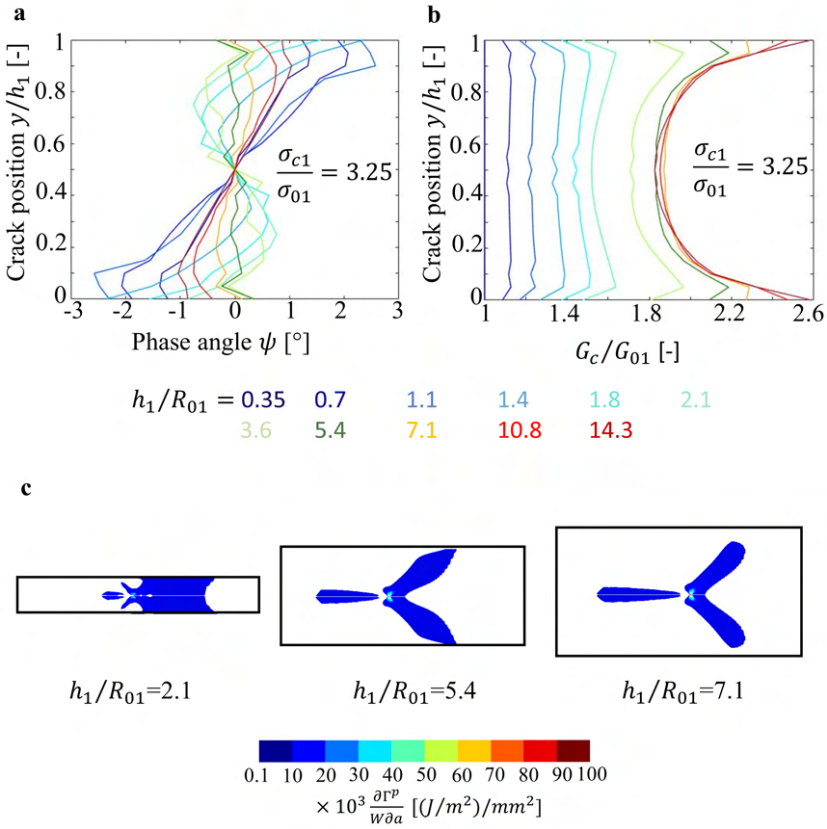


Figure 7.13 Variation of a) ψ and G_c across the adhesive thickness for different values of h_1/R_{01} if $\sigma_{c1}/\sigma_{01} = 3.25$. c) Plastic dissipation zones for h_1/R_{01} equal to 2.1, 5.4 and 7.1.

parabolic shape. G_c is then the lowest at the centre of the bondline. Progressively, the ψ profile is also modified, similar to case D in Figure 7.15. For $h_1/R_{01} \geq 10.8$, crack propagation can occur at the centre of the bondline if the crack initially propagates in the zone highlighted in green in Figure 7.15, but if outside this range, crack deflection towards the interface will take place. For $h_1/R_{01} = 14.3$, the crack tends to propagate towards the centre if $0.2 \leq y/h_1 \leq 0.8$. Figure 7.14c shows that for $y/h_1 = 0.8$, the plastic zone does not extend to the adherend while if $y/h_1 = 0.9$, it does. This change in crack propagation locus takes thus place when the plastic zone does not interact with at least one of the adherends.

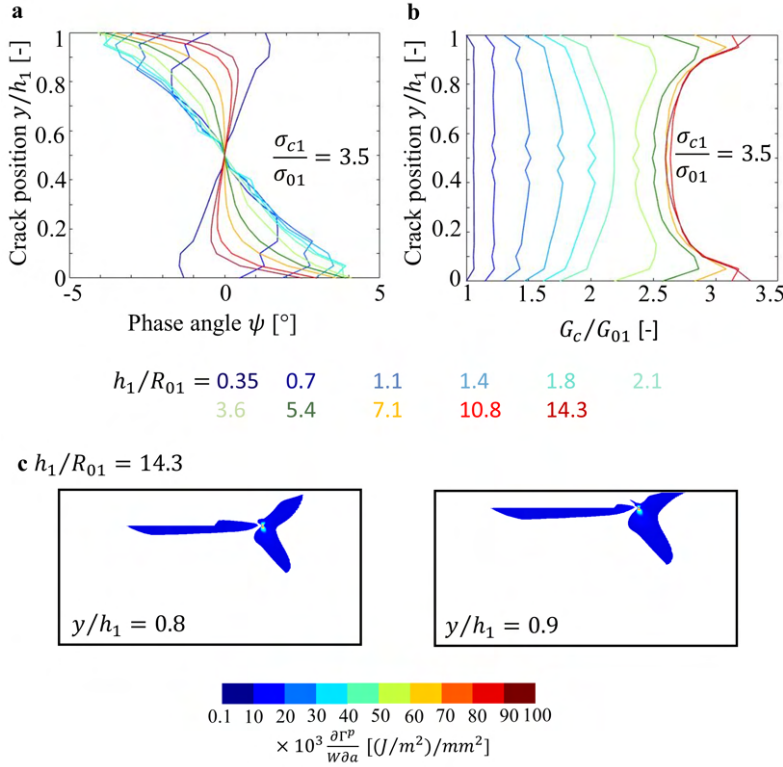


Figure 7.14 a-b) Variation of ψ across the adhesive thickness for different values of h_1/R_{01} for σ_{c1}/σ_{01} equal to a) 3.25 and b) 3.5. c-d) Variation of G_c depending on the crack location and h_1/R_{01} for σ_{c1}/σ_{01} equal to c) 3.25 and d) 3.5.

To conclude, crack path selection is governed by both the elastic mismatch and the plasticity, as summarized in Figure 7.15. Consequently, it depends on σ_{c1}/σ_{01} and h_1/R_{01} that dictates Γ^p . If plasticity remains limited, the crack tends to propagate at the centre of the bondline as predicted based on the elastic mismatch whatever the value of h_1/R_{01} . If plastic dissipation remains moderate ($\sigma_{c1}/\sigma_{01} \approx 3.25$) and if the adhesive layer is either thin or thick, the same conclusion is drawn. For intermediate adhesive thickness, there exists a crack path satisfying $K_{II} = 0$ and $\partial K_{II}/\partial y > 0$ away from the centre of the joint. If a large amount of plasticity is dissipated in the joint ($\sigma_{c1}/\sigma_{01} \geq 3.5$), the crack will tend to propagate towards the

adherends except for thick adhesive layers. In this last case, there exists a zone in which the crack will be deflected towards the centre of the bondline while outside this zone, it is deflected towards the interface (case D). As general conclusion, the elastic mismatch governs the crack path selection when plastic dissipation remains limited or if the plastic zone developing in the joint is smaller than the adhesive thickness.

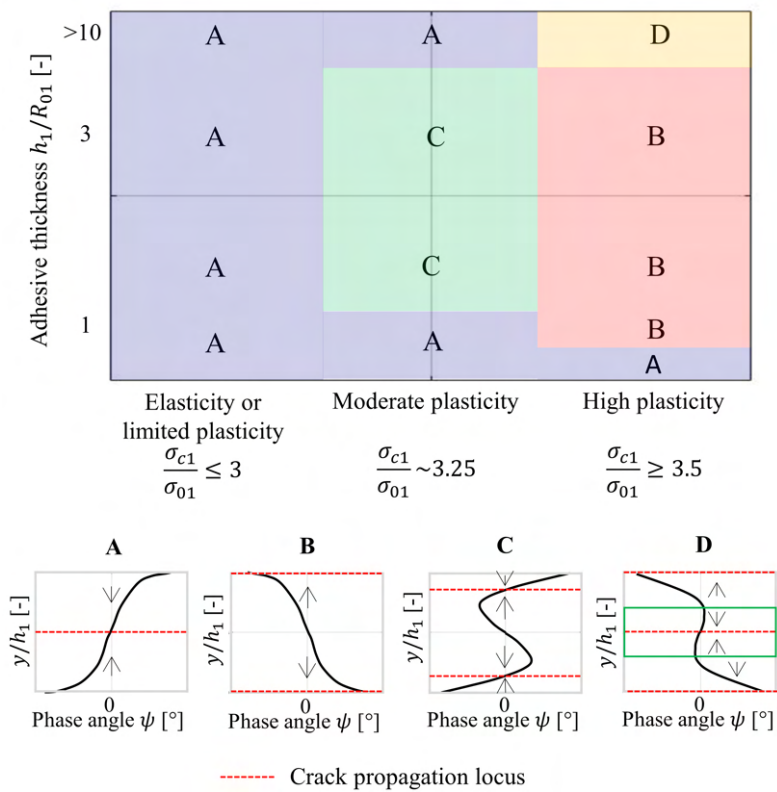


Figure 7.15 Summary of the different ψ profiles and crack path selection scenarios depending on the amount of plasticity in the bondline and the adhesive thickness.

7.4 Bilayer adhesive joint

In this section, the "adhesive stack" is made of two layers as shown in Figure 7.3. As a reminder, the idea is that one of these two layers would be purposely introduced to increase the toughness of the joint, so called "dissipative layer". The crack propagation locus and the joint toughness G_c is investigated depending on E_1/E_2 , σ_{01}/σ_{02} , h_1/R_{01} and h_2/R_{02} . However, before introducing plasticity, perfect elasticity is considered to determine the crack propagation locus depending on E_1/E_2 and h_1/h_2 .

7.4.1 Linear elastic layers

In the present case, the two interlayers shown in Figure 7.3 behave as linear elastic materials. The cohesive strength is set to 200 MPa for both materials while h_1 , h_2 and E_2 are modified.

The crack propagation locus in the joint is given by the position $y/(h_1 + h_2)$ depending on E_1/E_2 and h_1/h_2 as shown in Figure 7.16. For high h_1/h_2 , the crack tends to propagate in layer 1 even if $E_1 > E_2$, a case for which the elastic mismatch analysis would predict crack propagation in layer 2. If E_1/E_2 decreases, the crack tends to propagate at a higher position in the joint and thus in layer 1 which is the most compliant one, in agreement with the elastic mismatch theory.

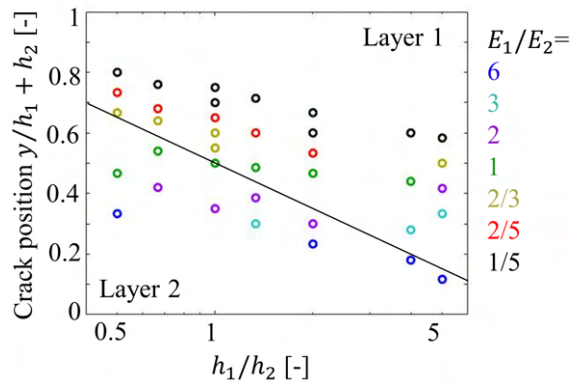


Figure 7.16 Crack propagation locus $y/(h_1 + h_2)$ for an elastic bilayer adhesive depending on h_1/h_2 and E_1/E_2 .

In the present case, it is assumed that the mode I (Γ_{cI}) and II (Γ_{cII}) cohesive energies are equal. Hence, whatever the mode-mixity, the energy dissipated in the cohesive zone is the same. Consequently, G_c is not influenced by the crack position in the stack. The influence of Γ_{cII}/Γ_{cI} is left for further investigations.

7.4.2 Elastoplastic layers

The two interlayers now behave as an elastoplastic material and their hardening law is given by Equation 7.4. The influence of σ_{ci}/σ_{0i} is first investigated before considering the influence of h_1/R_{01} and h_2/R_{02} .

Influence of σ_{c1}/σ_{01} , σ_{c2}/σ_{02}

Figure 7.17 shows the variation of the mode-mixity angle ψ and of G_c across the joint thickness for different values of σ_{c1}/σ_{01} and σ_{c2}/σ_{01} if $\sigma_{02}/\sigma_{01} = 2$ and $h_1/R_{01} = h_2/R_{02} = 2$. In what follows, we will first investigate the case of a crack propagating in the softer layer and then of a crack propagating in the harder layer.

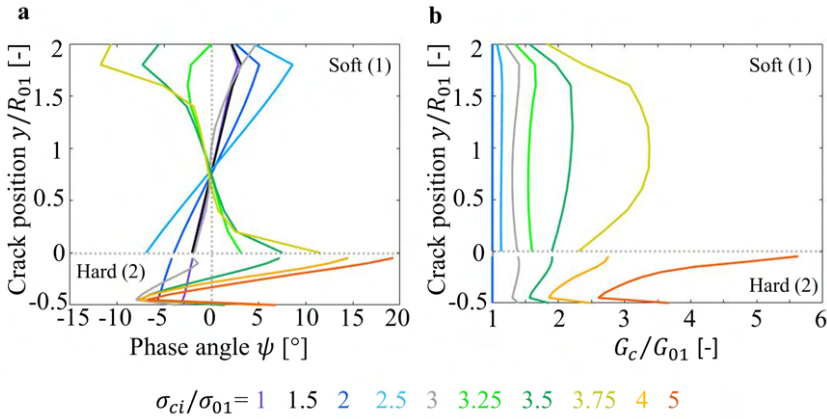


Figure 7.17 a) ψ and b) G_c profiles for different σ_{ci}/σ_{01} values if the adhesive stack is made of two elastoplastic layers such that $\sigma_{02}/\sigma_{01} = 2$ and $h_1/R_{01} = h_2/R_{02} = 2$.

□ Crack propagation in the softer layer (1)

If σ_{c1}/σ_{01} is lower or equal to 3, plastic dissipation remains limited or does not take place. In this case, as for a single elastoplastic layer, the ψ profile in

layer 1 is similar to the one found for a perfectly elastic joint. Consequently, the crack tends to propagate at the centre of the joint ($y = 0.75R_{01}$). This assumes that one can still use the criterion $\psi = 0^\circ$ to decide on the crack path which may not be correct, especially for widely varying G_{01} versus G_{02} or σ_{01} versus σ_{02} . We will get back to this complex question in the discussion.

For σ_{c1}/σ_{01} larger than 3, ψ varies similar to what is found for a single elastoplastic layer. In the present case $K_{II} = 0$ when $y = 0.6 - 0.8R_{01} = y_{KII=0-1}$, which is close to the centre of the joint but different from the centre of layer 1. If the crack propagates at a location such that $y > y_{KII=0}$, the crack will be deflected towards the interface with the adherend. At the opposite, if $y < y_{KII=0-1}$, the crack will be deflected towards the interface with the hard layer (2) and the crack path selection will then be governed by the properties of layer 2.

□ Crack propagation in the harder layer (2)

If σ_{c2}/σ_{01} is lower than 3, no plastic dissipation takes place in either the soft or hard layer and K_{II} increases when approaching the interface between the two interlayers. The crack propagates thus towards this interface with the soft layer. It can be assumed that if $h_1 < h_2$, the centre of the joint is located in the hard layer and the crack will remain in this layer. At the opposite, if $h_1 > h_2$, the crack propagates towards the interface with the soft layer and the crack path selection will then be governed by layer 1.

If σ_{c2}/σ_{01} is larger or equal to 3, crack propagation in the hard layer induces plasticity in the soft layer. However, the further the crack from the soft layer, the lower the joint toughness G_c as shown in Figure 7.17b. Away from the interface with the adherends, K_{II} increases as the crack approaches the interface with the hard layer. Depending on σ_{c2}/σ_{01} , there exists a position y such that $K_{II} = 0$ ($y_{KII=0-2}$) which does not correspond to y minimizing G_c . In the present case, it corresponds to $y = -0.3$ to $-0.35R_{01}$ while the lowest G_c is found for $y = 0.45R_{01}$. For $y > y_{KII=0-2}$, the crack is deflected away from the soft layer while for $y < y_{KII=0-2}$, the crack is deflected away from the interface with the adherend. In addition, plasticity also develops in the hard layer if $\sigma_{c2} \geq 3.5\sigma_{01}$, but it remains limited as Γ_2^p does not exceed 10% of Γ_1^p .

Influence of h_1/R_{01}

To investigate the influence of h_1/R_{01} , h_2/R_{02} is kept constant, equal to 2. The ψ and G_c/G_{01} profiles are shown in Figure 7.18 for h_1/R_{01} equal to 0.5, 1 and 4. It is observed that h_1/R_{01} does not affect the shape of the ψ and G_c profiles and the same conclusions can be drawn as mentioned here-above. Although, some additional informations can be extracted.

First, it is found in Figures 7.18b, d and f that for conditions in which plastic dissipation takes place, increasing the soft layer thickness, increases the toughness for a given location y/R_{01} . For a crack propagating in the soft layer, this is in agreement with the results found for a single elastoplastic layer while for a crack propagating in the hard layer by Bertholet et al. [97] and Dépinoy et al. [79].

The crack path selection is affected by h_1/R_{01} for given values of σ_{c1} and σ_{c2} . If σ_{c1} and σ_{c2} are such that the two layers behave elastically, the crack propagates in the soft layer if $h_1 > h_2$, in the hard layer if $h_2 > h_1$ and along the interface if $h_1 = h_2$. If σ_{c1} is such that $\Gamma_1^p \neq 0$, the ψ profile is inverted and there exists a position $y_{K_{II}=0-1}$ such that $K_{II} = 0$. This position varies and is equal to $0.3h_1$ if $h_1/R_{01} = 0.5$ and is around $0.45h_1$ if $h_1/R_{01} = 4$. Again, one can question the validity of the $\psi = 0^\circ$ criterion to decide on the crack path. For thin soft layers, this position is thus located close to the interface and there is a high probability that a crack propagating in the soft layer will propagate towards the interface with the adherends as it is the case when $y > y_{K_{II}=0-1}$. By increasing h_1 , this position tends towards $0.5h_1$.

Finally, in the previous subsection, a position such that $K_{II} = 0$ was found in the hard layer for high values of σ_{c2}/σ_{01} . However, it is found that for low values of h_1/R_{01} , it is not always the case. Consequently, for a thin soft layer, a crack propagating in the hard layer tends to propagate towards the adherend.

Influence of h_2/R_{02}

To investigate the influence of h_2/R_{02} , h_1/R_{01} is kept constant, equal to 4. The ψ and G_c profiles are shown in Figure 7.19 for h_2/R_{02} equal to 1, 4 and 8. It is observed that h_1/R_{01} does not affect the shape of the ψ and G_c profiles and the same conclusions can be drawn as mentioned here-above. Although, some additional informations can be extracted.

In agreement with the previous results, as long as $h_1 > h_2$, the crack propa-

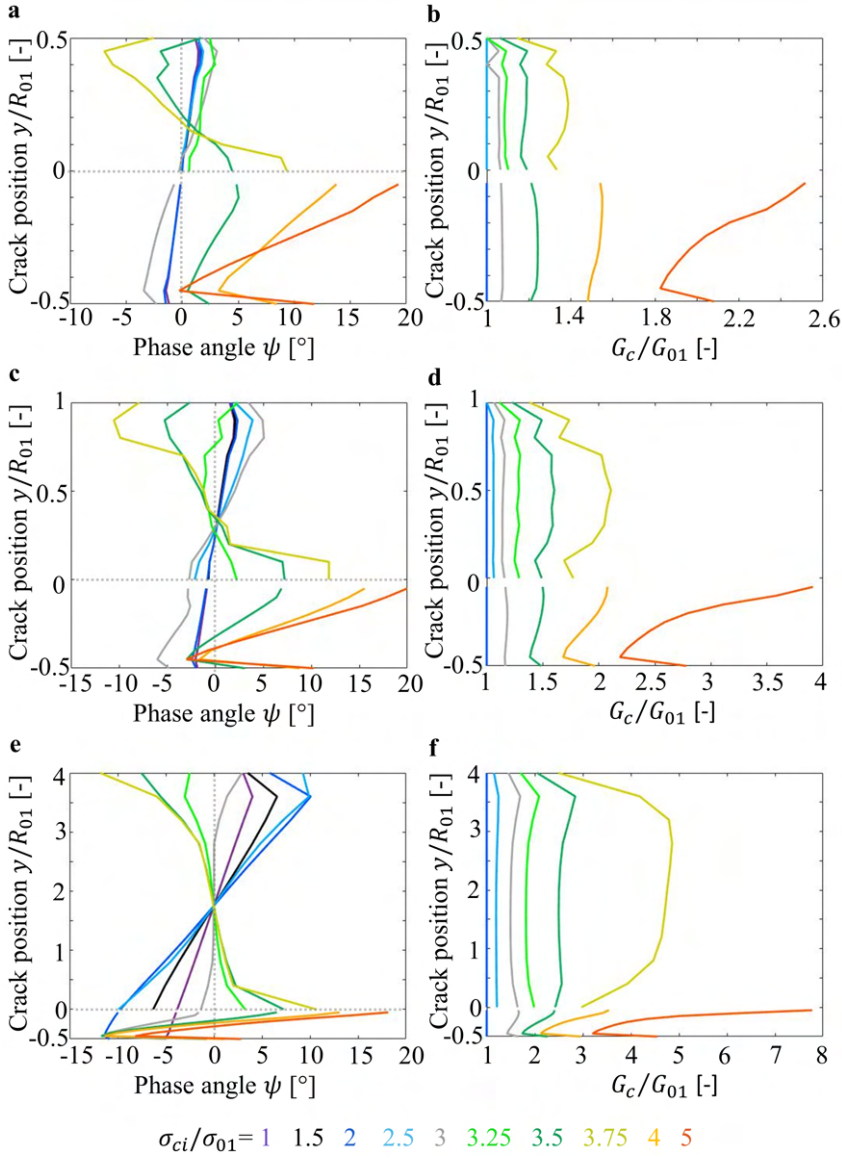


Figure 7.18 a-c-e) ψ and b-d-f) G_c profiles for different σ_{ci}/σ_{01} values if the adhesive stack is made of two elastoplastic layers such that $h_2/R_{02}=2$ and a-b) $h_1/R_{01} = 0.5$, c-d) $h_1/R_{01} = 0.1$ and e-f) $h_1/R_{01} = 4$.

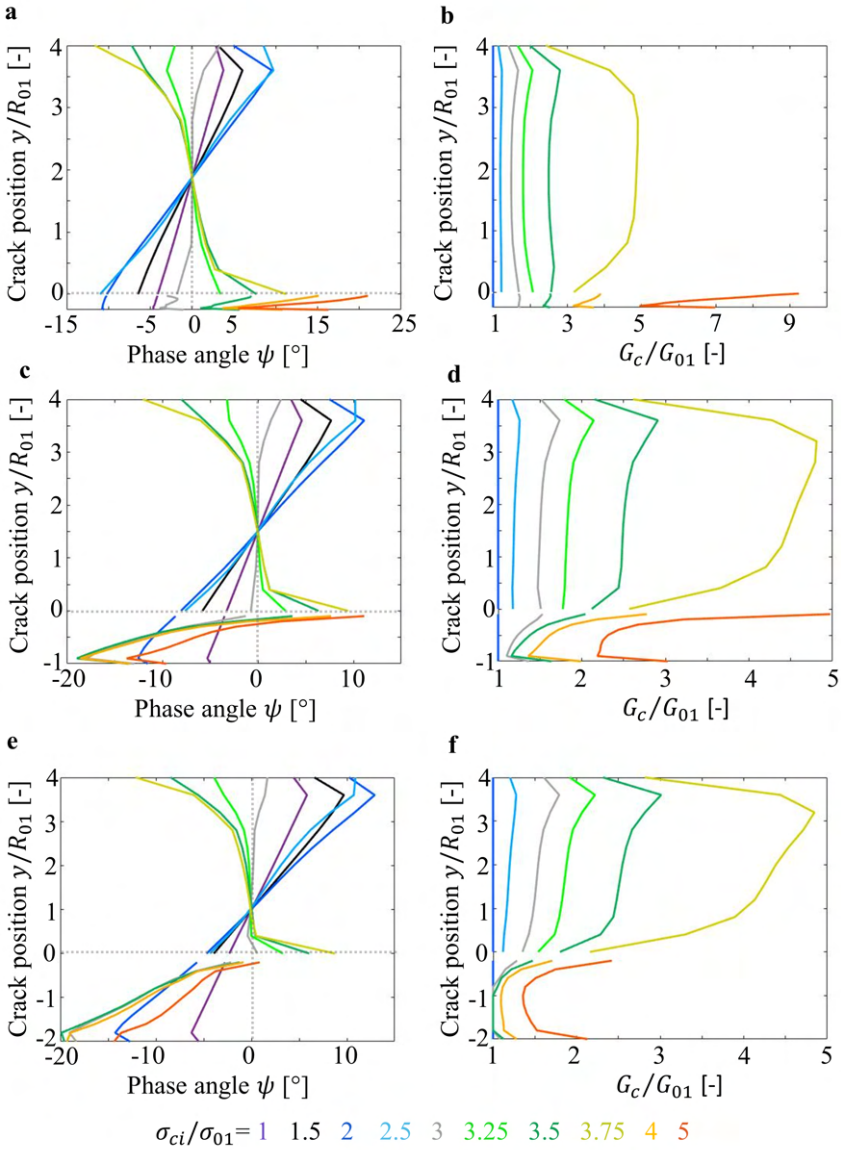


Figure 7.19 a-c-e) ψ and b-d-f) G_c profiles for different σ_{ci}/σ_{01} values if the adhesive stack is made of two elastoplastic layers such that $h_1/R_{01}=4$ and a-b) $h_2/R_{02} = 1$, c-d) $h_2/R_{02} = 4$ and e-f) $h_2/R_{02} = 8$.

gates in the soft layer if both layers behave elastically. A crack propagating in the soft layer generates plastic dissipation, $y_{KII=0-1}$ moves towards the interface between the two layers when h_2 increases.

h_2/R_{02} has an influence on G_c for a crack propagating in the hard layer, but not in the soft layer. The lower h_2/R_{02} , the higher the fracture toughness for a given crack position in the hard layer. However, if the crack propagates at a distance y from the interface larger than $R_{01} = 4R_{02}$, G_c is almost constant except close to the interface with the adherend. To conclude, if the crack propagates in the hard layer, h_2 must be minimized to maximize G_c , in agreement with the results of Bertholet et al. [97] and Depinoy et al. [79].

h_2 also influences the crack propagation locus. For thin hard layers, there is no locus such that $K_{II}=0$ when plastic dissipation takes place, indicating that the crack propagates towards the adherend. If h_2 increases, such locus exists but does not correspond to the lowest G_c point.

Crack path selection and discussion

Different scenarios can take place in the multi-layer joint depending on σ_{c1} , σ_{c2} , h_1 and h_2 . The scenarios are shown in Figure 7.20. Depending on σ_{c1} , σ_{c2} , a crack propagating in layer i results in plastic dissipation in the joint or not. This affects the crack propagation path selection. h_1/h_2 also influences the crack path selection. However, this is a simplified summary. Indeed, a soft polymer generally tends to be more compliant and tougher than a brittle one. And this is indeed the working principle expected for a dissipative interlayer: to trap the crack because of being softer and then dissipate a lot of energy because of high G_0 and hopefully high σ_c . The scenarios are discussed based on this comment.

If crack propagation in both layers (case 1) does not result in any plastic dissipation, the crack tends to propagate at the centre of the joint if $E_1 = E_2$. However for real systems, E_1 will tend to be lower than E_2 and $G_{01} > G_{02}$. Based on the elastic mismatch, crack propagation in layer 1 will be favoured. However, if the hard layer is significantly more brittle, the crack could also propagate in the hard layer.

Now, we consider a system in which crack propagation in the soft layer results in plastic dissipation while this is not true for a crack propagating in the hard layer. The crack is deflected towards the adherend or the hard layer. If it is deflected towards the hard layer and $h_2 > h_1$, crack propa-

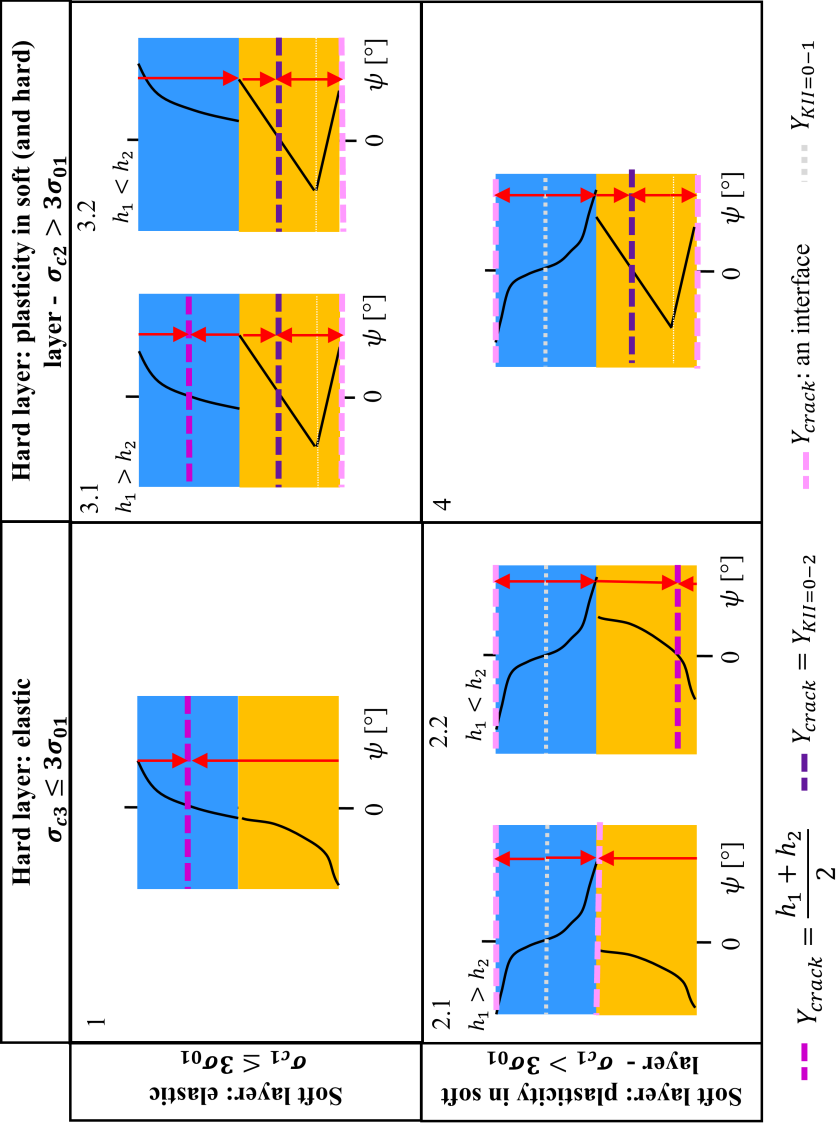


Figure 7.20 Summary of the crack path selection scenarios depending on σ_{c1}/σ_{01} , σ_{c2}/σ_{01} , h_1/h_2 for $\sigma_{02}/\sigma_{01} = 2$ and $E_1/E_2 = 1$. The red arrows indicate the crack propagation direction depending on the crack location.

gation takes place in the hard layer reducing G_c as $G_{c2} < G_{c1}$. If $h_1 > h_2$, crack propagation is more likely to take place at the interface between the two layers (case 2.1). Assuming a tough interface, the crack would remain in the hard layer which is the most brittle part of the joint.

The third case corresponds to a situation in which a crack propagating in the soft layer does not generate any plasticity while a crack propagating in the hard layer induces plastic deformation in the soft layer (case 3). If $h_1 > h_2$, depending on the initial crack location, the crack might propagate towards different positions. If initially located in the soft layer, it will remain in this layer. If it is initially located in the hard layer, it might propagate either in $y_{KII=0-2}$ or at the interface with the adherend. If $h_1 < h_2$, whatever the initial crack location, the crack will tend to propagate in the hard layer or at the hard layer/adherend interface. G_c is maximized if h_2 is minimized and if $\sigma_{c2} \geq 3\sigma_{01}$ for a given h_1 . If G_c exceeds G_{01} , the crack could select the path minimizing G_c which corresponds to crack propagation in the soft layer.

Finally, the propagation of a crack in both layers induces plasticity. In this case, the crack is less likely to stay in the soft layer and will either propagate at the soft layer/adherend interface or in the hard layer. The lowest G_c is reached if it propagates at the interface with the adherend. Once deflected in the hard layer, it might propagate at $y_{KII=0-2}$ or at the hard layer/adherend interface. G_c is maximized if h_2 is minimized and $\sigma_{c2} \geq 3\sigma_{01}$ for a given h_1 .

To conclude, an important parameter that controls the crack path selection is G_{01}/G_{02} . For case 1 and $h_1 > h_2$, additional finite element analyses should be conducted with G_{01}/G_{02} to determine whether the crack path selection is governed by the elastic mismatch or the weak path (lowest G_c). This also applies to case in case 3. In addition, the above scenarios do not consider the influence of the T-stress, which should be investigated for the different cases to determine the influence of σ_{01}/σ_{02} , E_1/E_2 and h_1/h_2 on its value. As a reminder $T < 0$ results in crack stability while $T > 0$ results in an unstable crack propagation [209].

Note on a crack approaching the interface

The propagation of a crack towards an interface and its deflection along the interface or its penetration in the material at the other side of the interface has been investigated by Pargmidiani and Thouless [217]. They

considered a crack propagating perpendicular to the interface. The interface and the second material are both characterized by a cohesive strength and a fracture toughness. Penetration in the second material takes place if the cohesive strength of the material is larger than the one of the second material. The critical ratio depends on the elastic mismatch. In the absence of elastic mismatch, it is about 3. If the second material is characterized by a lower Young's modulus than the first material, this ratio decreases. For a crack propagating from the hard to the soft layer, the crack is more likely to penetrate into the soft layer if $E_1 < E_2$ for given σ_{c1} and interface strength. The problem of kinking of a crack out of an interface has been investigated by He and Hutchinson [218]. For a crack propagating along an interface, the crack is trapped at the interface if the compliant material (soft layer) is tough and the stiff material (hard layer) is at least as tough as the interface. In the present case, it has been assumed that all interfaces are strong. However, this note shows that the interface properties might affect the crack path locations as well.

7.5 Discussion: design rules for tough multilayer adhesive joints

A crack propagating in the soft layer will tend to deflect towards either the interface with the adherend or towards the hard layer if plastic dissipation takes place when the crack propagates in the soft layer. At the opposite, crack propagation in the hard layer, even in the presence of significant plasticity, does not lead to the deflection of the crack towards the soft layer and the crack remains in the hard layer or propagates towards the interface with the adherend.

If one considers symmetric joints, two configurations are possible as shown in Figure 7.22. The soft layer is either located between the hard layer and the adherends (case A - Figure 7.22a) or between two hard layers bonded to the adherends (Case B - Figure 7.22b). It is assumed that $E_1 \leq E_2$, $G_{01} \geq G_{02}$ and $\sigma_{01} > \sigma_{02}$. We first consider a crack initiated in the soft layer and then a crack initiated in the hard layer.

7.5.1 A crack initiated in a soft layer

We first consider a crack initiated in the soft layer. Crack propagation in this layer results in plastic dissipation if $\sigma_{c1}/\sigma_{01} > 3$.

If crack propagation in the soft layer does not result in plastic dissipation,

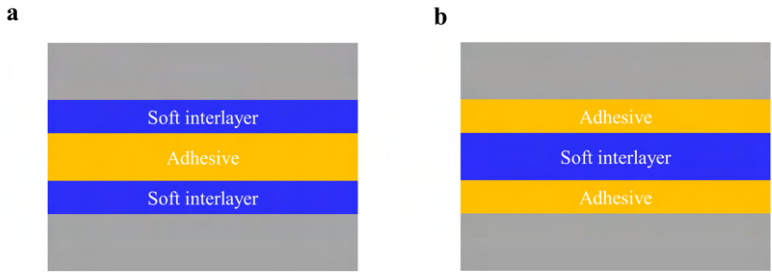


Figure 7.21 Possible joint configurations with a soft layer which is either placed a) close to the adherends or b) at the centre of the joint.

the crack will tend to propagate towards the centre of the joint. Hence, in Case A, it propagates towards the hard layer. However, depending on the elastic mismatch with the hard layer and on h_1 and h_2 , it could be possible to find a path such that $K_{II} = 0$ in the soft layer as found for an elastic bilayer joint. In Case B, it is trapped in the soft layer and propagates at the centre of it.

Alternatively, crack propagation in the soft layer generates a large amount of plasticity in the soft layer. Hence, the crack tends to propagate outside of this layer. In Case A, the position for which $K_{II} = 0$, $y_{KII=0-1}$, in the soft layer is close to the interface with the hard layer. If we consider the upper soft layer, there is thus a high probability of having a crack initiated at a position $y > y_{KII=0-1}$ and thus crack propagation towards the adherend. The crack will propagate towards the hard layer only if $y < y_{KII=0-1}$. In case B, the crack will propagate towards one of the hard layers. Depending on the plastic dissipation generated by a crack propagating in the hard layer (if it can cross the interface), several cases can be observed and are detailed here-below.

7.5.2 A crack initiated in a hard layer

We first consider a crack which is initiated in the soft layer. Crack propagation in this layer can result in plastic dissipation in the soft layer if $\sigma_{c2}/\sigma_{01} > 3$.

First, the crack propagation in the hard layer is assumed to not generate any plastic dissipation in the soft layer(s). In Case A, α and β are lower or

equal to 0. According to Fleck et al. [206], the crack will propagate towards the soft layer. In case B, the crack will propagate towards the centre of the joint which corresponds to the soft layer.

Plastic dissipation is now assumed to take place when a crack propagates in the hard layer. In case A, the crack tends to propagate towards the centre of the hard layer. At this position, the distance between the crack and the soft layer is maximized, minimizing the toughening effect resulting from plastic dissipation in the soft layers. In case B, there exists a position in each hard layer such that $K_{II} = 0$ ($y_{KII=0-2}$) if h_2 is large enough, otherwise it will propagate towards the adherend.

7.5.3 Generalization

In Case A, the crack could remain in the soft layer but is more likely to propagate towards the hard layer (with and without plastic dissipation) or the adherends (with plastic dissipation). If the crack is deflected towards the adherends, the presence of the hard layer is not beneficial as G_c will be maximized for a thick soft layer. If the crack propagates towards the hard layer and that plastic dissipation takes place when propagating in the hard layer, G_c will be maximized for a thin hard layer. This case seems not make sense as the crack will probably tend to propagate towards the closest adherend. G_c is maximized if the joint is only made of a single layer of the soft material in that case.

For Case B, the crack remains in the soft layer in the absence of plastic dissipation. If plastic dissipation takes place while the crack propagates in the soft layer, the crack is more likely to propagate towards one of the hard layers. If it reaches one of the hard layer, it will be deflected away from the soft layer towards the adherends. The fracture toughness will be maximized for the thin hard layers. This case is of particular interest for stiff/strong brittle adhesives for which a tough interface can be created with the adherends and with the soft layer. By propagating in the brittle adhesive, the crack is shielded by the plastic dissipation taking place in the soft layer and G_c is enhanced. The toughening effect is maximized if

- σ_{c2} is larger than σ_{01} by at least a factor 3,
- h_2 is minimized,
- h_1 is maximized,

- A strong interface between the hard and soft materials can be achieved.

Note that h_1 must be maximized to maximize G_c . However, a thick soft layer results in lower shear stiffness and shear strength of the joint [101]. Hence h_1 must be optimized such that it guarantees high enough joint G_c , stiffness and strength.

7.6 Conclusion

The goal of the study presented in this chapter is to investigate the potential toughening effect of the addition of a dissipative layer in an adhesive joint. This has been investigated relying on a cohesive zone model representation of the failure process into a steady-state framework. First, monolayer joints have been considered before exploring the response of bilayer joints in both cases with and without plasticity. These are the main conclusions.

- For a monolayer joint made of an elastic material, the results of Fleck et al. [206] are generally recovered. However, the distance from the crack tip at which ψ is evaluated influences its value, but not the position of $\psi = 0^\circ$ and its variation across the joint that dictates where the crack will propagate if this distance is shorter than the process zone length l_s . In addition to the results of Fleck et al. [206], it is found that the adhesive thickness slightly impacts the value of ψ for a given position y/h_1 .
- For an elastoplastic monolayer joint, if $\sigma_{c1} < 3\sigma_{01}$, plastic dissipation remains limited and the ψ profiles are similar to the elastic case, indicating crack propagation at the centre of the bondline. If plastic dissipation takes place, the crack tends to be deflected towards the adherends, except for very thin joints.
- For an elastic bilayer joint, the crack propagation locus depends on E_1/E_2 and h_1/h_2 . The crack will tend to propagate in the more compliant layer. However, there is an influence of h_1/h_2 . If h_1 is larger than h_2 , the crack might remain in layer 1 even if $E_1 > E_2$.
- For an elastoplastic bilayer joint, many scenarios might take place depending on σ_{c1}/σ_{01} , σ_{c2}/σ_{01} , σ_{02}/σ_{01} , h_1/R_{01} , h_2/R_{02} and h_1/h_2 . Based on these scenarios, the best joint configuration is the one including thin hard layers close to the adherends and in between with

a thick soft layer in between the two hard layers. In that case, G_c is maximized if $\sigma_{c2} > 3\sigma_{01}$.

The best joint configuration including a dissipative layer is interesting in presence of a stiff/strong brittle adhesive. The fracture toughness can indeed be increased by inserting a dissipative layer. However, this requires to have the right material properties: $\sigma_{02}/\sigma_{01} > 1$, $E_1/E_2 \leq 1$ and $\sigma_{c2} > 3\sigma_{01}$. However, these results do not consider the influence of the interfaces toughness and strength, of the T-stress and of the ratio G_{01}/G_{02} on the crack path selection. This is left for further investigation. Additional parameters could be investigated on the response of the joints such as σ_{01}/σ_{02} and the ratio between the mode I and mode II cohesive energies (Γ_{IIc}/Γ_{Ic}) and are left for further work.

In order to experimentally validate these results, joints could be manufactured with two different adhesives: a tough/soft adhesive and a stiff/brittle adhesive. The two adhesives can be stacked on top of each other to obtain a hard/soft/hard stack. However, this requires to have adhesives with matching curing cycles and which are under the form of films to avoid their spreading and mixing.

7.7 Appendix A: FE element model for comparison to LEFM

In order to compare the results obtained with the cohesive elements steady-state code with LEFM, static simulations are performed with the general purpose FE software Abaqus. The adherends are 5 mm thick while the adhesive layer thickness is set to 0.1, 0.25, 0.5 and 1 mm. Both the adhesive and adherend materials are modelled using perfect elasticity using the same properties as with the steady-state code model. The crack is modelled as a seam and its location along the joint thickness is varied. 4 nodes plane strain with reduced integration (CPE4R) are used. The size of the elements around the crack is about 2 μm around the crack tip before slightly coarsening the mesh as shown in

The mode mixity angle ψ is given by the ratio between K_{II} and K_I . Both are determined based on Equation 7.1 ahead of the crack tip. ψ varies depending on the distance at which it is evaluated from the crack tip. It is thus decided to evaluate it at a distance of 10 μm from the crack tip.

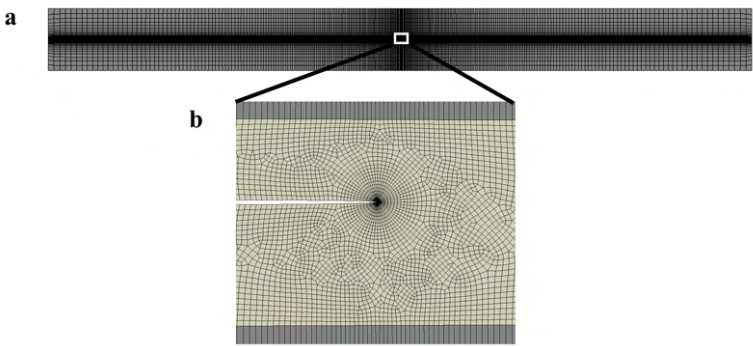


Figure 7.22 Typical mesh for LEFM simulation a) whole DCB and b) close to the crack tip.

PART III

Comparing architected joints to classical adhesive joints

8

Thickness-dependent selection of adhesive joints



This chapter is based on the article published in *The Journal of Adhesion*: Charline van Innis and Thomas Pardoën, *Thickness-dependent selection of adhesive joints*, J. Adhes., 110 (12), 2024.

8.1 Introduction

The manufacturing of complex structures such as windmills, planes, marine hulls and cars involves a vast number of joints, that are usually made by welding, mechanical fastening, and/or adhesive bonding. Adhesive bonding permits the assembly of both similar and dissimilar materials leading to mass savings and to homogeneous stress transfer across the joint [1]. However, the design of adhesive joints is a complicated task due to the large number of variables to be controlled [219–222] making complex the development of robust failure assessment frameworks as requested by

qualifying agencies.

The mechanical performances of adhesive joints depend upon the adhesive thickness t which influences the toughness, G_{Ic} , of the joint as well as the tensile strength, shear stiffness and shear strength. In general, the toughness increases with thickness until reaching a maximum at a thickness t^* corresponding approximately to the plane stress plastic zone size of the bulk adhesive equal to $2G_{Ic}E_a/(\pi\sigma_{0a}^2)$ where E_a and σ_{0a} are the adhesive Young's modulus and yield strength, respectively [13, 99, 102]. With increasing thickness, the degree of constraint in the bondline decreases, and the restriction on the development of the plastic zone is reduced. The degree of constraint is the highest for thin adhesive layers, leading to a high triaxial stress state ahead of the crack tip [103], which may lead to a change in failure mechanism [93, 98]. With increasing t , the plastic zone can expand into a larger volume and more energy can be dissipated by plastic deformation (Figure 8.1a). For large thicknesses, two different behaviors are reported. In some adhesive systems, once the maximum G_{Ic} is reached at $t = t^*$, the fracture toughness does not vary anymore with t , corresponding to adhesive A in Figure 8.1a [13, 104]. However, in other systems, after the optimum thickness has been attained, G_{Ic} decreases (adhesive B) as a result of the reduction of the plastic dissipation originating from shear stresses near the interface, to reach a lower plateau [13, 99, 102]. Note finally that in adhesive joints, K_{Ic} or G_{Ic} are usually not taken as the initiation fracture toughness, but as the steady state propagation value, knowing that the R-effect is usually weak or not observed at all [92, 104, 223–225].

Increasing the bondline thickness results in a reduction of the effective tensile and shear strength [143–147]. This is explained on the one hand by the fact that increasing t statistically leads to a larger number of larger microdefects (e.g. entrapped air) in the bondline [149] and, on the other hand, by the fact that when t increases, the stress state in the bondline varies and stresses tend to localize at the interface with the adherends near the joint edges (Figure 8.1b-c) [143, 145, 150] promoting adhesive instead of cohesive failure. This strength reduction is dependent upon the type of adhesive. Lap-shear tests have shown that this effect is less marked for ductile adhesives compared to brittle ones [147]. Finally, the stiffness also tends to decrease with increasing thickness [149]. Both stiffness and strength are influenced by the joint geometry and adherends material properties as these parameters influence the stress distribution in the bondline. The adhesive

thickness must thus be considered when searching for the optimum design of reliable adhesive joints.

The systematic materials selection approach in mechanical design pioneered by M.F. Ashby [201] starts by translating a given application/function into objectives and constraints. Next, “performance indices” are derived, also sometime called “figures of merit” or simply “material indices”. These indices are usually combinations of material properties, sometimes associated to geometry or loading parameters in more complex cases. This translation into objectives and constraints is followed by the elimination of the materials that do not meet the hard design requirements. This results into a list of potential materials among which the best choice is the one that maximizes the performance index or a weighted function of at least two indices. In the simple selection problems, the performance index is a group of material properties decoupled from the loading and geometry, making the final selection relatively straightforward and only based on materials properties. However, when properties depend on a dimension, as addressed here, the problem gets more complex and often requires a more advanced modelling effort.

The goal of the present study is to apply, for the first time, the materials selection approach to adhesive joints while systematically considering the impact of the adhesive layer thickness. Although bringing extra degrees of complexity in the analysis, this choice to include thickness effects will prove the existence of some non-obvious trade off and selection options depending on the constraints and objectives. Furthermore, we will not only address conventional joints, but also architected solutions, enlarging the window of options as also proposed by Ashby in the extension of the selection procedure to hybrids [201, 226]. During the past ten years, researchers indeed developed architected joints by inserting meshes [122, 123], stop holes [121, 123], metamaterials [84, 141] in the bondline or by patterning the adherend surface [82, 83] to improve the joint toughness, but generally without comparing the solution to classical adhesive joints or without addressing other constraints in parallel.

The mechanical models used to describe representative joint loading configurations are introduced in section 8.2. Then, the selection method is detailed in section 8.3 and applied to several case studies, in section 8.4, for conventional joints, and, in section 8.5, for architected joints. The limitations of the analysis are discussed in section 8.6. In order to apply this

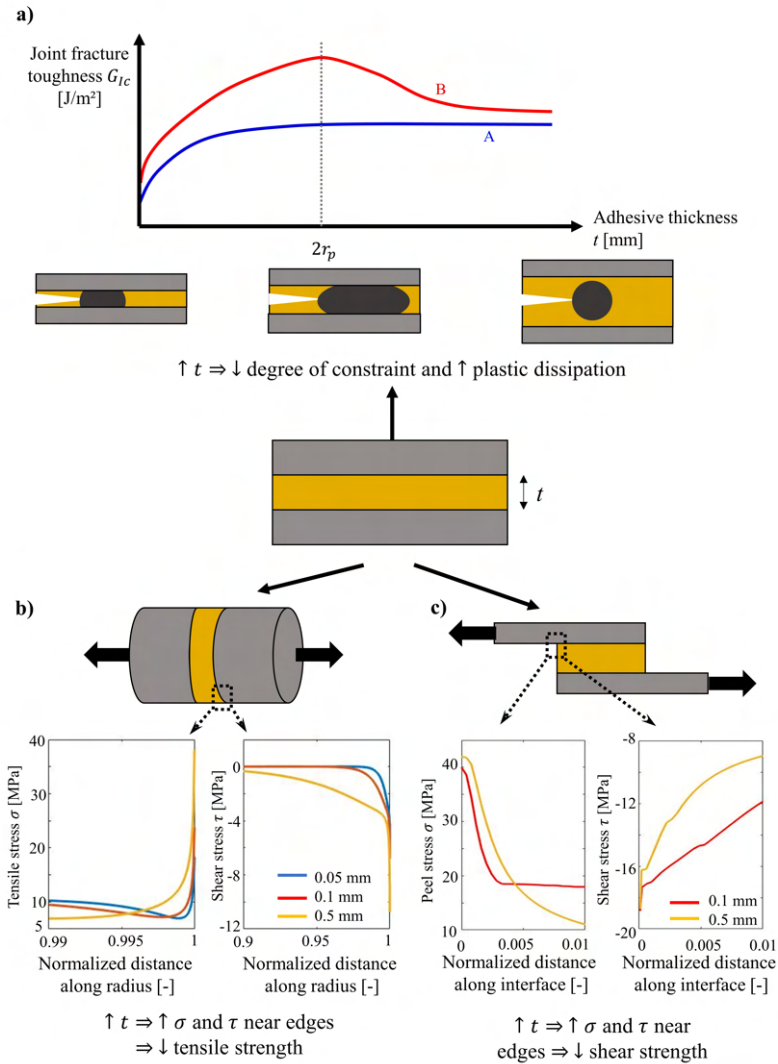


Figure 8.1 Impact of thickness on adhesive joint properties: a) fracture toughness (adhesive of type A (blue curve) and type B (red curve)); b) tensile and shear stress distribution in tensile butt tests close to interface; c) peel and shear stress distribution in lap shear joints close to interface (results from FE simulations with the software Abaqus, based on an elastoplastic adhesive behavior).

methodology to real adhesive joints, a wide range of literature data has been collected. These data include the fracture toughness and variation with adhesive thickness as well as bulk adhesive Young's modulus, Poisson ratio, tensile strength and, when available, density, type of adherends, bonding process, resistance to chemicals, humidity and maximum service temperature. The database is contained in .csv and .txt files which are called by a MATLAB app performing the adhesive selection based on the loading conditions. This information is made open to the community (Supplementary Material).

8.2 Mechanical models of adhesive joints

This section presents the analytical or semi-analytical models that will be used to relate the critical load applied to a structural joint, schematized in a generic way in Figure 8.2, to the mechanical properties of the adhesive joint in terms of fracture toughness and strength, taking the thickness effects into account. Two different loading conditions will be addressed in order to cover a range of realistic configurations involved in adhesively bonded components: the butt joint configuration that involves loads predominantly normal to the plane of the joint and the shear lap configuration to treat conditions where the loading is a macroscopically applied shear. Depending on the degree of complexity and assumptions that are selected, the analysis can go quite far and is then often not amenable to analytical treatment. Among others, the stiffness of the adherends can affect the mechanics of the bondline [13, 227–229]. Now, the goal of the study is primarily to look at the optimum choice of adhesive including its thickness, and not at predicting the failure load exactly. The objective is thus to use models that produce the right dependencies on the parameters playing the primary role in setting the magnitude of the failure load (i.e. fracture toughness, strength, stiffness, thickness) so that the ranking of the adhesives will be correct. Two levels of modelling are addressed. A simplified level neglects the effect of the adherend stiffness as well as other advanced effects that influence the stress distribution in the bondline such as related for instance to the adherend thickness, the adherend rounding and/or tapering. This first level is interesting when comparing, for example, different solutions for architected adhesive joints as done in subsection 8.5.1. A more advanced level of modelling is then addressed to produce a more accurate description of the mechanics of adhesive joints under dominantly

tensile or shear loading conditions relying on the literature [150, 151, 230, 231], as presented in Equation 8.2.1 and 8.2.2

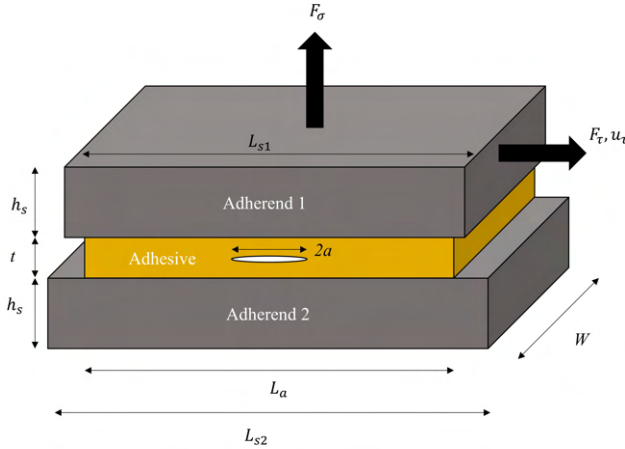


Figure 8.2 Schematic of the adhesive joint with its dimensions: width W , bonded length L_a , adherend lengths L_{s1} and L_{s2} , thickness of the adherends h_s and of the adhesive t . A crack of length $2a$ is present in the joint subjected to both tensile and shear loads.

8.2.1 Tensile butt joints

Tensile butt joints are often used to determine the tensile strength of an adhesive joint. Two large blocks made of the adherend materials, with Young's modulus E_s , are bonded with an adhesive layer of modulus E_a and thickness t as shown in Figure 8.2 with $L_{s1} = L_{s2} = L_a$. The joint contains a crack of length $2a$ at its centre, assuming that the only possible defects are voids developing during curing, treated as a crack, neglecting the fact that during in-service life, damage can also develop at the edges of the joint due to corner stress singularity. A damage tolerant approach is thus adopted to determine the joint tensile strength. The toughness G_{Ic} is thickness dependent, hence $G_{Ic} = G_{Ic}(t)$. The joint tensile strength is referred as the effective tensile strength σ^{f*} to be contrasted with the adhesive strength σ_a^f which would correspond to a material without major defects nor macro stress concentration (as measured in a clean uniaxial tensile test on bulk samples for instance). Experimentally, increasing t first

leads to an increase of σ^{f*} [232, 233] which is followed by a decrease of σ^{f*} [143, 145, 232, 233] once t is larger than the optimum thickness. This decrease has two main origins. First, when increasing t , the stress state in the bondline and, more precisely, near “the corners” (where the interface meets the external surface), changes involving larger tensile stresses and sometimes a transition towards adhesive failure. Second, increasing t statistically leads to a larger defect density (e.g. entrapped air)[149]. The adherends Young’s modulus can also have an influence on σ^{f*} [230].

Simple model for pre-cracked butt joint

The influence of the adherends is neglected. Depending on the crack length, the failure of the joint¹, dictated by cracking of the adhesive, will be governed by either the bulk adhesive tensile strength σ_a^f or by the adhesive fracture toughness $G_{Ic}(t)$ or $K_{Ic}(t)$. K_{Ic} is related to the critical energy release rate G_{Ic} by $K_{Ic} = \sqrt{G_{Ic} E_a^*}$ where E_a^* is equal to E_a under plane stress conditions and equal to $E_a / (1 - \nu_a^2)$ under plane strain conditions, and ν_a is the adhesive Poisson ratio. If the crack is short with respect to the joint dimensions, the adhesive strength governs failure and the effective tensile strength is bound by σ_a^f , hence:

$$\sigma^{f*} = \sigma_a^f. \quad (8.1)$$

For cracks longer than the critical crack length given by $a_c(t) = G_{Ic}(t) E_a / \pi \sigma_a^{f2}$, the resistance of the joint is set by the fracture toughness of the adhesive. To avoid failure, the remote stress intensity factor K^∞ must be lower than $K_{Ic}(t)$ of the adhesive leading to

$$\sigma^{f*} = \frac{K_{Ic}(t)}{Y \sqrt{\pi a}}, \quad (8.2)$$

where Y is a geometric factor equal to 1 if $2a \ll L$ and W . Equation 8.2 is equivalent to

$$\sigma^{f*} = \frac{1}{Y} \sqrt{\frac{G_{Ic}(t) E_a}{\pi a (1 - \nu_a^2)}}. \quad (8.3)$$

The transition from one regime to the other takes place at the critical crack length $a_c(t)$. Depending on the magnitude of $2a$, failure may be governed

¹The term “failure” means that at least macroscopic damage is initiated in the adhesive. Under some conditions, unstable crack propagation can take place leading to complete adhesive fracture while in other cases, the crack may start propagating without complete failure.

by the strength for some range of thickness and by the toughness for other thicknesses.

More advanced model for pre-cracked butt joint

The effect of adherend stiffness is now taken into account. Van Loock et al. [230] investigated the influence of crack length, fracture toughness and elastic modulus mismatch ratio on the adhesive tensile strength. A fracture process zone model was introduced through a traction separation law, characterized by a finite peak strength σ_{peak} set equal to σ_a^f (while the second characteristic parameter of the law is related to G_{Ic} as the work of separation). The fracture process zone has a length l_s which sets an important length scale in the analysis. In the limit of very high σ_{peak} , one falls back onto a single parameter (G_{Ic}) problem for which the authors proposed a simplified analytical treatment³. Four regimes were evidenced by Van Loock et al. instead of two. The two first regimes (A and B in Figure 8.3) correspond to the short crack case for which the failure of the joint is only governed by the adhesive properties and for which

$$a + l_s(t) < 0.15t, \quad (8.4)$$

with the fracture process zone length l_s (for which thickness dependence is introduced here) given by

$$l_s(t) = \frac{E_a^* G_{Ic}(t)}{\pi \sigma_a^{f2}}. \quad (8.5)$$

In regime A, joint failure is governed by the adhesive strength while in regime B, it is governed by the fracture toughness. Strength-controlled fracture takes place when

$$l_s > 3.37a, \quad (8.6)$$

and the effective tensile strength is obtained by solving

$$\frac{2l_s}{t} = \frac{8}{\pi^2} \frac{2a}{t} \ln \left[\sec \left(\frac{\pi \sigma^{f*}}{\sigma_a^f} \right) \right]. \quad (8.7)$$

²This is an assumption that considers σ_a^f as the “intrinsic” strength of a bulk adhesive material with no defect, and disregards the crack tip constraint effect (e.g. [94]).

³When only considering the crack length with respect to the adhesive thickness, Van Loock et al. suggests as simplified analytical treatment $\sigma^{f*} = \sqrt{(E_a^* t)(E_s 2\pi a)}$ which is valid for large σ_{peak} , falling back on one parameter despite of the fracture resistance.

For the two other regimes (C and D in Figure 8.3), one considers cracks of intermediate length as well as long cracks. The influence of the substrates can no longer be neglected, with a potential shielding effect on the crack. If it is assumed that both the adherends and the adhesive have the same Poisson ratio, the strength of the joint is obtained by solving

$$\frac{2l_s}{t} = \frac{8}{\pi^2} \frac{E_s^*}{E_a^*} \frac{2a}{t} \ln \left[\sec \left(\frac{\pi \sigma^{f*}}{\sigma_a^f} \right) \right] + \frac{1 - \bar{\nu}^2}{\pi} \left(\frac{\sigma^{f*}}{\sigma_a^f} \right)^2 \left(1 - \frac{E_s^*}{E_a^*} \right) \left(1 - \frac{2a}{L_a} \right)^{-2}, \quad (8.8)$$

with $\bar{\nu} = \nu / (1 - \nu)$ and L_a , the bonded length. The transition between the strength-governed regime D and the toughness-governed regime C is given by

$$\frac{2l_s}{t} = 3.37 \frac{E_a^*}{E_s^*} \frac{2a}{t} + 0.31 \left(1 - \bar{\nu}^2 \right) \left(1 - \frac{E_a^*}{E_s^*} \right) \left(1 - \frac{2a}{L_a} \right)^{-2}. \quad (8.9)$$

This analysis considers thus four non-dimensional parameters: a/t , l_s/t , E_a/E_s and a/L_a as shown in Figure 8.3. First, the transition between the short crack regime and the intermediate and long crack regime given by Equation 8.4 is independent of E_a/E_s , as well as of σ^{f*} (Equation 8.7). In the intermediate and long crack regimes, the transition from regime C to D and the magnitude of σ^{f*} for given l_s and a depend on E_a/E_s . By decreasing E_a/E_s , the shielding effect of the stiff adherends is more pronounced, resulting in a larger σ^{f*} for given a , l_s and t .

The model derived by Van Loock et al.[230] does not consider the fact that, sometimes, when increasing the adhesive thickness, the joint tends to fail by adhesive failure or cohesive failure close to the interface, leading to a decrease of the effective tensile strength. This arises from the stress singularity at the interface corner. Reedy and Guess [150] derived a model based on an interface corner stress intensity factor to predict σ^{f*} , assuming that failure only takes place near the interface and that the joint is free from any defect. In tensile butt joints, the interface corner stress singularity order is $\lambda - 1$. Hence, the stress components near the corner can be expressed as $\sigma_{ij} = K_f r^{\lambda-1} f_{ij}$ with K_f the interface corner stress intensity factor and r the distance from the corner. For a semi-infinite layer of thickness t , the effective tensile strength is then given by

$$\sigma^{f*} = \frac{(1 - \nu)}{\nu} \frac{K_f}{A_p(\nu)} \left(\frac{t}{2} \right)^{\lambda-1} \quad (8.10)$$

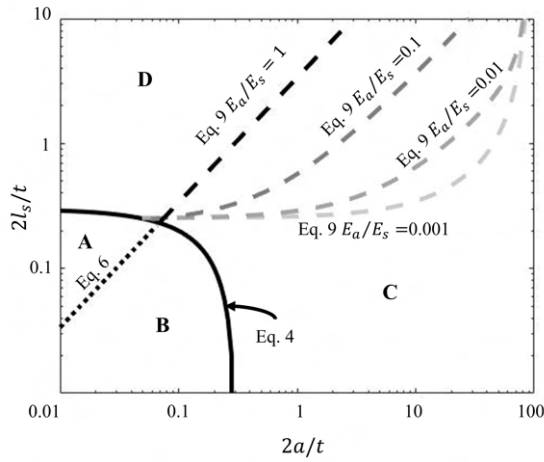


Figure 8.3 Failure regime maps of joints subjected to tensile stresses, based on Van Loock et al. [230]. The transition between regimes A, B, C and D are shown for mismatch ratios E_a/E_s from 1 to 0.001. A and B are controlled by the adhesives only with A being the strength-controlled regime and B the toughness-controlled one. C and D are the two regimes for which the adherends have an influence with C being the toughness-controlled regime and D the strength-controlled regime.

where $A_p(\nu)$ and λ are derived in Appendix A. A complete model must thus consider the presence of a crack, but also of the stress singularity at the interface corner.

8.2.2 Lap shear joints

The single lap-shear test is the most common test used by the industry to determine the joint shear strength. However, in single lap-shear, stresses do not uniformly develop in the bondline and the bending of the adherends lead to the development of peel stresses near the edges making the analysis difficult [228, 234]. Hence, it provides an “apparent” shear strength which is strongly dependent upon the adherend properties (Young’s modulus and yield strength), adherend thickness, overlap length, and distance between the loading point and the bonded part. The shear strength is also influenced by the adhesive thickness due to the change in stress state in the bondline and the increasing presence of defects as t increases. Here, a thick adherend shear test (TAST) configuration is considered to avoid ex-

tra complexity associated to regular single lap-shear test. In addition, we neglect here the mode II fracture mechanics problem associated to the centred crack in the adhesive layer used for the case of butt tensile joints is neglected.

Simple mechanical description of shear lap configuration

For the elementary analysis, the geometrical parameters and adherend properties are neglected. Hence, it is simply assumed that failure occurs when

$$\tau^{f*} = \tau_a^f, \quad (8.11)$$

with τ_a^f being the “ideal” shear strength of the bulk adhesive as measured on as perfect as possible system. Equation 8.11 has no influence of the thickness, leading to an overestimation of the effective shear strength.

Advanced model for shear lap configuration

For this second level of analysis, the effect of the adherend properties and of the thickness are now taken into account. Models outside fracture mechanics formalism have been derived in order to predict the lap-shear strength. However, these models fail to correctly capture the evolution of τ^{f*} with t , predicting an increase instead of a decrease as t is reduced [148]. Models based on finite fracture mechanics are more successful but require numerical simulations which would make the selection analysis cumbersome [235]. Some authors suggested to use a Generalized Stress Intensity Factor (GSIF) approach [151, 236–239] to predict the joint shear strength when adhesive failure occurs or cohesive failure takes place close to the interface. If we consider of lap-shear or TAST configuration, the presence of corners, see Figure 8.2 when $L_{s1}, L_{s2} > L_a$, at the joint edge induces a singular stress field. The order of the singularity is denoted as $\lambda - 1$ and the magnitude of the stress field is scaled by the parameter H . Stresses at a distance x from the corner and in the direction given by the angle θ are then expressed as $\sigma_{ij}(x, \theta) = Hx^{\lambda-1}f_{ij}(\lambda, \theta)$. It was postulated that the joint failure occurred when H is equal to a critical value H_c . However, several studies have shown that H_c is both dependent on material properties and on the joint geometry. Regarding the joint geometry, increasing both the adhesive and the adherend thickness results in a decrease of H_c while increasing the overlap length and the free length (unbonded length between the loading point and the overlap) increases H_c [239]. In addition, a higher joint shear strength is not correlated with a higher H_c . An expression for H has been

given by Askarinejad et al. [151]: $H = \tau^\infty t^{1-\lambda} b(\alpha, \beta)$ which depends both on material properties (λ, α, β) and on the joint geometry (t and b). More details on the determination of λ for the TAST configuration is given in Appendix B. Based on this GSIF approach, we heuristically assume that the effective shear strength is given by

$$\tau^{f*}(t) = \begin{cases} \tau_a^f & \text{for } t \leq 0.1 \text{ mm} \\ At^{\lambda-1} & \text{for } t > 0.1 \text{ mm} \end{cases}, \quad (8.12)$$

with A such that $\tau^{f*}(t = 0.1 \text{ mm}) = \tau_a^f$. The adhesive selection can be affected by this arbitrary choice, but its only purpose is to provide examples in Figure 8.4.1 when addressing the influence of t and E_s on τ^{f*} . Note that the power law variation will involve an exponent -0.35 (assuming $\lambda = 0.65$) for a corner not so far from the exponent -0.5 for a sharp crack under mode II. It is important to note that there exists an optimum thickness maximizing the joint shear strength, although it is not often reported in the literature [109]. Below this thickness, the shear strength tends to increase as t increases while any increase in thickness leads to a decrease in τ^{f*} at larger thicknesses as considered by Equation 8.12. This optimum thickness for instance depends on surface roughness. As this optimum thickness is dependent on several parameters and is often not reported, it has been decided to keep it as equal to 0.1 mm for all adhesives and the dependence of τ^{f*} below the optimum thickness is neglected by assuming a constant shear strength. This equation thus not necessarily predict the joint shear strength with high accuracy, but is sufficient to correctly rank the adhesives of the database by considering a decreasing strength as thickness increases and the elastic mismatch between the adhesive and adherends.

It is important to note that the GSIF approach has been developed for brittle adhesives and has been extrapolated to ductile adhesives in the present analysis. Using a brittle adhesive model for ductile adhesives can lead to an underestimate of the joint shear strength as ductile adhesives tend to be tougher and that an increase in toughness often results into an increase in joint shear strength [229]. Note also that we are not treating in this study the case of a lap-shear configuration with a pre-crack undergoing a complex mixed mode loading. We will discuss this limitation at the end of the paper.

8.2.3 Summary of the models

The different key equations of the models presented before that will be used next to describe the representative loading configurations are summarized in Table 8.1.

Table 8.1 Equations used for the different loading configurations and levels of analysis.

Loading	Elementary level	Advanced level
Tension	Eq. 8.1 and 8.3	- Eq. 8.4 is satisfied: is Eq. 8.6 satisfied? - o Yes: - o No: Eq. 8.7 - Eq. 8.4 is not satisfied: - o Eq. 8.8
Shear	Eq. 8.11	Eq. 8.12

8.3 Adhesive selection methodology

The geometry shown in Figure 8.2 is considered. A shear force F_τ is applied parallel to the bondline and results in a transverse displacement u_τ while a normal force F_σ acts perpendicular to the bondline in which a crack of length $2a$ is present. The joint width is W , the adhesive length L_a while the top adherend length is L_{s1} and the bottom adherend length L_{s2} with $L_{s1}, L_{s2} \geq L_a$ to generate both 180° and 90° corners. It refers thus to a situation combining a tensile load similar to that applied on tensile butt joints and a shear load corresponding to a TAST shear lap joint. As mentioned in Equation 8.2.1 and 8.2.2, the corners lead to stress singularity (its order depends on E_a , E_s and corner angle) in the adhesive layer that can cause joint fracture. The impact of geometrical parameters such as the adherend's thickness and overlap length in lap-shear joints are thus omitted in order to limit complexity and focus on adhesive performances. For several applications, the adhesive thickness must remain within certain bounds set by a minimum thickness $t_{min} > 0 \mu m$ and a maximum thickness t_{max} . This range must be considered during the selection procedure. It can either result from manufacturing constraints such as in windmills blades for tolerance or geometry reasons or by compactness requirements such as in laptops or appliances. Hence t_{min} and t_{max} enter as two extra parameters involved in the selection process. The Ashby selection approach starts with

the statement of the objectives and constraints (step 1 in Figure 8.5).

8.3.1 Objectives

The objective(s) depend on the application and on the function of the joint in the application. Several objectives are contemplated to cover a variety of options. The two first objectives are classical for applications driven by a combination of economic and environmental concerns: (1) minimization of mass and/or (2) minimization of cost. The third objective (3) is the maximization of the fracture toughness. Recently, this objective motivated the design of tough joints through bondline architecturing, interface patterning and more generally through triggering additional toughening mechanisms [11, 123]. Directly linked to this objective, the fourth one (4) is the maximization of the tensile force that can be applied to the joint. Finally, in some applications focused on small dimensions, thickness minimization can also be an objective (5). The reasons that can push to one or a combination of these are addressed in what follows.

Mass minimization

For some applications, lightweight is required like in aeronautics or automotive to decrease fuel consumptions. However, adhesives must be tough, strong and stiff enough to comply with the structural constraints. As G_{Ic} increases with t , a trade-off must be found between a thicker adhesive and cost of a mass increase. For instance, the Aston Martin DBX contains 23 kg of adhesives [240]. If, for the sake of increasing toughness, the thickness is increased by 50%, the extra mass would be ≈ 12 kg and the extra cost, assuming 100 €/kg, would be >1000 € without taking into account additional fuel consumption costs. Another field where adhesive thickness is adding significant mass to the structure is in windmills blade. Adhesive thicknesses up to 30 mm are reported [7]. Mass minimization can thus, in some cases, be a relevant objective (sometimes with a second objective). The mass of the structure is given by

$$m \equiv \rho_a L_a W t + \rho_s W h_s (L_{s1} + L_{s2}), \quad (8.13)$$

where ρ_a and ρ_s are the adhesive and adherend's density, respectively. We assume that the nature and geometry of the adherends have been selected based on constraints put on the overall structure independent of the adhe-

sive choice. Hence, one is left with minimization of the adhesive mass:

$$m \equiv m_a = \rho_a L_a W t. \quad (8.14)$$

Cost minimization

Cost comprises the cost per unit mass of the adhesive, but also the manufacturing cost:

$$C \equiv C_{adhesive} + C_{manufacturing} = C_a \rho_a L_a W t + (C_m t L_a W + C_{fixed}), \quad (8.15)$$

where C_a is the cost of adhesive per kg and the manufacturing cost involves both fixed costs C_{fixed} (moulds, fixtures ...) and variable costs which are related to the curing of the adhesive, C_m . When the costs are known, the analysis made for the mass can directly be used.

Maximization of fracture toughness of the joint

Knowing that a remnant bottleneck to the widespread use of structural adhesives is the real, or perceived, limited fracture toughness, one can consider as an objective per se. Recently, several research groups worked on adhesive toughening by triggering several dissipation or shielding mechanisms by playing with the joint architecture (see review in Chapter 33 of [11]). For example, Maloney and Fleck [123], Heide-Jørgensen and Budzik [121, 122] inserted holes in the bondline to blunt the crack tip to increase G_{Ic} . Others suggested to pattern the interface to induce mechanical interlocking and/or mode II components, at the microscale [195, 241] or at the macroscale [80, 106, 107]. Another toughening mechanism is crack bridging which can be triggered by playing on surface treatments [83] or by inserting meshes in the adhesive [81, 123]. The insertion of metamaterials within the bondline has been suggested [84, 141]. For this objective (3), the selection statement is "the best adhesive is the toughest one that meets all constraints for the considered range of adhesive thicknesses".

Maximization of applied tensile force

An alternative to the maximization of G_{Ic} is the maximization of the remote tensile stress applied on the joint σ^∞ . Compared to the previous objective, it considers or not the presence of cracks in the adhesive and the influence of the adherends on the selection. In this case, the selected adhesive is the one with the largest effective tensile strength σ^{f*} for a given crack length and range of adhesive thickness that meets the constraints.

Minimization of adhesive thickness

Several applications are constrained by compactness (in appliances, laptop, etc). In this case, the selected adhesive will be the one meeting all the constraints for the lowest possible thickness.

8.3.2 Constraints

Three constraints are envisioned:

- a . The joint must support a prescribed tensile force without failure.
- b . The joint must support a prescribed shear force without failure.
- c . The joint must exhibit a prescribed level of shear stiffness (this is equivalent to stating that the shear displacement under the prescribed shear force cannot be larger than a threshold value, e.g. for dimensional tolerance).

These constraints can be combined or not. In real applications other constraints must of course be imposed in terms of resistance to fluids, thermal degradation, or fatigue which will be addressed in future extensions of the present study. The three constraints are now connected to the mechanical models presented in section 8.2. Finally, one can see the crack length a also as a geometric constraint.

Tensile loading constraint

The first constraint is to resist the tensile force F_σ , resulting in a remote applied tensile stress $\sigma^\infty = F_\sigma / L_a W$. Depending on the level of analysis, either Equation 8.1 to Equation 8.3 or Equation 8.4 to Equation 8.9 are considered. Hence, failure is assumed to occur by a cohesive mechanism only (e.g. fracture inside the adhesive) as it originates from the centred crack lying in the middle of the adhesive layer. The joints are assumed free from any other defect than the pre-existing crack of length $2a$. Stress singularity at the joint edge due to material mismatch, as considered by Equation 8.10, is thus neglected in what follows. Generally speaking, σ^∞ must remain lower or equal to the effective tensile strength of the joint as stated by

$$\sigma^\infty \leq \sigma^{f*}. \quad (8.16)$$

Depending on the model, the analysis changes as explained in subsection 8.2.1. In the elementary approach, σ^{f*} is given by Equation 8.1 or Equation 8.3 depending on the crack length. In the more advanced description,

the interactions between the adherends and adhesive are considered. In this last case, if the fracture toughness and the crack length are such that Equation 8.4 is satisfied, σ^{f*} is equal to σ_a^f if Equation 8.6 is satisfied and if not, it is given by solving Equation 8.7. If Equation 8.4 is not satisfied, Equation 8.8 is used to determine σ^{f*} .

Shear loading constraints

The second constraint is related to the shear load, F_τ , resulting in a mean shear stress $\tau^\infty = F_\tau/LW$ in the bondline and in an overall longitudinal displacement u_τ . This displacement cannot exceed a maximum value u_τ^{max} , set by the minimum shear stiffness admissible for the component. Under a pure shear load, the shear stiffness is governed by the adhesive. The shear stiffness is expressed by

$$\frac{\tau^\infty}{u_\tau^{max}} < \frac{\mu_a}{t}, \quad (8.17)$$

where μ_a is the adhesive shear modulus. This assumption gives an upper limit for the joint shear stiffness which corresponds to cases for which the adherends are stiff and/or thick [242, 243]. In addition, F_τ must not induce any shear failure so that the shear strength τ^f , of the joint which is dependent upon t , cannot be exceeded:

$$\tau^f \leq \tau^{f*}. \quad (8.18)$$

With the elementary model, the change in stress state in the bondline as t increases is not considered and it is assumed that τ^{f*} is constant and equal to the bulk adhesive shear strength approximated by

$$\tau^{f*} = \frac{\sigma_a^f}{\sqrt{3}}. \quad (8.19)$$

In the advanced description, the influence of the thickness and the adherends' elastic properties is considered through Equation 8.12.

8.3.3 Selection scenarios

Depending on the considered set of objective(s) and constraints (as emerging from different types of structural applications), several selection scenarios are possible. One can contemplate a single objective and a single constraint resulting in a method similar to the most basic approach, see also [201]. However, it is also possible to consider several objectives with

one or more constraints. Here, for the sake of illustrating the approach and revealing several interesting non-trivial, trade-off problems with only one objective but with several constraints will be addressed:

- Scenario 1: Mass minimization with constraint on tensile force, shear force and shear stiffness;
- Scenario 2: Cost minimization with constraints on tensile force, shear force and shear stiffness;
- Scenario 3: Fracture toughness maximization with constraints on shear force and shear stiffness;
- Scenario 4: Tensile stress maximization with constraints on shear force and stiffness;
- Scenario 5: Thickness minimizations with constraints on tensile and shear forces and shear stiffness.

This leads to different objective functions and, depending on the scenario, the objective function is combined or not with the constraints. If not, the constraints will correspond to a limit which will be translated into a possible range of thickness that can be selected. The analysis can be directly extended to several objectives [201] requiring then the introduction of weights to balance the impact of each objective on the optimization function.

The logic of the selection is schematically explained in Figure 8.4. The considered constraints will lead, for each adhesive, to a range of thickness inside which the adhesive can be selected as G_{Ic} , τ^{f*} and the shear stiffness all depend upon t . As G_{Ic} increases with t , the constraint will lead to a minimum thickness that can be selected (Figure 8.4a') to avoid failure. If the dependence of G_{Ic} is similar to that of adhesive B in Figure 8.2, it can also lead to a maximum thickness (Figure 8.4a''). At the opposite, as the effective shear strength and stiffness decrease with t , these two constraints result in setting a maximum thickness that can be selected (Figure 8.4b and c). The combination of the three constraints sets an acceptable thickness range inside which one can select the best thickness with respect to the objective function (Figure 8.4e) and this for each adhesive; and of course, if the constraints are too severe, some adhesives will end up never meeting these whatever the thickness. The best adhesive (chosen at its optimum thickness) is the one that maximizes or minimizes the objective.

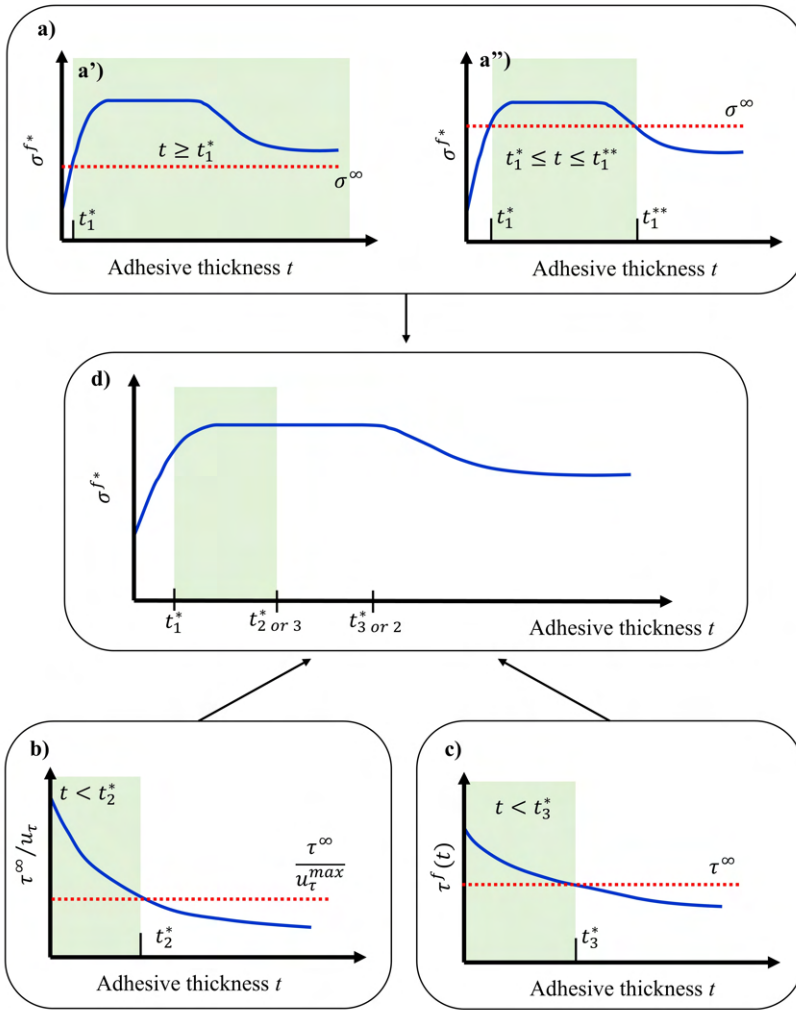


Figure 8.4 Determination of the admissible thickness range for a given adhesive from a) the tensile constraint when the constraint gives a') a lower bound or a'') a lower and an upper bound; b) the shear stiffness constraint and c) the shear strength constraint. d) is the combination of the possible thickness ranges reported on a graph giving the maximum tensile stress that can be applied to the joint depending on the adhesive thickness.

If the constraints do not enter the objective function as for scenarios 3 to 5, the selection will be made based on the constrained thickness range, as long as it is comprised in the range between t_{min} and t_{max} . Maximizing the fracture toughness is thus similar to selecting the adhesive characterized by the largest fracture toughness in the acceptable regime of thicknesses. When the tensile force is maximized, failure is governed either by the strength or by the fracture toughness, the selection is now dependent on the crack length. In this case, for a given crack length, the adhesive characterized by the largest effective tensile strength is considered. Finally, if the thickness must be minimized, the adhesive which can be selected with the lowest possible thickness is chosen. In this case, it is possible to have several adhesives that can be selected and that meet the mechanical requirements.

For scenario 1 and 2, depending on the constraints, a range of thickness will be determined for each adhesive, but the objective function is also dependent upon the constraints. Hence, the objective functions must be derived, requiring to isolate $L_a W$ in the constraint equations to be replaced then into Equation 8.14) when minimizing mass and into Equation 8.15 when minimizing cost. Only the objective function for the scenario 1 will be derived. Indeed, adhesive suppliers do not always make their price publicly available and the manufacturing cost is often unknown. Hence, it is difficult to give practical examples for this scenario. Nevertheless, the methodology is exactly similar to that of mass minimization. Regarding the derivation of the objective functions, one objective function is derived for each constraint. Regarding the tensile force constraint, the objective function, m_{TC} , is given by

$$m_{TC} = \frac{\rho_a t}{\sigma f^*}. \quad (8.20)$$

The shear stiffness constraint leads to Equation 8.21 (objective function denoted as m_{sc1}) while the objective function corresponding to the shear force constraint, denoted as m_{sc2} , is given by Equation 8.22.

$$m_{sc1} = \frac{\rho_a t^2 F_\tau}{\mu_a u_{tau}^{max}}, \quad (8.21)$$

$$m_{sc2} = \frac{\rho_a t F_\tau}{\tau^f(t)}. \quad (8.22)$$

For each adhesive that meets the constraints, m_{TC} , m_{sc1} and m_{sc2} are com-

puted. If at least two adhesives minimize the total mass, the adhesive characterized by the lowest maximum between m_{TC} , m_{SC1} and m_{SC2} is selected.

It is also important to note that when tensile and shear stresses are jointly applied to the joint, a combined failure criterion should ideally be used. Several failure criteria have already been developed such as a maximum stress or strain over a certain distance, an energy criterion, a global yielding criterion or a criterion based on failure envelopes [85, 87, 244]. Here, it has been decided to not apply here a combined failure criterion in order to not further complexify the model and because of the absence of relevant data on most of the considered adhesive systems. Hence, limits for tensile and peel stresses are evaluated separately. Except in the case of pure shear or tensile loading, this assumption leads to an overestimation of the maximum tensile and shear stresses that can be applied to the joint. The implementation of an interactive failure criterion is left for further deployment of the methodology, although one does not expect significant changes in the final rankings.

The assumptions mentioned previously regarding the geometry and the parameters having an influence on the joint shear and tensile strengths and on the shear stiffness are summarized here-below:

- Only two macroscopic loads are considered: tensile and shear loads; macroscopic peel is neglected.
- Assumptions regarding the tensile loading condition:
 - A through-thickness, centred crack of length $2a$ is assumed in the adhesive layer.
 - Failure is assumed to be cohesive only as it originates from the centred crack and not from a stress singularity caused by material mismatch and due to the presence of a corner at the joint edges (Equation 8.10 is not considered).
 - With the simple model, the influence of the adherends Young's modulus and of the crack length with respect to the adhesive thickness is neglected.
- Assumptions regarding the shear loading condition:
 - The presence of the centred crack is neglected due to the lack of data for mode II fracture toughness variations with thickness.

- With the simple model, the joint shear strength is equal to the adhesive shear strength.
- Advanced model: failure originates from the stress singularity due to the presence of the corner at the overlap edges. Failure is thus assumed to be cohesive near the interface.
- Advanced model: the model has been developed for brittle adhesives, but it is also applied to ductile adhesives in the following examples.
- Shear stiffness: the influence of the adherends Young's modulus and thickness is neglected while the thinner and the more flexible the adherends, the lower the shear stiffness [242, 243].
- Assumption regarding the combination of the shear and tensile loads: no interactive failure criterion is used and the limits for both loading conditions are assessed separately.

8.3.4 Database

Literature data regarding the influence of the thickness on adhesive joint properties have been collected from different sources, as reported in Table 8.2. However, when data were found for the fracture toughness, most often no data were given for the evolution of the shear strength with thickness. The opposite is also true. Consequently, literature data do not allow estimating the influence of thickness on fracture toughness and shear strength simultaneously. For the fracture toughness data, tests are usually performed only for a few adhesive thicknesses. Hence, for each adhesive, the data have been extrapolated to consider a broader range of thickness and extend the window of investigation (see Figure 8.5 step 3). Experimental data are fitted using piecewise cubic hermite interpolating polynomial (PCHIP). For thickness larger than the maximum available one, G_{Ic} is considered to be constant, equal to the last experimental data point. For thicknesses smaller than the available range, the two first data are considered and linearly extrapolated. This is done for each adhesive independently of the others. Finally, the tensile strength and the shear modulus are set to the bulk adhesive values. There is thus much room left for extending and refining the database in the future.

In addition to these constraints linked to the mechanical loading, constraints regarding the process conditions, the adherend materials (metal or polymer-

based) and the operating environment can be introduced. Similar to the software EduPack [179], the constraints can be applied through checkboxes in the developed app.

Finally, the fact that thickness influences the adhesive properties does not lead to readable charts similar to those one found in Ashby's book [201]. Hence, in the present case, the selection graph (in the app) shows the effective tensile strength of the joint as a function of the thickness with the allowed thickness range corresponding to the green part of the graph in Figure 8.4d.

The selection procedure can be divided into 4 steps, schematized in Figure 8.5. The first step is the definition of the objective(s) and of the constraint(s) (see subsection 8.3.1 and in subsection 8.3.2), then performance indices and objective functions are derived (see subsection 8.3.3). Then, the adhesive database is screened involving the extrapolation of the experimental data to determine for which range of thickness each adhesive meets the requirements (see subsection 8.3.4). Finally, based on the objective and on the derived indices, the adhesives are ranked to select the best one and its corresponding thickness.

Table 8.2 Adhesives entering the selection app for which literature data regarding the evolution of G_{Ic} can be found.

Adhesive	Maximum toughness (thickness) [J/m^2] (mm)	fracture (adhesive) [J/m^2] (mm)	Fracture toughness – plateau value [J/m^2]	Young's modulus (GPa)	Tensile strength (MPa)	Source
Araldite GY260	810 (0.5)		810	3.1	82	Daghyani et al. [223]
Betamate 1493	6240 (0.4)		6240	1.8	40	Martiny et al. [13]
Betamate 73455	220 (0.5)		210	6	25	Martiny et al. [13]
Betamate XW1044-3	1690 (1)		1500	2.9	35	Carlberger et al. [245]
BP907	800 (0.25)		520	3.2	90*	Chai [100]
ESP110	1480 (0.88)		1480	5.7	70	Martiny et al. [13]
F185	3445 (0.25)		3445	2.9	50	Chai [100]
FM1000	5280 (0.05)		4860	1.2	50*	Chai [100]
Loctite Hysol 9460	760 (1)		760	2.8	30	Ji et al. [14]

Lord 320/322	1020 (4.5)	1020	2.5	28	Ranade et al. [104]
Rubber modified epoxy	3100 (0.42)	2350	2.6	70*	Kinloch and Shaw [99]
Rubber modified epoxy	240 (2)	240	2.6	50*	Lee et al. [246]
Sikaforce 7752 L60	5240 (2)	5240	0.5	12	Campilho et al. [225]
SikaPower 498	6960 (2)	6960	2	30	Marzi et al. [247]

* Data not available: assumed from typical values found in the literature or estimated based on the knowledge of the glass transition temperature.

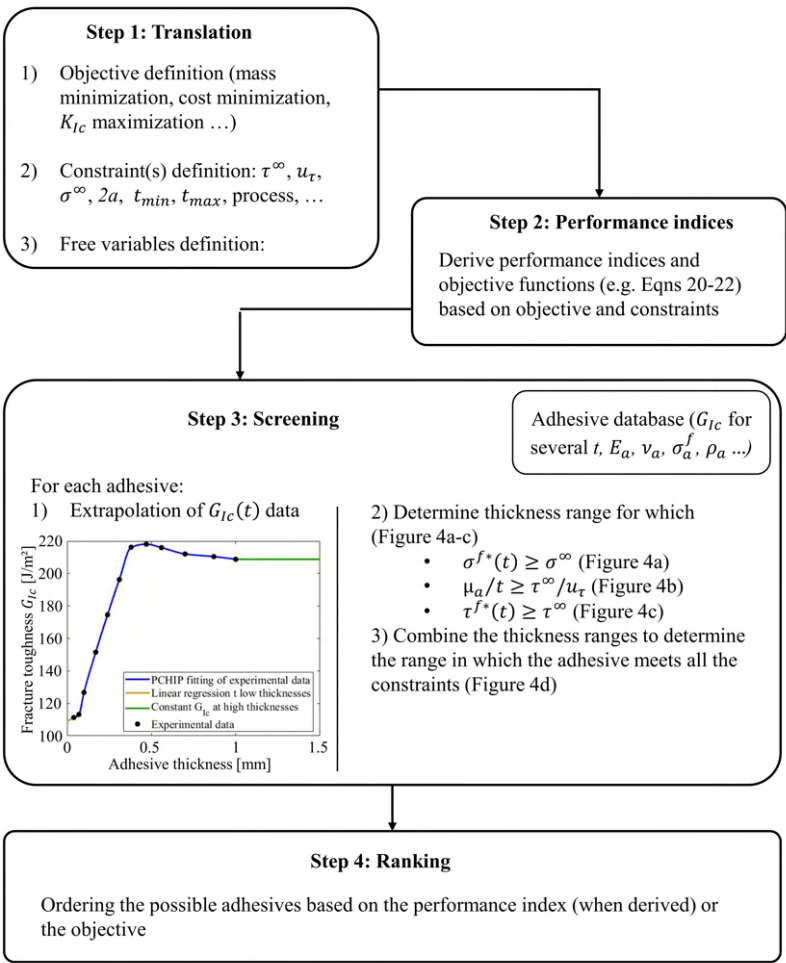


Figure 8.5 Graphical workflow of the selection process: step 1 translation into objective and constraints, step 2 derivation of the performance indices, step 3 screening of the database with the extrapolation of the experimental data and determining the thickness range for which each adhesive meets the constraints and step 4 ranking of the adhesive based on the objective and performance indices.

8.4 Case studies for classical adhesive joints

Several scenarios are investigated and applied to mostly commercial adhesives for which reliable data regarding the evolution of the fracture toughness with t can be found in the literature, see subsection 8.3.4 and Table 8.2. For the sake of illustration, we describe in details the first scenario to minimize mass, and give in Appendix C and D examples for the maximization of K_{Ic} and σ^∞ . The first scenario is to minimize mass. But, first, Figure 8.6 shows the Ashby maps giving $K_{Ic}(t \geq 0.2 \text{ mm})$ versus (a) the adhesive tensile strength and (b) the Young's modulus for all the adhesives of the database. As a general trend, increasing the K_{Ic} is associated to a decrease of the adhesive Young's modulus while no obvious trend is observed regarding the tensile strength.

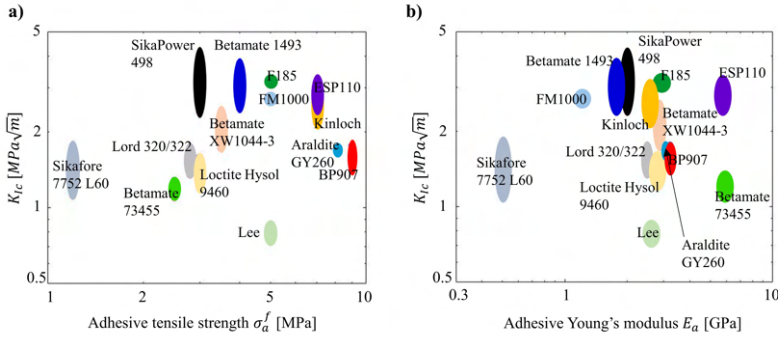


Figure 8.6 Ashby maps for the adhesives of the database a) $K_{Ic}(t \geq 0.2 \text{ mm})$ versus adhesive tensile strength and b) $K_{Ic}(t \geq 0.2 \text{ mm})$ versus adhesives Young's modulus.

8.4.1 Mass minimization: elementary model

When minimizing mass, three constraints are considered: to support the applied shear and tensile forces and to meet a prescribed shear stiffness. This leads to a selection involving 6 parameters: $2a$, σ^∞ , τ^∞ , u_τ^{max} , t_{min} and t_{max} . First, the constraints on the tensile and shear loading are considered separately before analyzing their combined effect using here the elementary model. For all analyses, t_{min} is set to 200 μm simply because G_{Ic} data

are not available for all adhesives ⁴ below this thickness.

Impact of the applied tensile load on the selection

Equation 8.7 addresses the influence of the remote tensile stress σ^∞ , as well as the effect of crack length $2a$ without any applied shear load $\tau^\infty (= 0)$. For each crack length, the remote tensile stress is varied, so that for each combination of σ^∞ and $2a$, the lightest joint meeting the requirements is selected. In some cases, several adhesives meet the constraints and lead to joints of similar mass. In this case, the one with the lowest m_{TC} (Equation 8.20) is selected, corresponding to the largest σ^{f*} . In addition, the failure regime (strength or toughness-dominated) is highlighted for each selected adhesive in Figure 8.7.

Depending on the magnitude of $2a$, σ^{f*} varies with the adhesive strength or with the fracture toughness. Zones corresponding to strength and toughness-governed regimes alternate in Figure 8.7 crack length such that $2a = 2a_c$. For very short cracks, the failure of all adhesives is governed by strength, leading to the selection of the strongest adhesive, i.e. BP907. With increasing crack length, failure of BP907 starts to be dominated by fracture toughness although it remains the strongest adhesive for $t = t_{min}$ as shown in Figure 8.7c. Further increasing $2a$ leads to the selection of ESP110 which appears to be the strongest one at $2a=1$ mm for $t = t_{min}$ (Figure 8.7d). As the crack length is progressively increased, it exceeds $2a_c$ for all adhesives. Hence, for large crack lengths, failure is dominated by fracture toughness only and the selected adhesive is the one characterized by the largest K_{Ic} for $t = t_{min}$ which is the adhesive F185. This first analysis already shows that the selection of the best adhesive significantly varies with the assumed/known crack size.

The magnitude of the applied tensile load plays a minor role in the selection compared to the crack length. For a given crack length, one indeed rarely sees in Figure 8.7 that the choice of the adhesive changes when changing the applied stress except for some small ranges of crack lengths (gray zones). These correspond only to conditions for which the adhesive minimizing the mass meets the requirements for a thickness larger than t_{min} . This is highlighted in Figure 8.7e where the maximum σ^{f*} for $t = t_{min}$

⁴This limit is set to ensure that the influence of t on G_{Ic} corresponds to the two types of influences shown in Figure 8.1 to not consider the fact that at very small thickness, a slight increase of G_{Ic} when decreasing t has been sometime reported due to the change of failure mechanism from propagation at the centre of the bondline to propagation near the interface and failure by shear yielding [98, 248, 249].

is equal to 25 MPa. However, some adhesives can carry larger stresses at larger t . For instance, if σ^∞ is equal to 30 MPa (gray dashed line), the lightest joint meeting the requirements is with Betamate 1493 at $t = 0.4$ mm. This means that the selected adhesive is the one able to carry σ^∞ for the lowest possible thickness in order to generate the lightest joint.

Impact of the applied shear load on the selection

The influence of the applied shear stress τ^∞ and of the shear stiffness τ^∞ / u_t^{max} is shown in Equation 8.8 when imposing minimum adhesive thickness and applied tensile stress to 0.2 mm and 0 MPa, respectively. The shear strength dominates for low imposed stiffness and high applied shear load resulting in the selection of BP907, the strongest adhesive. Stiffness dominates the selection when high shear stiffness is required leading to the selection of Betamate 73455, the stiffest adhesive. For intermediate cases, the adhesive offering the best combination of strength and stiffness is selected which is here ESP110.

Combined applied tensile and shear loads

The simultaneous influence of the four parameters - $2a, \sigma^\infty, \tau^\infty, u_t^{max}$ - is investigated based on the selection charts in Figure 8.9 which depend on $2a$ and τ^∞ . In Figure 8.9a, the shear stiffness and the remote tensile stress are set equal to 1000 MPa/mm and 5 MPa, respectively. The selection depends significantly on $2a$ and τ^∞ . For small crack lengths, the selection is dominated by stiffness for low values of τ^∞ while the shear stress constraint dominates for large τ^∞ . For large crack lengths, whatever the magnitude of τ^∞ , the tensile constraint dominates, pointing towards the selection of the toughest adhesive. In between, there exists a zone where the shear stress and tensile constraints compete. For a given shear stress, increasing the crack length leads to a dominant tensile constraint instead of a dominant shear stress constraint. For a given $2a$, the shear stress constraint tends to dominate if τ^∞ is increased.

The influence of σ^∞ is investigated by comparing Figure 8.9a and b for σ^∞ equal to 5 and 20 MPa. The maximum allowed crack length decreases as σ^∞ increases. In addition, the zones over which the strongest adhesive is selected is reduced, highlighting the fact that, in this case, tensile load dominates over the shear load.

The influence of the shear stiffness is investigated in Figure 8.9a (1000 MPa/mm), 9c (100 MPa/mm) and 9d (10000 MPa/mm). For a low stiff-

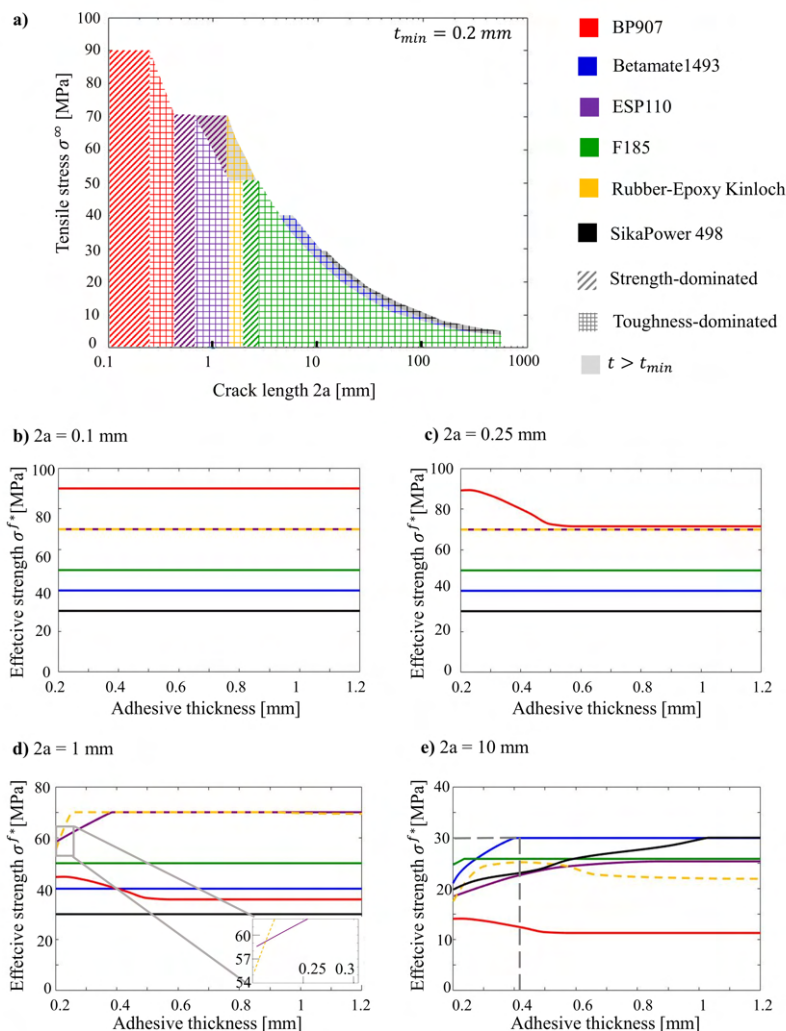


Figure 8.7 a) Constraint chart for light adhesive joints for different applied tensile stress σ^∞ and crack length $2a$ without shear loading. b)-d) Evolution of the apparent tensile strength of the adhesives with the thickness for several crack lengths b) 0.1 mm; c) 0.25 mm; d) 1 mm and e) 10 mm.

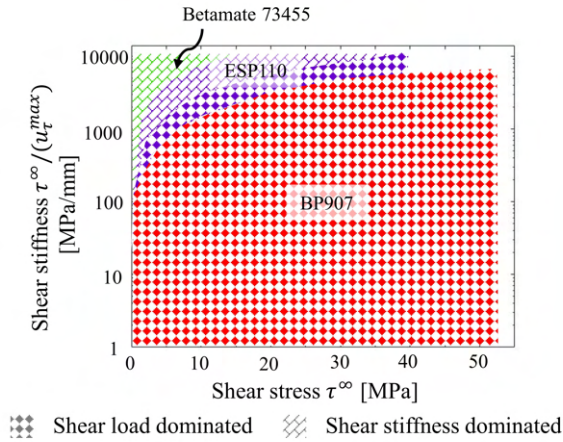


Figure 8.8 Influence of the shear stiffness and the applied shear stress on the adhesive selection for $t_{min} = 0.2$ mm.

ness (9c), the selection is dominated by the shear and tensile constraints only. At large stiffness (9d), stiffness dominates the selection. As a stiffer adhesive tends to be more brittle, long cracks and high tensile stresses are not allowed for these joints indicating a gap in the available performances of current adhesives (more details in subsection 8.4.4). At intermediate stiffness (9a), the three constraints compete.

Influence of t_{min} and t_{max}

As the fracture toughness varies with the adhesive thickness, the choice of t_{min} and t_{max} affects the selection of the best adhesive. Considering a tensile load only (Figure 8.10a), if t_{min} is set to 5 mm (a quite large value as used sometimes for geometrical tolerance reasons in ship building or windmills – although much thicker joints exist up to 30 mm or higher [7]) for instance, at large crack length, SikaPower 498 is selected while when set to 200 μm , F185 is preferred. This arises because the G_{Ic} , and consequently σ^{f*} , of the SikaPower 498 is larger than that of F185 if $t_{min} > 0.6$ mm (Figure 8.7e). If now t_{max} is varied and is lower than t^* , for a given crack length, the maximum tensile stress that can be applied, considering all adhesives of the database, is reduced, limiting the domain in which an adhesive can meet the constraint. Regarding, the shear stress and stiffness constraints only, decreasing t_{min} increases the maximum shear stress that

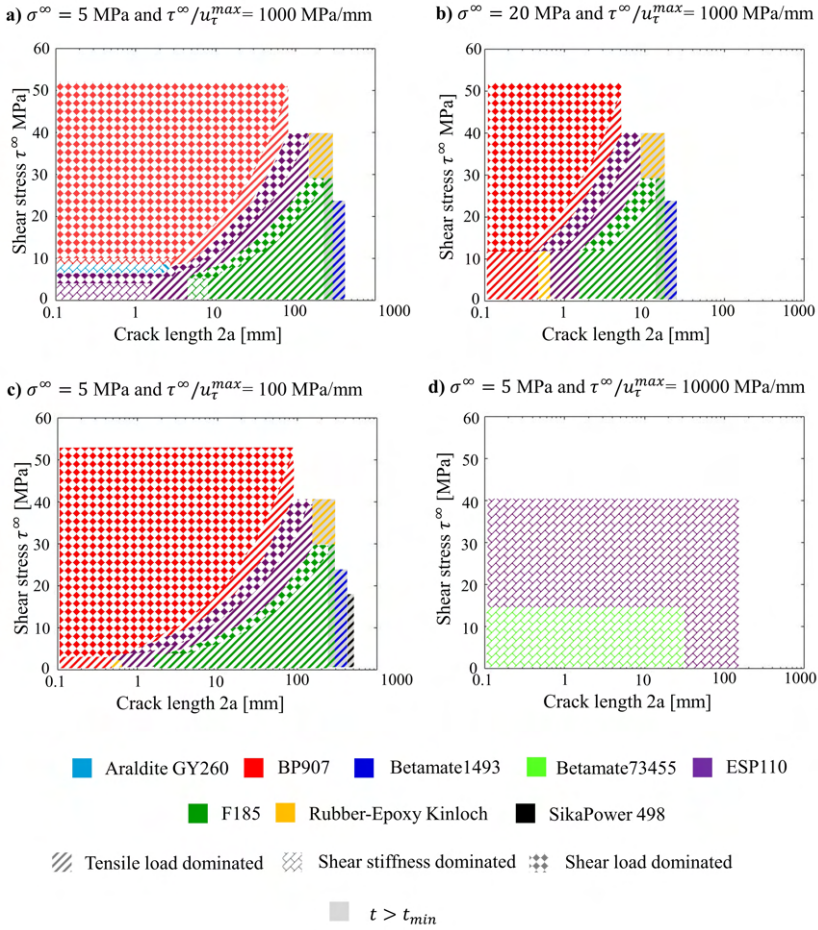


Figure 8.9 Investigation of the combined effect of tensile and shear loading on the choice of best light adhesive for different remote tensile stress σ^∞ and imposed shear stiffness: a) $\sigma^\infty = 5 \text{ MPa}$ and $\tau^\infty/u_\tau^{max} = 1000 \text{ MPa/mm}$; b) $\sigma^\infty = 20 \text{ MPa}$ and $\tau^\infty/u_\tau^{max} = 1000 \text{ MPa/mm}$; c) $\sigma^\infty = 5 \text{ MPa}$ and $\tau^\infty/u_\tau^{max} = 100 \text{ MPa/mm}$ and d) $\sigma^\infty = 5 \text{ MPa}$ and $\tau^\infty/u_\tau^{max} = 10000 \text{ MPa/mm}$.

can be reached while increasing it has the opposite effect. Hence, if t_{min} is increased, the softest adhesives will tend to no longer be selected, as shown in Figure 8.10b-d. When, the three constraints are combined, increasing t_{min} leads to a selection that is more and more dominated by the

shear stiffness constraint, leading to the selection of more brittle adhesives. These effects arise from the dependence of G_{Ic} on t and due to the fact that tough adhesives tend to be flexible while stiff adhesives tend to be more brittle as shown in Figure 8.6.

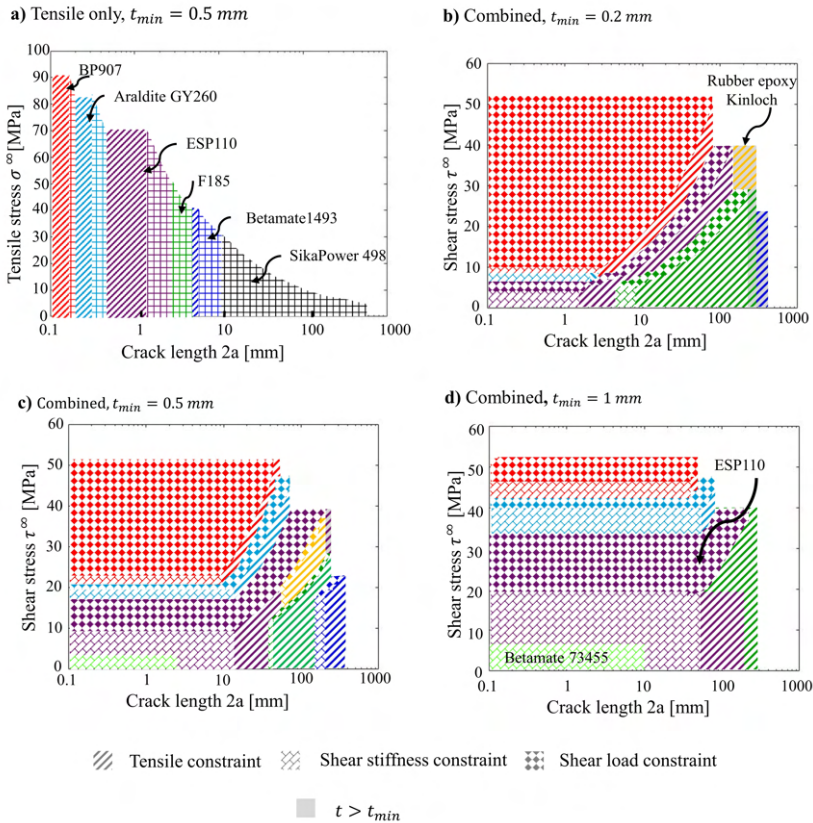


Figure 8.10 Influence of the imposed minimum thickness on the selection of best light adhesive for a) constraint on the tensile load only with $t_{min}=0.5 \text{ mm}$, and b) to d) for a combination of tensile load (5 MPa), shear stiffness (1000 MPa/mm) and varying shear loads and crack length with t_{min} equal to b) 0.2 mm; c) 0.5 mm and d) 1 mm.

8.4.2 Mass minimization: effect of adherend-adhesive stiffness mismatch

Tensile load

The results of Figure 8.11 are obtained taking into account the adherend-adhesive interaction based on the more advanced analysis of Van Loock et al. [230] and Askarinejad et al. [151]. A first key difference that emerges when compared to Figure 8.7 produced with the elementary level, is that adhesives can now be selected (and thus meet the imposed constraint) for larger crack lengths. Indeed, if $E_s=200$ GPa, BP907 is selected up to a crack length of 70 mm instead of 0.5 mm. When the adherends Young's modulus is lower, the crack length for which one adhesive can be selected decreases as the shielding effect is reduced. Indeed, for E_s equal to 200, 100 and 10 GPa, the maximum $2a$ for which BP907 can be selected is equal to 70, 20 and 2 mm, respectively. In addition, compared to Equation 8.7, the selected adhesives are no longer the same. Indeed, ESP110 is no longer dominant and instead of selecting F185, FM1000 is now preferred. Indeed, F185 is two times stiffer than FM1000 but is characterized by a process zone of similar length. Van Loock et al. showed that if two adhesives of same σ_a^f and l_s are considered, the selected adhesive for given l_s/h and a/h will be the less stiff. For this reason, FM1000 is selected instead of F185 because, as shown in Figure 8.6, these are characterized by similar σ_a^f and K_{Ic} , but FM100 is the most compliant.

Shear constraints only

Figure 8.12 shows the influence of the imposed shear load and stiffness constraints on the selection if E_s is equal to 200 GPa. The selection is not drastically modified compared to the case where the interactions with the adherends are neglected. The strongest adhesive is still selected when a high shear load is applied while the stiffest one is selected when a high shear stiffness is imposed. When varying E_s to 10 GPa or 100 GPa, no difference was observed in the selection, except when increasing E_s , then the maximum shear strength that can be met slightly increases as λ in Eq. (B1) increases. In addition, the thickness effect is now introduced, leading to a lower maximum shear stress that can be carried when comparing Figure 8.12 to Figure 8.8.

Combined effects

Figure 8.13 shows selection maps for varying crack length and applied shear load, for different values of E_s and imposed shear stiffness. By in-

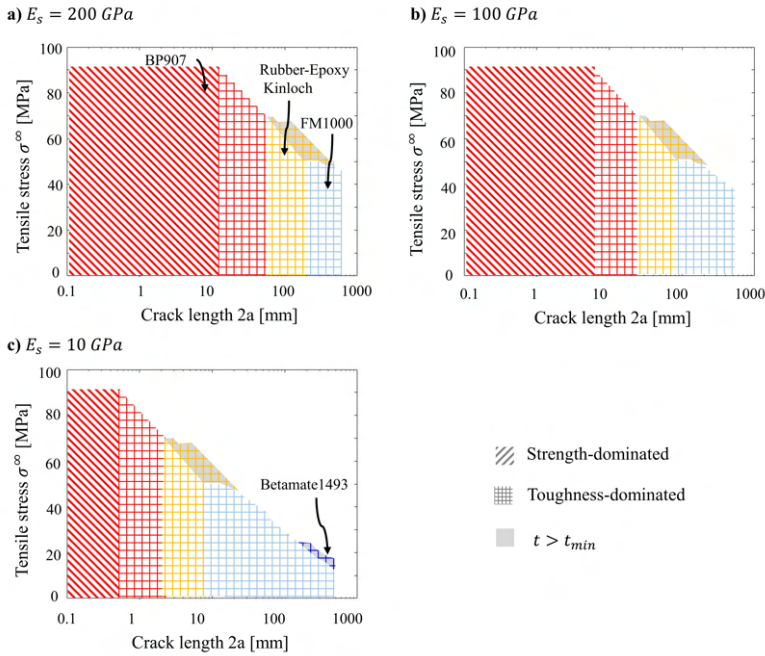


Figure 8.11 Adhesive selection maps when considering the tensile constraint only and the effects of the adherends with Young's modulus of a) 200 GPa, b) 100 GPa and c) 10 GPa for $t_{min}=0.2$ mm.

creasing the adherends stiffness, the maximum shear strength and the maximum crack length for which an adhesive can be selected increase due to more pronounced shielding. In addition, increasing the adherend Young's modulus results in a selection more dominated by the applied shear load than by the tensile load.

Comparison with the elementary model

Regarding the tensile constraint, a simple “adherend independent” model leads to the selection of the adhesive with the largest K_{Ic} while the advanced analysis indicates a competition between the adhesive toughness and the shielding effect of the adherends that can lead to the selection of a slightly more brittle adhesive but softer. In any case, the simple approach captures the main trends which is to select tougher adhesives as $2a$ increases and stiffer and stronger adhesive when the imposed shear load and stiffness increase.

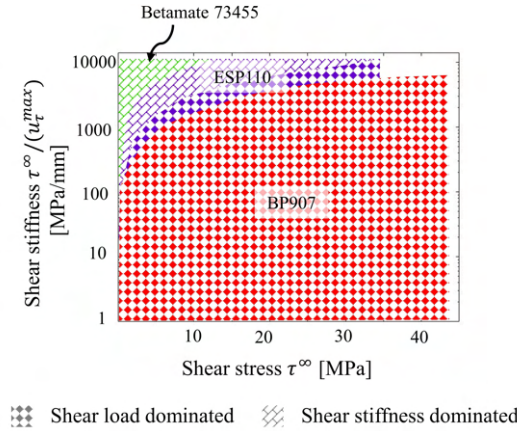


Figure 8.12 Adhesive selection maps when considering the shear stiffness and strength constraints only and the interactions with the adherends if E_s equal 200 GPa and if t_{min} equals 0.2 mm.

8.4.3 Other objectives

The same procedure has been followed for the maximization of K_{Ic} or the maximization of σ^∞ . The detailed results are shown in Appendix C and D. In both cases, the selection is strongly dependent on the constraints imposed for the shear load and stiffness. Increasing the magnitude of these constraints results in the selection of stiffer and stronger adhesives that are characterized by a lower fracture toughness. The main difference between these two cases, is the dependence of the selection on the crack length when maximizing σ^∞ while maximizing K_{Ic} results in a selection independent on the crack length. When maximizing σ^∞ , the maximum tensile force that can be carried by the joints decreases as the crack length increases. As for mass minimization, t_{min} and t_{max} also influence the selection due to the dependence of G_{Ic} on the adhesive thickness.

8.4.4 Gaps in adhesive property charts

Depending on the application and selection problem, gaps appear in the selection chart of adhesives as schematically drawn in Figure 14. This schematic chart represents in 3D the different constraints and the domain that can be met with existing adhesives. As shown previously, these gaps depend on thickness. For low t (t_1 in Figure 8.14), adhesive joints are generally characterized by high shear strength and stiffness, but lower fracture

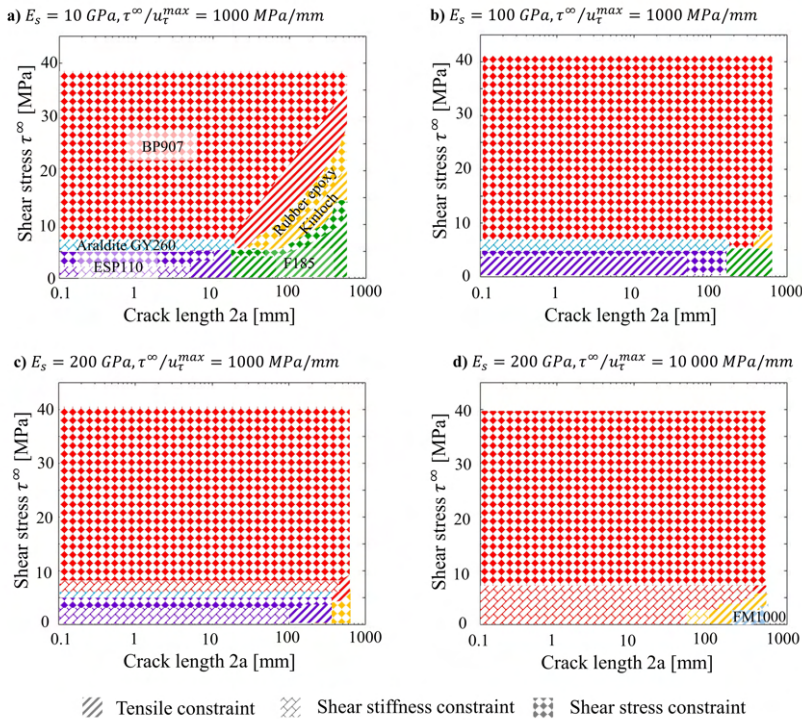


Figure 8.13 Investigation of the combined effect of tensile and shear loading on the choice of best light adhesive for different adherends Young's modulus if the remote tensile stress is set to $\sigma^\infty = 5 \text{ MPa}$ and the shear stiffness to a)-c) 1000 MPa/mm or d) 10 000 MPa/mm: a) $E_s = 10 \text{ GPa}$, (b) $E_s = 100 \text{ GPa}$ and c)-d) $E_s = 200 \text{ GPa}$.

toughness. This leads to the definition of two gaps corresponding (1) to stiff and tough adhesives region and (2) to tough adhesives with high shear strength region. For high t (t_2 in Figure 8.14), the fracture toughness increases, but the shear strength and shear stiffness tend to decrease. Hence, here, three gaps are revealed: (1) high shear strength and stiffness region; (2) high shear strength and high toughness region; (3) stiff and tough region.

These gaps in the selection of adhesive materials have two origins: the adhesive thickness and the adhesive material. The influence of thickness has already been discussed. In addition to this, adhesives are character-

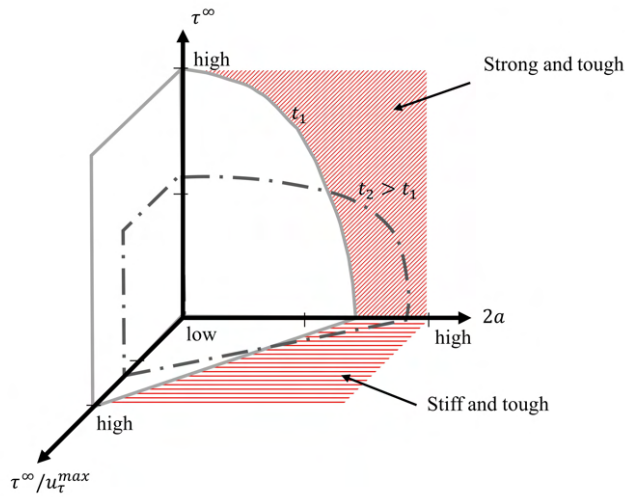


Figure 8.14 Schematic of the gaps in a constraint-based charts and influence of the thickness.

ized by antagonist performances “stiff, strong and brittle” and “soft, flexible and tough” as also shown in Figure 8.6. Applications requiring high shear strength and stiffness will preferentially end up with thin adhesive bondlines, limiting the fracture toughness of the joint while thick-adhesive joints are better suited for applications with high fracture toughness, but allowing for limited shear stiffness and strength.

8.5 Case studies for architected joints

Bondline architecturing is one of the strategies that has been recently investigated by different research groups to increase the joint fracture toughness [11, 84, 121, 123, 135, 141]. The question here is to assess if the potential of such solutions to indeed push the envelope of the selection leads to more optimal solutions compared to traditional joints taking into account the fact that other constraints play a role. The joint architecture can involve the addition of a mesh, stop holes or more complex metamaterials. Covering all possibilities goes beyond the scope of this work. Here, to illustrate the interest of the selection approach, one considers architected joints made of stop holes having a length D . D is determined based on the process zone

length l_p which is equal to $l_p = 1/\lambda_w^5$ [121]. λ_w is the elastic foundation wave number given by

$$\lambda_w = \sqrt[4]{\frac{m E_a W}{4 t E_s I}} \quad \text{with} \quad m = \frac{1 - \nu_a}{(1 - 2\nu_a)(1 + \nu_a)}, \quad (8.23)$$

where I is the second inertia moment equal to $Wh_s/12$. It is thus dependent on the adhesive and adherend Young's moduli E_a , E_s and on the geometric parameters t , W , h_s . In the present case, 3D plane strain was assumed to derive the expression of m , a parameter depending on the crack tip stress/strain state. Heide-Jørgensen and Budzik[121] showed that the joint fracture toughness is maximized when D is about $2l_p$.

In order to apply the materials selection approach to this class of architected joints, the maximum principal stress criterion developed by Martiny et al. [92] for the adhesive Betamate 73455 is implemented in Abaqus. Both Double Cantilever Beam (DCB) and lap shear joints are modeled to investigate the influence of the stop holes and of the adhesive thickness on the joint properties. More details are given in Appendix E. The height of the cavities is set equal to the adhesive thickness while their spacing is set to $2l_p$, $4l_p$ and $6l_p$, leading to joints made of 50%, 33% and 25% of cavities. The spacing is known to not influence the fracture toughness if it is large enough [123]. However, they did not investigate the impact of the holes on the adhesive shear stiffness and strength. One anticipates that the shear stiffness decreases by increasing the number of cavities, and by decreasing their spacing, affecting then directly the selection of best solution depending on the problem. G_{Ic} and shear stiffness are computed for the different stop holes distributions using FE simulations. The shear and tensile strength are roughly estimated as given by

$$\sigma_{archi}^f = \frac{S - S_{holes}}{S} \sigma_a^f, \quad (8.24)$$

$$\tau_{archi}^f = \frac{S - S_{holes}}{S} \tau_a^f, \quad (8.25)$$

where σ_a^f and τ_a^f are the bulk tensile and shear strength of the plain ad-

⁵The process zone denoted by l_p is different from the l_s process zone. The l_p process zone refers to the length corresponding to the zone that extends from the crack tip to the zone where no deformations take place in the adhesive and the DCB arms. For more information, the reader is referred to [250].

hesive. S is the real surface area of the joint, equal to $L_a W$, while S_{holes} correspond to the surface area occupied by the stop holes when orthogonally projected. $S - S_{holes}$ is thus the effective area covered by the adhesive over which the forces act. More advanced models for the strength of architected joints is left for future investigation, but one can already foresee that in stiffness and strength dominated problems, an architected joint will not outperform the traditional joints.

8.5.1 Objective on mass minimization

Four joints are considered, all made of the adhesive Betamate 73455 and 0%, 25%, 33% and 50% stop holes. The goal is to determine under which conditions an architected joint must be preferred to its classical counterpart.

Tensile dominated loading

The influence of σ^∞ and $2a$ on the adhesive selection is shown in Figure 8.15. The range of forces and crack lengths for which an adhesive with stop holes can be selected is modified compared to the plain adhesive joints considered before. The architected joint is associated to a larger effective tensile strength for long cracks. However, this depends on the amount of stop holes inserted in the bondline. Indeed, as shown in Figure 8.15b and c, the more the stop holes in the bondline, the lower the σ^{f*} as the effective cross-section area over which the tensile force acts decreases. Conversely, a reduction of the number of stop holes decreases the number of crack arrestors. In the current analysis, the crack tip is assumed to correspond to a stop hole. However, if the crack tip does not correspond to a stop hole, the toughness is first equal to the plain adhesive joint G_{Ic} [123] and when the crack reaches the next stop hole, the toughness is the one of the architected joint. The considered crack length is equal to $\max(2a, nD)$ with n being an integer and $n \geq 1$. The joint must be made of less than 33 % of cavities to improve the damage tolerance (Figure 8.15c). For small cracks, when the imposed tensile load increases, the selection points towards an adhesive with the lowest number of cavities as it corresponds to the joint with the larger effective cross-section area. Previously, the adhesive with the highest σ^{f*} was selected while in the present case it is no longer true because the weight saving coming from the introduction of holes dominates the selection for an objective based on mass minimization.

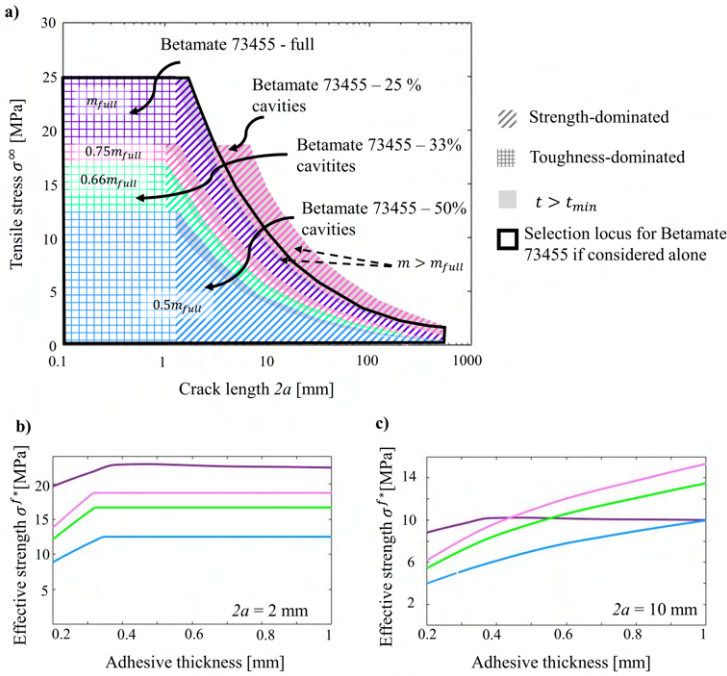


Figure 8.15 Influence of the remote tensile stress σ^∞ and the crack length $2a$ on the selection of the lightest adhesive without shear loading. The joints are made of Betamate 73455, but contain 0%, 25%, 33% and 50% of stop holes. t_{min} is set to 0.2 mm.

Shear loading

The joint with the largest proportion of cavities is always selected as long as its shear stiffness and strength are not exceeded as shown in Figure 8.16. Further increasing the imposed shear stress and stiffness leads to the selection of a joint with a lower amount of stop holes, until the plain adhesive joint remains the only one that can be selected. Indeed, decreasing the number of cavities in the bondline improves the shear strength and stiffness. When considering only classical adhesive joints, regions where the shear strength or stiffness dominate are both predicted, while here it is no longer the case because the lightweight of the architected joint dominates. When using an architected joint with stop holes, the required shear stiffness and strength are limited to lower values compared to conventional joints. Hence, inserting stop holes limits the joint shear properties.

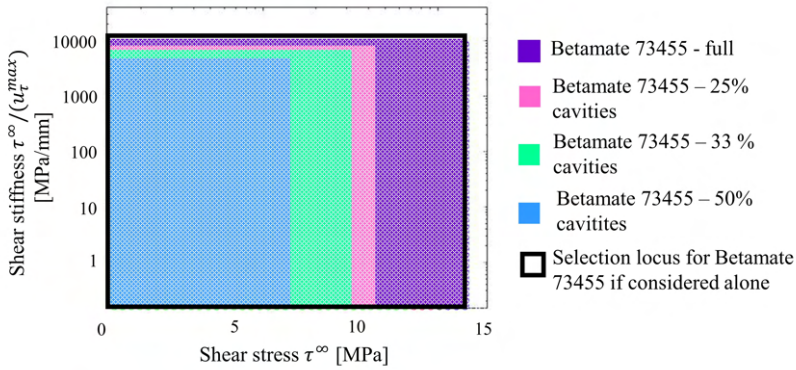


Figure 8.16 Influence of the shear stiffness and the applied shear stress on the selection of light adhesive for joints made of Betamate 73455 made of 0%, 25%, 33% and 50% of stop holes. t_{min} is set to 0.2 mm.

Combined effects

The combined effects of $2a$, τ^∞ and $\tau^\infty / u_\tau^{max}$ are investigated. No novel conclusion emerges compared to the former analysis. The damage tolerance is improved only if the required level of shear strength and stiffness remains moderate. Indeed, if the constraints on shear stress and stiffness are too large, a plain adhesive joint is preferred. Another advantage of architected joints is the associated weight saving owing to the presence of the cavities which act positively on density and toughness. This weight saving dominates the selection and not the tensile and shear constraints as for classical adhesive joints.

8.5.2 Other objectives

As for the classical adhesive joints, two other objectives have been applied to the selection of architected joints: K_{Ic} maximization (Appendix C) and maximization of applied load σ^∞ (Appendix D). Compared to the mass minimization objective, when an architected joint can be selected among other architected joints with stop holes, in these two cases, it is the one containing the less stop holes. This arises from the fact that by increasing the amount of stop holes in the bondline, the effective cross-section area over which the tensile and shear forces act decreases resulting in lower effective tensile strength, shear strength and shear stiffness. These two examples show that there is thus a trade-off to find when designing architected

joints between the gain in fracture toughness and the loss in shear strength and stiffness and that the optimal design depends on the objective.

8.5.3 Filling gaps in adhesive property space

Introducing stop holes leads to an increase of the fracture toughness, but reduces the strength and stiffness. Hence, gaps left when considering classical adhesive joints are partly filled owing to architecturing (Figure 8.17). The spacing between the stop holes offers some degree of freedom to fill in this domain. In addition to toughening, bondline architecturing with stop holes results in lighter joints. But, for applications requiring high shear strength and stiffness, joints with stop holes may not be suitable. Bondline architecturing with stop holes is thus interesting for applications requiring low weight and high fracture toughness, but moderate strength and stiffness.

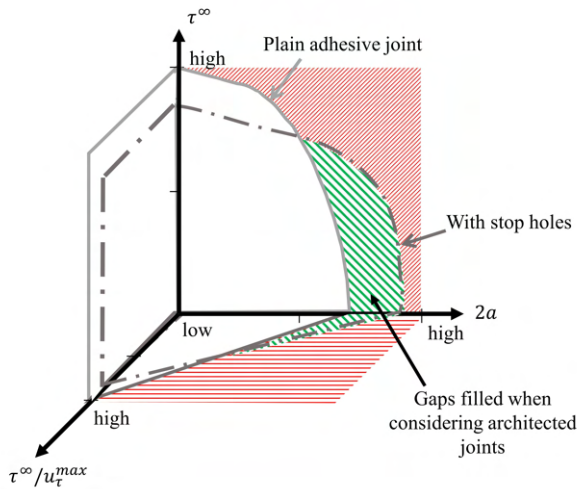


Figure 8.17 Influence of bondline architecturing with stop holes on the joint properties.

8.6 Limitation of the adhesive selection analysis

The selection analysis of the best adhesive system setup in this study although addressing many aspects, in particular with the incorporation of

adhesive thickness effects, is still missing several elements while also relying on a few restrictive assumptions. Once again, the goal in a materials selection analysis is not to predict the exact failure load or exact joint performance, but to use models that are sufficiently correct to lead to the right ranking among different adhesives. In a few cases, an advanced model is required to avoid missing some effects, such as the adherend stiffness mismatch that may modify the selection; while in several other instances, the selection with an elementary model is appropriate. We review here the main limitations of the current study, aimed at paving the road for future works.

First of all, the database that has been compiled for the purpose of this study is incomplete both in terms of adhesive systems and in terms of specific data. There is a need to find and/or generate more extensive data on thickness effects on strength and toughness. Also, novel systems may exist or will emerge that will modify the conclusion made regarding best adhesive choice. These conclusions should thus be seen as illustrative and not definitive.

Then, although the database already accounts for other properties when available such as the temperature resistance, the resistance to humidity and the chemical and bonding processes, these have not been addressed in the present selection analysis. Many of these constraints, depending on the type of application, can be easily introduced simply through a “go – no go / yes - no” choice (already implemented in the MATLAB application).

Another key remnant issue in adhesive bonding is the importance of the surface treatment. Depending on the adherends and the adhesive, there exists an optimum roughness [110, 111]. Failure can be either cohesive or adhesive [14], affecting very much the strength of the joint. For example, in peel tests, the roughness can have an influence up to large thicknesses. The rougher the surface, the more the roughness influences the results [251]. The proposed analysis does not consider the adherends surface treatment and roughness. As a matter of fact, it is assumed that the failure of the joint is always cohesive as often considered in the field of adhesive bonding.

Adhesive joints are subjected in some applications to dynamic loading, not addressed by the current selection procedure dealing only with static loading. When subjected to fatigue loading, fatigue life is also dependent upon the adherend properties, dimensions and joint geometry [252, 253]. This would require collecting a large amount of data which are not always

available for the adhesives for which toughness data can be found in the literature.

On the mechanical description side, the first element of generalization that would improve the relevance of the analysis, but at the price of more complexity, is to account for mixed-mode cracking. Here, only mode I cracking from an internal pre-crack of length $2a$ is considered. The problem of Figure 8.2 should ideally account for the impact of the shear load on generating a K_{II} component that combines with K_I to set the crack driving force. Taking mixed-mode into account is possible if toughness values are known for different mode-mixities, which is rarely the case (at least for publicly available data), going back to a previous comment on the availability of rich mechanical databases for adhesives. The second element is regarding the architected joints, to develop models to predict the shear stiffness and thickness dependent strength for failure initiating at corners or collect experimental data, in order to provide a more realistic view on the potential of such solutions.

Many results show that the selection very much depends on crack length. A tacit assumption in our analysis, is that the crack size is material independent. This might not be true. The representative crack length could potentially be adhesive dependent due to the different type of chemistry and curing schedule. The problem of identification of the proper crack size is a remnant problem in all structural integrity assessment problems, especially with structural adhesive joints and very much connected to the capacity to estimate/measure the size of internal defects with non-destructive methods.

Regarding the considered crack configuration (centered crack through the thickness), one extension of the study would be to address other configurations or several crack positions in the joints: near the interface, in the bondline, at the centre of the joint, at the edges. Indeed, several types of defects and damages can be found in adhesive joints. Some defects develop during the manufacturing phase such as voids due to entrapped air or kissing bonds which are located in the bondline and not especially close to the edges. Some damage also develops the in-service life due to mechanical loading or environment leading to crack developing along the edges. Depending on the crack position, the strength of the joint will vary as the expression of the geometric factor Y in Equation 8.3 is geometry dependent. This could possibly influence the ranking process by shifting the

transition between the strength- and toughness-dominated regimes.

In order to set the bases of the selection approach, several arbitrary choices have been made regarding the models used to predict the tensile strength, the shear strength and the shear stiffness that affect the results. However, other models have also been developed in the literature and could be used. Alternatively, the selection tool could be coupled with finite element analyses. The choice of the different models has been based on the information available for the adhesives of the database but also on their predictive potential. In addition, some models are relevant for brittle adhesives but not for ductile ones and the opposite is also true. Introducing other models is thus left for further development of the selection procedure.

8.7 Conclusion

A method for selecting the best adhesive system has been worked out following Ashby's approach, accounting for the impact of the bond line thickness on the performance of the joint. The objectives that have been considered include: mass minimization, maximization of fracture toughness or maximization of the tensile load applied to the assembly. The constraints include: no failure under prescribed tensile and shear loading as well as an imposed minimum shear stiffness. Different loading configurations have been analyzed taking into account the presence of an internal crack and the presence of corners at the interface between adhesive and adherend. Both elementary and advanced models have been used to predict the effective strength of the joint including or not adherend effects as well as a fracture process zone effect. A database of adhesive properties with thickness dependence has been compiled to effectively apply the selection analysis. The key findings of the work are the following:

- The selection is very much dependent on crack length, leading to non-trivial choices with some adhesive performing better for short cracks and other for long cracks. This raises the remnant difficulty in structural integrity assessment based on fracture mechanics: How to precisely determine a relevant crack length? Progress in non-destructive analysis of adhesive joints [222] have been made to determine the defect sizes, but not all defects, such as weak bonds, can be detected [254].
- Working at the top fracture toughness requires the optimum adhe-

sive thickness. But the optimum thickness is sometimes large, especially in very tough adhesives. A large thickness leads to a lower resistance to fracture from corners under tensile and shear loads. Also, thicker joints are less stiff in shear which is sometimes a constraint in some applications. Subtle trade-offs have been found showing that the best adhesive choice varies from one problem to another.

- The use of advanced models taking the adherend stiffness into account is important [151, 230]. Indeed, a given adhesive tends to be selected for a larger crack length and a larger shear strength if E_s increases. In addition, taking the adherend stiffness into account can change the selection when having adhesives with almost equal K_{Ic} and σ_a^f . With the elementary model, the adhesive with the largest K_{Ic} is selected while, with a more advanced model, the one with the lowest E_a is selected.
- The selection method reveals gaps in the adhesive property charts: (i) a region with both stiff and tough adhesives and, (ii) a region with both strong and tough adhesives indicating the need for further possible improvements.
- The selection approach has been applied to architected joints with stop holes. Stop holes induce larger G_{Ic} , extending the range of adhesive performances. Such joints offer thus improved damage tolerance at lightweight, but at lower shear strength and stiffness. Several directions have been proposed to enrich the present selection analysis to make it more versatile and more general.

8.8 Appendices

8.8.1 Appendix A: Derivation of $A_p(\nu)$ and λ in butt tensile joints

Reedy and Guess [150] derived a model based on an interface corner stress intensity factor to predict σ^f , assuming that failure only takes place near the interface and that the joint is free from any defect. In tensile butt joints, the interface corner stress singularity order is $\lambda - 1$. The joint tensile strength is given by Equation 8.10 which depends on $A_p(\nu)$ and λ . Values of $A_p(\nu)$ have been tabulated by Reedy and Guess and can be found in [150]. Regarding the order of the singularity, it depends upon the Dundur's parameters α and β . λ can be determined by solving the Eq. A9.1

derived by Akisanya and Fleck [231]:

$$\begin{aligned}
& 16(\mu - 1)^2 \left[\lambda^2 - \sin^2 \left(\frac{\lambda\pi}{2} \right) \right]^2 \\
& - 4 [\mu(\kappa_s + 1) - (\kappa_a + 1)]^2 \left[\lambda^2 - \sin^4 \left(\frac{\lambda\pi}{2} \right) \right] \\
& + 16(\mu - 1) \sin^2 \left(\frac{\lambda\pi}{2} \right) [\mu(\kappa_s + 1) - (\kappa_a + 1)] \left[\lambda^2 - \sin^2 \left(\frac{\lambda\pi}{2} \right) \right] \\
& + [\mu(\kappa_s + 1) + (\kappa_a + 1)]^2 \sin^2(\lambda\pi) = 0 \quad (8A.1)
\end{aligned}$$

where $\mu = \mu_a / \mu_s$ with $\mu_i = E_i / 2(1 + \nu_i)$ the shear modulus of material i and the shear coefficient $\kappa_i = 3 - 4\nu_i$.

8.8.2 Appendix B: Derivation of λ for TAST configuration

Askarinejad et al. [151] developed a fracture mechanics methodology for the failure of elastic-brittle lap-shear joints based on the TAST configuration. This model depends on two parameters: b and λ . The value of b depends on the joint geometry and on the adhesive and adherends shear modulus and Poisson ratio while α and β are the Dundurs parameters. The authors determined b by calibrating the model on experimental data. Hence, b can be obtained based on the shear strength of the joint for at least two different thicknesses. However, these data are not available for the adhesives entering our database (subsection 8.3.4). The singularity order is also dependent upon adherends Young's modulus and Poisson ratio and can be determined by finding the root of

$$\begin{aligned}
F(\lambda) = & \left[\left(4 \left(\kappa_a^2 - 4\lambda^2 + 1 \right) + 8\kappa_a \cos(\pi\lambda) \right) \sin^2(\pi\lambda) \right. \\
& - 2\mu^2 \left(\left(\cos(\pi\lambda) + 2\lambda^2 \right) - \left(4\kappa_s \cos^2(\pi\lambda) + (\kappa_s - 1)^2 \right) \right) \\
& - 4\mu \left((\kappa_s - 1) \left(4\lambda^2 \sin^2(\pi\lambda) - (\kappa_s - 1) \cos^2(\pi\lambda) \right) \right. \\
& \left. \left. + (\kappa_a \kappa_s + 1) (1 - \sin(2\pi\lambda) \sin(\pi\lambda)) - \kappa_a - \kappa_s \right) \right] / \left[4 \sin^2(\pi\lambda) \right] \quad (8B.1)
\end{aligned}$$

where $\mu = \mu_a / \mu_s$ with $\mu_i = E_i / 2(1 + \nu_i)$ the shear modulus of material i and the shear coefficient $\kappa_i = 3 - 4\nu_i$. For given geometric parameters and adherend and adhesives properties, the shear strength decreases thus as $t^{\lambda-1}$ and this decrease depends on the properties of both adherends

and adhesive. The use of this relationship requires computing λ and b for several geometries and adherend-adhesive properties which is time consuming. For typical metal-adhesive systems, λ will tend to 0.63-0.65. The dependence of b is neglected in the current work.

8.8.3 Appendix C: Maximization of K_{Ic}

Classical adhesive joints

This scenario involves the maximization of the joint fracture toughness, with two constraints being put on the applied shear stress and the required shear stiffness. The two constraints are put as limits. Hence, the selected adhesive is the one that meets the shear strength and stiffness requirements and that maximizes K_{Ic} for t comprised between t_{min} and t_{max} . t_{min} is set to 200 μm while no limit is put on t_{max} . Figure 8C.1 shows the influence of τ^∞ and $\tau^\infty / u_\tau^{max}$ on the selection. First, it is important to note that the selection is independent of the crack length. Figure 8C.1 shows that increasing the applied shear stress leads to the selection of a stronger adhesive, but which is characterized by a lower maximum critical stress intensity factor (lighter color). If the required shear stiffness is now increased, stiffer adhesives tend to be selected. In addition, when the stiffness is increased, the maximum K_{Ic} that can be reached decreases. These two effects arise from the fact that stiff and strong adhesives tend to be brittle while soft and flexible ones are tougher (see Figure 8.6). For a given stiffness and for a given adhesive, the maximum K_{Ic} that can be reached is independent upon τ^∞ as in the elementary level of analysis when minimizing the mass. However, for a given adhesive and a given required shear strength, increasing the stiffness leads to a reduction of the maximum K_{Ic} that can be reached. Indeed, as the stiffness increases, the maximum thickness for which the adhesive can be selected is decreased. If this thickness is lower than the thickness maximizing K_{Ic} , t^* , decreasing the stiffness leads to a decrease of the maximum K_{Ic} .

The parameters t_{min} and t_{max} also influence the selection as the adhesive fracture toughness evolves with the adhesive thickness. Increasing t_{min} results in a decrease of the maximum stiffness that can be reached in general and for each adhesive (Figure 8C.1a-b). Hence, stiffer adhesives tend to be selected for lower required stiffnesses. For given conditions, as long as t_{min} is below t^* and that the adhesive is of type A (Figure 8.2), the maximum K_{Ic} that can be reached is not affected. If now, there is a limit on

t_{max} , the selection can be affected. Indeed, in Figure 8C.1c, SikaPower 498 cannot longer be selected because in the range going from t_{min} to t_{max} , the Betamate 1493 adhesive is the toughest. Due to the dependence of G_{Ic} on the adhesive thickness, t_{min} and t_{max} influence the selection of the adhesive maximizing K_{Ic} .

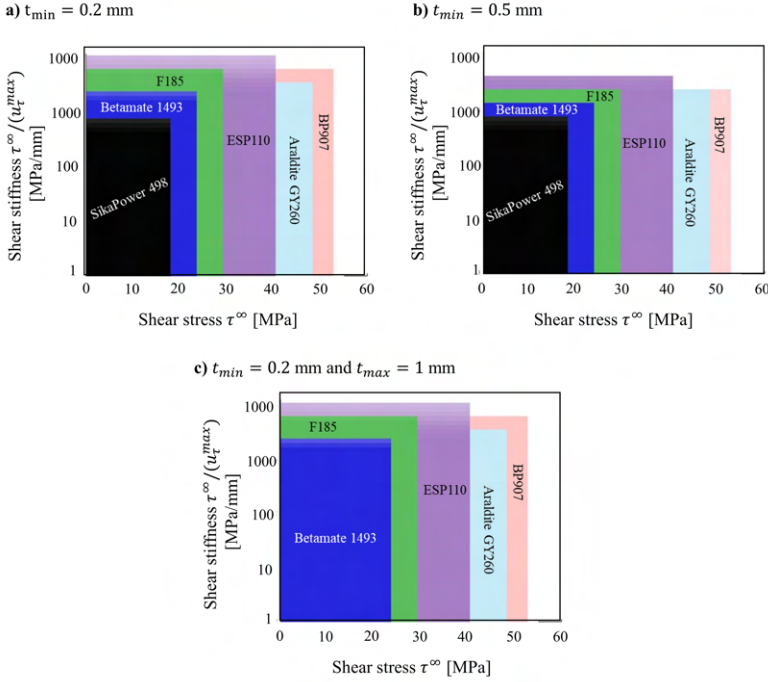


Figure 8C.1 Selection map for the maximization of K_{Ic} depending on the constraints on shear stiffness and shear strength for a) $t_{min} = 0.2$ mm, b) $t_{min} = 0.5$ mm and c) $t_{min} = 0.2$ mm and $t_{max} = 1$ mm.

Architected joints with stop holes

The objective is now to maximize K_{Ic} for architected joints with stop holes. This objective is the one that motivated the design of such joints by Maloney and Fleck [123] and Heide-Jørgensen and Budzik [122]. The selection map is shown in Figure 8C.2. depending on the constraints on the shear stress and the shear stiffness. For low imposed shear stress and stiffnesses, the three architected joints containing 50%, 33% and 25% of cavities can be selected. Indeed, they all lead to the same fracture toughness. As the

stiffness is increased, joints containing a large number of stop holes tend to no longer be selected. Indeed, the larger the number of cavities, the lower the true surface area over which the shear force acts, resulting in a lower stiffness. Consequently, for a given thickness, the joint containing 50% of cavities is less stiff than the one containing 33% and the maximum thickness for which it can be selected tends to be lower than t^* for a lower stiffness constraint. This results in a lower fracture toughness while the joints containing 33% and 25% can still have a thickness t^* , maximizing K_{Ic} . At some point, only the adhesive containing 25% of stop holes can be selected. However, when further increasing the stiffness, even if the selection thickness is lower than t^* , it remains tougher than the plain adhesive joint. At this point, it is observed that the maximum achieved K_{Ic} decreases. At some point, the required stiffness is high enough such that the thickness for which the 25% joint can be selected leads to a joint less tough than the plain adhesive joint. Regarding, the influence of the shear stress, when it is increased, joints containing large amount of cavities can no longer be selected and for large shear stress, only the plain adhesive joint can be selected resulting in more brittle joint. To conclude, architected joints with stop holes are an alternative to plain adhesive joints for limited strength and stiffness constraints.

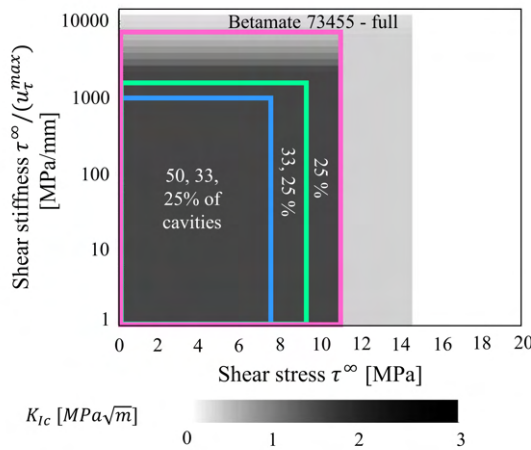


Figure 8C.2 Selection map for architected joints with stop holes when the objective is to maximize the fracture toughness.

8.8.4 Appendix D: Maximization of the remote tensile stress σ^∞ *Classical adhesive joints*

The remote tensile stress is now maximized for classical adhesive joints. As in the previous case, the two constraints are the shear stress and stiffness which are set as limits. The parameters entering the selection are thus $2a$, τ^∞ , u_τ^{max} , t_{min} and t_{max} . Figure 8D.1 shows the influence of the crack length and the required shear stress on the selection for different required shear stiffnesses. The maximum tensile stress that can be reached depending on τ^∞ , $\tau^\infty / u_\tau^{max}$ and $2a$ is shown based on the color scale, the lighter the color, the lower the maximum σ^∞ that can be applied. For short cracks and high shear strength, the strongest and stiffest adhesives are selected. For low shear strengths, if the crack length is longer, tougher adhesives start to be selected leading to the selection of the SikaPower 498 adhesive. However, these tough adhesives are often softer than the brittle one. Hence, when a high shear stress is applied, stronger adhesives such as the BP907 and the Araldite GY260 adhesives are selected. However, this is accompanied by a decrease in the maximum remote tensile load that can be applied on the joint as shown in Figure 8D.1d. The influence of the shear stiffness is considered by comparing Figure 8D.1a to c. By increasing the stiffness from 100 MPa/mm to 1000 MPa/mm, the SikaPower 498 adhesive cannot be selected due to its low shear modulus. Hence the Betamate 1493 adhesive is selected. Further increasing the required stiffness leads to the selection of only the ESP110 adhesive. The Betamate 73455 also meets the stiffness requirements but it is less strong and tough and is thus never the optimal adhesive to select. As the other adhesives do not meet the stiffness requirement, no adhesive can be selected for a required shear strength larger than 40 MPa.

As for the other scenarios, t_{min} and t_{max} can influence the selection as shown in Figure 8D.2. First t_{min} influence the maximum stiffness that can be reached. If t_{min} is increased, for a given required stiffness, the softest adhesive in Figure 8D.1b, Betamate 1493, is no longer selected and the F185 adhesive is then selected, as shown in Figure D.1a. In addition, when comparing Figure 8D.1b and Figure 8D.2a, BP907 is selected for a lower range of thicknesses if t_{min} is increased. Figure 8D.2b shows the influence of t_{max} when compared to Figure 8D.1a. The SikaPower 498 adhesive is no longer selected being replaced by the Betamate 1493 adhesive. Due to the dependence of G_{Ic} on t , the selection is changed when varying t_{min} and t_{max} .

The objectives of maximizing σ^∞ or K_{Ic} can now be compared. When K_{Ic} is maximized, the selection is independent on crack length. In both cases, increasing the imposed shear stress and the shear stiffness results in the selection of less tough adhesives. This arises from the fact that adhesives are either characterized by high strength and stiffness, but low toughness or high toughness and low strength and stiffness.

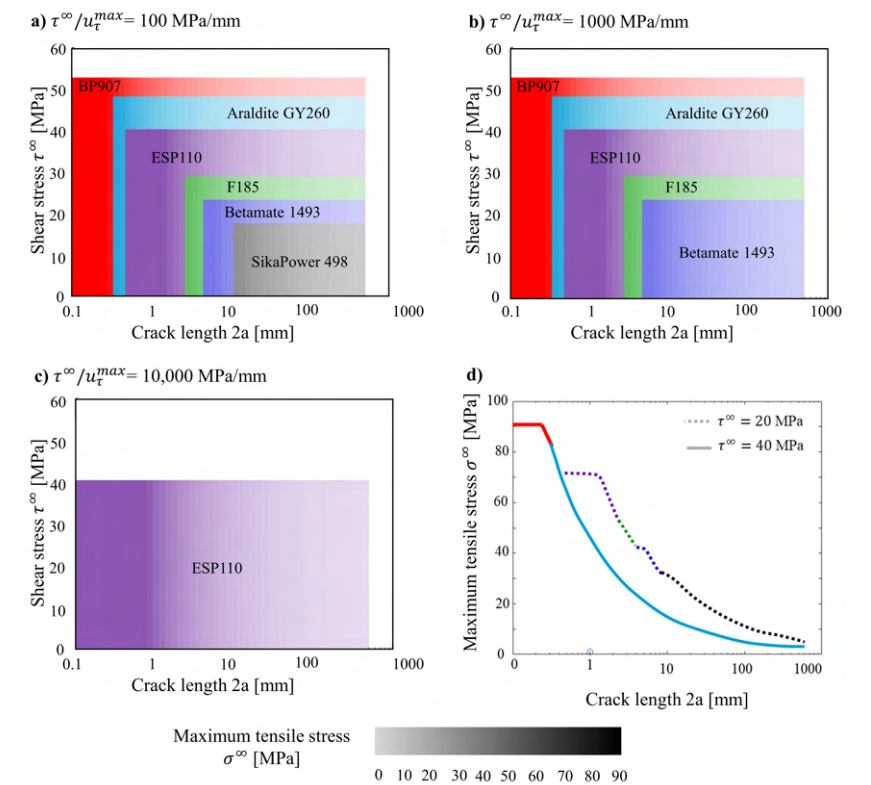


Figure 8D.1 Selection map for σ^∞ maximization depending on the crack length and the shear strength constraint for shear stiffnesses of a) 100 MPa/mm, b) 1000 MPa/mm and c) 10000 MPa/mm. d) Cuts in a) for τ^∞ equal to 20 and 40 MPa to show the variation of σ^∞ maximum with crack length.

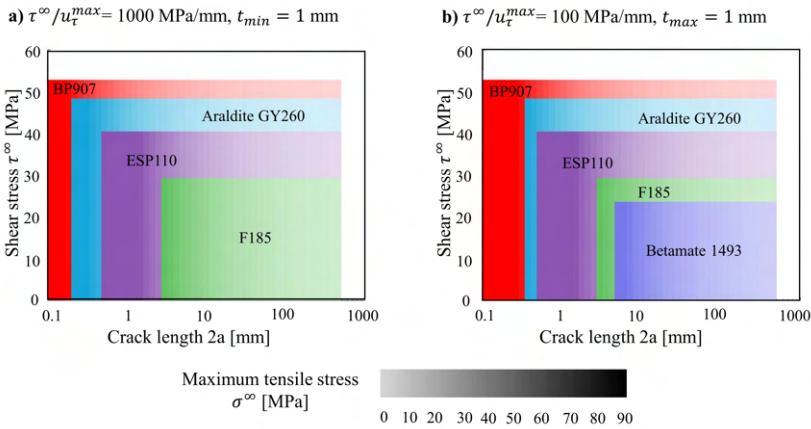


Figure 8D.2 Selection for σ^∞ maximization depending on the shear strength constraint and the crack length and depending on $t_{\min}$ and $t_{\max}$ with a) a stiffness of 1000 MPa/mm, $t_{\min} = 1 \text{ mm}$ and no limit on $t_{\max}$ and with b) a stiffness of 100 MPa/mm, $t_{\min} = 0.2 \text{ mm}$ and $t_{\max} = 1 \text{ mm}$.

Architected joints with stop holes

As for classical adhesive joints, the goal is to maximize the applied tensile stress as a function of the applied shear and stiffness constraints and of the crack length. The selection map for shear stiffnesses lower than 4000 MPa/mm is shown in Figure 8D.3. For small cracks, the plain joint is selected. Indeed, at small crack length, the adhesive is first insensitive to defect size and the effective surface over which the tensile force acts is lower with cavities. Hence, the effective tensile strength is larger for a plain adhesive joint. When increasing crack length, the joint with 25% of stop holes is selected as the reduction of effective cross-section area is balanced by the improved fracture toughness coming from the crack blunting by the stop holes. However, joints with more stop holes tend not to be selected because they are characterized by a similar fracture toughness and lower effective cross-section resulting in a lower σ^{f*} . Due to this reduction in effective cross-section, the shear strength of the joint also decreases when inserting stop holes leading to the selection of the plain joint for applications involving large applied shear stresses. If the required shear stiffness exceeds 4000 MPa/mm, only the plain adhesive joint is selected.

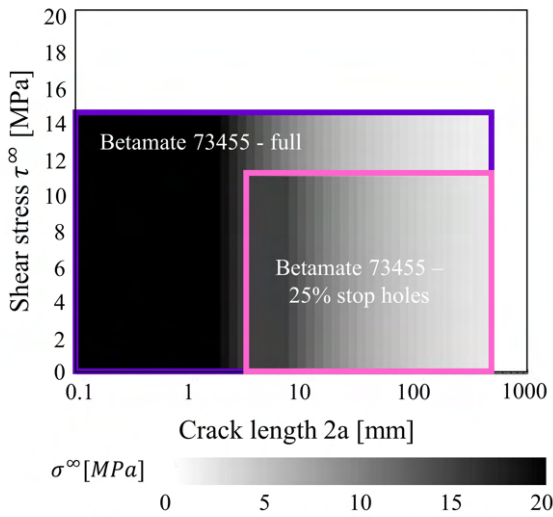


Figure 8D.3 Selection map for architected joints containing stop holes when maximizing the remote tensile force and for a shear stiffness not exceeding 4000 MPa/mm.

8.8.5 Appendix E: Finite element model for architected joints with stop holes

In order to apply the selection approach to architected joints, simulations are run based on the design rules of Heide-Jørgensen and Budzik [121], using the finite element software Abaqus. The influence of the spacing between the stop holes, S_{archi} , and of the thickness is investigated. The length of the stop holes D is thus set to $2l_p$ (Equation 8.23) while the spacing S_{archi} between them is set to $2l_p$, $4l_p$ and $6l_p$ to vary the number of cavities in the joint while t is varied from 0.1 to 1 mm. The influence of D and t on the joint fracture toughness is investigated by modelling a DCB specimen while a lap shear joint is modelled to consider their influence on the shear properties (Figure 8E.1). The implemented constitutive laws and failure criterion correspond to those of Martiny et al. [92]. The 4 mm thick adherends are thus modelled as mild steel and the adhesive as Betamate 73455. The crack propagates when the maximum principal stress ahead of the crack tip reaches 98 MPa over a distance of 18 μm . For both models, the mesh is generated using a Python script and the Gmsh API and is made of

four nodes plane strain elements (CPE4R).

The DCB is 150 mm long with a pre-crack length of 50 mm (Figure E.1). The crack tip corresponds to the end of the first stop hole while two additional stop holes are inserted ahead of the crack tip. The edges of the stop holes are slightly rounded with a filet radius equal to the adhesive thickness to avoid large stress concentrations at the corners near the interface. The mesh is refined near the crack tip over a distance equal to t_a with elements of length between 1 and 5 μm . The simulations are run under controlled displacement with a displacement imposed on the left upper corner as shown in Figure 8E.1a. When the failure criterion is met, G_{Ic} is computed based on Equation 8E.1 from the ISO 25217 standard [172], using the single beam theory and is given in Figure 8E.2b.

$$G_{Ic} = \frac{4P^2}{E_s W^2} \left(\frac{3a^2}{h_s^3} + \frac{1}{h_s} \right). \quad (8E.1)$$

Symmetric lap shear joints (Figure 8E.1b) are modelled with 100 mm long arms overlapping over a distance of 12.7 mm. The number of cavities is adapted depending on the stop hole length and spacing. A horizontal displacement is prescribed at the left side of the joint while the right edge is clamped. The shear stiffness and is given in Figure 8E.2a.

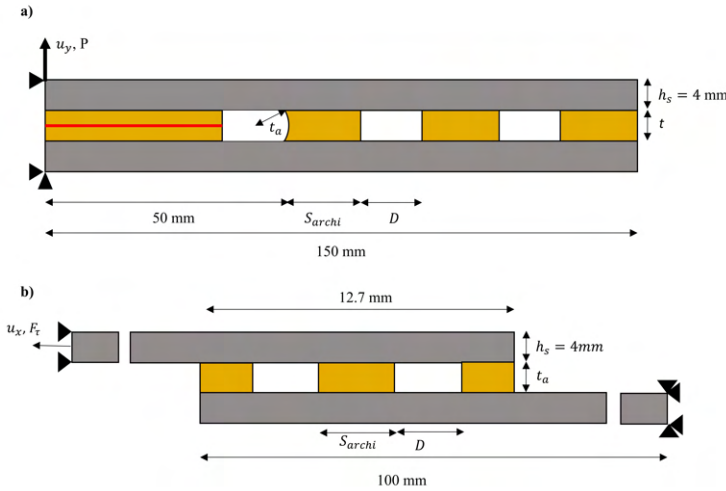


Figure 8E.1 Finite element model for a) Double Cantilever Beam samples and b) Lap shear joint for architected joints with stop holes.

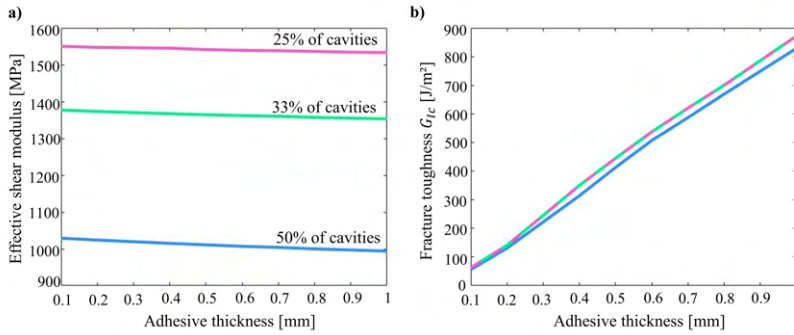


Figure 8E.2 FE simulations results for architected joints containing 25%, 33% and 50% of cavities: evolution of a) the effective shear modulus and b) the fracture toughness G_{Ic} with the adhesive thickness.

8.8.6 Supplementary material: MatLab application

As supplementary material, a MATLAB application is available following the link: <https://www.dropbox.com/sc/lfo/guxcginsvkhyfqfqpnc1w/h?dl=0&rlkey=r14190wb1p3bpu7stcuhsehlv>. This allows the determination of the adhesive to select depending on the loading configuration. It runs based on the .csv and .txt files containing the information of Table 1. The current version only considers the elementary analysis in order to let it run for reasonable amount of time. Including the advanced models results in computations lasting for more than 1h instead of maximum 2 minutes.

9

Conclusion

The objective of this thesis is to investigate different strategies that would favour further deployment of adhesive bonding in high performance applications such as in airplanes, windmills or cars involving CFRP components. Although adhesive joints offer lightweight and homogeneous stress transfer across the joint, adhesive bonding suffers from (i) time- and labour-intensive processes and (ii) limited fracture toughness (G_{Ic}). In this conclusion, we come back to the three questions that motivated the work performed in this thesis and summarize the answers and findings related to these questions. Specific perspectives are listed after the key conclusions regarding each research question.

9.1 Co-curing of composite joints with a TP interlayer

Co-curing is an alternative to secondary bonding which involves simultaneous curing and bonding of composite adherends. This is only possible with a limited number of adhesives and with the use of pre-pregs as resin injection would lead to flow out of the adhesive. In addition, co-cured joints tend to be more brittle than secondary bonded joints [26–28]. Different scientists suggested to replace the adhesive layer by a thermoplastic film. This strategy showed interesting results in the literature when using

pre-pregs [15, 29–32]. In this thesis, Resin Transfer Moulding (RTM) was used to manufacture the co-cured composite joints.

9.1.1 Failure of TP-epoxy interphases

PEI is a TP that is often investigated to manufacture co-cured joints owing to the interphase developing upon curing with an epoxy resin. For this reason, the failure of PEI-epoxy systems has been investigated in recent years [12, 49, 51]. Farooq et al. [49] investigated the failure mechanisms taking place for cracks perpendicular to the interphase. Opposite, Voleppe et al. [12] focused on cracks paralleling the interphase. They reported a tough crack trapping mechanism. Here, we try to understand this trapping mechanism to further mobilize and generalize in other contexts. It has been shown in Chapter 3 that the high toughness results from crack initiation in the tough PEI-rich zone of the morphological gradient. The crack trapping mechanism taking place during the propagation results from the presence of a weak interface between the PEI- and epoxy-rich zones. Hence, the interphase fracture toughness is lower than the one determined by Voleppe et al. [12] but could not be determined in the present work. This conclusion highlights the need to develop an appropriate testing method to determine the fracture toughness of interphases which considers both crack initiation and propagation. Regarding future work investigating the fracture of interphases, it is suggested in Appendix B of Chapter 3 to use wedge-peel tests. However, the manufacturing of the samples and more particularly the insertion of the crack is the first challenge to overcome.

9.1.2 TP co-cured composite joints

Chapters 3 and 4 investigate several TP materials as interlayer for the manufacturing of co-cured composite joints. PEI, PES, PSU, PC, PMMA and PA6 have been tested. PEI and PES co-cured joints have already been investigated in the literature but not the other systems. In addition, autoclave curing with pre-pregs is usually used while here, for the first time, co-curing is achieved through Resin Transfer Moulding (RTM). Several conclusions have been drawn and pave the road for further developments of such joints.

First, it has been found that PA co-cured joints are characterized by a very low G_{Ic} . The PA-epoxy system was the only system exhibiting no morphological gradient upon curing indicating the absence of interdiffusion

during the curing process. Interdiffusion and the development of a morphological gradient seem to be a necessary condition to potentially obtain a tough co-cured joint.

For systems exhibiting a morphological gradient (PEI, PES, PSU, PMMA and PC), some of them present a weak interface between the TP- and epoxy-rich regions such as PEI-epoxy and PES-epoxy systems. In this case, the TP-rich zone must be wider than the distance between the TP film and the carbon fibres. If it is too short, the crack propagates along the weak interface resulting in a brittle joints as found for the PES co-cured joints. The interdiffusion length is influenced by the curing cycle. Several curing cycles could thus be investigated to avoid the issue encountered with PES co-cured joints.

When a morphological gradient is observed and the interdiffusion length is long enough, a strong interface can be achieved. Due to crack propagation along the interface between the fibres and the PEI-rich zone, PEI co-cured joints compete with composite delamination. PMMA co-cured joints even better performed than composites due to a crack propagating in the interphase between the PMMA film and the carbon fibres. PC co-cured joints were characterized by the highest fracture toughness resulting from crack deflection from one interface to the other due to the formation of voids at the interfaces during the manufacturing process. These are thus not as reliable as PMMA and PEI joints. The PMMA joints were thus considered as the joints exhibiting the highest performances in terms of G_{Ic} and the most reliable ones.

Morphological gradients that develop during co-curing depend on the curing cycle. In this thesis, only one curing cycle has been investigated. Several curing cycles could be investigated for each TP-epoxy system to determine their influence on the developed morphologies, on the length of the morphological gradient and on the failure mechanism of the interphase.

For PEI co-cured joints, a higher G_{Ic} than the one associated to composite delamination was expected from the literature in which the 8552 epoxy resin is used instead of the RTM6 resin. In both systems, similar morphological gradients develop. However, in this thesis, RTM was used instead of autoclave curing. This change of process could be at the origin of the lower G_{Ic} . For further work, joints made of same materials could be manufactured through different manufacturing processes to investigate their influence on the joint performances. In addition, the 8552 resin is harder

than PEI while RTM6 is softer. This could modify the failure mechanism taking place in the joint. This could be assessed by investigating the influence of the epoxy resin properties on the failure of the interphase (as suggested in Appendix B of Chapter 3) and the co-cured joint.

The toughness of PA joints was improved by roughening the film surface. Architecturing the TP film surface is thus an attractive toughening strategy to improve the toughness of co-cured composite joints. However, in a first time, it is more important to focus on the manufacturing and the characterization of simple TP co-cured joints.

To conclude, interdiffusion is key to obtain a strong TP-epoxy interphase. However, in some cases, a weak interface can develop due to the lack of chemical interaction between the TP and the epoxy. The interest for TP co-cured joints started 10 years ago [5, 12, 15, 29–33, 68]. Research is thus still needed, in particular regarding the manufacturing of the joints with comparison of the performances of joints manufactured via different processes under different curing cycles and with different TP. Also the durability of the joints in terms of thermal resistance, hygrothermal ageing, fatigue resistance or chemical resistance should be addressed.

9.2 Extrinsic toughening strategies

Several extrinsic toughening strategies have already been investigated for classical adhesive joints [11] such as crack bridging, crack blunting or crack deflection. The goal here was to investigate toughening strategies to be implemented in the context of co-cured composite joints.

9.2.1 Crack bridging

Chapter 5 investigated the toughening of PEI co-cured composite joints by inserting fishing lines between the PEI film and the composite adherends to trigger crack bridging.

This strategy proved to be efficient with an improvement of the toughness by a factor 3 depending on the strength and spacing of the fishing lines. Experimental testing was combined to analytical modelling to elucidate the role of the key parameters. G_{Ic} is improved by increasing the tensile strength of the fishing line and by reducing the spacing between them. The analytical model revealed also the importance of the tensile elongation of the fishing lines. The higher the tensile elongation, the tougher the

joint. However, it also governs the distance over which the R-effect takes place. A longer ligament results in a longer crack extension before attaining the maximum G_{Ic} . The two important parameters for the joint design are the allowed crack extension and the toughness that must be reached to arrest this crack. These two parameters determine the adequate ligament strength and elongation for a given line spacing.

For the moment, the analytical model requires the tensile elongation and the tensile strength to determine the distance over which the R-effect takes place and the final fracture toughness. For joint design, the inverse problem must be solved, requiring the maximum crack extension and the required toughness as inputs to get as output the tensile strength and elongation.

Finally, inserting fishing lines by hand is not reliable and not feasible at large industrial scales. An alternative could be to insert a thin polymeric or metallic mesh between the TP film and the composite. This would act as a carrier, commonly found in commercial adhesives. This carrier could be manufactured via 3D printing for example.

9.2.2 Architected TP film

3D printing is an interesting technology to architecture joints made of TP interlayers. This was investigated in Chapter 6 by designing an architected PA film with channels.

Playing with the film architecture was found to give many degrees of freedom to positively alter the joint properties. By closing some channels on their edges, it is possible to prevent their filling by the composite resin during the RTM process. Controlling this filling triggers different toughening mechanisms: plastic dissipation and mechanical interlocking if the channels are filled and crack deflection in the tough TP and crack blunting if one channel is empty. Aeronautic certifications requires to demonstrate that the crack can be arrested [8]. Hence, the presence of the large channels that could be detected is interesting. In addition to tune the toughness, FE analysis showed that the shear strength of the joint can be tuned by controlling the amount of filled/empty channels in the joint and their location. The novelties of this work are (i) the design of an architected film for co-curing and (ii) the large range of properties that can be achieved from a generic film.

A patent is pending (submission number EP23161330.8) and additional development are thus required. These are listed here-below:

- Modifying the TP material to investigate whether the same toughening mechanisms are triggered or not. For example, one could use a PEI, PMMA or PSU architected film. Depending on the final application, the requirements in terms of durability, chemical, thermal resistance are different, requiring to adapt the choice of the TP.
- Testing of lap-shear joints similar to the one simulated with FE model to confirm the fine tuning of the joint properties with the amount of filled/empty channels and their position.
- Investigating the influence of the geometrical parameters such as film thickness, cavity height, channel spacing and channel length of the joint mechanical properties. This can be assessed through FE simulations (ongoing work).

9.2.3 Addition of a dissipative layer

The addition of a dissipative layer in the joint has been investigated in Chapter 7 using a Finite Element model. It was found to be an efficient toughening strategies. To maximize the joint toughness, the soft layer (i.e. the dissipative layer) must be inserted between two thin hard layers. The softer and the thicker the dissipative layer, the higher the joint fracture toughness as it maximizes plastic dissipation. Minimizing the hard layer thickness improved the joint toughness also by maximizing the interactions between the crack and the soft layer.

First, the results should be experimentally validated by designing adhesive joints involving a soft layer stacked between two hard layers. One idea, could be to insert as TP film between two adhesive layers and to cure the adhesive-TP-adhesive stack during the bonding step. However, a weak interface could be develop as reported in Chapters 3 and 4. Another alternative could be to use two different adhesive films with different properties and to replace the TP film by the softer adhesive. However, this requires to have matching curing cycles for the two adhesives.

Then, the mechanical behaviour under other loading conditions should be investigated like under mode II, under shear (lap-shear tests). Indeed, it must be investigated if the increase in fracture toughness is not detrimental for other aspects.

9.3 Selection of adhesive joints

Chapter 8 dealt with the comparison between classical and architected joints. Toughening strategies have been developed while joints are most of the time loaded under shear. Hence, their shear properties must also be considered before using them in real applications. In Chapter 8, an adhesive selection methodology is developed based on the Ashby materials selection approach. It considers the influence of adhesive thickness on joint fracture toughness and shear strength and stiffness. Two gaps in the adhesive property space are found corresponding to stiff/strong and tough adhesives. The same selection approach has been applied to compare an architected joints with stop holes to its classical counterpart. It is shown that architecturing allows to partly fill this gap. It is the first time that the Ashby selection method is extended to adhesive joints and it proved to be interesting to compare architected/toughened solutions to classical joints to determine for which kind of application they represent an interesting alternative.

The main advantage of the proposed methodology is to easily compare adhesives together by considering a simple joint configuration. It allows for a first screening of the adhesives and to select some potential ones. This tool has not the vocation of being an accurate design tool.

One of the main limitation of the methodology is the need for experimental data for all adhesives which are not always available. To avoid this issue, the best option would be to determine for each adhesive its constitutive law and a failure criterion. With these data, it is possible to link the selection tool with a FE code that could predict joint failure depending on the loading and the configuration. However, this will require large computation time.

Finally, the initial goal was to be able to compare architected/toughened joints with their classical adhesive joint counterpart. In the literature, researches only focus on the mode I toughness of such joints. In order to compare them to classical adhesive joints, it would be necessary to collect data about the toughness of the joints but also on other mechanical properties such as the shear strength or the shear stiffness. Gathering experimental data under different loading conditions and the use of the proposed methodology would allow determining the range of applications for which a toughened/architected joint can replace its classical counterpart.

9.4 General perspectives

Specific perspectives have been provided above, but it is now interesting to take some distance and look at the thesis from a broader perspective.

First, metal-composite bonding is a challenging and very important subject for many applications going from sport equipment to larger structures such as airplanes and wind turbine blades. The complexity comes from the need to combine chemistry (surface chemistry, rheology, physico-chemical properties) and mechanics (deformation and fracture). These aspects must be simultaneously considered to improve the joint performances as illustrated in this thesis. For instance, co-cured composite joints showed limited fracture resistance but architecturing the joints resulted in improved fracture resistance.

Regarding the further deployment of co-curing in the industry, research is still needed to convince industries to move to such process. Indeed, it requires to redesign moulds and change the manufacturing processes resulting in high costs for the industries. Hence, the barrier to the market is important and industries must be convinced by the benefits brought by such change in terms of process efficiency, long term cost reductions and in terms of resulting joints properties.

Regarding the improvement of architected joints, two aspects appear to be important for further development. First, toughness is not the only essential property of a joint as stiffness and strength also matter. Strength and stiffness are negatively affected by adhesive thickness while toughness is improved. It is important to consider this aspect to avoid designing thick tough joints but which lack of strength and stiffness. For thin joints, this is a real limitation in the design possibilities (e.g. 3D printing cannot achieve structures at the order of 100-300 μm). Secondly, the maximum allowed defect size in the joint is key to design the joint such that the defect is arrested before reaching this size. The best situation would be to have a critical crack length corresponding to the resolution of non-destructive control techniques. The pattern would then be designed to stop all defects smaller than the critical defect size.

Variability in terms of toughness is reported in classical adhesive joints. For example, a crack can propagate in the adhesive and then along one of the interfaces. By introducing an architecture, the crack path can be con-

trolled, removing this variability. Architecturing can thus lead to a more predictable behaviour.

Classical adhesive joints are difficult to introduce in some applications such as in aeronautics as certification agencies challenge their reliability. We do not believe that architected joints will be used in such applications on a short or medium term. But, windmills constitute an attractive field for the deployment of architected joint. Adhesives are indeed already widely used and certification requirements are less severe. A key limitation of adhesive bonding is the lack of non-destructive techniques to control their quality. One has to probably wait for integration of health monitoring to see adhesive bonding to be used at larger scales. Architected joints constitute maybe a path towards integration of micro-sensors without negative effect.



Thermoplastic welding: towards composite-metal joints

Composite parts are not only welded to composite parts but also to some metallic parts. This appendix is a step forward the co-curing of composite-metal joints through the addition of a thermoplastic layer. This appendix investigates the influence of the welding conditions on the tensile strength of Ti-TP-Ti joints. Part of the results have been generated during the master thesis of G. Lejeune.

A.1 Introduction

Carbon fibre reinforced polymer composites (CFRPs) are broadly used in advanced structures such as in airplanes, wind turbine blades or marine structures, owing to a high strength and stiffness to weight ratio. Manufacturing these structures often requires assembling CFRPs together or with other materials. This can be achieved through adhesive bonding that offers lightweight and homogeneous stress transfer opposite to mechanical fasteners [1]. However, one of the main disadvantages of adhesive bond-

ing are the time and labour-intensive bonding processes. An alternative to the classical bonding process is co-curing. If bonding is achieved through co-curing without inserting any adhesive, the resulting joint is characterized by a low fracture toughness [255] due to the lack of adhesion between the composite resin and the metal plate. In Chapters 3 and 4, it has been shown that composite-composite joints can be manufactured through co-curing by inserting a TP layer. The idea of the current chapter is to insert a TP layer between the metallic adherend and the composite. In this case, joining is achieved through welding of a thermoplastic layer with a metal. Two strategies could be investigated to manufacture composite-metal joints using a TP layer as adhesive. As shown in Figure A.1, either the TP layer is first welded on the the metal and then this assembly is co-cured with the composite or the TP is first co-cured with the composite and then welded to the metal. However, this requires to generate a good adherence between the TP layer and the metal part. On the road towards the manufacturing of composite-titanium joints, the present chapter investigates the manufacturing of titanium-TP-titanium joints through thermoplastic welding using a static hot press. For this purpose, the tensile strength of titanium-thermoplastic-titanium joints is investigated by modifying the titanium surface treatments and the welding conditions.

Opposite to thermosetting polymers, TPs have the advantage to soften above a certain temperature T_m . Above this temperature, their viscosity drops and they can easily flow leading to the penetration of surface asperities to achieve bonding. Several processes have been developed to perform thermoplastic welding (see Figure 2.2 in chapter 2)[3, 5, 6, 18–21]. One of the most investigated process is ultrasonic welding. The most important processing parameters are the vibration amplitude, the supplied power and energy, the welding time, the holding pressure and the holding time after welding during solidification [18, 256]. Ultrasonic welding has the advantage of not inserting any component between the two parts to join. However, large areas cannot be bonded in a single step [257]. In this case, several spot welds must be performed. Another process is induction welding [20, 258] which requires to insert magnetic particles in the thermoplastic. This process enables the welding of large surface areas by progressively moving the coils [3]. Another possibility is to insert an electric resistance between the two parts to join to perform resistance welding [6, 259, 260]. Other alternatives are laser beam welding [21] and friction stir welding [19]. Welding of thermoplastic composite and metallic parts has

been proved to be efficient using induction and ultrasonic welding [3, 20]. Marinosci et al. [261] also showed that autoclave consolidation is possible.

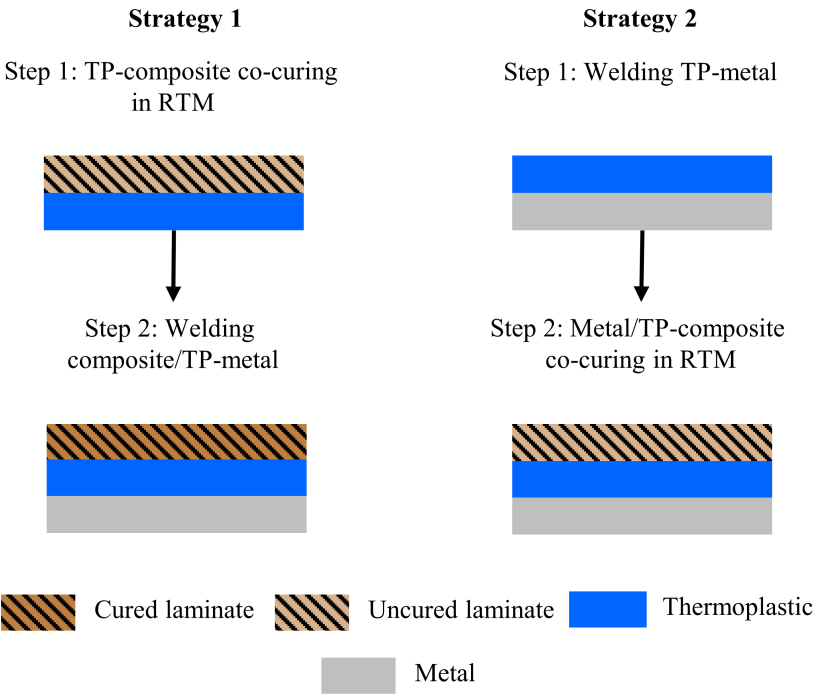


Figure A.1 Strategies to joint CFRP to metal using a thermoplastic inter-layer. Strategy 1: welding to metal after co-curing of CFRP and thermoplastic. Strategy 2: welding of metal with thermoplastic before composite co-curing.

In addition to the manufacturing process and its parameters, adhesion must be ensured between two components two bond. Several adhesion theories exist [1, 262]. First, in the case of adhesive joints, the adhesive must wet the surface to ensure intimate contact between the adhesive and the substrate. Adhesion can also be favoured by mechanical interlocking resulting from surface roughness, by electrostatic interactions, by chemical bonding (formation of covalent or hydrogen bonds), by acid-base interactions and finally by the diffusion of species to create an interphase. Surface treatments have been developed to ensure good adhesion between parts to bond as they create surface roughness and modify the surface chemistry

[1, 14]. Depending on the material, several treatments can be envisaged. Both for polymeric and metallic materials, mechanical treatment such as grit blasting and abrasion are possible. These remove some surface contaminations and create macroscopic roughness. Joint strength tends to increase with roughness up to an optimum roughness that depends on several parameters such as adhesive viscosity [109–113]. These mechanical treatments are avoided by some industries such as aeronautic due to the difficulty of producing homogeneous surfaces. For CFRPs, peel-ply and tear-ply are two other alternatives to create macroscopic roughness. The peel/tear-ply is stacked on the top of the composite before curing and can be removed upon curing, before bonding. Other treatments such as corona discharge, plasma, flame or laser treatment are known to be effective by inducing some roughness and modifying the surface chemistry. These treatments change the surface chemistry by creating carbonyls, hydroxyls or carboxylic acid groups on the surface. For metals, acid and alkaline etching are two common treatments. For titanium chromic and NaOH anodisation have also been proved to be efficient surface treatments. Due to environmental concerns with these treatments, Boeing developed the solgel treatment. The solgel treatment is a water-based treatment creating an hybrid organic/inorganic coating on the surface to treat [263] as shown in Figure A.2. Organic groups can be chosen to have a good compatibility with the adhesive to improve chemical adhesion.

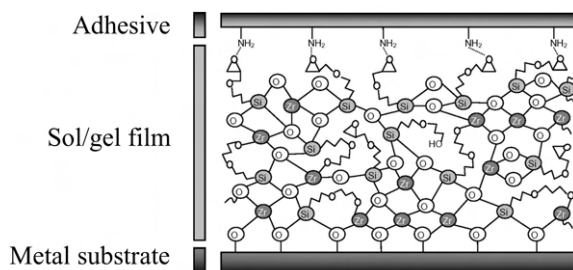


Figure A.2 Scheme of a solgel coating between a metallic substrate and an epoxy adhesive using an organofunctional silane. Image taken from [263].

A.2 Materials and methods

A.2.1 Manufacturing of the joints

The manufacturing of the joints involves two steps. First the titanium surfaces must be treated to remove the contaminations. Then, welding can take place using a hot press. Prior to surface treatment, Titanium studs have been manufactured from titanium rods with a diameter of 20 mm. The studs (see Figure A.3) are made of a flat surface that can be bonded while at the other side, a screw is machined in order to be able to perform the butt tensile tests based on the methodology used in the thesis of Y. Bertholet [264]. As thermoplastic materials, PEI and PA6 films supplied by GoodFellow have been used. For PEI, films of 50, 175 and 500 μm have been used while for PA6, only 200 μm thick films have been used.

In order to be able to compare these joints to classical joints, an epoxy film commonly used in aeronautic is used.

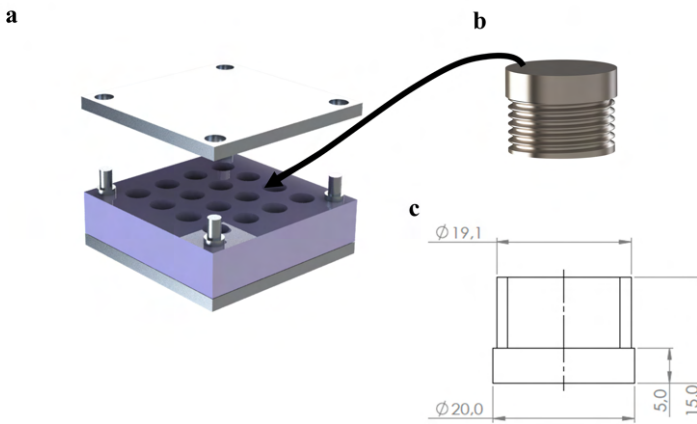


Figure A.3 a) Mould to perform welding in which the titanium studs and the thermoplastic film are inserted. The cover can be equipped with springs; b) titanium stud and c) dimensions.

Surface treatments

First, the surfaces have been treated mechanically. The titanium studs have been polished using 80, 320 and 800 SiC papers. For the 80 SiC paper, only one step is needed while for the two other the SiC paper grade is progres-

sively increased. For every steps, the surface has been polished for 30 seconds. In order to avoid roughness variability due to the wear of the SiC paper, studs are never polished at the same location and the papers have been replaced after six samples. In addition to polishing, sandblasting (SB) and beadblasting (BB) have been used as mechanical surface treatments. Upon surface treatment, right before welding, the surfaces to weld are cleaned using acetone and rinsed using distilled water before being dried using compressed air.

In the industry, mechanical surface treatments such as polishing or blasting are not used as it is difficult to obtain a uniform quality of the surface over large areas. Hence, other treatments are preferred such as solgel treatments (SG)¹. Some of the titanium studs have been treated using an industrial protocol. For the titanium studs joined with the adhesive, this is the only treatment that has been used as these are the optimal industrial conditions for titanium joints manufactured using the epoxy adhesive.

The thermoplastic films have also been treated with a O₂ plasma treatment with a pressure of 8 mTorr during five minutes.

Welding with hot press

In order to perform Ti-TP welding using the hot press, a specific mould has been designed and machined. The mould is made of 25 cavities having a diameter of 20 mm to ensure the good alignment of the two Ti studs to be welded (Figure A.3). A first Ti stud is inserted on which a thermoplastic film is stacked before adding the second Ti stud. The mould is then closed. The cover is perforated in order to insert springs to control the pressure applied on the studs. When the springs are used, a pressure about 0.3 MPa is applied on each Ti-TP-Ti stack. The pressure applied using the hot press and the one applied to the springs are thus not related to each other. The goal was to avoid to apply a high pressure that could lead to the squeezing of the thermoplastic. Once the mould is closed, it is introduced between the two stages of the hot press which has been previously heated up to a temperature T_w . When, the mould is introduced, the temperature decreases before increasing again. Once T_w is reached again, this temperature is held for a time that can vary. After this time, the moulds is removed from the hot-press and is placed in a second press which is not heated for 15 minutes.

¹Any other information cannot be disclosed due to a non-disclosure agreement.

The different parameters are thus:

- the thermoplastic material: PEI or PA6
- the initial thickness of the thermoplastic film
- the surface treatment applied on the Titanium studs
- the welding temperature
- the welding pressure: cover with (P) or without springs (NP).

The nomenclature of the different conditions is as follow: TP-Thickness-SurfaceTreatment-Temperature-Time-Pressure.

Adhesive bonding

The same mould has been used for hot press welding, except that the adhesive film is inserted instead of the thermoplastic film. However, instead of a hot press, an Heraeus oven with controlled temperature cycle has been used. Bonding is achieved through following the adequate curing cycle. The cover of the mould was equipped with the springs resulting in a pressure of 0.3 bars.

A.2.2 Butt tensile tests

The butt tensile tests are performed following the methodology developed in the PhD thesis of Y. Bertholet [264]. However, in the present case, the two studs are welded/bonded prior to insert the studs in the machine. Each stud is screwed in one of the two supports shown in Figure A.4 that can be gripped by the testing machine. A screw driven Zwick/Roell Machine equipped with a 50kN cell is used. The tests are performed at a constant cross-head speed of 1 mm/min. Upon testing, only the tensile strength is computed based on the peak load. For example, the elastic modulus and tensile elongation could not be computed due to some compliance effects in the mounting. The tensile strength was the only reproducible property from one test to another. The tests are performed in a ventilated room at a constant temperature of 23°C and relative humidity content of 50%. For each condition, seven samples are tested.

A.2.3 Microscopy

A VHX optical microscope from Keyence and an Ultra 55 Field-Emission SEM (Zeiss, Germany) are used to image the fracture surfaces of the welds.



Figure A.4 Ti-TP-Ti weld and the fixtures in which they are mounted to perform the butt tensile tests.

For SEM imaging, the samples are coated with a 1 μm thick gold layer prior to observation. The samples are imaged at 5 keV.

A.3 Results and discussion

Butt tensile tests are performed on Ti-TP-Ti welds. The strength of the joints is known to be affected by many parameters such as the surface roughness of the adherends or the adhesive thickness. The performance of thermoplastic welds is also known to be influenced by the maximum temperature that is reached during the welding process, the pressure, the welding time or even the holding time during consolidation [18, 20]. In order to determine the conditions leading to the best welds, different surface treatments, welding conditions (temperature, pressure) and initial film thickness are investigated as presented here-below. In addition, some statistics have been done based on the Weibull distribution and the results are given in Appendix A.

As reference, titanium studs treated with the solgel treatment are first bonded using the adhesive. A high strength is achieved as all joints failed cohesively. The value is taken as the reference to evaluate the performances of the titanium studs welded using a thermoplastic film. It is denoted as τ_a^f .

A.3.1 Influence of the surface treatment

Several surface treatments have been investigated. As for the adhesive joints, the titanium studs have been treated with a solgel treatment. In addition, the titanium studs have been mechanically treated through sand-blasting, beadblasting and polished using 80, 320 and 800 grade SiC papers to create surface roughness.

Figures A.5 and A.6 show the strength of the solgel treated titanium studs welded with PEI and PA. The PEI-Ti welds are characterized by a tensile strength equal to $0.25\tau_a^f$. However, only 65% of the welds could be tested as the others failed upon unmoulding. Failure is found to always be adhesive. The PA-Ti welds are characterized by a tensile strength of $0.22\tau_a^f$ with two populations: a brittle population with a strength lower than 1 MPa and a stronger population with a strength exceeding $0.26\tau_a^f$ up to 52% of τ_a^f , but in both cases adhesive failure takes place. However, for the strong population, PA is present on both sides indicating the propagation of a wavy crack. For the PA-Ti welds, only 70% of the welds survived to the unmoulding. Owing to these low tensile strengths, additional tests are performed by treating the thermoplastic film with a O_2 plasma to favour chemical bonding. For the PEI joints with a plasma treatment, the weld success rate is about 30% and the strength is equal to $0.04\tau_a^f$ with systematic adhesive failure. 85% of the PA-Ti welds are successful and characterized by a tensile strength equal to $0.34\tau_a^f$ which is better than without the plasma treatment. By increasing the welding temperature from 235°C to 255°C for the PA-Ti joints, a success rate of 100% is achieved and the tensile strength is increased to $0.59\tau_a^f$. For both the PA treated welds and the welds performed at 255°C , mix-failure is observed. The solgel treatment leads to low strength and the welds are not highly reliable as some failed before testing. PEI and PA are respectively characterized by tensile strength ranging between 90 and 100 MPa and 50 and 80 MPa [179]. As the attained values are way lower than these values, higher performances could be attained by triggering cohesive failure rather than adhesive failure. Upon welding, the titanium studs change of colour, from a silver colour to a golden colour. In addition, as shown in Figure A.5d, burning traces appear, indicating degradation of the solgel coating. For this reason, mechanical surface treatments are applied to the titanium studs although these are avoided by the industry as homogeneous surface conditions are difficult to obtain over large surface areas.

First, mechanical surface treatments affect the surface roughness in terms of depth and wavelength. The surface roughness profiles are shown in Figure A.7. Sandblasting, beadblasting and SiC80 paper polishing result in similar average roughness R_a about 1.74 ± 0.7 , 1.34 ± 0.2 and 1.74 ± 0.6 μm . However, it can be seen in Figure A.7a-c that the wavelength (distance between two consecutive peaks) is the largest for sandblasting and the lowest

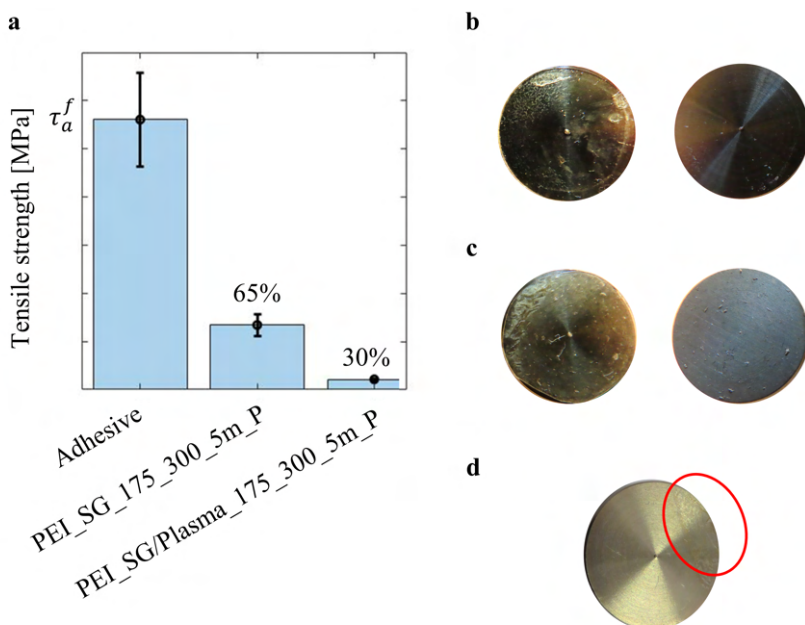


Figure A.5 a) Strength of welds made of the Titanium studs treated with the solgel with and without plasma treatment on PEI and the corresponding fracture surfaces b) without and c) with the plasma treatment. d) Titanium studs with solgel coating after welding and burning traces highlighted by the red circle.

for beadblasting. The average roughness obtained with the 320 and 800 SiC papers are about 0.2 and 0.13 μm , respectively.

Figure A.8 shows the tensile strength of welds achieved by inserting a PEI film. Maximum tensile strength (71 ± 4.6 MPa) is achieved by sandblasting the titanium studs while beadblasting results in the lowest strength (30.4 ± 7.7 MPa), still higher than with the solgel treated surfaces. This difference can be explained by the failure mechanism taking place in the welds. For the sandblasted welds, cohesive failure takes place while adhesive failure is observed for the beadblasted welds. For the polished studs, the tensile strength is about 39 ± 12.6 , 53.1 ± 7.2 and 53.2 ± 9.6 MPa for the 80, 320 and 800 SiC papers, respectively. For the samples polished with the 80 SiC papers, adhesive failure is dominant compared to cohesive failure close to the interface while for the two other polishing conditions, it is the

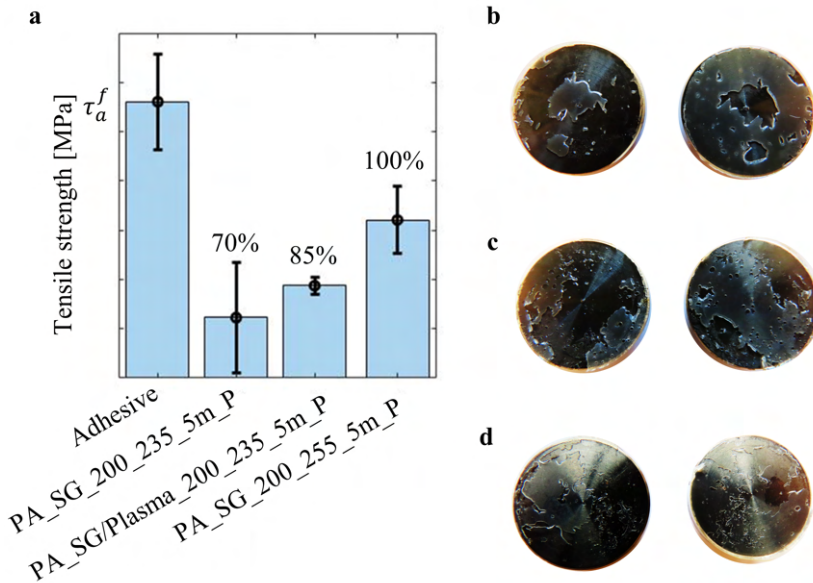


Figure A.6 a) Strength of welds made of the Titanium studs treated with the solgel with and without plasma treatment on PA and the corresponding fracture surfaces b) and d) without and c) with the plasma treatment. Welding temperature is set to 235°C for b) and 255°C for d).

opposite and cohesive failure close to the interface is more often observed than adhesive failure resulting in a higher strength. The reliability of the welds is statistically investigated using the Weibull distribution. The most reliable conditions is the one with the highest Weibull modulus which is sandblasting (22.1). It is important to note that no weld failed before being tested compared to welds performed on solgel treated titanium studs.

Figure A.9 shows the results for the PA joints with different surface treatments. Beadblasting and sandblasting result in the strongest joint with tensile strengths equal to 46.9 ± 14.3 MPa and 45.6 ± 4.9 MPa, respectively. However, the sandblasted joint is the most reliable owing to its lower standard deviation and its highest Weibull modulus (13.8 compared to 5.6). In both cases, cohesive failure is shown in Figure A.9b and c. Polishing the titanium studs result in lower tensile strength, not exceeding 30 MPa with cohesive failure close to the interface and adhesive failure being the dominant failure mechanisms. Again, no weld failed prior to testing.

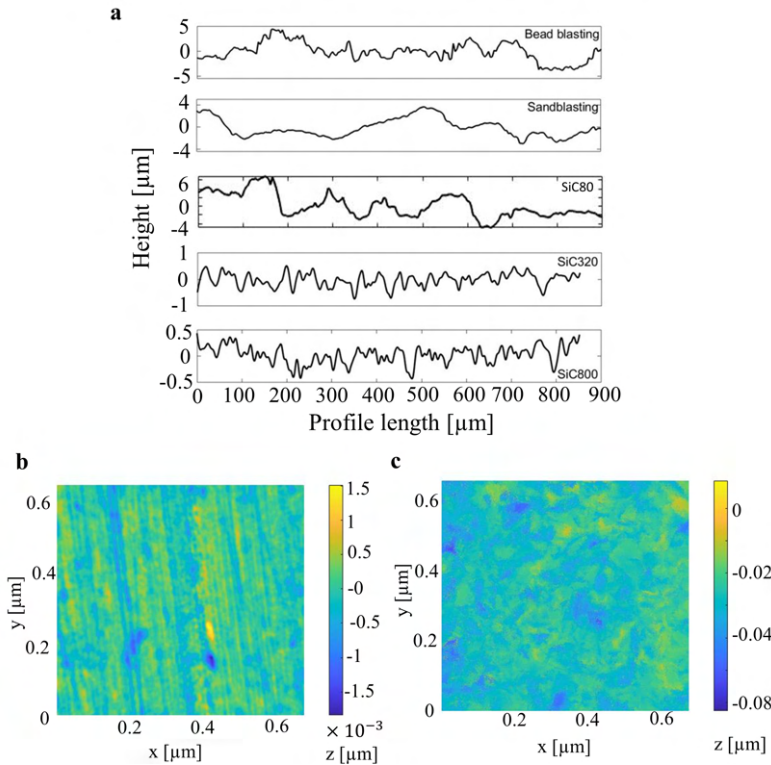


Figure A.7 a) Roughness profiles for the different mechanical surface treatments. b-c) Image surfaces for studs treated with b) 320 SiC paper and c) sandblasting.

The evolution of the tensile strength with the average surface roughness is shown in Figure A.10 for both PEI and PA welds. For PEI, there is no major increase in strength. Indeed, a R_a about 1.5 μm is achieved with three surface treatments and, the tensile strength is strongly affected by the selected treatment. The surface roughness induced by the three treatments can be compared in terms of wavelength. The longer wavelength is achieved with sandblasting and the shortest one with beadblasting. Hence, the surface asperities in which PEI has to penetrate are narrower with beadblasting. Based on the theoretical analysis of De Bruyne [113, 265], it can be argued that a longer wavelength favours adhesive penetration into the pits. The lower the wavelength, the more pressure must be applied to force adhesive penetration into the pits. Faizi [112] experimentally showed that narrow-

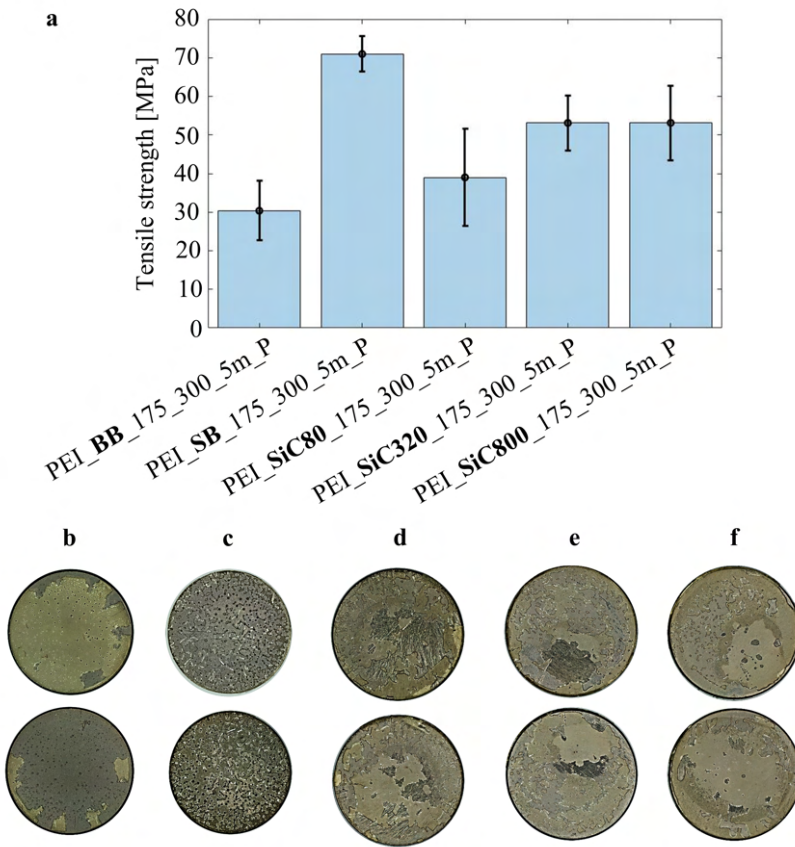


Figure A.8 Influence of the surface treatment on the strength of PEI-Ti welds and the fracture surfaces after b) beadblasting, c) sandblasting, d) 80 SiC, e) 320 SiC and f) 800 SiC polishing.

spaced valleys contribute to poor wettability and that there exists an optimum wavelength which is not as often investigated with respect to R_a . Hence, PEI penetrates more easily into the surface asperities of the sandblasted studs resulting in enhanced mechanical interlocking and tensile strength. For the PA welds, the tensile strength increases with roughness. In this case, beadblasting and sandblasting both result in similar tensile strengths indicating a good wetting by PA in both cases. The lower viscosity of PA compared to PEI could explain this difference [111]. Based on this, a similar strength could be expected for the studs polished with the

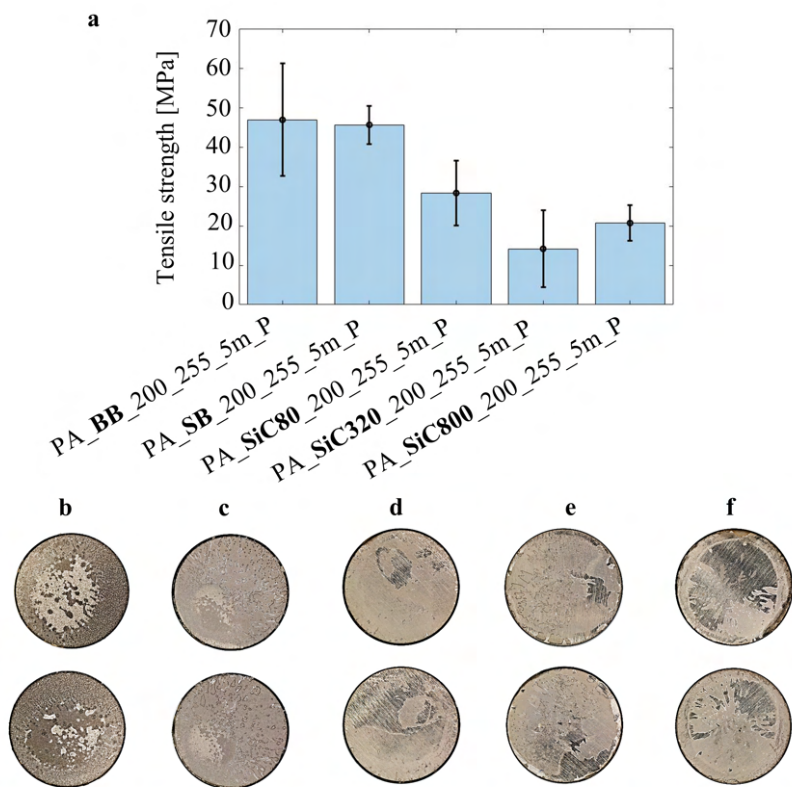


Figure A.9 Influence of the surface treatment on the strength of PA-Ti welds and the fracture surfaces after b) sandblasting, c) beadblasting, d) 80 SiC, e) 320 SiC and f) 800 SiC polishing.

80 SiC papers. The lower strength could be explained by the difference in surface pattern. Indeed, polishing results in grooves all oriented in the same direction. The profiles given in Figure A.7 are taken perpendicular to the groove orientation. Parallel to the groove orientation, a more flat profile can be expected (Figure A.7b) while with bead- and sandblasting, the roughness profile is similar in all directions (Figure A.7c) resulting in more interlocking and larger surface area.

This first part showed that the surface treatment strongly influences the joint tensile strength. It is often argued that the larger the average rough-

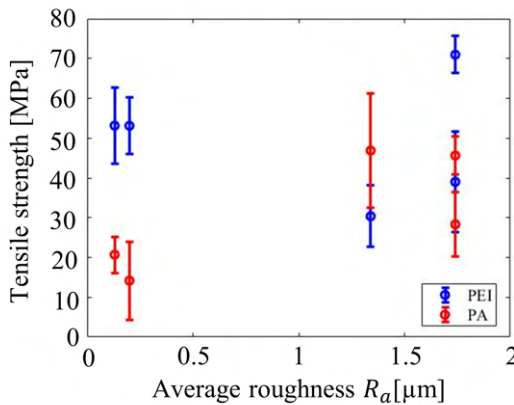


Figure A.10 Evolution of the tensile strength with the average surface roughness R_a for the PEI-Ti (blue) and PA-Ti welds (red).

ness, the higher the tensile strength. However, it has been shown here that the wavelength is also important. In addition to the wavelength, the polymer viscosity is an important parameter: the less viscous the polymer, the more easily it can penetrate into the pits.

A.3.2 Influence of welding temperature

The influence of the welding temperature on the tensile strength for PEI-Ti welds is shown in Figure A.11 for sandblasted studs and titanium studs polished with 80 SiC papers. For the sandblasted joints, the temperature does not affect the tensile strength. For the 80 SiC polished studs, a slight influence of the temperature is found. However, the temperature affects the size distribution of the voids that develop during welding in the thermoplastic layer. By increasing the temperature, the probability to have small voids decreases while larger voids tend to form. If the voids are approximated by a single void of equivalent surface area, the diameter of the equivalent void increases from 3 mm at 300 and 320 °C to 6 mm at 340 °C. For the polished samples, at 300 °C, the voids could not be localized while the equivalent diameter increases from 2.5 to 5.6 mm if the temperature is increased from 320 to 340 °C. Van Loock et al. [230] summed up all voids to define an equivalent void of diameter ϕ_{eq} . This equivalent void is then treated as a crack of length $2a$ equal to ϕ_{eq} . For a given thickness, a lower tensile strength is expected with ϕ_{eq} increases, opposite to the current re-

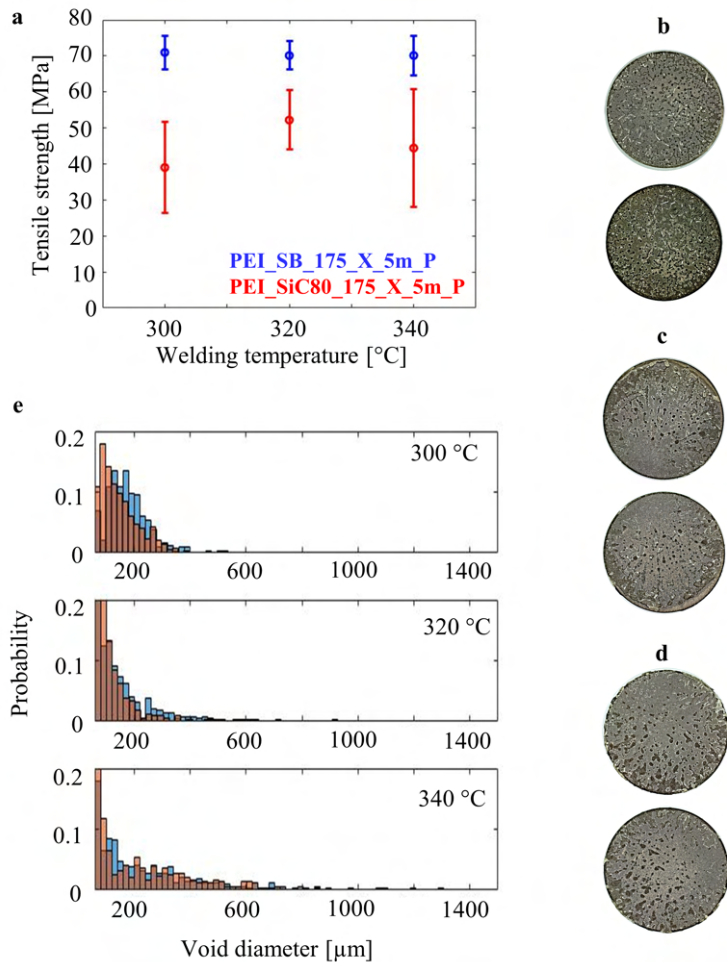


Figure A.11 a) Influence of welding temperature for PEI-Ti welds sandblasted (blue) and polished with 80 SiC paper (red) and the fracture surfaces for the sandblasted studs welded at b) 300, c) 320 and d) 340°C. e) Probability distribution of the size of voids in the welds depending on the temperature (2 samples per temperature).

sults. Hence, temperature should also play on other parameters such as the wetting of the surface and the penetration of PEI into the surface asperities. Indeed, wetting is known to be favoured by increasing the temperature and the viscosity decreases with temperature resulting in a better

flow of the polymer. However, this could not be analysed in the context of the present experiments.

The influence of the welding temperature on the strength of sandblasted and 80 SiC polished samples is shown in Figure A.12 for Ti-PA welds. In this case, increasing the temperature tends to reduce the joint tensile strength. Increasing the temperature may increase the size of the voids in the joint, improve the wetting of the titanium surface and decrease the polymer viscosity. The decrease of the strength can be partly explained by the increased defect size as shown in Figure A.12 as the equivalent defect diameter increases from 1.6 to 3 mm if the temperature is increased from 235 to 255 °C. In addition, by looking at the cross-sections of unbroken joints, it is found that increasing the temperature results in a lower remaining PA thickness. By increasing the temperature, the polymer flows more easily and can be squeezed out of the studs. For the sandblasted welds, the thickness decreases from $73 \pm 28 \mu\text{m}$ to $26 \pm 6 \mu\text{m}$ by increasing the temperature from 255 to 300 °C. Hence, the ratio between the equivalent crack length and the thermoplastic layer thickness decreases resulting in a decrease of the strength as explained by Van Loock et al. [230]. In addition, for the welds performed at 300°C, burning traces can be observed on the PA squeezed out of the studs indicating some degradation of the polymer.

To conclude, by affecting the viscosity and the development of voids, the welding temperature influences the tensile strength. Increasing the temperature results in improved wettability due to lower viscosity resulting in better penetration into the pits and thus enhanced mechanical interlocking. It also leads to improved polymer flow that can be squeezed out of the weld resulting in a lower polymer thickness which is not always beneficial. Finally, increasing the temperature results in the development of larger voids which can be seen as longer cracks. The optimum temperature must thus be found which is a compromise between a good wettability of the surface and small voids.

A.3.3 Influence of initial film thickness

For titanium studs welded through the insertion of a PEI layer, the influence of the PEI layer thickness on the joint strength is investigated. Sandblasted titanium studs are welded at 300°C using 50, 175 and 500 μm thick PEI films. Figure A.13 shows that the maximum tensile strength is achieved for a thickness of 175 μm . At low thickness, an increase in joint

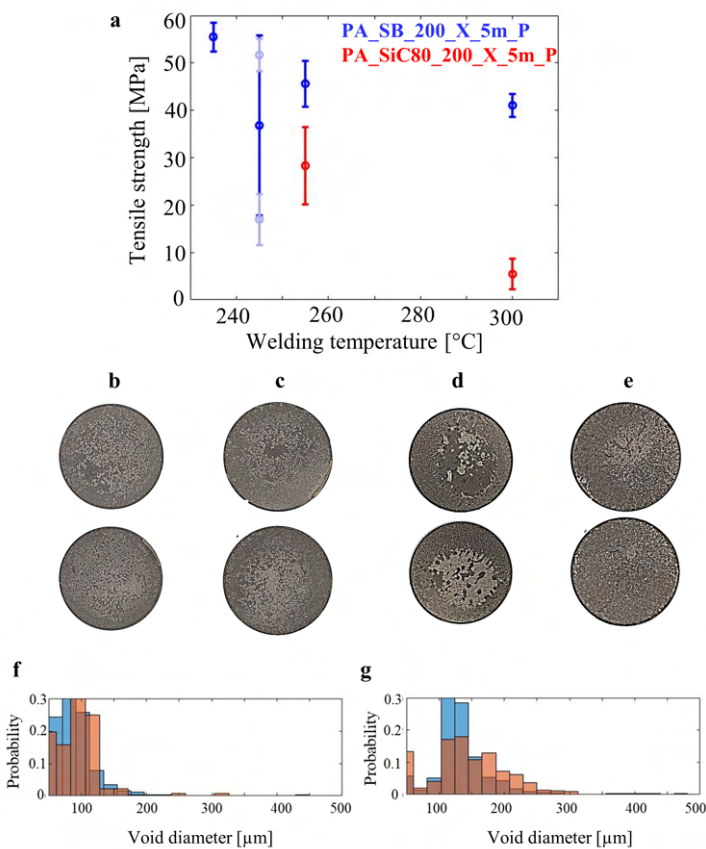


Figure A.12 a) Influence of welding temperature for PA-Ti welds sand-blasted (blue) and polished with 80 SiC paper (red) and the fracture surfaces for the sandblasted studs welded at b) 235, c) 245, d) 255 and e) 300°C. Probability distribution of the size of voids in the welds depending on the temperature (2 samples per temperature) for PA-SB-200-X-5m-P welds for temperatures of f) 235°C and g) 255°C.

strength is often reported due to the decreasing degree of constraint in the adhesive layer. At large thickness, the strength decreases due to stress singularity developing at the joint corners close to the interface with the adherends resulting in adhesive failure instead of cohesive failure. The fracture surfaces shown in Figure A.13c and d shows this shift in failure mechanism. For the 50μm thick film, the degree of constraint is too high

resulting in lower failure strength. In DCB joints, the optimum adhesive thickness is about twice the plane stress plastic zone size, corresponding to a thickness about 200 μm in agreement with the current results.

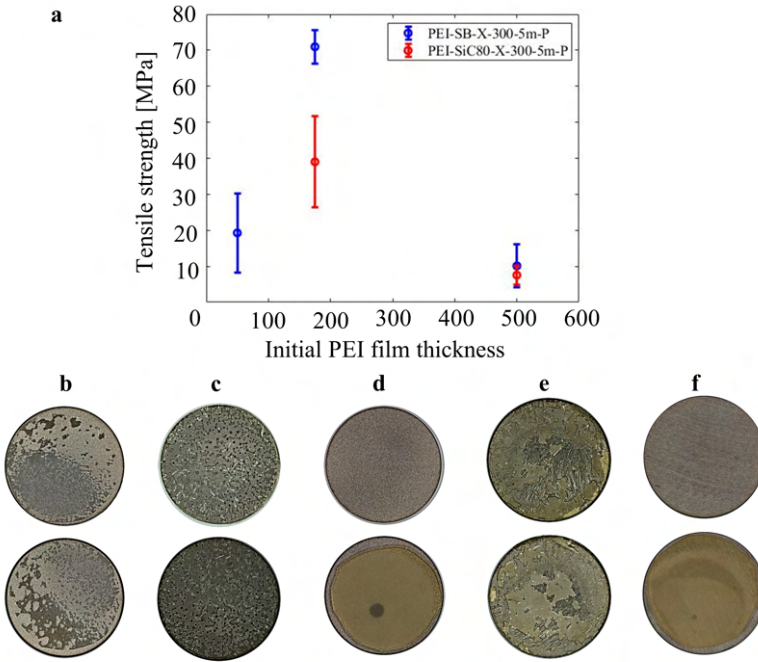


Figure A.13 a) Influence of initial film thickness for PEI-Ti welds sandblasted (blue) and polished with 80 SiC paper (red) and the fracture surfaces for the sandblasted studs welded with b) 50, c) 175 and d) 500 μm thick film. Fracture surfaces for the SiC 80 polished samples welded with a PEI film of e) 175 and f) 500 μm .

A.3.4 Influence of welding pressure

Pressure has been applied during the welding of the titanium studs through springs inserted in the mould cover. Adding the springs results in a pressure about 0.3 bar. The influence of the presence of the springs or not is shown in Figure A.14 for three different conditions. Adding the springs results in a stronger joint except for the polished samples. First, the probability to have larger voids increases by removing the springs, increasing

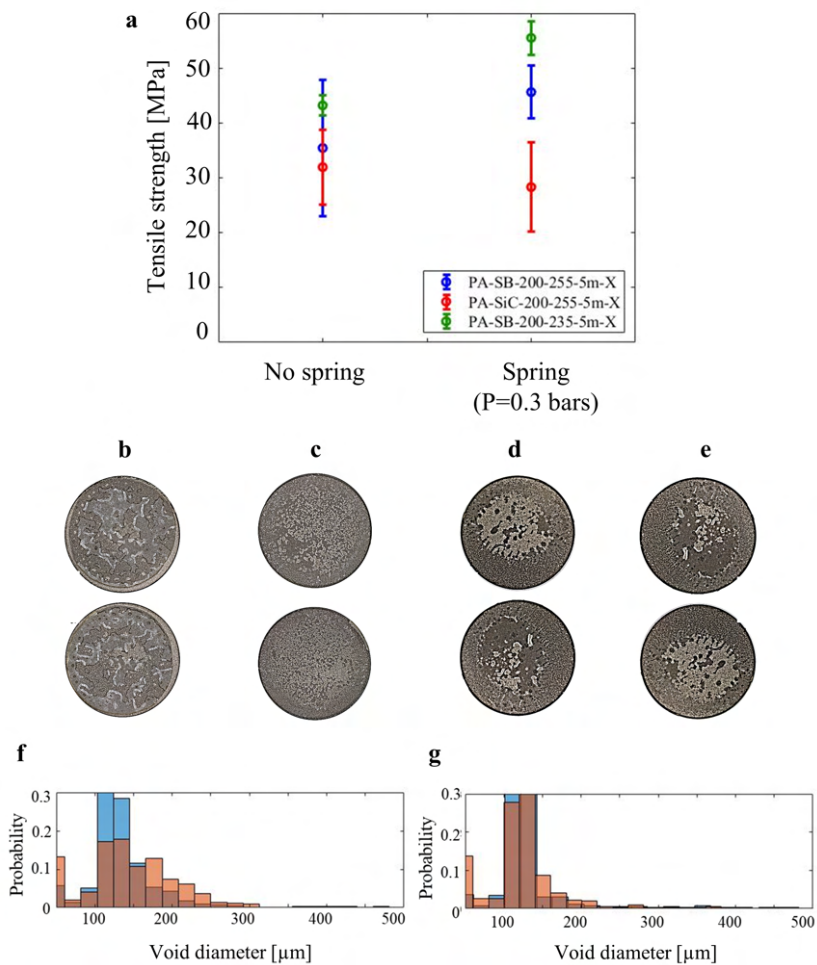


Figure A.14 a) Influence of the welding pressure for PA-Ti welds and the corresponding fracture surfaces for the sandbasted studs at b) 235°C without spring, c) 235°C with springs, d) at 255°C without spring and e) 255°C with springs. f-g) Probability distribution of the size of voids in the welds depending on the temperature (2 samples per temperature) for PA-SB-200-255-5m-X welds f) without spring and g) with springs.

the equivalent defect size that can result in a decrease in strength. In addition, surface wetting is favoured by pressure that helps the thermoplastic to penetrate into the surface asperities, increasing mechanical interlock-

ing and strength. At the opposite, increasing the pressure may reduce the thermoplastic thickness. Depending on the optimum thickness, this can be an advantage or not. To conclude, adding pressure should in most of the cases improves the joint tensile strength, in agreement with [258, 266]. The pressure must be high enough to have intimate contact between the two parts to join and it must be low enough to avoid squeezing out the thermoplastic from the weld.

A.3.5 Influence of remaining thermoplastic layer thickness

The surface roughness, temperature, pressure and initial film thickness all influence the thickness of the remaining thermoplastic layer between the titanium studs. Table A.1 give the remaining thickness for several conditions. Increasing the roughness increases the remaining thickness while increasing the temperature has the opposite effect. Roughness acts as a barrier to the spreading of the polymer while temperature favours its flow by lowering the viscosity. Increasing the pressure tends to squeeze the polymer out of the weld, resulting in a smaller thickness. Figure A.15 shows the evolution of the strength with the remaining thermoplastic thickness for both PA and PEI. For a thickness lower than 200 μm , the tensile strength tends to increase with the thickness while above it decreases. Increasing the thickness results in a lower degree of constraint in the bondline, lower stress triaxiality and more plastic deformation for elastoplastic materials. All these effects tend to increase the tensile strength. At larger thickness, the stress singularity at the joint edges arising due to material mismatch dominates the failure [150]. Consequently, adhesive failure takes place instead of cohesive failure reducing the strength. The influence of the thermoplastic thickness is in agreement with observations for adhesive joints [143, 150].

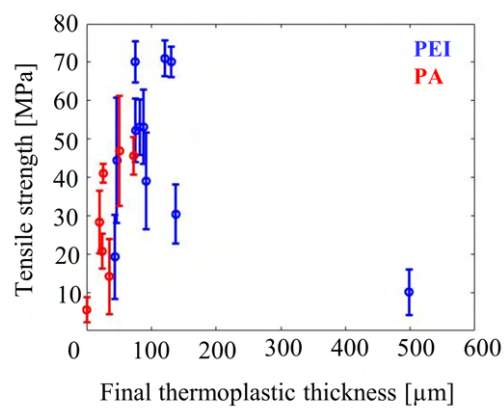


Figure A.15 Influence of the remaining thermoplastic thickness in the weld on the tensile strength for the PEI (blue) and PA (red) welds.

Table A.1 Remaining thermoplastic thickness for different welding conditions.

Sample	Thickness [μm]	Sample	Thickness [μm]
PEI-SB-175-300-5m-P	121 $\pm$ 18	PA-SB-200-255-5m-P	73 $\pm$ 28
PEI-BB-175-300-5m-P	138 $\pm$ 8	PA-BB-200-255-5m-P	51 $\pm$ 6
PEI-SiC80-175-300-5m-P	92 $\pm$ 11	PA-SiC80-200-255-5m-P	20 $\pm$ 9
PEI-SiC320-175-300-5m-P	82 $\pm$ 12	PA-SiC320-200-255-5m-P	35 $\pm$ 16
PEI-SiC800-175-300-5m-P	88 $\pm$ 10	PA-SiC800-200-255-5m-P	24 $\pm$ 7
PEI-SB-50-300-5m-P	44 $\pm$ 4	PA-SB-200-300-5m-P	26 $\pm$ 6
PEI-SB-500-300-5m-P	498 $\pm$ 4	PA-SiC80-200-300-5m-P	$\rightarrow$ 0
PEI-SiC80-500-300-5m-P	Broken		
PEI-SB-175-320-5m-P	131 $\pm$ 25		
PEI-SiC80-175-320-5m-P	76 $\pm$ 4		
PEI-SB-175-340-5m-P	75 $\pm$ 28		
PEI-SiC80-175-340-5m-P	47 $\pm$ 7		

A.3.6 Perspective: welding of larger parts

Several attempts have been made to weld squares of 50 by 50 mm using the hot press. The goal was to extrapolate the best conditions identified for butt tensile welds to the welding of larger parts. For this purpose, a 175 μm thick PEI film was inserted between two sandblasted steel plates spaced by steel wires of a diameter of 0.1 mm. The same is done with 500 μm thick films and 0.4 mm spacers. Welding time, temperature and pressure have been changed. The joints are then broken by hand. The fracture surfaces reveal many voids which would lead to joints with low performances as shown in Figure A.16. Several strategies have been investigated to circumvent this issue:

1. Inserting PEI pellets instead of a film in order to add a large quantity of PEI. In this case, PEI is squeezed out of the weld, but many voids are still found on the fracture surface.
2. Use of PEI powder leads to the same results as with PEI pellets.
3. A two step process: each steel plate is first covered with a thin PEI layer by inserting a 175 μm film between the steel plate and a teflon film. No voids could be observed on both covered plates after this step. Then, the two plates are welded together by adding a spacer and a PEI film. Joints are also made of many voids. To determine if the added film could be at the origin of these voids, no film has been added and the two plates have been directly welded together, resulting in a joint with less voids but with a thickness lower than 50 μm which would lead in low fracture toughness and strength.
4. Progressive increase of the pressure: the steel-PEI fil-steel assemblies are first inserted in the press at 300°C without pressure for 10 minutes to ensure melting of PEI. Then the pressure is increased to 1, 2, 3 and 5 bar every two minutes. As for the initial attempts, the joints are made of many voids.
5. Cutting the TP film into circles: As the adhesive deposition pattern can have an influence on the formation of voids, PEI films have been cut into circles as suggested by Kaufmann et al. [267].

Many strategies have been investigated to limit the number of voids in larger metal thermoplastic welds but without any success. It is assumed that the voids are coming from the air trapped between the steel plates

and the PEI film creating void growing during the welding process and that due to the high viscosity of PEI cannot escape as shown in Figure A.16. With pellets and powder, air is also trapped between the initial PEI particles. Other welding processes should be investigated such as induction or resistance welding. Ultrasonic welding is another alternative but it is only possible to weld small areas at a time require to perform several spot welds. The main limitation of these techniques is the availability of the equipment. Hence, another alternative be hot-pressing but by putting the mould under vacuum and progressively increasing the pressure and the temperature as done by Marinosci et al. [261] in an autoclave. These options have not been tried in the context of this thesis.

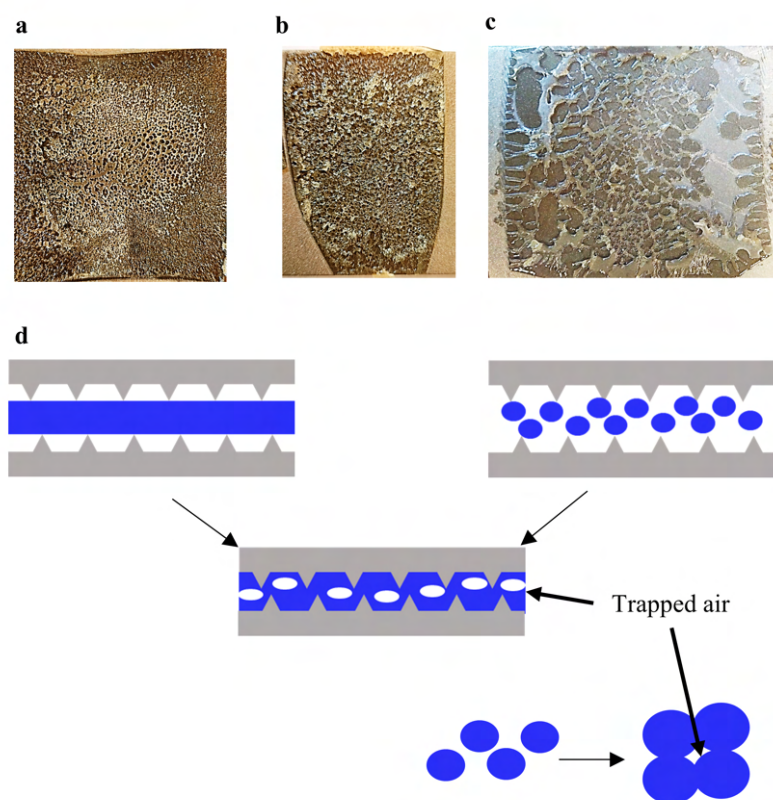


Figure A.16 Welding of large samples with a) a PEI film, b) PEI powder and c) PEI film. d) Scheme of air entrapment with a film and with powder/pellets.

A.4 Conclusion

To conclude, determining the optimum welding conditions for metal/T-P/metal joints is a complicated task as it involves a large number of parameters such as surface roughness, thermoplastic thickness, thermoplastic viscosity and welding temperature, welding time and welding pressure. Figure A.17 summarizes the parameters identified as entering into the determination of the best welding conditions. For PEI, the best conditions are a temperature between 300 and 340 °C, welding time of 5 minutes, pressure of 0.3 bar, initial film of 175µm and sandblasted titanium. For PA, the conditions are the same except that the initial film thickness is about 200 µm and that the welding temperature must be about 235°C. These conditions are the optimum for welding small surface. For larger surface areas to be joined, more research is required and probably another welding process should be investigated as hot-pressing results in a large number of voids in the bondline.

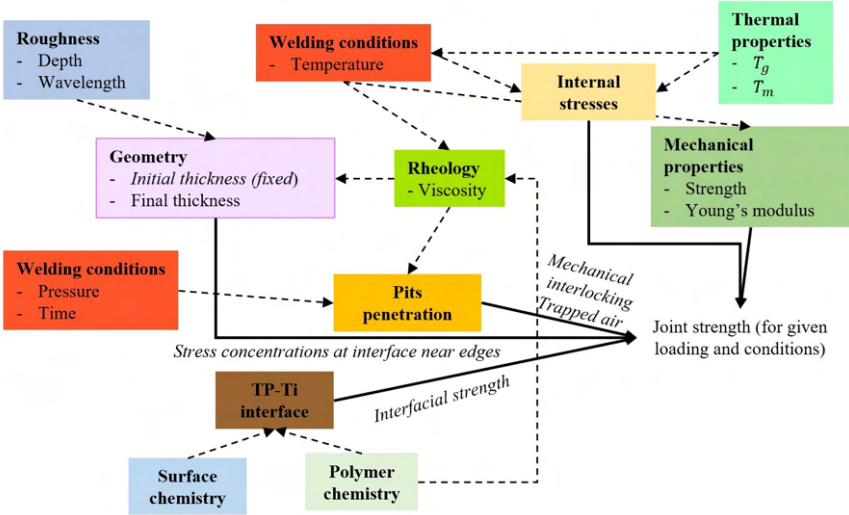


Figure A.17 Parameters influencing the joint strength.

A.5 Supplementary: Weibull distribution - probability of failure

The Weibull distribution is often used to characterized the strength distribution and the probability of failure. The probability density function is given by

$$f(x) = \frac{k}{\lambda} \left(\frac{x}{\lambda} \right)^{k-1} e^{-(x/\lambda)^k} \quad (\text{A.1})$$

x is the variable, in our case the remote tensile stress, k is the shape parameter also called Weibull modulus while λ is the scale parameter. Reliable welding conditions are characterized by a large Weibull modulus while a strong weld is characterized by a high λ value. These two parameters have been computed for all conditions and are given in Table A.2.

Table A.2 Weibull modulus and scale factor for the different welding conditions.

Condition	Weibull modulus	Scale factor
PEI-SG-175-300-5m-P	6.3	14.7
PEI-SG/Plasma-175-300-5m-P	not enough data	not enough data
PEI-SB-175-300-5m-P	22.1	73.8
PEI-BB-175-300-5m-P	5.3	34.7
PEI-SiC80-175-300-5m-P	7.3	46
PEI-SiC320-175-300-5m-P	11.6	57.3
PEI-SiC800-175-300-5m-P	8.7	58.8
PEI-SB-50-300-5m-P	2.2	23.8
PEI-SB-500-300-5m-P	2.1	12.4
PEI-SiC80-500-300-5m-P	16.7	9.1
PEI-SB-175-320-5m-P	47.9	72.4
PEI-SiC80-175-320-5m-P	8.2	57.1
PEI-SB-175-340-5m-P	21.5	73.3
PEI-SiC80-175-340-5m-P	4.9	53.4
PA-SG-200-235-5m-P	0.8	12.2
PA-SG/Plasma-200-235-5m-P	11.5	19.8
PA-SG-200-255-5m-P	7.5	36
PA-BB-200-255-5m-P	5.6	55
PA-SB-200-255-5m-P	13.8	48.6
PA-SiC80-200-255-5m-P	6.2	33
PA-SiC320-200-255-5m-P	1.9	17.7
PA-SiC800-200-255-5m-P	8.6	23.3
PA-SB-200-300-5m-P	17	42.5
PA-SiC80-200-300-5m-P	2.3	6.9
PA-SB-200-255-5m-NP	3.8	41.9
PA-SiC80-200-255-5m-NP	14.5	35.6
PA-SB-200-235-5m-P	27.8	57.3
PA-SB-200-235-5m-NP	45.1	44.4
PA-SB-200-245-5m-P	3	45.7

List of publications

★ List of publications in journal

C. van Innis, M.K. Budzik and T. Pardoen, *Ultra-tough architected adhesive joints for integrated composite processing and bonding*, Composites Part A: Applied Science and Manufacturing, 177, 2024 [268].

C. van Innis, T. Pardoen, *Thickness-dependent selection of adhesive joints*, The Journal of Adhesion, 110 (12), 2024 [101].

C. van Innis, M.K. Budzik and T. Pardoen, *Bridging in co-cured composite joints*, to be submitted to the International Journal of Solids and Structures.

★ Patent submission

C. van Innis, T. Pardoen, *Film for joining parts*, patent submission EP23161330.8.

★ Communications in conferences

[Oral] C. van Innis, M.K. Budzik, T. Pardoen, *Ultra tough architected joints through single manufacturing step*, 9th International Conference on Fracture of Polymers, Composites and Adhesives (Les Diablerets, Switzerland, 03/24/2024 - 03/27/2024).

[Oral] C. van Innis, M.K. Budzik, T. Pardoen, *Ultra tough architected joints through single step bonding process with tunable properties*, AB2023 7th International Conference on Adhesive Bonding 2023 (Porto, Portugal, 07/13/2023 - 07/14/2023).

[Oral] C. van Innis, M.K. Budzik, T. Pardoen, *Fracture toughness of architected joints involving crack instabilities*, CECAM 3D cracks and crack instability (Lausanne, Suisse, du 06/14/2023 - 06/16/2023).

[Poster] C. van Innis, T. Pardoen, *Mechanics of PEI-expoxy interfaces*, 3rd Benelux Network Meeting & Workshop on Damage & Fracture Mechanics (BDFM-2023) (Antwerp, Belgium, 05/25/2023).

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[Poster] C. van Innis, W. Ballout, C. Bailly, T. Pardoen, *Bonding polymer Composites with PEI film: crack trapping and enhanced fracture resistance*, Les Assemblages mécaniques – Evolutions récentes et perspectives (Saint-Ouen, France, 07/01/2021 - 07/02/2021).

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