



Process modeling and simulation

ECCM19-ABS-433

Vacuum infusion of thick glass-fibre reinforced methacrylic composites: computationally efficient modelling of temperature profiles and kinetics during in-situ polymerization

Sarah F. Gayot¹, Frederik Van Loock¹, Pierre Gérard², Thomas Pardoën¹, Christian Bailly³

¹Institute of Mechanics, Materials, and Civil Engineering (iMMC), Université Catholique de Louvain, Louvain-la-Neuve, Belgium, ²Groupe de Recherche de Lacq, Arkema, Lacq, France, ³Institute of Condensed Matter and Nanosciences (iMCN), Université Catholique de Louvain, Louvain-la-Neuve, Belgium

Preferred presentation method: ORAL

Second topic: 5.9 Manufacturing: Liquid composite molding

Third Topic: 7.1 Recycling and sustainability: Sustainability - Resource efficient technologies and supply chain

Abstract text:

Recycling composite materials is a key challenge in the context of energy efficiency and climate change mitigation. Among other strategies, the use of thermoplastic matrices has emerged as an effective way to produce partially recyclable composite parts. Ensuring the compatibility of such new resins with pre-existing processes – in particular liquid moulding techniques - is of prime importance for current manufacturers of polymer-based composite structures.

In this perspective, dedicated monomers have been specifically formulated to enable the vacuum-assisted resin infusion through a fibrous preform, before undergoing in-situ radical polymerization. However, in some cases, the inherent exothermic nature of polymerization reactions can lead to thermal runaway - often compromising the quality of the final part. This is especially true for thick composite plates infused with methyl methacrylate (MMA)-based formulas, which are prone to an acute auto-acceleration phenomenon during polymerization known as the “gel (or Trommsdorff [1]) effect”. While ensuring optimal conversion values, this also gives rise to a sudden and non-homogenous temperature rise in the reaction medium, leading to unpredictable properties and/or the presence of voids in the polymerized matrix.

In order to design a relevant manufacturing method for thick MMA-based composite plates, it is thus crucial to understand and model the influence of the (numerous) process parameters on the heat release pattern induced by polymerization across the thickness of the part. To this end, a one-dimensional finite difference numerical model was developed in MATLAB, combining heat transfer and free-radical polymerization kinetics. Using either an empirical approach [2] or a semi-empirical approach [3]–[5] based on the free-volume theory [6] for the reaction kinetics, the framework is used to predict the temperature and monomer conversion profiles achieved during the in-situ polymerization of a MMA-based resin infused into a 7-cm thick glass fibre preform. Experimental validation was conducted by comparing the calculated temperature and conversion profiles with those measured during the corresponding real-scale infusion experiments. The predictive capacity, robustness and computation time of both empirical and semi-empirical approaches were assessed and compared for various sets of boundary and initial conditions involving isothermal vs. non-isothermal external temperatures (thermal triggering of the reaction by heating up the bottom part of the plate), and different resin formulations. Both models successfully capture the non-linear behaviour characteristic of the gel effect across the thickness of composite plates.

- [1] V. E. Trommsdorff, H. Köhle, and P. Lagally, « Zur polymerisation des methacrylsäuremethylesters », *Die Makromolekulare Chemie*, vol. 1, no 3, p. 169-198, Jan. 1948.
- [2] P. Hayden and H. Melville, « The kinetics of the polymerization of methyl methacrylate. I. The bulk reaction », *Journal of Polymer Science*, vol. 43, no 141, p. 201-214, Mar. 1960.
- [3] D. S. Achilias and C. Kiparissides, « Development of a general mathematical framework for modeling diffusion-controlled free-radical polymerization reactions », *Macromolecules*, vol. 25, no 14, p. 3739-3750, Jul. 1992.
- [4] D. S. Achilias and I. D. Sideridou, « Kinetics of the Benzoyl Peroxide/Amine Initiated Free-Radical Polymerization of Dental Dimethacrylate Monomers: Experimental Studies and Mathematical Modeling for TEGDMA and Bis-EMA », *Macromolecules*, vol. 37, no 11, p. 4254-4265, Jun. 2004.
- [5] A. Zoller, D. Gimes, and Y. Guillaneuf, « Simulation of radical polymerization of methyl methacrylate at room temperature using a tertiary amine/BPO initiating system », *Polymer Chemistry*, vol. 6, no 31, p. 5719-5727, 2015.
- [6] J. S. Vrentas and J. L. Duda, « Diffusion in polymer—solvent systems. I. Reexamination of the free-volume theory », *Journal of Polymer Science: Polymer Physics Edition*, vol. 15, no 3, p. 403-416, Mar. 1977.

Disclosure of Interest: None Declared