



THE UNIVERSITY *of* EDINBURGH

Edinburgh Research Explorer

Multi-stage, in-situ polymerisation for low-exotherm, liquid resin infusion of thick thermoplastic laminates at room temperature

Citation for published version:

Obande, W, O'Rourke, K, Stankovic, D, Lykkeberg, A, Garden, JA, Ó Brádaigh, CM & Ray, D 2023, 'Multi-stage, in-situ polymerisation for low-exotherm, liquid resin infusion of thick thermoplastic laminates at room temperature', *Composites Communications*. <https://doi.org/10.1016/j.coco.2023.101788>

Digital Object Identifier (DOI):

[10.1016/j.coco.2023.101788](https://doi.org/10.1016/j.coco.2023.101788)

Link:

[Link to publication record in Edinburgh Research Explorer](#)

Document Version:

Peer reviewed version

Published In:

Composites Communications

General rights

Copyright for the publications made accessible via the Edinburgh Research Explorer is retained by the author(s) and / or other copyright owners and it is a condition of accessing these publications that users recognise and abide by the legal requirements associated with these rights.

Take down policy

The University of Edinburgh has made every reasonable effort to ensure that Edinburgh Research Explorer content complies with UK legislation. If you believe that the public display of this file breaches copyright please contact openaccess@ed.ac.uk providing details, and we will remove access to the work immediately and investigate your claim.



Multi-stage, *in-situ* polymerisation for low-exotherm, liquid resin infusion of thick thermoplastic laminates at room temperature

Winifred Obande ^a, Kit O'Rourke ^a, Danijela Stankovic ^a, Anna Lykkeberg ^b, Jennifer A Garden ^b, Conchúr Ó Brádaigh ^a, Dipa Ray ^{a, *}

^a School of Engineering, Institute for Materials and Processes, The University of Edinburgh, Sanderson Building, Robert Stevenson Road, Edinburgh EH9 3FB, Scotland, United Kingdom

^b EaStCHEM School of Chemistry, The University of Edinburgh, Joseph Black Building, David Brewster Road, Edinburgh EH9 3FJ, Scotland, United Kingdom

* Corresponding author. E-mail address: dipa.roy@ed.ac.uk (D. Ray)

ABSTRACT

This study exploits the unique attributes of a reactive thermoplastic acrylic resin system (room-temperature liquid-resin infusibility, *in-situ* polymerisability, and room-temperature weldability) to achieve additive-free mitigation of exothermic heat generation during thick laminate production. Additives are typically required when thicknesses exceed 12 mm, but their use often compromises the degree of polymerisation and mechanical performance. An innovative multi-stage manufacturing scheme has been used to achieve a 55°C reduction in exothermic peak temperature during the production of a 16-mm-thick laminate compared to the use of a standard resin infusion (89°C) for the same thickness. Laminates produced using the multi-stage scheme were also found to exhibit 24% higher short-beam shear strengths than those obtained via standard resin infusion, suggesting improved part quality as an additional benefit. Further demonstrating the applicability of the proposed method, a 40-mm-thick laminate was successfully produced with a peak temperature of only 60°C. This work highlights the potential of room-temperature welding for practical and low-cost production of ultra-thick laminates at room temperature.

1. Introduction

The production of thick laminates (>12 mm) is notoriously challenging due to the high potential for runaway exotherm during resin *in-situ* polymerisation or curing processes. The susceptibility of reactive acrylic resins to auto-acceleration (or the Trommsdorff effect) combined with their boiling temperatures of 100°C increases the risk of exothermic peak temperatures leading to boiling [1–3]. As low-viscosity monomeric resins, these novel liquid acrylic resins offer comparable room-temperature infusibility to certain common epoxy resins without requiring expensive, specialised tooling. Their popularity continues to increase, as shown by the growing research interest in their processibility [4–6], applicability [7–12], and recyclability [13–16]. However, a significant drawback is their high heat of polymerisation, which can be up to three times that of epoxies due to concurrent increases in viscosity and decreases in termination rates after the induction period for polymerisation [17]. Issues arising from the sudden and uncontrolled heat generation within the resin during thick laminate production include inadequate resin saturation, void formation, and geometric distortion.

With the growing popularity of thermoplastic acrylic monomers in sectors like wind energy and shipbuilding, exotherm-related thermal management has become crucial for manufacturing and production operations. Specialised equipment, additives, and processes to control exothermic temperatures and reaction rates can be relatively costly, posing challenges for manufacturers seeking cost-effective production. While the use of exotherm-control additives (ECA) can limit peak exothermic temperatures below 70°C for 37-mm-thick laminates [2], direct comparisons between 12.7-mm-thick laminates with and without ECA revealed a slight decrease in flexural strength (-8%) in the ECA-containing laminate. Moreover, there are additional concerns about the use of ECA to achieve consistent degrees of polymerisation and hardening in parts with thickness variations [15].

In a recent patent application [18], we explored the room-temperature weldability of reactive acrylic resins. Unlike conventional joining techniques like ultrasonic welding [19,20] used for acrylic (and other thermoplastic) composites, this method does not require external heat application or specialised tooling, making it an excellent route for sequentially building thick laminates. The weldability of these resins offers immense potential to effectively *weld* preconsolidated laminates together via a secondary infusion stage for the fabrication of thick-section laminates. During this infusion, dry plies of fabric may be added between the layers and infused with the same resin used in the primary infusion stages for producing the preconsolidated laminates. Given the reduction in reactive resin volume, i.e., liquid resin undergoing *in-situ* polymerisation, such a scheme may enable the mitigation of the risk of polymerisation-induced exothermicity. Some efforts have been made in the Layer-by-Layer (LbL) Curing Project [21,22], which has been reported to enable concurrent layer deposition and cure of a highly exothermic prepreg material to reduce cure temperature overshoot by 70-75% during the production of thick and ultra thick laminates. Nonetheless, there are limited studies investigating additive-free routes for mitigating exothermic heat generation during the production of thick, acrylic-matrix laminates beyond 12 mm. Two schemes are examined herein to produce 16-mm-thick laminates at room temperature. The production of a 40-mm-thick demonstrator using the most effective scheme of the two, highlights its viability for thick-section laminate production without the need for specialised additives and low-exotherm resin grades.

2. Materials and Methods

2.1. Thick-section laminate fabrication

Two fabrication schemes were employed to produce 16-mm-thick laminates, targeting reductions in the exothermic temperatures during liquid resin infusion and *in-situ* polymerisation. Both processes were carried out under vacuum at room temperature. The materials used and infusion schemes for each 16-mm-thick laminate are summarised in Table 1.

Table 1.

Laminate ID	Scheme and description	Resin:Initiator	Infusions	Comparisons with Ref.
SS-16-BPO3	Reference (Ref.) <i>single-stage infusion</i>	100:3	1	–
SS-16-BPO2	Scheme 1 <i>Single-stage infusion;</i> <i>lower initiator content</i>	100:2	1	Exothermic thermal profile

MS-16-BPO3	Scheme 2 <i>Multi-stage infusion</i>	100:3	2	Exothermic thermal profile NMR and SEC analyses
<p>SS→Single-stage; MS→Multi-stage; 16 → laminate thickness; BPO# → # parts BPO per 100 parts resin. Resin: Elium® 188 O, a reactive, liquid thermoplastic acrylic resin – Arkema GRL. Initiator: BP-50-FT, a dibenzoyl peroxide (BPO) initiator – United Initiators GmbH. Glass fabric: TEST2594-125-50, a quasi-unidirectional NCF – Ahlstrom-Munksjö Glassfibre OY. NCF – non-crimp fabric with 0° fibres (600 gsm); 90° fibres (36 gsm); and polyester stitching (10 gsm). NMR: Nuclear magnetic resonance spectroscopy SEC: Size exclusion chromatography</p>				

All resin infusions and *in-situ* polymerisation processes were performed at room temperature with no specialised exothermic-control tooling or additives required. Specifically, each resin infusion process involved carefully assembling the required preform onto a rigid, flat tool base with the relevant auxiliary infusion aids (Figure S1 in Supplementary Material S1). The preform was subsequently sealed under a flexible vacuum bag and evacuated. Following a successful leak test over 10 minutes (leak rate of <3 mbar/min), resin infusion was performed under vacuum at room temperature with the liquid resin-initiator mixture into the preform assembly. Once fully saturated with no air bubbles visible at the outlet tubing (Figure S1), the infusion was terminated, and the part was left under vacuum at room temperature to *in-situ* polymerise before demoulding after 24 hours. For the production of the 16-mm-thick reference laminate (SS-16-BPO3) 3 parts per hundred resin (phr) of BPO initiator was used, whereas 2 phr was used for the Scheme 1 laminate, SS-16-BPO2. The MS-16-BPO3 laminate for Scheme 2 was produced in two resin infusion stages. During the primary infusion, a 6-mm-thick preconsolidated laminate (PL) was prepared following the standard resin infusion process described for SS-16-BPO3. After demoulding, the PL was cut using a wet saw into smaller pieces measuring 110 mm × 110 mm as shown in Figure 1a. A new preform was prepared for the secondary infusion comprising two PL pieces as inserts within an otherwise dry preform (Figure 1a and Figure 1b). Once assembled, the preform was infused following the same procedure as previously described.

As detailed in Table 1, all comparisons were made between 16-mm-thick laminates only, using the SS-16-BPO3 as the reference. In the interest of comparability, the stacking sequence employed for SS-16-BPO2 and SS-16-BPO3 were strategically matched with that of the MS-16-BPO3 (shown in Figure 2). For clarity, the lay-up strategies are summarised below:

- **SS-16-BPO3 [0]32 and SS-16-BPO3 [0]32** (refer to Figure 2a):
 - **Plies 1-8** → upward-facing 90° fibre faces.
 - **Plies 9-16** → downward-facing 90° fibre faces.
 - **Plies 17-24** → upward-facing 90° fibre faces.
 - **Plies 25-32** → downward-facing 90° fibre faces.
- **Preconsolidated laminate [0]12** (refer to Figure 2b):
 - **Plies 1-6** → upward-facing 90° fibre faces.
 - **Plies 7-12** → downward-facing 90° fibre faces.
- **Preconsolidated laminate [0]32** (refer to Figure 2b and Figure 2c):
 - **Plies 1-8** → upward-facing 90° fibre faces.
 - *Plies 1-2 as dry NCF plies*

- *Plies 3-8 as Plies 1-6 of a preconsolidated laminate*
- **Plies 9-16 → downward-facing 90° fibre faces.**
 - *Plies 9-14 as Plies 7-12 of a preconsolidated laminate*
 - *Plies 15-16 as dry NCF plies*
- **Plies 17-24 → upward-facing 90° fibre faces.**
 - *Plies 17-18 as dry NCF plies*
 - *Plies 19-24 as Plies 1-6 of a preconsolidated laminate*
- **Plies 25-32 → downward-facing 90° fibre faces.**
 - *Plies 25-30 as Plies 7-12 of a preconsolidated laminate*
 - *Plies 31-32 as dry NCF plies*

The adoption of the outlined lay-up strategy was important in this work due to the quasi-unidirectional nature of the non-crimp fabric employed in this work, which comprised faces with fibres in aligned in the 0° or 90° direction, and also dissimilar polyester stitch patterns as shown in Figure 2d.

A 40-mm-thick demonstrator laminate (MS-40-BPO3) was produced using the multi-stage infusion scheme (Figure 1a and Figure 1b).

Exothermic peak temperatures were measured during resin infusion using a single embedded k-type thermocouple at the midplane. A Pico Technology TC-08 data logger was used to record each thermal profile for 144 minutes.

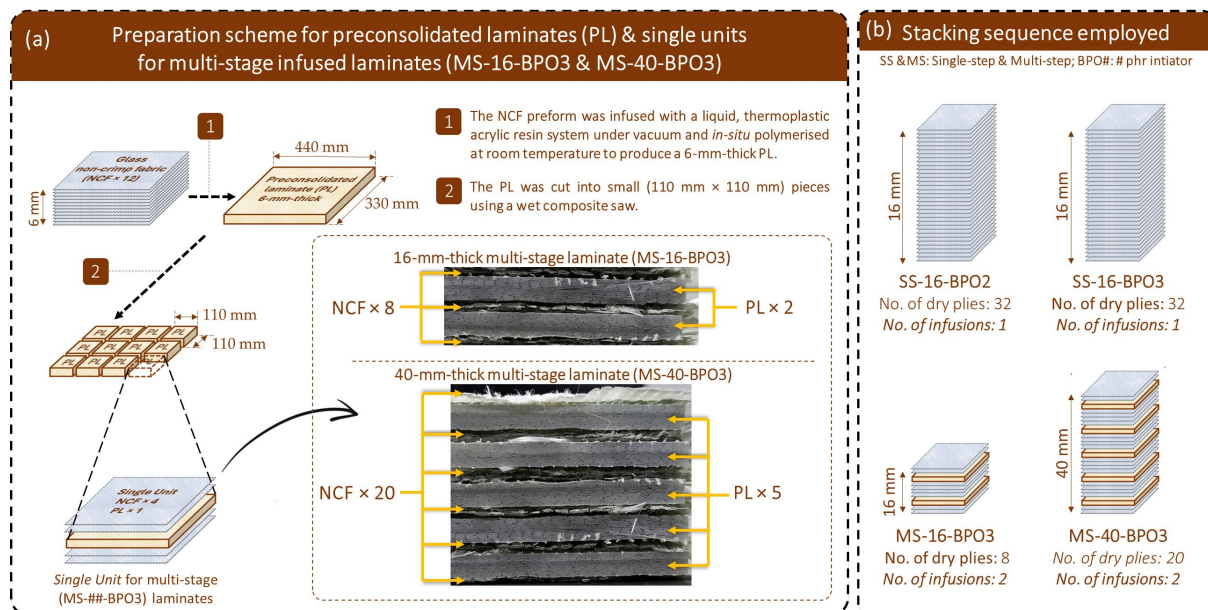


Figure 1.

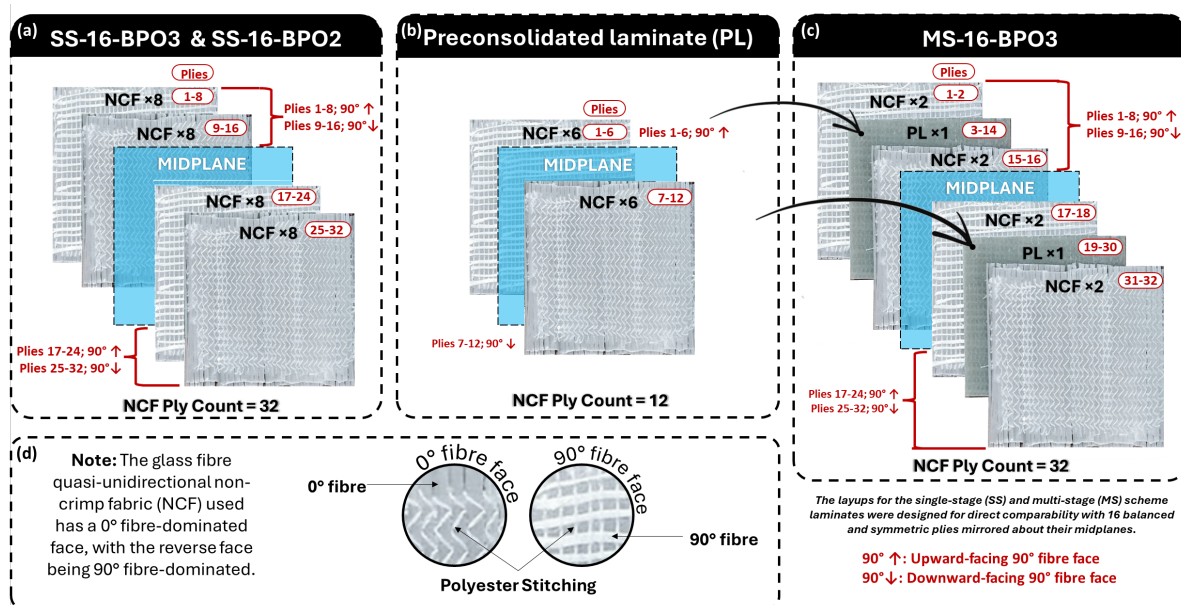


Figure 2.

2.2. Solution-state nuclear magnetic resonance (NMR) spectroscopy and size exclusion chromatography (SEC)

NMR spectroscopy was performed on the polymer matrices from the MS-16-BPO3 laminate, along with the 6-mm-thick preconsolidated laminate (PL) and the SS-16-BPO3 laminate as baselines, to investigate any changes in the chemical structure resulting from the multi-stage process. Samples from each laminate were immersed in chloroform and glass fibre fractions were removed by filtration. The solvent was then evaporated to obtain films for analysis using solution-state ^1H NMR spectroscopy.

For SEC, gel permeation chromatography-grade tetrahydrofuran (THF) was used for the previously recovered acrylic matrices from the PL, SS-16-BPO3, and MS-16-BPO3 samples.

Further details are contained in Supplementary Material S1.

2.3. Comparative mechanical testing of laminates prepared using single- and multi-stage resin infusion processes

2.3.1. Thin laminate fabrication for mechanical characterisation

Thin (4 mm) test laminates were produced using single- and multi-stage processing schemes as described in S.1.4 to study the effects on interlaminar shear performance. This thickness was selected due to the relative ease of applying standard test methods, which are not currently applicable to the thicknesses demonstrated in this work. For clarity, the single-stage-infused laminate (SS-4-BPO3) was comparatively assessed with its multi-stage counterpart (MS-4-BPO3) using short-beam shear testing as described in Section 2.3.2.

The interested reader may refer to S.1.5 in Supplementary Material S1 for further details on the materials and laminate fabrication.

2.3.2. Short-beam shear testing

Short-beam shear tests were performed in accordance with BS EN ISO 14130 to determine the apparent interlaminar shear strengths (ILSS) for these laminates. Six samples measuring 40 mm × 20 mm were loaded to failure using the Instron test frame fitted with a 10-kN load cell at a displacement rate of 1mm/min. Span- and width-to-thickness ratios were 4:1 and 2:1, respectively, and 6-mm-diameter loading and support pins were used.

3. Results and discussions

3.1. Thermal profile and optical micrographs of thick-section laminates

The thermal profiles obtained during room-temperature liquid resin infusion and *in-situ* polymerisation of the laminates within this work are presented in Figure 3a. Beginning with a direct comparison for Scheme 1 (reduction of initiator content), the exothermic peak for the laminate containing a lower initiator content SS-16-BPO2 was 94°C, which was higher than the reference laminate SS-16-BPO3 (89°C). Thus, this scheme was ineffective at mitigating polymerisation reaction-induced exotherm as it pushes the temperature even closer to the resin's boiling temperature (100°C). For Scheme 2 (multi-stage infusion using preconsolidated inserts), a similar comparison revealed a 55°C reduction from the single-stage scheme to 34°C for the multi-stage-infused 16-mm-thick laminate (MS-16-BPO3), indicating that this scheme offers the most effective means of achieving non-additive exothermic control with respect to the standard infusion of the SS-16-BPO3 laminate.

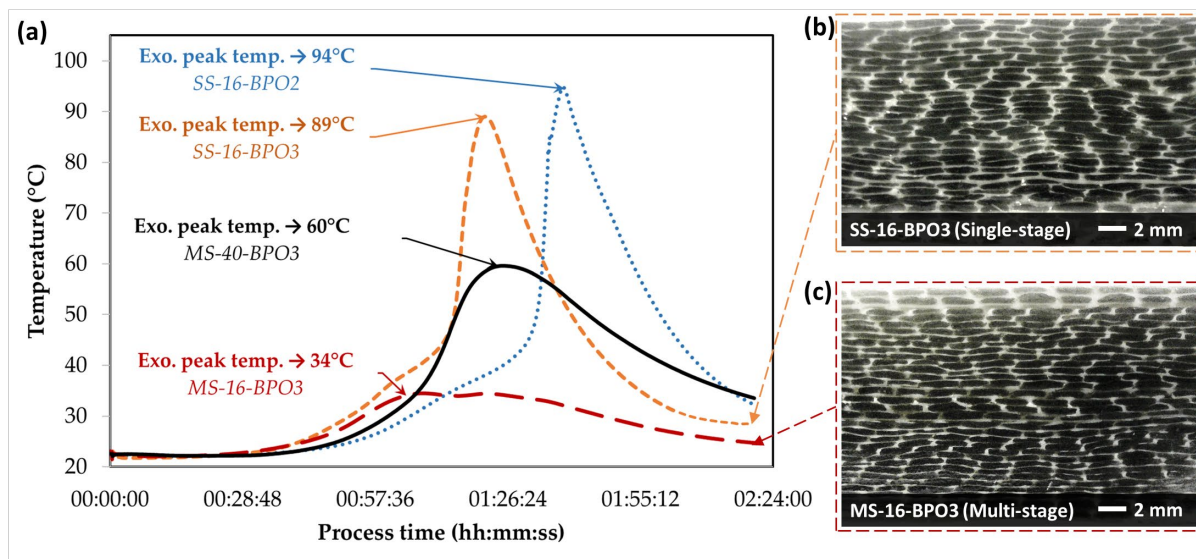


Figure 3.

From Figure 3b, it can be seen that the use of a multi-stage infusion scheme yields comparable laminate quality at this thickness range; there are no negative compaction or saturation effects, suggesting that this is a viable scheme for further investigation to establish extended applicability beyond 16 mm.

Owing to the resin's ability to act as a reactive diluent that can locally dissolve the surface layer of preconsolidated acrylic polymer. This enabled the creation of reactive surface sites and increases the permeability of the network in the surface layers to growing polymer chains during the secondary *in-situ* polymerisation stage. These growing

chains can effectively permeate through the original polymer network and interact with newly exposed reactive sites within the preconsolidated laminate, forming a semi-interpenetrating polymer network [23–28]. The formation of a semi-IPN as graphically shown in Figure 4 is the premise for the aforementioned weldability of this resin system, and facilitates the production of complex and thicker composite parts, with strong interfaces due to the presence of highly entangled polymer chains traversing the interface to effectively produce a region that may be even stronger than the bulk matrix itself.

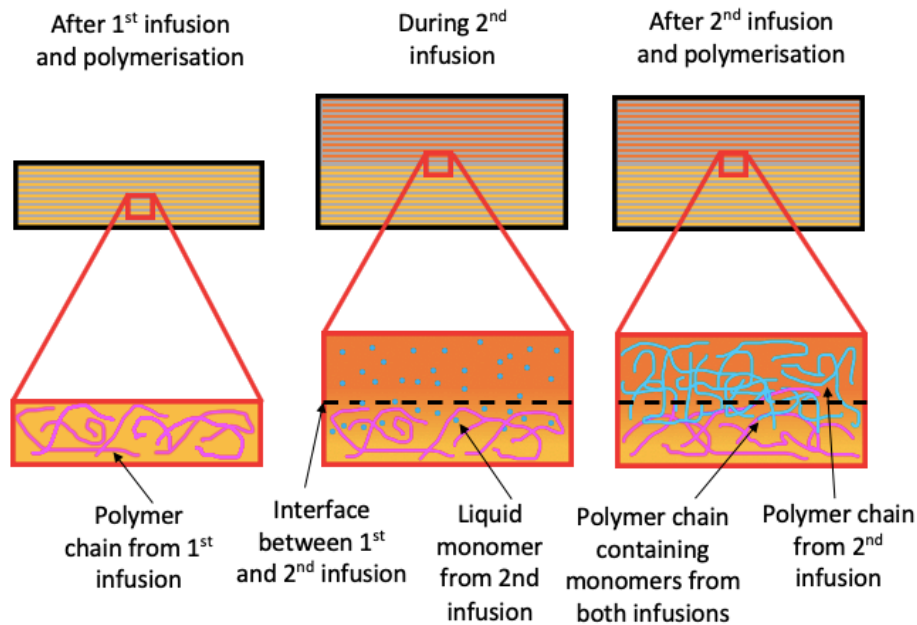


Figure 4.

Informed by these results, a 40-mm-thick laminate (MS-40-BPO3) was produced to further demonstrate the scheme. Five 6-mm-thick preconsolidated laminates were used as inserts within a dry fabric preform as depicted in Figure 1a. Favourably, the use of this scheme results in an exothermic peak temperature of 60°C, which remains much lower than the peak temperature recorded for the 16-mm-thick SS-16-BPO3 and comparable to the range reported for exothermic-control additive based laminates (~37-mm-thick; ~70°C with additive [2]). This is an important step in demonstrating the viability of the proposed scheme beyond the established thickness limit for additive-free infusion where dramatic polymerisation-induced exothermic reactions do not ensue. More importantly, such a scheme provides a route for reusing thin laminates as inserts for thick laminate infusion, a route to minimising manufacturing scrap and fostering circularity via end-of-life re-use.

3.2. Solution-state nuclear magnetic resonance (NMR) spectroscopy and size exclusion chromatography (SEC)

From complementary NMR and SEC analyses on the 16-mm-thick samples (SS-16-BPO3 and MS-16-BPO3), multi-stage infusions do not result in molecular alteration to the matrix with respect to a standard single-stage scheme. This can be clearly seen in Figure 5 and Table S1, where all materials appear to exhibit comparable ^1H

NMR spectra and molecular weights, respectively. The interested reader is referred to S1.1-S1.3 for extended discussions on the NMR and SEC results.

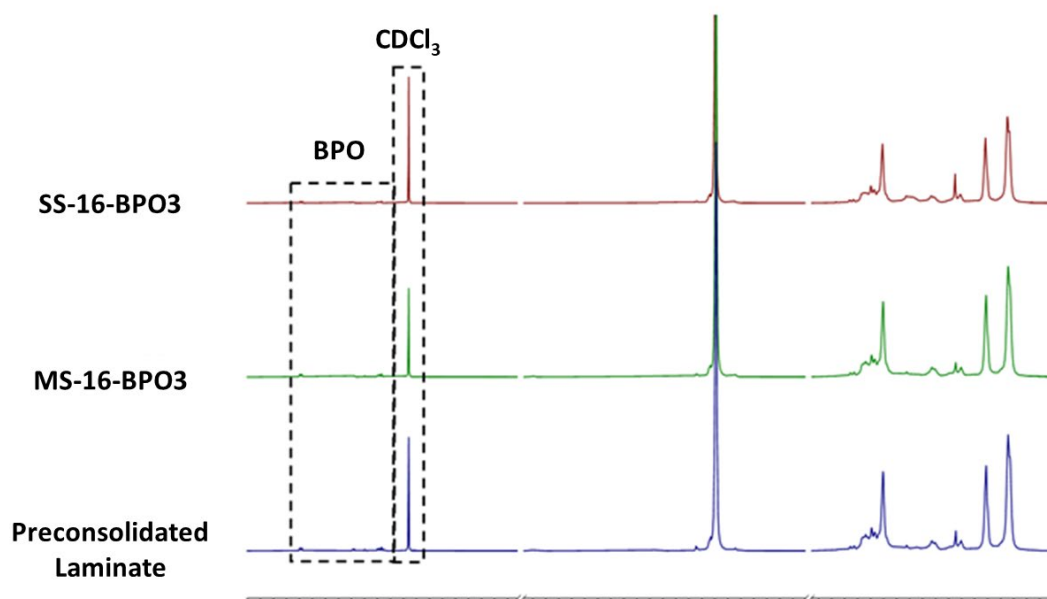


Figure 5.

3.3. Comparative short-beam shear properties of multi-staged and single-stage infused laminates

Short-beam shear testing revealed that the laminate produced using a multi-stage infusion scheme (MS-4-BPO3) had a much higher apparent interlaminar shear strength (52 ± 0.9 MPa), compared to a shear strength of 42 ± 1.4 MPa for its single-stage counterpart (SS-4-BPO3), as shown in Figure 6b. Both materials exhibit similar pre-peak load evolution behaviour (Figure 6a and Figure S3) with the characteristic inelastic deformation and absence of sudden load drops associated with thermoplastic-matrix composites. MS-4-BPO3 samples appear to have high areas bounded under the curve, which may be indicative of higher energy absorption and interlaminar fracture energy than the SS-4-BPO3 samples. While complementary SEC and $^1\text{H-NMR}$ analyses (see Section 3.2 and S1.1 – S.1.3) indicate comparable composition and molecular weights, it must be noted that the likely formation of a semi-IPN during the secondary infusion process (Figure 4) may enable favourable interactions between primary polymer and secondary polymer chains at the interface within the multi-stage resin-infused material system. Such interactions can serve to improve the resistance to shear deformation and failure along the midplane (relative to a standard infusion process). Given the sensitivity of short-beam shear testing to the presence of voids, these results suggest that the use of the multi-stage scheme may contribute positively to the low-exotherm, production of high-quality thick laminates for sectors and applications where exothermic control additives are not practical solutions.

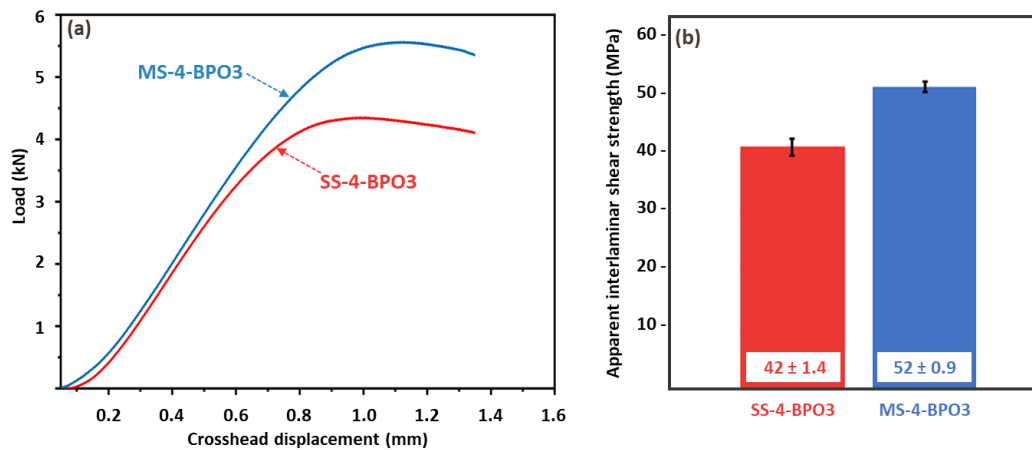


Figure 6.

4. Conclusions

In this work, two strategies were compared as routes for achieving additive-free, exothermic control during *in-situ* polymerisation of thick (16 mm) acrylic-matrix laminates. The following are the key findings and insights:

- A standard single-stage infusion of a 16-mm-thick laminate with 3 phr benzoyl peroxide initiator was used as a reference with an exothermic peak of 89°C.
- Scheme 1 also produced a 16-mm-thick laminate with reduced initiator content to 2 phr, but was ineffective, raising the exotherm temperature to 95°C.
- Scheme 2 produced a 16-mm-thick laminate using 6-mm-thick preconsolidated laminate inserts within a dry preform for secondary infusion as part of a multi-stage infusion scheme and achieved a 55°C temperature reduction relative to the single-stage reference.
 - Matrix molecular weight and polydispersity were preserved with respect to the reference.
 - Compaction and resin saturation were unaffected by the adopted scheme.

Scheme 2 was used to successfully produce a 40-mm-thick laminate with an exotherm peak temperature of 60°C — considerably lower than the single-stage-infused, 16-mm-thick reference and the range reported for infusions where additives have been employed in other works (~70°C for ~37-mm-thick laminates).

Further demonstrating the viability of this scheme, standard test samples from thin (4-mm-thick) laminates produced using the multi-stage scheme exhibited improved apparent interlaminar shear strength (52 MPa) than their single-stage counterparts (42 MPa). These improvements may be attributed to the favourable formation of a semi-IPN at the midplane. This represents a notable advancement in overcoming the current thickness limitations for reactive acrylic resin infusion without compromising mechanical performance and part quality. This approach eliminates the need for specialised additives and low-exotherm grades, which can potentially lower operational costs and simplify the adoption of recyclable thermoplastic materials. Furthermore, it enables the reuse of thin laminates as inserts for creating thicker ones, which can promote circular manufacturing practices.

5. Acknowledgements

The authors wish to thank the Supergen ORE Hub for funding received through the Flexible Fund Award FF2021-1014. We would also like to acknowledge the following funding sources: EPSRC SOFI2 Centre for Doctoral Training and Croda (A. L. EP/S023631/1); UKRI Future Leaders Fellowship (J. A. G. MR/T042710\1); and Royal Society (J. A. G. Grant RSG/R1/180101).

6. References

- [1] W. Obande, C.M. Ó Brádaigh, D. Ray, Continuous fibre-reinforced thermoplastic acrylic-matrix composites prepared by liquid resin infusion – A review, *Compos. Part B Eng.* (2021). <https://doi.org/10.1016/j.compositesb.2021.108771>.
- [2] R.E. Murray, D. Penumadu, D. Cousins, R. Beach, D. Snowberg, D. Berry, Y. Suzuki, A. Stebner, Manufacturing and flexural characterization of infusion-reacted thermoplastic wind turbine blade subcomponents, *Appl. Compos. Mater.* 27 (2019) 945–961. <https://doi.org/10.1007/s10443-019-9760-2>.
- [3] N. Han, I. Baran, J.S.M. Zanjani, O. Yuksel, L.L. An, R. Akkerman, Experimental and computational analysis of the polymerization overheating in thick glass/Elium®acrylic thermoplastic resin composites, *Compos. Part B Eng.* (2020). <https://doi.org/10.1016/j.compositesb.2020.108430>.
- [4] S.K.K. Bhudolia, P. Perrotey, S.C.C. Joshi, Optimizing polymer infusion process for thin ply textile composites with novel matrix system, *Materials (Basel)*. 10 (2017). <https://doi.org/10.3390/ma10030293>.
- [5] O. de A. Raponi, L.C.M. Barbosa, B.R. de Souza, A.C. Ancelotti Junior, Study of the influence of initiator content in the polymerization reaction of a thermoplastic liquid resin for advanced composite manufacturing, *Adv. Polym. Technol.* 37 (2018) 3579–3587. <https://doi.org/10.1002/adv.22142>.
- [6] O. de Andrade Raponi, B.R. de Souza, L.C.M. Barbosa, A.C. Ancelotti Junior, Thermal, rheological, and dielectric analyses of the polymerization reaction of a liquid thermoplastic resin for infusion manufacturing of composite materials, *Polym. Test.* 71 (2018) 32–37. <https://doi.org/10.1016/j.polymertesting.2018.08.024>.
- [7] W. Obande, C.M. Ó Brádaigh, D. Ray, Thermoplastic hybrid-matrix composite prepared by a room-temperature vacuum infusion and in-situ polymerisation process, *Compos. Commun.* (2020). <https://doi.org/10.1016/j.coco.2020.100439>.
- [8] W. Obande, W. Gruszka, J.A. Garden, C. Wurzer, C.M. Ó Brádaigh, D. Ray, Enhancing the solvent resistance and thermomechanical properties of thermoplastic acrylic polymers and composites via reactive hybridisation, *Mater. Des.* 206 (2021) 109804. <https://doi.org/10.1016/j.matdes.2021.109804>.
- [9] G. Gohel, S.K. Bhudolia, K.F. Leong, P. Gerard, On the structural damping response of hollow carbon composite shafts with room temperature curable novel acrylic liquid thermoplastic resin, *Compos. Commun.* 29 (2022). <https://doi.org/10.1016/j.coco.2021.100990>.
- [10] A.K. Kadiyala, T. O’Shaughnessy, S. Lee, A. Portela, A. Comer, Influence of environmental conditioning on mechanical properties of carbon dry fibre preformed thermoplastic matrix composites manufactured via automated placement-resin infusion process, *Compos. Commun.* 27 (2021). <https://doi.org/10.1016/j.coco.2021.100864>.
- [11] M.E. Kazemi, L. Shanmugam, S. Chen, L. Yang, J. Yang, Novel thermoplastic fiber metal laminates manufactured with an innovative acrylic resin at room temperature, *Compos. Part A Appl. Sci. Manuf.* 138 (2020) 106043. <https://doi.org/10.1016/j.compositesa.2020.106043>.
- [12] S.K. Bhudolia, S.C. Joshi, A. Bert, Y.B. Di, R. Makam, G. Gohel, Flexural characteristics of novel carbon methylmethacrylate composites, *Compos. Commun.* 13 (2019) 129–133. <https://doi.org/10.1016/j.coco.2019.04.007>.
- [13] M. Gebhardt, I. Manolakis, G. Kalinka, J. Deubener, S. Chakraborty, D. Meiners, Re-use potential of carbon fibre fabric recovered from infusible thermoplastic CFRPs in 2nd generation thermosetting-matrix composites, *Compos. Commun.* 28 (2021). <https://doi.org/10.1016/j.coco.2021.100974>.

- [14] W. Obande, D. Stankovic, A. Bajpai, M. Devine, C. Wurzer, A. Lykkeberg, J.A. Garden, C.M. Ó Brádaigh, D. Ray, Thermal reshaping as a route for reuse of end-of-life glass fibre-reinforced acrylic composites, *Compos. Part B Eng.* (2023) 110662. <https://doi.org/10.1016/J.COMPOSITESB.2023.110662>.
- [15] R.E. Murray, D. Snowberg, D. Berry, R. Beach, S. Rooney, D. Swan, D. Snowberg, D. Berry, R. Beach, S. Rooney, Manufacturing a 9-meter thermoplastic composite wind turbine blade, *32nd Tech. Conf. Am. Soc. Compos.* 2017. 1 (2017) 29–43. <https://doi.org/10.12783/asc2017/15166>.
- [16] S. Allagui, A. El Mahi, J. luc Rebiere, M. Beyaoui, A. Bouguecha, M. Haddar, Thermoplastic Elium Recycling: Mechanical Behaviour and Damage Mechanisms Analysis by Acoustic Emission, in: *Appl. Cond. Monit.*, Springer Science and Business Media Deutschland GmbH, 2021: pp. 53–61. https://doi.org/10.1007/978-3-030-76517-0_7.
- [17] Y. Suzuki, D. Cousins, J. Wassgren, B.B. Kappes, J. Dorgan, A.P. Stebner, Kinetics and temperature evolution during the bulk polymerization of methyl methacrylate for vacuum-assisted resin transfer molding, *Compos. Part A Appl. Sci. Manuf.* 104 (2018) 60–67. <https://doi.org/10.1016/j.compositesa.2017.10.022>.
- [18] D. Roy, C. Ó Brádaigh, K.N. O'Rourke, O.W. Obande, Method for joining thermoplastic articles, WO/2023/017267, 2023. https://patentscope.wipo.int/search/en/detail.jsf?docId=WO2023017267&_cid=P11-LGFRA7-07899-1 (accessed April 14, 2023).
- [19] G. Gohel, S.K. Bhudolia, J. Kantipudi, K.F. Leong, R.J. Barsotti, Ultrasonic welding of novel Carbon/Elium® with carbon/epoxy composites, *Compos. Commun.* 22 (2020). <https://doi.org/10.1016/j.coco.2020.100463>.
- [20] R.E. Murray, A. Plumer, R. Beach, P. Broome, Validation of a lightning protection system for a fusion-welded thermoplastic composite wind turbine blade tip, *Wind Eng.* (2021) 0309524X2110246. <https://doi.org/10.1177/0309524x211024642>.
- [21] A. Skordos, J. Kratz, Layer by layer curing (LbL) Feasibility study final report (Executive Project Summary), 2023.
- [22] EPSRC CIMComp, Development, Underpinning the Composites, of next generation For, manufacturing processes Report), exploitation by industry (Annual, 2023).
- [23] X.-H. Yu, J.G. Homan, T.J. Connor, S.L. Cooper, UV-Curable Polyurethane-Methacrylate Co-Networks and Interpenetrating Networks, 1989. <https://doi.org/https://apps.dtic.mil/sti/tr/pdf/ADA210445.pdf>.
- [24] T.J. Gutiérrez, R. Ollier, V.A. Alvarez, Surface Properties of Thermoplastic Starch Materials Reinforced with Natural Fillers, in: *Funct. Biopolym.*, 2018. https://doi.org/10.1007/978-3-319-66417-0_5.
- [25] C. Bandl, W. Kern, S. Schlögl, Adhesives for “debonding-on-demand”: Triggered release mechanisms and typical applications, *Int. J. Adhes. Adhes.* (2020). <https://doi.org/10.1016/j.ijadhadh.2020.102585>.
- [26] N. Zoratto, P. Matricardi, Semi-IPNs and IPN-based hydrogels, in: *Polym. Gels*, 2018: pp. 94–95. <https://doi.org/10.1016/b978-0-08-102179-8.00004-1>.
- [27] N. Karak, Vegetable oil-based polymers, 2012. <https://doi.org/10.1533/9780857097149>.
- [28] L. Kleintjens, 9.7 High pressure polymer processing, in: *Ind. Chem. Libr.*, 2001: p. 577. [https://doi.org/10.1016/S0926-9614\(01\)80033-2](https://doi.org/10.1016/S0926-9614(01)80033-2).

Figure Captions

Figure 1. Graphical representations of (a) the preparation of PL pieces for the MS-16-BPO3 and MS-40-BPO3 laminates; and (b) the stacking sequence employed for the SS-16-BPO2, SS-16-BPO3, MS-16-BPO3, and MS-40-BPO3 laminates. Note: All laminates were manufactured using a liquid resin infusion method at room temperature under vacuum. SS, MS, BPO, NCF, and PL stand for single-stage, multi-stage, benzoyl peroxide initiator, non-crimp fabric, and preconsolidated laminate, respectively. The suffixes -BPO2 and -BPO3 indicate the initiator content, i.e., 2 phr or 3 phr, respectively.

Figure 2. Illustrative representation of the balanced and symmetric layup adopted to match the 16-mm-thick laminates produced using single-stage (SS-16-BPO3 and SS-16-BPO2) and multi-stage (MS-16-BPO3) schemes for direct comparability.

Figure 3. (a) Liquid resin infusion and *in-situ* polymerisation thermal profiles were recorded using a TC-08 thermocouple data logger with a mid-plane embedded k-type thermocouple at room temperature. Exothermic peak temperatures are annotated for each glass fibre/acrylic laminate. (b) and (c) Comparative full-thickness, cross-sectional micrographs for SS-16-BPO3 and MS-16-BPO3 laminates, respectively.

Figure 4. A schematic showing the formation of a semi-IPN at the joining plane within the multi-stage resin-infused and *in-situ* polymerised laminates.

Figure 5. ¹H NMR spectra of recovered polymer matrices in chloroform. Note: Samples are from the SS-16-BPO3, MS-16-BPO3 and 6-mm-thick preconsolidated laminates (PL). The MS-16-BPO3 was produced with two pieces of the PL as inserts as part of the additive-free exothermic control scheme under investigation.

Figure 6. (a) Representative short beam shear load-displacement curves and (b) mean apparent interlaminar shear strengths for 4-mm-thick test laminates SS-4-BPO3 and MS-4-BPO3, produced using single- and multi-stage resin infusion scheme, respectively.

Table Captions

Table 1. Summary of the materials, laminate IDs and key comparisons made in this work for assessing the non-additive exothermic control scheme. The reference laminate (SS-16-BPO3) serves as a baseline for comparative analyses to assess the effectiveness of the proposed strategies.