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Self-stratifying epoxy vitrimer/fluoropolymer coatings: Thermally induced detachment for substrate recovery

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ABSTRACT

In this study, we propose an innovative approach to improve the sustainability of polyvinylidene fluoride (PVDF) coatings by incorporating an epoxy vitrimer primer. The dynamic nature of the vitrimer allows for controlled coating removal under specific conditions, facilitating end-of-life management and recyclability. The self-stratifying coating, developed through a one-step application and curing process, consists of an epoxy vitrimer bottom layer and a PVDF top layer. This system was designed to spontaneously form two distinct layers during curing. Two vitrimer resins containing disulfide dynamic bonds were evaluated as base layers: polysulfide epoxy (EPS35) and diglycidyl ether of vanillyl alcohol (DGEVA).

The epoxy/fluorinated systems were applied on different substrates (carbon coated aluminum or copper foils) and at different thicknesses (from 25 µm to 230 µm) and then characterized. All systems present two distinct coating layers with the epoxy on the substrate and the PVDF on top. Cross hatch cutter test results confirmed the perfect adhesion of the coating at room temperature. The epoxy layers self-healing was proved, highlighting the achievement of a dynamic network. Finally, the coating films were removed easily from the substrate under thermal stimuli of at least 100 °C.

Finally, the influence of key parameters on stratification, applied wet thickness, substrate and epoxy formulation was evaluated. In all cases, a well-defined stratification (type I), perfect adhesion at room temperature (type "0") and a total removal at 100 °C was consistently observed.

1. Introduction

Fluoropolymers are largely employed in industrial applications due to important chemical resistance, low surface energy, and electrochemical stability [1–3]. These properties make them particularly valuable in fields such as energy storage, corrosion protection, and membrane technology. However, despite their outstanding performances, fluoropolymers belong to the class of per- and polyfluoroalkyl substances (PFAS) [4], which persist in the environment and present potential health risks. Growing regulatory restrictions on PFAS have led to increased efforts to find sustainable alternatives or improve their end-of-life management. While replacing fluoropolymers is a long-term goal, improving the recyclability of coatings is a more practical short-term solution to reduce environmental impact.

One of the key limitations of fluoropolymer-based coatings is poor adhesion [5] to substrates, requiring a primer layer to ensure durability and mechanical stability, and thus requiring two formulation, application and curing steps. This limitation has driven interest in self-stratifying coatings, which enable the formation of a structured bi-layered system, with excellent interlayer adhesion, from a single application and curing step. Self-stratification not only simplifies the coating process but also reduces solvent usage, processing time, and energy consumption, in line with the increasing focus on sustainable manufacturing [6,7].

Beyond adhesion, recyclability of crosslinked coatings remains a major challenge. While epoxy primers provide strong adhesion, permanent crosslinked structures make separation from substrates at end-of-life nearly impossible without extensive chemical treatments [8]. Vitrimers, a class of covalently crosslinked polymers with dynamic

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exchangeable bonds, offer a promising solution. Unlike traditional thermosets, vitrimers maintain network integrity while allowing bond rearrangement under stimuli, enabling controlled delamination from substrates and potential recyclability [9,10]. Thus, replacing conventional thermoset primers with vitrimers could address recycling challenges of crosslinked coatings without compromising functional properties. While self-stratified bi-layers have been extensively studied with formulation of a thermoplastic and conventional thermosets [11,12], their realization using vitrimers has been explored in only one study [13], adhesion being ensured by a conventional thermoplastic epoxy as base layer, but with a vitrimer polydimethyl siloxane resin as the top layer for hydrophobic properties.

In this work, the objective is to design a self-stratifying coating system based on an epoxy vitrimer as base layer and PVDF as top layer. This approach aims to combine strong adhesion at room temperature, simplified processing, and improved end-of-life management through high temperature-induced coating removal, contributing to a more sustainable coating technology.

In this work, epoxy vitrimer resins were synthesized as described in studies of *Canadell* et al. and *Guggari* et al. [14,15], using a 1:1 epoxy/hardener ratio based on their epoxy equivalent weight (EEW), and in case of the EPS35 formulations, an addition of 1%$_{wt}$ catalyst. These vitrimers were incorporated as primer layers in self-stratifying coating systems to ensure adhesion and recyclability.

The prepared epoxy/PVDF coatings were applied on different substrates and the influence of key parameters for self-stratification to occur such as applied wet thickness, substrate type, and epoxy formulation was investigated. In this system, adhesion properties were assessed using cross-hatch cutter tests and recyclability of the system was evaluated by applying a thermal stimuli.

2. Materials and methods

2.1. Materials

In this study, three commercials resins were used. The aliphatic epoxy-terminated polysulfide polymer Thioplast EPS35 (EPS35, blend of ≈ 90% of liquid polysulphide polymer with epoxy end groups and ≈ 10% of 1.4-bis(2,3-epoxypropoxy)butane, Fig. 1.a., EEW of 700–800 g/eq) was graciously supplied by Nouryon Chemicals (Mons, Belgium). Diglycidyl ether of vanillyl alcohol (DGEVA, Fig. 1.b., EEW of 139 g/eq,) was bought from Specific Polymer (Castries, France). Fluoroethylene vinyl ether resin Lumiflon LF200 (Lumiflon, Fig. 1.f.) was obtained from AGC Chemicals (Amsterdam, Netherlands). EPS35 was crosslinked by pentaerythritol tetrakis(3-mercaptopropionate) (PTMP) (Fig. 1.c.) and with 4-diméthylaminopyridine (DMAP) (Fig. 1.e.) as catalyst both purchased from Sigma-Aldrich (St. Louis, USA). DGEVA was crosslinked by cystamine dihydrochloride and purified following the procedure by *Khalafi* et al. [16] (Fig. 1.d.). Solvents used are Ortho-Xylene (O-xylene, 99%, $T_{Boiling}$ = 144 °C) and Methyl IsoButyl Ketone (MIBK, ⩾ 99%, $T_{Boiling}$ = 116 °C) that were purchased from Sigma-Aldrich (St. Louis, USA) and were used as received.

All of chemicals are illustrated on Fig. 1.

Coatings were applied either on carbon coated aluminum foils of 12 µm thickness (with carbon metallization of 0.5 to 2.0 g m^{-2}, notified as Al-C) or on copper foil of 12 µm thickness. Both substrates were supplied by Xiamen Tob new energy technology CO., LTD. Among the various epoxy vitrimer systems reported in the literature, two commercial resins based on disulfide bonds were selected, due to their potential low vitrimer transition temperature (T_v) : one oil-based and one bio-based. This selection was driven by the need of vitrimer compatible with coating formulations while ensuring processability, strong adhesion, and potential recyclability. The first epoxy vitrimer was synthetized using EPS35, an oil-based epoxy monomer, pentaerythritol tetrakis(3-mercaptopropionate) (PTMP) as crosslinker and 4-(dimethylamino)pyridine (DMAP) as catalyst, as reported by *Canadell* et al. In this system, *Canadell* et al. observed a glass transition temperature (T_g) of −35 °C and a T_v of 100 °C. Furthermore, this system was cured at 60 °C for 2 h [15]. The second system consisted of DGEVA and cystamine, based on the work of *Guggari* et al. For this system, cured at 90 °C for 2 h, a T_g of 53 °C was noted, along with a reprocessability temperature of 150 °C at 1 metric ton [14].

Table 1
Different formulations investigated.

	SS_E_Al-C	SS_D_Al-C	SS_E_Cu	SS_E_Al-C_230
Epoxy	EPS35	DGEVA	EPS35	EPS35
Substrate	Al-C	Al-C	Cu	Al-C
Thickness (µm)	25 ± 1	34 ± 4	36 ± 3	230 ± 7

2.2. Coatings' preparation

Self-stratifying systems were prepared as described hereafter, this coating preparation is adapted from *Beaugendre* et al. [12,17].

3 g of EPS35 were dissolved in a first beaker containing 3.5 g of o-xylene and 3.5 g of MIBK. In a second beaker 5 g of Lumiflon LF200 were dissolved in a blend of 1.5 g of o-xylene and 3.5 g of MIBK (Lumiflon LF200 is composed of 60%$_{wt}$ of PVDF and 40%$_{wt}$ of o-xylene). Both mixtures were magnetically stirred until homogenization at 300 rpm. They were then mixed together at a 1:1 mass ratio and stirred until homogenization at 300 rpm. Catalyst and crosslinker were added one by one and mixed till obtaining an homogeneous mixture. Depending on the dried thickness expected, two application methods were used. In the first case, solutions were sprayed on the chosen substrates using a spraying gun FPRO G from Sames Kremlin (2.5 bar of N_2 pressure). In this case, an average wet thickness of 150 µm was targeted, that leads to relatively thin dry coatings (20 to 50 µm). In an attempt to increase the dry thickness, the solutions were also poured on the substrate (without control of the thickness). After application, the coated samples were let at room temperature for 15 min before being placed in a ventilated oven to simultaneously evaporate the solvents and cure the epoxy resins. EPS35/PTMP samples were cured at 60 °C for 2 h whereas DGEVA/cystamine samples required a curing at 90 °C for 2 h.

2.3. Formulations

Four different formulations were prepared (Table 1), in order to study the impact of the epoxy used (EPS35 — named (E) or DGEVA — named (D)), type of substrate (Al-C or copper) and the dry-thickness on the stratification process. The sample thickness was characterized using a Mitutoyo Digimatic IP65 micrometer. The average and standard deviations were calculated based on a data set of ten points mapped across the sample.

The SS_E_Al-C system, used as a reference, was obtained by spraying a Lumiflon/EPS35 formulation onto Al-C foil, achieving a dry thickness of 25 µm. To assess the impact of vitrimers natures on stratification, EPS35 was replaced by DGEVA in SS_D_Al-C, still applied on the Al-C substrate and achieving a 34 µm thickness. The SS_E_Cu formulation allowed to evaluate the substrate influence by applying the formulation onto copper foil at a 36 µm thickness. Finally, SS_E_Al-C_230 explored the maximum achievable coating thickness, reaching 230 µm on Al-C.

2.4. Discs for self-healing characterization

In a vial, 3 g of EPS35, 0.52 g of PTMP and 0.03 g of DMAP were magnetically stirred together at 300 rpm until homogenization. Mixture was poured on a disc mold of 3 mm of thickness and 3 cm of diameter and cured at 60 °C for 2 h. Surfaces were cross cut with razor blade.

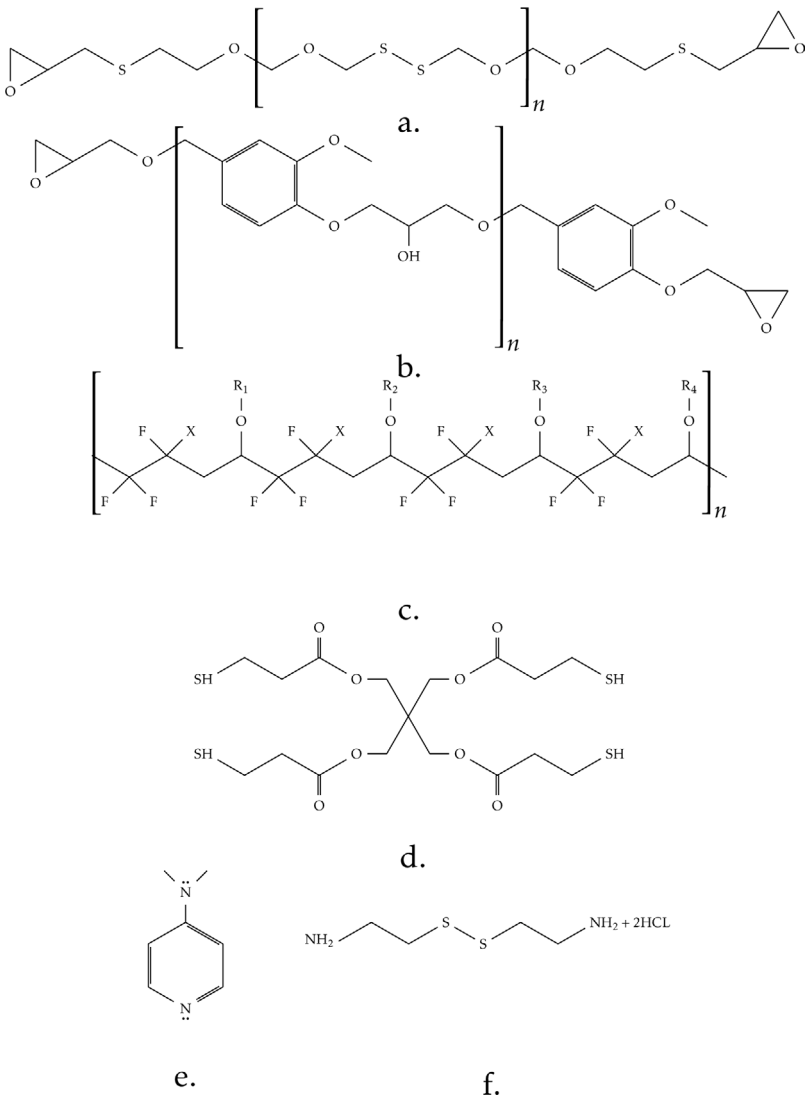


Fig. 1. Chemical structure of reactants used in this study : a. EPS35, b. DGEVA, c. Lumiflon LF200, d. PTMP, e. DMAP, f. Cystamine dihydrochloride.

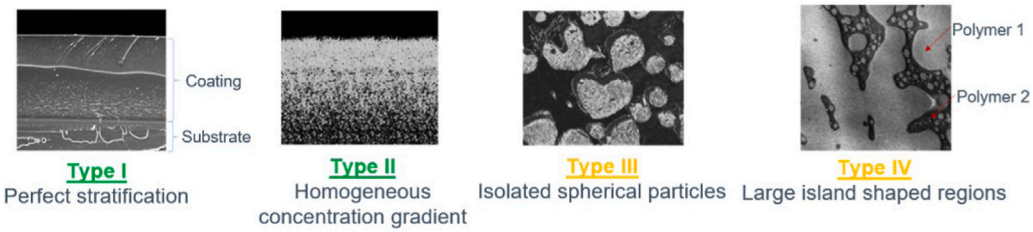


Fig. 2. The four main types of stratification [12].

2.5. Characterization methods

Scanning Electron Microscope (SEM) analyses coupled with elemental analyses (EDX) were carried out to characterize the degree of stratification achieved after curing. To characterize the distribution of the two polymers within the coating film, the characteristic elements of the polymers were chosen. The level of stratification was then described using the classification established by *Toussaint* et al., who categorizes stratification levels from type I to type IV, as presented in Fig. 2 [18].

The elements present in our system are H, C, O, F, Al, S and Cu. Since the detection window is made of beryllium, the energy of the hydrogen Kα line is too low to be visible. Carbon and oxygen are present in both polymers, making their mapping irrelevant. Therefore, the atoms used as probes in our systems were aluminum or copper to locate the substrate, sulfur to trace the presence of the epoxy resin, and fluorine to identify the PVDF resin. If type I stratification occurs, sulfur and fluorine should be detected in two different layers. It is expected to map the fluorine in the top layer and the sulfur in the bottom one.

The adhesion of the coatings was evaluated at room temperature using the cross-hatch cutter test (ISO2409:2020) that enables to classify the adhesion from 5 (poor adhesion) to 0 (perfect adhesion) depending on the percentage of coating removed during the test, as depicted in Fig. 3.

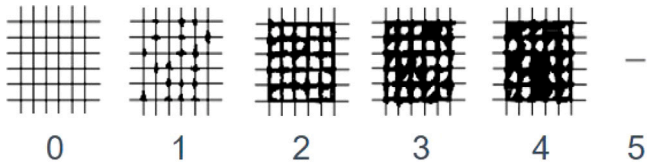


Fig. 3. Cross hatch cutter test type results, classification (from best to worse adhesion) (ISO2409:2020).

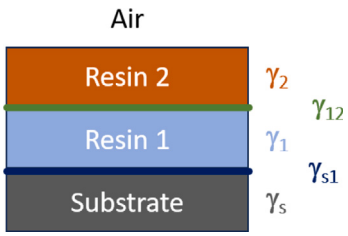


Fig. 4. Interface and surface free energy associated to a bilayer coating.

Removal tests were conducted to evaluate the ability to remove the coating when heated. Above T_v, the vitrimer epoxy gains malleability and should therefore be removable from the substrate, whether Al-C or copper. According to the state of the art, the T_v of the EPS35/PTMP and DGEVA/Cystamine systems is approximately 100 °C. Removal tests, performed simply using a spatula, were carried out above 100 °C. Microscopic analyses using an optical microscope (Keyence) were conducted on the samples to observe the thickness evolution after removal.

Surface Free Energy (SFE) Experimentally, surface tensions of the resins were obtained using the sessile drop method performed with a Drop Shape Analyzer (DSA100) from Krüss GmbH (Hamburg, Germany). Each test was performed on discs, prepared with the same protocol one as the discs prepared for self-healing tests. Contact angles formed by different solvents at the surface of each resin were determined using the "Advance" software provided by Krüss. Three solvents with different polarities were used to calculate the surface energy: distilled water (γ_{tot} = 72.8; γ_d = 21.8; γ_p = 51 mN m^{-1}), diiodomethane (γ_{tot} = 50.8; γ_d = 50.8; γ_p = 0 mN m^{-1}) and formamide (γ_{tot} = 58.2; γ_d = 39.5; γ_{tot} = 18.7 mN m^{-1}). Wu method (ASTM D 2578–67) was used to determine the total surface energy γ_{tot} as well as the dispersive γ_d and polar γ_p components [19]. This method is widely used for the calculation of the surface energy of polymers with low surface energy (below 40 mN m^{-1}).

3. Results and discussion

Before presenting the designed self-stratified resins, it is important to check if already published model can succeed in predicting the stratification.

3.1. Theoretical aspect

Based on the literature, stratification could be predicted by using a model based on surface free energy. *Funk* et al. demonstrated that surface free energy difference between the two resins is the primary force driving stratification [7].

As shown in Fig. 4, different surface energies have to be taken in account, i.e. γ_s, γ_1 and γ_2 that correspond to the total surface free energy of the substrate, of the base layer and of the stratifying layer, respectively. γ_{s1} and γ_{12} are the interfacial surface energies that are calculated following Harmonic mean method of Wu described in (Eq. 1), with γ^d and γ^p the disperse and polar surface free energies, respectively.

$$\gamma_{12} = \gamma_1 + \gamma_2 - \frac{4\gamma_1^d\gamma_2^d}{\gamma_1^d + \gamma_2^d} - \frac{4\gamma_1^p\gamma_2^p}{\gamma_1^p + \gamma_2^p} \quad \text{(Eq. 1)}$$

Table 2
Dispersive (γ_d), polar (γ_p) and total (γ_{tot}) SFEs of crosslinked epoxy resins, thermoplastic fluoropolymer and substrates used in next self-stratifying systems.

Components		γ_d (mN m^{-1})	γ_p (mN m^{-1})	γ_{tot} (mN m^{-1})
Substrate	Al-C	35 ± 1.9	13 ± 1.7	48 ± 3.6
	Copper	38 ± 1.0	1.5 ± 0.5	39 ± 1.5
Resins	Lumiflon LF200	27 ± 0.3	8.0 ± 0.3	35 ± 0.6
	EPS35	18 ± 1.8	7.0 ± 1.0	25 ± 2.8
	DGEVA	26 ± 2.2	0.08 ± 0.2	26 ± 2.4

Table 3
Interfacial surface free energy calculated following (Eq. 1).

Interfaces	Interfacial surface tensions (mN m^{-1})
Al-C/EPS35	6.8 ± 2.0
Cu/EPS35	10.6 ± 2.3
Al-C/DGEVA	13.9 ± 5.1
EPS35/Lumiflon	1.6 ± 0.6
DGEVA/Lumiflon	7.6 ± 0.9

Table 4
Calculated predict stratification equations.

	(Eq. 2)	(Eq. 3)	(Eq. 4)	Theory
SS_E_Al-C	3 ± 2.3	13 ± 3.0	18 ± 2.9	✓
SS_D_Al-C	10 ± 5.3	20 ± 5.7	12 ± 5.7	✓
SS_E_Cu	3 ± 2.5	16 ± 3.1	6 ± 3.2	✓

To predict stratification, the three following conditions must be fulfilled [12]:

$$\gamma_{s1} - \gamma_{s2} - \gamma_{12} \geqslant 0 \quad \text{(Eq. 2)}$$

$$\gamma_{s1} - \gamma_1 - \gamma_{s2} + \gamma_2 > 0 \quad \text{(Eq. 3)}$$

$$\gamma_s - \gamma_{s2} - \gamma_{12} - \gamma_1 > 0 \quad \text{(Eq. 4)}$$

In case of the systems tested in this paper, Surface Free Energies (SFEs) are summarized in the following Table 2.

To solve (Eq. 2) to 4, it is necessary to calculate the interfacial surface energies. These were calculated using (Eq. 1) and are listed in Table 3.

For the three systems tested, the three conditions are met (Table 4) indicating that stratification should occur. As validated by the theory, those systems were prepared and characterized.

3.2. Reference system (SS_E_Al-C)

The system SS_E_Al-C was characterized using SEM-EDX, as shown in Fig. 5. It reveals three distinct layers. The bottom layer, in orange, represents the Al-C substrate. The 12 µm thickness of the Al-C foil aligns with the value provided by the supplier. The second layer, in violet, maps sulfur attributed to the epoxy layer, with a thickness around 15 µm. The top layer, in red, corresponds to the fluorine from the PVDF layer, with an approximate thickness of 10 µm. These mappings of the reference system highlight consequently a type I stratification as defined by *Toussaint* et al.

Cross-hatch cutter tests were conducted to evaluate the adhesion of the bilayer to the aluminum substrate at room temperature. The sample after testing is shown in Fig. 6. A close-up of the cut, presented in Fig. 6.b, reveals a cut depth of 26 µm. Considering the total thickness of the coating, the cut extends throughout the entire coating up to the substrate. This demonstrates perfect adhesion between the coating and the Al-C foil, as no detachment from the substrate is observed, corresponding to the highest adhesion score, "0".

Considering that the adhesion is perfect, tests for the removal of the coating were conducted at different temperatures between 80 °C

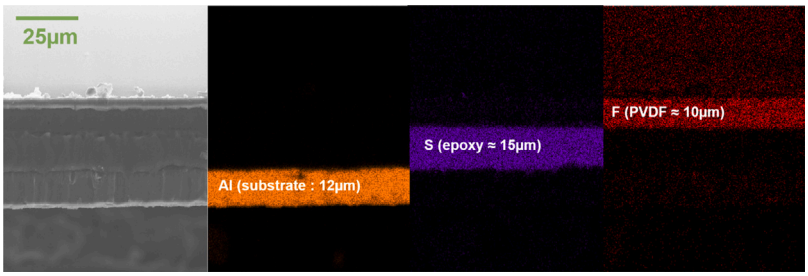


Fig. 5. Cross-section SEM-EDX analyses of the SS_E_Al-C system with Al-C as substrate, EPS35 as epoxy and Lumiflon LF200 as PVDF layers.

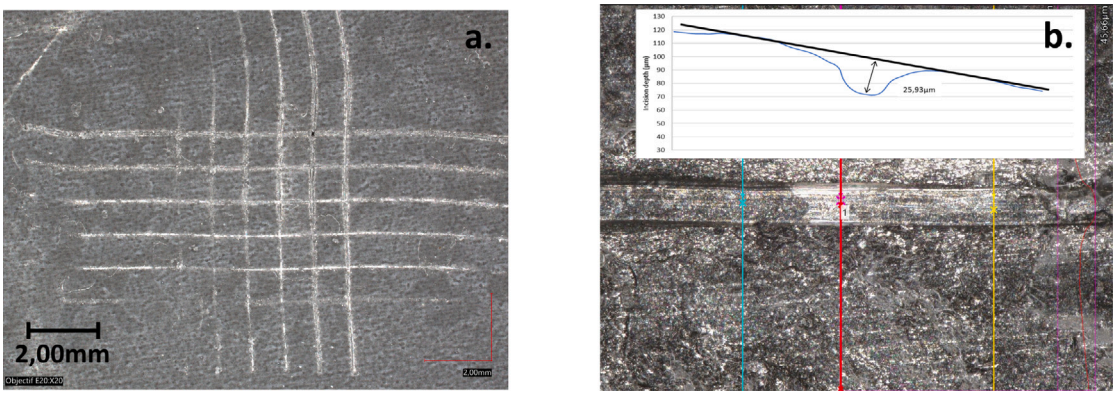
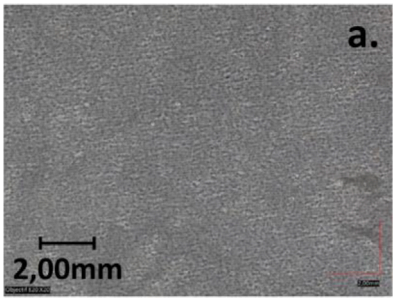
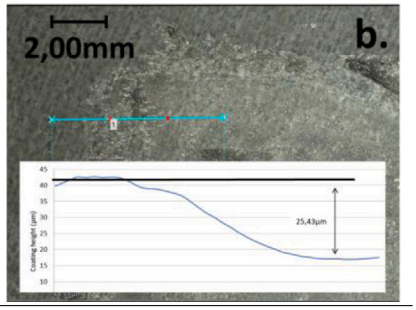
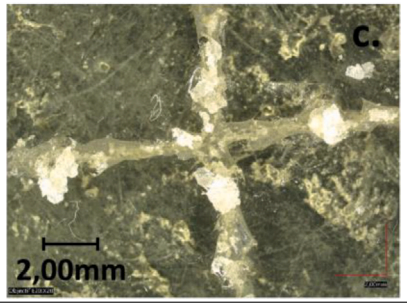
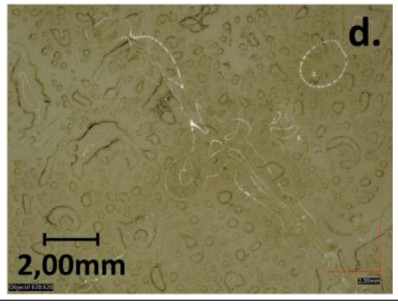


Fig. 6. a.: Cross hatch cutter test on system SS_E_Al-C. b.: Focus on the cut and cutting profile to highlight the depth of the cut, 26 µm.

Table 5
Properties linked to the vitrimer resin: a. Cross cut on a disc of EPS35/PTMP cured as self-stratifying systems of 3 mm of thickness. b. Same cut after 12 h at 100 °C. c. Virgin SS_E_Al-C system. d. Removal of the coating heated up to 100 °C.

	Before test	After test
Removal test Heated up to 100 °C		
Healing test 100 °C, 12 h		

and 140 °C. It appears that both coating layers can be removed in the 100-140 °C temperature range, which is in agreement with our hypotheses. Since the T_v is at 100 °C, the exchange reactions occur, making the epoxy malleable and thus enabling a clean detachment.

Coating removal at 100 °C is illustrated in the first row of Table 5. Images, observed under an optical microscope, before (Table 5.a.) and after (Table 5.b.) removal clearly demonstrate the efficiency of the process at 100 °C. An evolution of thickness is measured with a displacement on the blue line that goes from a zone with the untouched coating to a zone were the removal test has been performed (Table 5.b.). A drop of 25 µm, a value within the standard deviation of the coating thickness, is observed in the tested zone, indicating the complete removal of the coating.

The second row of Table 5 shows the results of the self-healing test. The optical microscope picture on the left (5.c.) shows the scratch made on one of the discs, while the picture on the right (5.d.) shows the sample after 12 h at 100 °C. The scratch is no more visible, demonstrating the complete healing after 12 h.

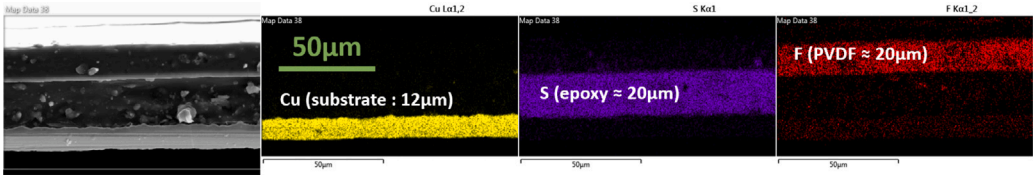


Fig. 7. Cross section SEM-EDX of the system SS_E_Cu.

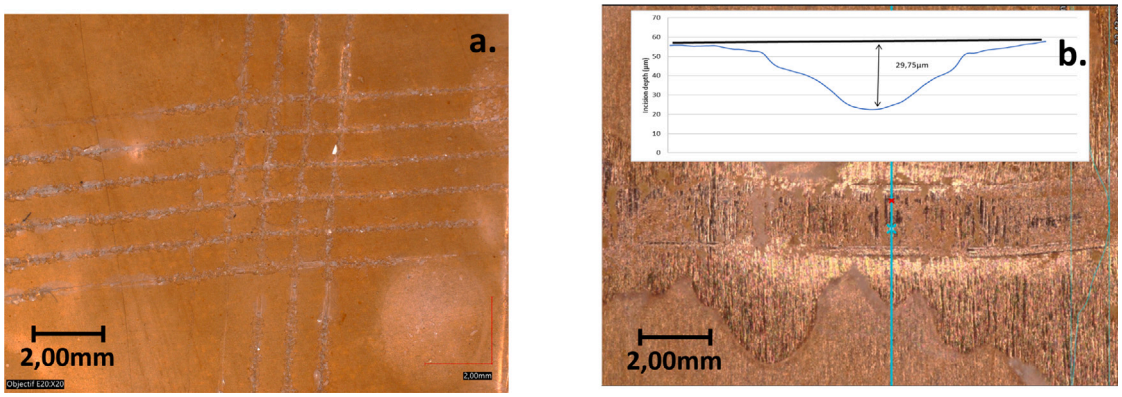


Fig. 8. a.: Cross hatch cutter test on SS_E_Cu system. b.: Focus on the cut and cutting profile to highlight the depth of the cut, 30 µm.

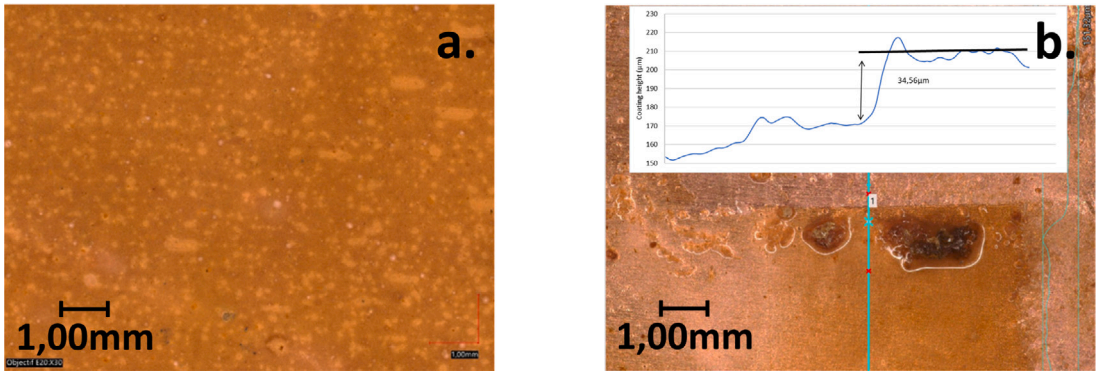


Fig. 9. a. Virgin SS_E_Cu system. b. Removal of the coating after a heating up to 100 °C and profile highlighting a removal of 35 µm.

These findings further support the proposed self-stratifying behavior and the reversibility of the vitrimer network under thermal activation.

Obtained results show that it was possible to design a type I self-stratifying coating made from a base layer of a vitrimer epoxy resin and a top layer of PVDF, showing excellent adhesion to carbon-coated aluminum foils and removable at 100 °C. The influence of the substrate was then studied and Al-C was replaced with a copper foil and is presented in the next part.

3.3. Influence of the substrate (SS_E_Cu)

For a coating to stratify, one of the most important parameters is the surface tension of the substrate. A change in substrate surface tension could thus induce a different stratification mechanism or completely prevent stratification to occur. Hereafter are presented the results of the application of SS_E_Cu coating on copper. The SEM-EDX images of the coating (Fig. 7), reveal a two-layers coating on the copper substrate.

As in the reference system, the bottom layer contains sulfur (violet) and is consequently identified as the epoxy resin, while the top layer (red) corresponds to the fluorine from the PVDF resin. A clear type I stratification degree is thus obtained.

Fig. 8.a. illustrates the cross-hatch cutter test performed on the SS_E_Cu system, revealing no detachment of the coating and confirming excellent adhesion. A closer examination of the cut in Fig. 8.b., along with the cutting profile, indicates a cut depth of 30 µm throughout the total thickness of the system. This confirms that the incision fully penetrates the coating without compromising adhesion. The highest adhesion score of "0" is consequently achieved.

The thermally triggered removability was then evaluated between 80 °C and 140 °C. Fig. 9 shows the test carried out at 100 °C. Delamination was observed starting from 100 °C, indicating that like the SS_E_Al-C system, the vitrimer's dynamic bond exchange is activated above its T_v, enabling again the detachment of the coating.

The substrate difference showing no impact, the influence of dry-thickness of the coating was investigated.

3.4. Influence of the coating thickness, (SS_E_Al-C_230)

A thick sample was obtained by directly pouring the coating on the substrate. Fig. 10 shows a cross-section SEM-EDX mapping of this coating. It shows a perfect Type I stratification, with a final thickness of 230 µm. This experiment shows that the system stratifies, whatever its thickness.

Finally, an alternative partially bio-based epoxy vitrimer (DGEVA), was tested instead of the fully oil-based EPS35.

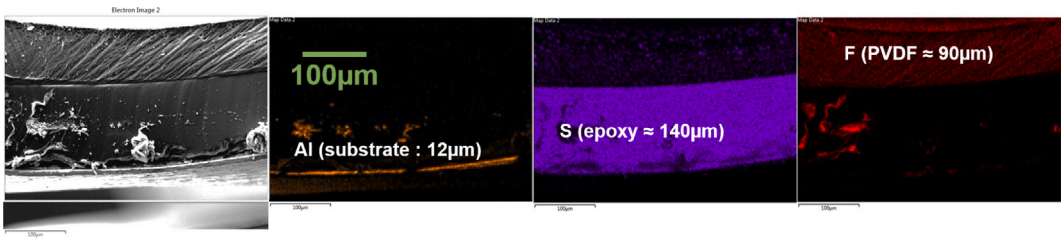


Fig. 10. Self-stratifying system SS_E_Al-C_230.

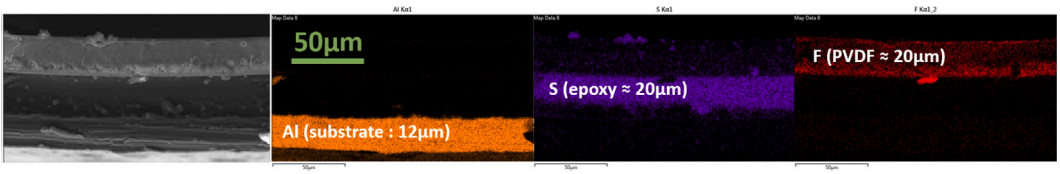


Fig. 11. Self-stratifying system SS_D_Al-C.

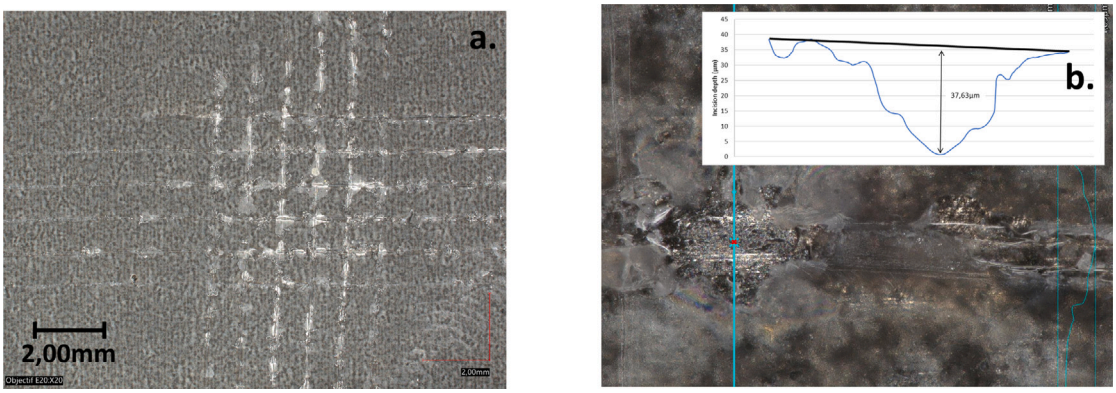


Fig. 12. a.: Cross hatch cutter test on SS_D_Al-C system. b.: Focus on the cut and cutting profile to highlight the depth of the cut, 38 µm.

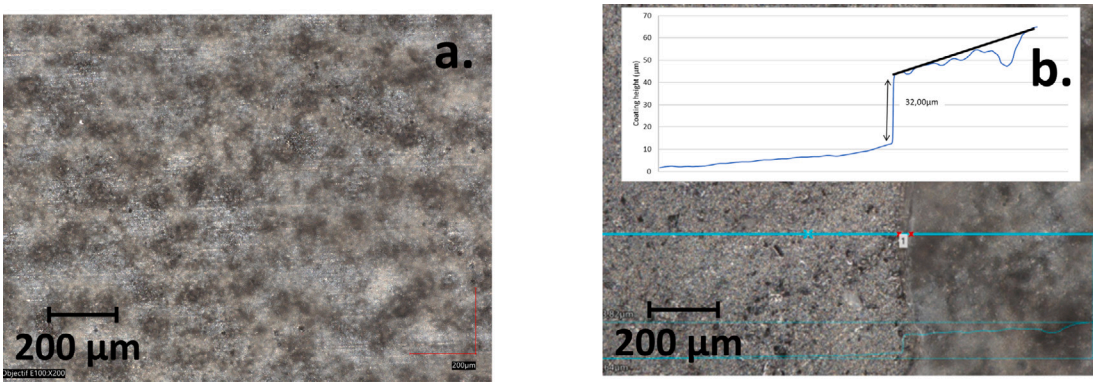


Fig. 13. a. Virgin SS_D_Al-C system. b. Removal of the coating after a heating up to 100 °C and profile highlighting a gap of 32 µm.

3.5. DGEVA based coating, (SS_D_Al-C)

DGEVA-containing self-stratifying coatings were made following the same protocol than that used with EPS35. DGEVA was crosslinked with cystamine, that contains sulfur atoms. Sulfur was thus mapped by SEM-EDX to identify the epoxy vitrimer. Fig. 11 shows the cross section of a self-stratifying coating containing DGEVA and PVDF, applied on carbon-coated aluminum.

This system shows a clear type I stratification, with balanced thicknesses (around 20 µm for each layer). As for the SS_E_Al-C system, adhesion cross-hatch cutter test was carried out and also shows the best adhesion rate "0" at room temperature (see Fig. 12).

Removal tests were then carried out. *Guggari* et al. highlight a reprocessability of the DGEVA when it is hot pressed at 100 °C under 1 metric ton of pressure [14]. In this paper, coatings were heated up to 100 °C. In Fig. 13.b, the displacement of the blue line highlights the

coating thickness. It indicates a total thickness of 38 µm, within the standard deviation, which decreases to zero, demonstrating successful removal down to the substrate at this temperature.

4. Conclusions

Novel self-stratifying systems containing a vitrimer base resin (DGEVA or EPS35) together with PVDF have been successfully developed and characterized. Those system consist in a dual-layer structure, the base layer being a self-healable vitrimer epoxy resin promoting adhesion and allowing the removal of the whole coating from the substrate at 100 °C. In addition, the self-stratifying epoxy vitrimer/PVDF systems, exhibit a highly effective stratification, classified as Type I, over a wide range of thicknesses from 25 µm to 230 µm.

Cross-hatch cutter tests confirmed excellent adhesion of all systems (adhesion score "0") at room temperature. The vitrimer properties of EPS35 or DGEVA allow the entire self-stratifying system to be removed from the substrate with ease at 100 °C, whether with carbon-coated aluminum or copper foil as substrate. This combination of properties offers significant potential for applications requiring durable, reusable and recyclable coatings with strong adhesion and self-healing characteristics.

CRediT authorship contribution statement

Alban Kerrache: Investigation, Data curation, Writing – original draft, Formal analysis, Methodology. **Clémentine Roche:** Formal analysis, Data curation. **Jean-François Gohy:** Writing – review & editing, Resources, Project administration, Funding acquisition, Conceptualization, Supervision, Methodology. **Fabienne Samyn:** Writing – review & editing, Methodology, Conceptualization, Supervision, Investigation. **Maude Jimenez:** Supervision, Methodology, Writing – review & editing, Resources, Conceptualization, Validation, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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