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Interlayer bonding between thermoplastic composites and metals by in-situ polymerization technique

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Abstract

Fiber-metal laminates (FMLs) offer the superior characteristics of polymer composites (i.e., light weight, high strength and stiffness) with the ductility and fracture strength of metals. The bond strength between the two dissimilar materials, composite and metal, dictates the properties and performance of the FMLs. The bonding becomes more critical when the polymer matrix is thermoplastic and hydrophobic in nature. This work employed a novel bonding technique between thermoplastic composites and a metal layer using six different combinations of organic coatings. The flexural, and interlaminar shear strength of the thermoplastic fiber metal laminates (TP-FMLs) were examined to investigate the bond strengths in the different cases along with fracture characteristics revealed from the tested samples using scanning electron microscopy. The viscoelastic performance of the fabricated TP-FMLs were also investigated using the dynamic mechanical thermal analysis method.

KEYWORDS

manufacturing, surfaces and interfaces, thermoplastics

1 | INTRODUCTION

Thermoplastic fiber-metal laminates (TP-FMLs), where thin metal sheets and fiber-reinforced composites are integrated, offer the superior characteristics of the composites combined with the ductility of the metals. Typical examples of such FMLs include glass fiber/aluminum, aramid fiber/aluminum, and carbon fiber/aluminum, which are attracting interest from a wide range¹ of engineering sectors.

The mechanical properties of epoxy-based fiber-metal laminates, manufactured mostly by prepreg/autoclave technology, have been investigated in a number of studies.¹ The experimental results indicated that the damage threshold energy of such hybrid laminates is significantly higher than those of traditional engineering materials. In recent times, out-of-autoclave vacuum assisted resin transfer molding (VARTM) has been demonstrated as

a successful technology to manufacture FMLs with thermoset epoxy resins.² Despite the superior properties, thermoset FMLs possess some distinct drawbacks, such as low interlaminar shear strength and challenges associated with reparability and recyclability.

TP-FMLs can exhibit higher interlaminar fracture toughness, easier reparability, reshapability and recyclability compared to their thermoset counterparts. TP matrices such as polyetherimide, polyetheretherketone and polypropylene have been investigated by researchers,^{3–5} but the obvious limitations of such thermoplastic matrices are their high processing temperatures, which are likely to introduce thermal stresses at the composite/metal interface due to their different coefficients of thermal expansion. Cortés et al.⁵ manufactured TP-FMLs with titanium alloy foil and a woven glass-fiber-reinforced polyetherimide thermoplastic material at a processing temperature of 320°C. Abdullah

et al.³ investigated the interfacial fracture toughness of self-reinforced polypropylene and glass fiber reinforced polypropylene-based FMLs using aluminium alloy metal layers. PP adhesive film was used to bond the composite and the metal layers at a temperature of 135°C. Kalyanasundaram et al.⁶ manufactured a 2/1-aluminum/composite TP-FML where the inner layer of the laminate comprised self-reinforced polypropylene, Curv™. Two layers of thick hot-melt polypropylene adhesive (Glucobond) were applied to each bi-material interface. The laminate was placed in a hot press and heated to the Glucobond bonding temperature range of 155–165°C with 0.4 MPa applied pressure. The cooling rate of the composite layer and the adhesive layer controlled their crystallinity and played an important role in dictating their mechanical properties.

The development of liquid acrylic-based TP resin systems (Elium®),^{7–12} which are designed for room temperature processing by the liquid resin infusion route, could perform as an attractive candidate for manufacturing low-cost TP-FMLs. Room temperature processing would avoid the generation of any thermal stress at the metal-composite interface during manufacturing. In combination with appropriate metal surface treatments and suitably selected bonding agents, the low viscosity and in situ polymerization mechanism of the acrylic resin could be exploited for the fabrication of new-generation TP-FMLs achieving good interfacial bond strengths. This has been demonstrated in our previous work¹³ where various metal surface treatments and epoxy acrylate coatings were used as the bonding agents between aluminium and the composite layers. Moreover, recent works by Shanmugam et al.¹⁴ and Kazemi et al.¹⁵ further demonstrate the success of in situ polymerizable TP-FMLs. Given their heterogenous makeup, the ultimate performance of the FMLs is determined not only by the individual constituents alone, but by their interfacial bond strength. The structural integrity of the interface between the metal and the composite layer plays a significant role in the FML's performance.⁵ Various work has been published on metal surface treatment techniques and coatings to enhance composite adhesion with metals. Most studies to-date have focused on the use of metal surface activation with subsequent application of a coating or adhesive to promote adhesion between the organic matrices to the metal substrates.^{16–23} This interfacial bonding is critical in the performance of FMLs. Bonding with metals is even more complex with hydrophobic matrices such as acrylics. The in situ polymerisability of the liquid acrylics has been exploited in this work to introduce a novel bonding in the acrylic-based TP-FMLs.

This paper investigates the quality of bond formation between metal and the composite layers. Anodised Al alloy was used as the metallic constituent and acrylic resin/glass fiber (GF-acrylic) was used as the composite

layer. The bonding agents, which were acrylic adhesives (without hardener) mixed with epoxy acrylates, were polymerized in-situ with the liquid acrylic resin during composite fabrication, bonding the metal to the composite via covalent bonding. This unique bonding technique via in-situ polymerization has been employed for the first time for this new generation of acrylic-based TP-FMLs. Mechanical characterization of the TP-FMLs in terms of flexural and short beam shear strength was performed to assess their structural integrity. The fracture surfaces were examined under a scanning electron microscope (SEM) to study the adhesion between composite and the metal layers.

2 | EXPERIMENTAL PROCEDURE

2.1 | Materials

In this study, Elium® 180 liquid thermoplastic acrylic resin (supplied by ARKEMA, France) with viscosity and density of 100 mPa.s and 1.01 g/cm³, respectively, was used as the matrix material for the fabrication of the FMLs. An organic peroxide powder (BP-50-FT) was used as the initiator (3 wt %), supplied by United Initiators GmbH & Co. KG. Unidirectional (UD) E-glass fabric with an areal density of 646 g/m², supplied from Ahlstrom-Munksjö, was used as the reinforcement. The fabric consisted of 600 g/m² E-glass fibers in the 0° direction and 36 g/m² in the 90° direction and used a multi-compatible fiber sizing. Industrial grade aluminum (Al) alloy 6082-T6 (T6; solution heat-treated and artificially aged) in the form of a sheet with a thickness of 0.71 mm was used as the metal constituent, all details are given in Table 1. Given the vital role played by the interface in the performance of FMLs,⁵ a range of organic coatings were explored to enhance interfacial bonding (with the surface treated metal surface) during the in-situ polymerization of the TP-resin. Two methacrylate-based adhesives, SAF30-5 and SAF30-MIB, were used from Bostik (UK) and two epoxy acrylate resins (difunctional bisphenol A-based epoxy acrylate oligomer), CN104 and CN120Z, supplied by Sartomer (Arkema Group) were used. These adhesives were applied alone and in different combinations to enhance the bonding at the metal/composite interface. Anodisation was chosen for the activation/cleaning of the Al alloy sheets, since it is recognized as a highly effective technique and allows for easily transferable industrial scale usage. All Al alloy samples were electrochemically etched in a sulfuric acid (H₂SO₄) bath using an anodising technique, provided by NPI-SOLUTIONS (UK). The Al alloy sheets were anodised by immersing them in the acid solution (electrolyte) and applying DC (direct current) electricity in accordance with Def Stan 2003–25/4.²⁴

TABLE 1 (Al) alloy 6082-T6 composition and mechanical details²⁵

| Composition | 98% Al, 0.9% Si, 0.49% Mn, 0.38% Fe and 0.23% others (wt%) |
|-------------------|--|
| Modulus | 70 GPa |
| Yield strength | 260 MPa |
| Ultimate strength | 310 MPa |
| Strain to failure | 10% |

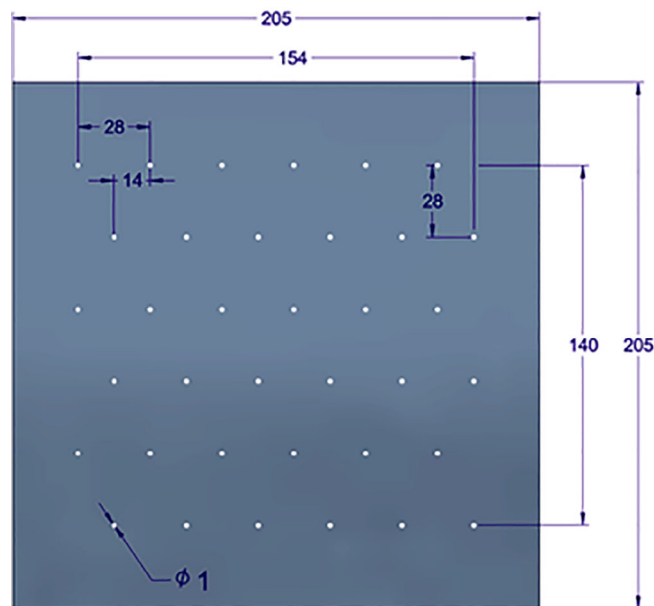


FIGURE 1 Hole-map design of the Al alloy sheet [Color figure can be viewed at wileyonlinelibrary.com]

2.2 | Manufacturing of TP-FMLs

TP-FMLs were manufactured using a standard VARTM technique. Holes were drilled in the Al alloy sheets, as shown in Figure 1, in order to produce flow paths for the liquid resin to permeate through the metal layer. The holes were drilled using a Computer Numerical Control (CNC) machine, based on Ortiz de Mendibil et al.² High dimensional precision and low roughness of the hole-map were obtained with the use of a CNC machine.

TP-FMLs were manufactured using a central aluminium sheet with UD glass fabric/acrylic resin based composite on either side. A thickness of 3.4 ± 0.2 mm was achieved. The dimensions of the fabricated plates were 210 mm \times 210 mm. Six sets of laminates (more than three laminates for each case) were manufactured with different combinations of bonding agents (methacrylate adhesives and epoxy acrylates) as shown in Table 2.

TABLE 2 Different bonding agents used to manufacture TP-FMLs

| Laminate type | Bonding agent |
|---------------|---------------------------------|
| 1 | Epoxy Acrylate; CN 104 |
| 2 | Epoxy Acrylate; CN 120Z |
| 3 | Adhesive SAF30-5 |
| 4 | Adhesive SAF30-MIB |
| 5 | 25 wt% CN104 & 75 wt% SAF30-MIB |
| 6 | 25 wt% CN104 & 75 wt% SAF30-5 |

Abbreviation: TP-FMLs, thermoplastic fiber metal laminates.

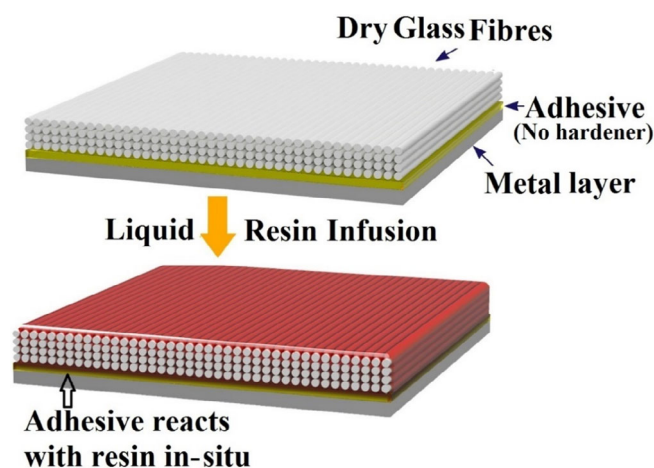


FIGURE 2 A schematic diagram showing the basic principle of bonding a composite layer with a metal layer via in-situ polymerization during composite manufacturing. Glass fiber/adhesive layers were applied on both sides of the aluminium sheet [Color figure can be viewed at wileyonlinelibrary.com]

A novel technique was employed for in-situ bonding of the metal with the composite layer. The adhesives were used without their hardeners. They were applied on the anodised Al sheets simply as organic coatings, on their own or in combination with epoxy acrylate resins, as shown in Figure 2. They were hardened when they came in contact with the infused resin containing the peroxide catalyst during composite fabrication. The latter allowed an in-situ covalent bond formation between the polymerizing resin and the bonding layer during manufacturing via in-situ polymerization. This technique is novel and has been employed for the first time to in-situ bond a thermoplastic composite with a metal layer introducing covalent bonding. This mechanism is shown schematically in Figure 2.

Prior to resin infusion, the applied adhesive does not cure or polymerize, as no initiator or hardener is present in the adhesive. During the resin infusion process, the applied adhesive mixes with the peroxide catalyst in the infused resin and polymerizes, forming strong covalent bonds

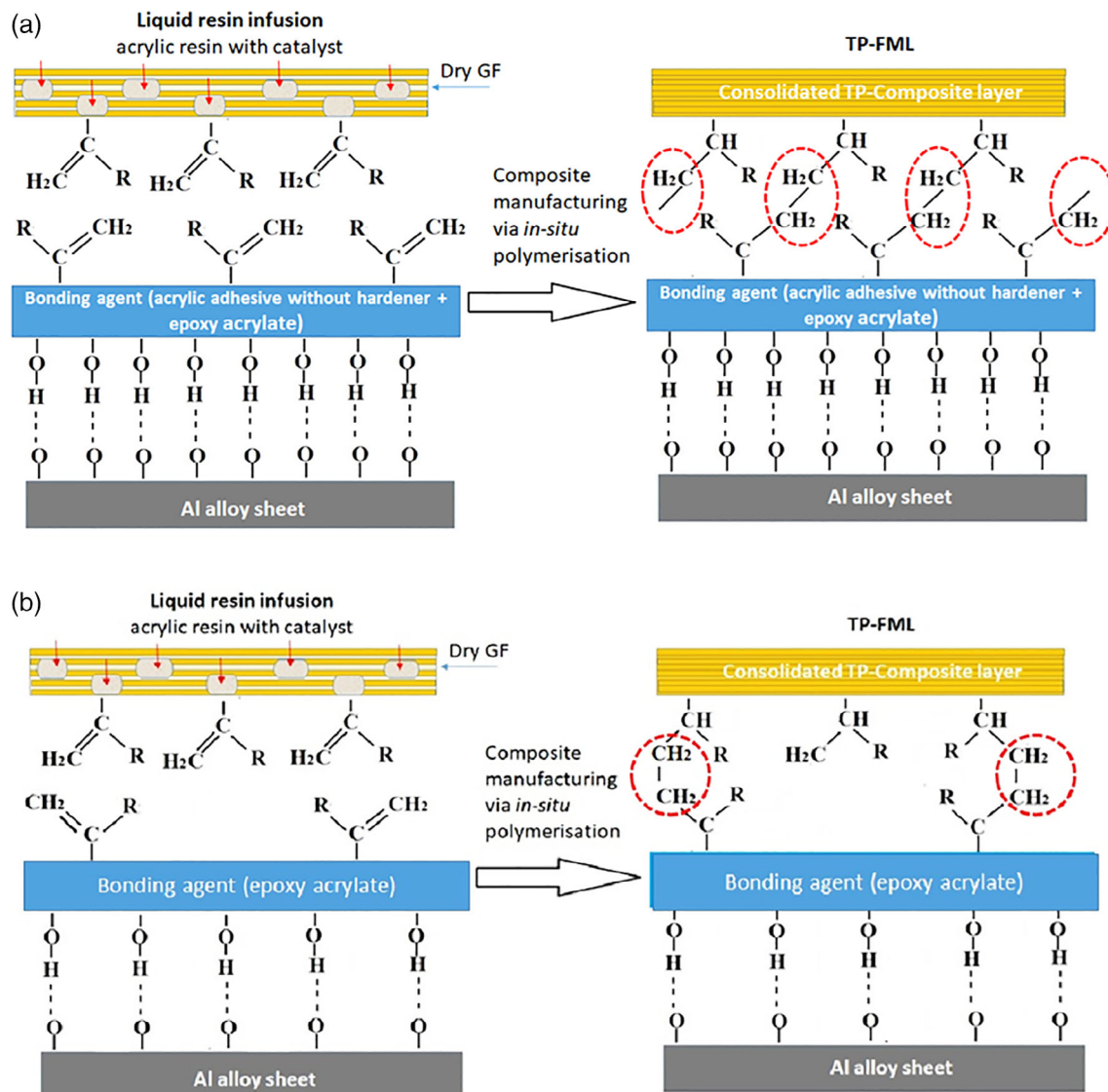


FIGURE 3 A schematic drawing depicting the proposed bonding mechanism for a thermoplastic composite with a metal layer via in-situ polymerization. The red dotted circles represent the covalent bond formation between the adhesive and the matrix via free radical polymerization during composite manufacturing. The adhesive bonds with the anodised metal surface occur via H-bonding. The bonding agents employed are (a) a methacrylate adhesive (without hardener) mixed with epoxy acrylate resin and (b) epoxy acrylate alone [Color figure can be viewed at wileyonlinelibrary.com]

between the composite and the activated aluminium layer. The in-situ polymerization was allowed to take place for 24 h. The chemical bonding is illustrated schematically in Figure 3.

2.3 | Mechanical testing

2.3.1 | Flexural testing

The flexural properties of the manufactured TP-FMLs were investigated on a universal testing machine (Zwick/Roell, model Z010) using a four-point bending fixture at a crosshead speed of 1 mm/min, according to ASTM

D7264 (2007). The standard specimen width was 13 mm and the span-to-thickness ratio was 32:1. The loading nose and supports are made from hardened steel pins of 6 mm diameter. The flexure strength and modulus were determined by loading five or more specimens to failure for each test case.

2.3.2 | Interlaminar shear strength

The interlaminar shear strengths (ILLS) of the TP-FMLs were determined by a short beam shear test in accordance with EN ISO 14130 (1997). Specimens measuring 34 mm × 13 mm were tested based on the nominal

TABLE 3 Flexural properties of TP-FMLs

| Type of bonding agent | Flexural modulus (GPa) | Standard deviation modulus (GPa) | Flexural strength (MPa) | Standard deviation strength (MPa) |
|-----------------------|------------------------|----------------------------------|-------------------------|-----------------------------------|
| CN104 | 35.7 | 2.5 | 357.3 | 50 |
| CN 120Z | 32.6 | 1.4 | 230.9 | 55.1 |
| SAF 30-5 | 37.2 | 3.5 | 614.1 | 20.1 |
| SAF 30-MIB | 35 | 1.5 | 464.5 | 31.2 |
| 25% 104 & 75% 30-5 | 40.7 | 3.4 | 630.8 | 70.2 |
| 25% 104 & 75% 30-MIB | 39.5 | 2.4 | 501.9 | 25.4 |

Abbreviation: TP-FMLs, thermoplastic fiber metal laminates.

laminates thickness of 3.4 mm. The radius of the loading member was 5 ± 0.2 mm and that of the two supports was 2 ± 0.2 mm.

2.3.3 | Microscopic observations

The fracture surfaces of the TP-FMLs were examined with a Carl Zeiss SIGMA HD VP field emission scanning electron microscope (FE-SEM). Prior to the examination, the fracture surfaces were sputter-coated with a thin evaporated layer of gold for 5–8 min for a target thickness of approximately 200 Å. Flexural tests were continued by further loading/deflecting the samples until complete failure, to obtain two halves from the FML specimens to examine under FE-SEM.

2.3.4 | Dynamic mechanical thermal analysis

A Tritec 2000 dynamic mechanical thermal analyzer was used for the characterization of the fabricated TP-FMLs. Samples measuring 35 mm × 10 mm × 3.7 mm (nominal) were dried for 48 h at 70°C and oscillated at a frequency of 1 Hz to a maximum displacement of 0.03 mm using a four-point bending configuration (BS ISO 6721-11 standard). Tests were carried out at a heating rate of 3°C/min from 25 to 185°C.

3 | RESULTS AND DISCUSSION

3.1 | Flexural properties

The overall flexural properties of TP-FMLs are summarized in Table 3.

The flexural stiffnesses of TP-FMLs presented in Figure 4a displayed a light trend upward from epoxy

acrylates to methacrylate adhesives and then to the mixed ones. This behavior is connected to better interfacial adhesion between the aluminium plate and the composite layers in the case of mixed bonding agents. The flexural modulus values of the TP-FMLs are in agreement with our previously published work on GF/acrylic composites.³²

Figure 4b presents the flexural strength of TP-FML samples. It is worth noting that the epoxy acrylate-only samples performed significantly inferior to the other cases, also manifesting a larger standard deviation, suggesting some extent of bonding inhomogeneity.

The methacrylate adhesive samples displayed higher mechanical properties in comparison to the epoxy acrylate ones. SAF 30-5 performed better than SAF 30-MIB (strengths of 614.1 and 464.5 MPa, respectively). The standard deviations of both samples were lower than the epoxy acrylate samples, highlighting a better homogeneity of the bonding layer.

Finally, the results for mixed epoxy acrylate/methacrylate adhesive samples were marginally higher than their pure methacrylate adhesive-based counterparts, with a comparative improvement of 5% for SAF-30 MIB and 4% for SAF 30-5 over the pure methacrylate adhesive samples. The mechanical properties of the two adhesives when polymerized or hardened with their respective hardeners are given below in Table 4 (values taken from the technical data sheets^{26,27}). It is evident that the SAF 30-5 is a stronger, stiffer adhesive with lower percentage elongation than SAF 30-MIB. This might be attributed to their inherent structural characteristics, which are not known, as they are commercial materials. The same trend was observed in the flexural properties of the TP-FMLs bonded with these adhesives, although for the materials presented herein, polymerization was initiated by the same peroxide catalyst used for acrylic resin.

A few examples of FML flexural properties can be found in the literature: Ostapiuk et al. presented an (Al/GFRP/Al) epoxy based FML study where their

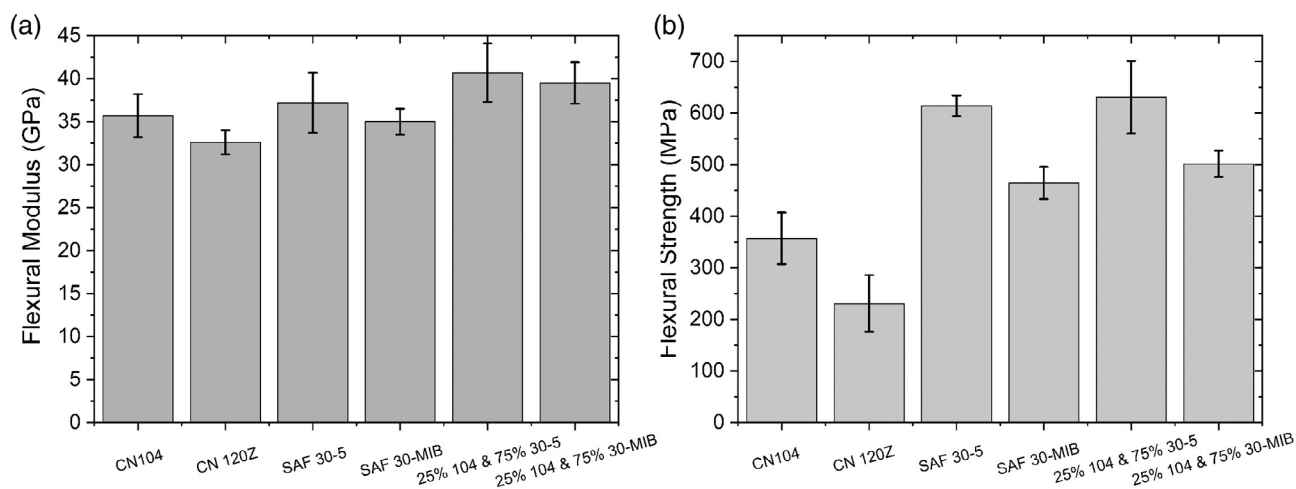


FIGURE 4 (a) Flexural modulus and (b) strength according to ASTM D7264

TABLE 4 Mechanical properties of methacrylate adhesives^{26,2}

| | Tensile strength (MPa) | Percentage elongation | Tensile modulus (MPa) | Hardness shore (D) |
|------------|------------------------|-----------------------|-----------------------|--------------------|
| SAF 30-5 | 12-16 | 30 | 250-400 | 70-80 |
| SAF 30-MIB | 10-12 | 60 | 150-250 | 60 |

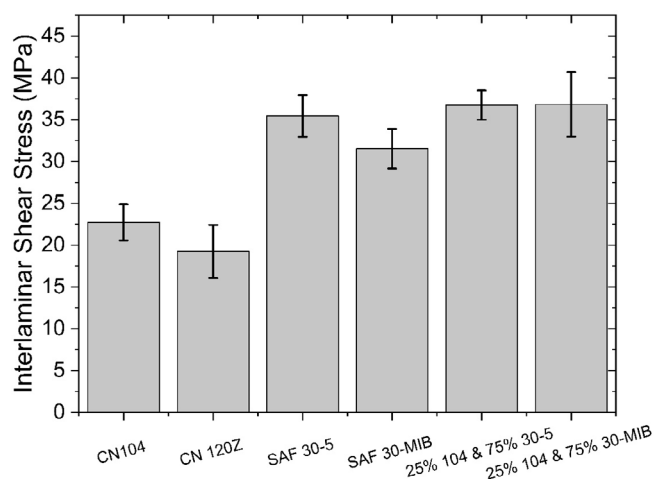


FIGURE 5 Interlaminar shear strength according to ILSS ISO 14130 (short beam shear)

unidirectional configuration reached 700 MPa strength²⁸ and similar observations were made by Cepeda Jimenez²⁹ (epoxy matrix) and Hu³⁰ (polyimide matrix). It is interesting to notice that commercial prepreg materials were used in their studies.²⁸⁻³⁰ In the present study, the in situ polymerization allowed the authors to obtain flexural strengths which are close to the above commercial prepreg-based FMLs, while only using a standard infusion process. It is also worth noting that Rajkumar et al. only obtained a

bending strength of about 170 MPa (epoxy based) in the same unidirectional configuration, layout and mechanical testing parameters while using a hand layout technique.³¹ This highlights the versatility of the process and bonding mechanisms detailed in this study.

3.2 | Interlaminar shear strength

While the standard four-point bending test involves tension, compression and shear properties, the short beam shear test is mainly driven by shear behavior, due to a reduced span to thickness ratio. This test gives an insight into the interlaminar adhesion between the constituents.

Figure 5 presents the interlaminar shear strengths (ILSS) of all samples. As per the four-point bending tests, the epoxy acrylate bonded samples, CN 120Z and CN 104, exhibit the lowest ILSS properties (with shear strengths between 19 and 23 MPa). They were followed by the methacrylate adhesive bonded ones, SAF 30-MIB and SAF 30-5 (with shear strengths between 31 and 36 MPa). Finally, the mixed bonding agent samples 25% CN 104 & 75% SAF 30-5 and 25% CN 104 & 75% SAF MIB showed the best performance, with mean ILSS values of approximately 37 MPa. These results (summarized in Table 5) clearly show that the mixed bonding agents at the metal/composite interface provided excellent adhesion in these cases.

TABLE 5 ILSS properties of TP-FML

| Type of bonding agent | ILSS (MPa) | Standard deviation (MPa) |
|-----------------------|------------|--------------------------|
| CN104 | 22.7 | 2.2 |
| CN 120Z | 19.2 | 3.2 |
| SAF 30-5 | 35.4 | 2.5 |
| SAF 30-MIB | 31.5 | 2.4 |
| 25% 104 & 75% 30-5 | 36.7 | 1.7 |
| 25% 104 & 75% 30-MIB | 36.8 | 3.9 |

Abbreviations: ILSS, interlaminar shear strength; TP-FML, thermoplastic fiber metal laminates.

TABLE 6 $\tan \delta$ peak intensity for the TP-FML samples with different bonding agents

| Type of bonding agent | $\tan \delta$ peak intensity |
|--------------------------|------------------------------|
| Epoxy acrylate, CN 104 | 0.357 |
| Epoxy acrylate, CN 120 Z | 0.293 |
| SAF 30-5 | 0.472 |
| SAF 30-MIB | 0.428 |
| 25% CN104-75% SAF 30-MIB | 0.452 |
| 25% CN104-75% SAF 30-5 | 0.513 |

Abbreviation: TP-FML, thermoplastic fiber metal laminates.

The enhanced interlaminar shear strength with the mixed bonding agents can be attributed to the combined effects of the (i) inherent structural rigidity of the methacrylate adhesives; (ii) presence of additional —OH groups contributed by the epoxy acrylate moieties; and most importantly, (iii) the covalent bonds formed between the in-situ polymerized matrix and the bonding layer.

3.3 | Dynamic mechanical thermal analysis

The thermomechanical properties of TP-FMLs evaluated by DMA tests are summarized in Table 6.

As seen in Figure 6, the epoxy acrylate-bonded samples displayed lower damping properties than the methacrylate adhesive bonded samples (–27.8% $\tan \delta$ peak value on average). The stronger interface in the methacrylate adhesive bonded TP-FMLs might have contributed to a higher viscous dissipation, increasing the damping potential in the samples.

Figure 7 shows the damping behavior of the TP-FML samples bonded with the methacrylate adhesives and with mixed bonding agents. The second type of epoxy acrylate CN 120Z was not included in the DMA study as

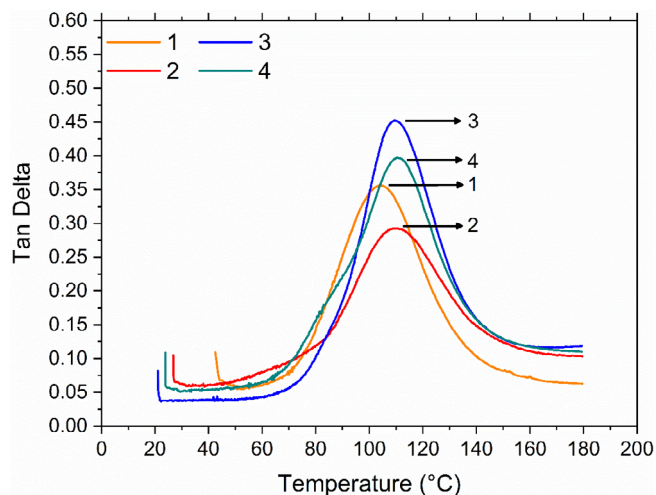


FIGURE 6 Damping behavior of thermoplastic fiber metal laminates (TP-FMLs) with individual bonding agents. 1: Epoxy acrylate CN 104. 2: Epoxy acrylate CN 120Z. 3: Methacrylate adhesive SAF 30-5. 4: Methacrylate adhesive SAF 30-MIB [Color figure can be viewed at wileyonlinelibrary.com]

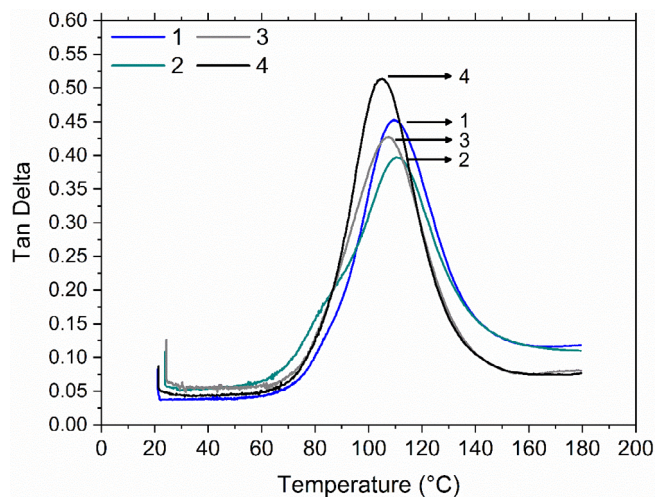


FIGURE 7 Damping behavior of thermoplastic fiber metal laminates (TP-FMLs) with methacrylate adhesives and mixed bonding agents. 1: Methacrylate adhesive SAF 30-5. 2: Methacrylate adhesive SAF 30-MIB. 3: 25% epoxy acrylate CN 104 + 75% acrylate adhesive SAF 30-MIB. 4: 25% epoxy acrylate CN 104 + 75% methacrylate adhesive SAF 30-5 [Color figure can be viewed at wileyonlinelibrary.com]

the results given by CN 120Z were inferior when compared to CN 104. The methacrylate adhesive SAF 30-5, mixed with epoxy acrylate CN 104, resulted in the highest $\tan \delta$ peak. The results are well aligned with the flexural strength and ILSS findings. The excellent interfacial bonding in this sample gave rise to an enhanced viscous dissipation and increased the $\tan \delta$ value.

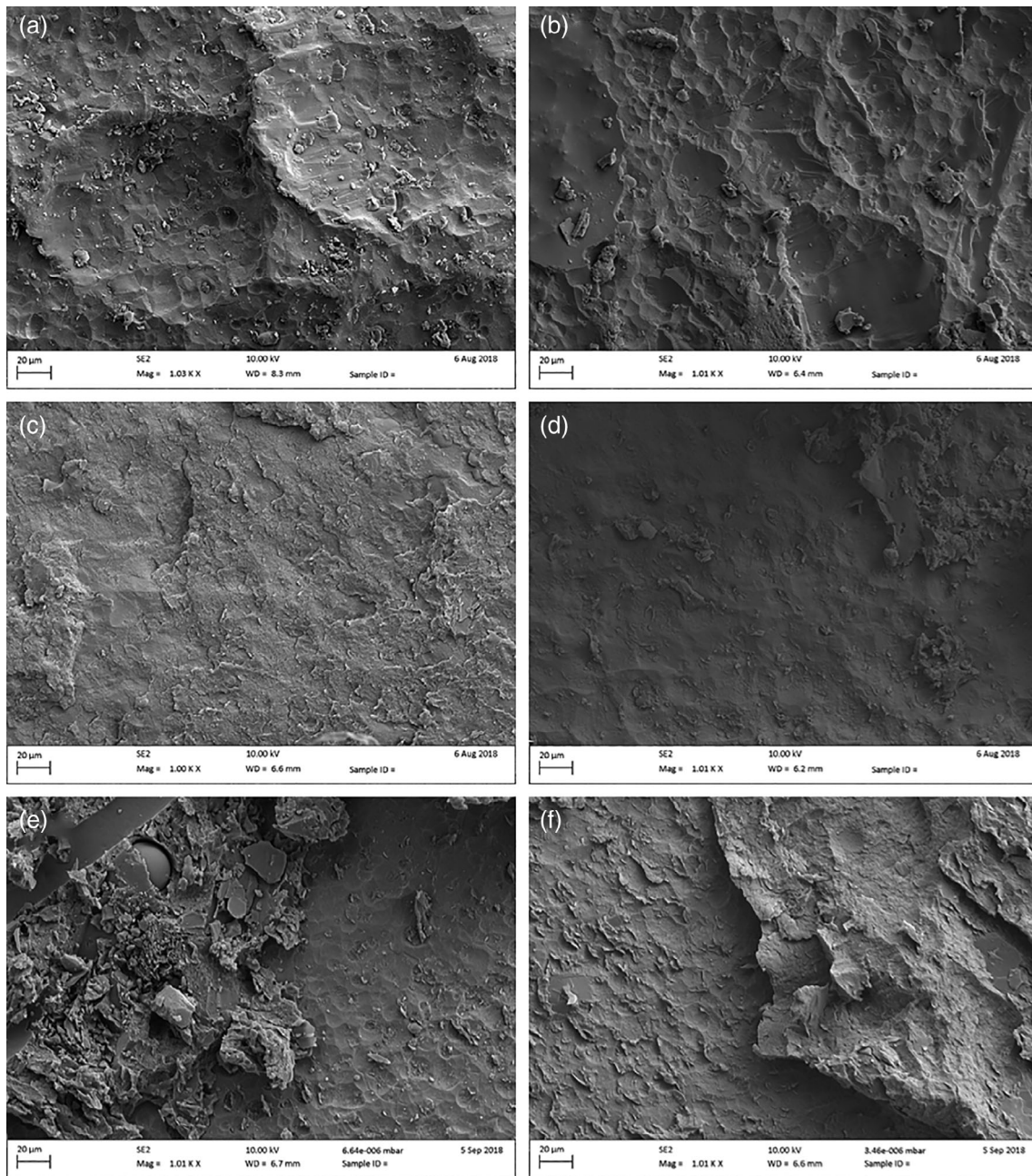


FIGURE 8 Scanning electron microscope (SEM) micrographs of representative metal/composite faying surfaces showing cohesive failure of the bonding layer after breakage (flexure samples) for (a) epoxy acrylate CN104, (b) epoxy acrylate CN120Z, (c) methacrylate SAF30-5, (d) methacrylate SAF-30-MIB, (e) 25% epoxy acrylate CN104 + 75% methacrylate SAF30-5 and (f) 25% epoxy acrylate CN104 + 75% methacrylate SAF-30-MIB

3.4 | Fractographic characteristics

The flexure samples all failed by delamination at the metal/composite interface. The de-bonded metal surface from each set of TP-FML was examined under SEM, as shown in Figure 8. The de-bonded metal surfaces were seen to be uniformly covered with the bonding agents, which shows good wetting of the metal surfaces during manufacturing. All the bonding layers showed cohesive

failure at the interface, indicating a strong bonding with both the metal and the composite layers. However, the mixed bonding agents with the methacrylate adhesives and epoxy acrylate exhibited the largest residues of the de-bonded and broken composite layers on the metal surfaces (Figure 8e,f). This indicates that the mixed bonding agents were most effective in bonding the metal to the composite layer and supports the previous mechanical test results. Ultimately, these results

confirm the fact that the mixed bonding agents resulted in the formation of the strongest bonds between the metal and the composite layers during in situ polymerization.

4 | CONCLUSIONS

In this study, commercial methacrylate adhesives and epoxy acrylate resins were investigated as bonding agents to manufacture TP-FMLs based on aluminium alloy and glass-fiber/acrylic composites. The manufacturing was carried out by liquid resin infusion at room temperature and in situ polymerization was employed as the key step to bond the metal with the composite. The combination of methacrylate adhesives with epoxy acrylate resins resulted in the highest interfacial bond strength between the metal and the composite layers. The presence of acrylate groups and hydroxyl groups in the mixed bonding agents facilitated covalent bond formation with the acrylic resin and H-bonding with the anodised metal surfaces, respectively. ILSS values for the TP-FMLs with the mixed bonding agents were in almost 37 MPa. The flexural strength and modulus of the TP-FMLs with the mixed bonding agents were measured to be in the range of 230–630 MPa and 33–41 GPa, respectively.

The highest damping behavior was observed in the TP-FMLs with the mixed bonding agents. A stronger metal/composite interface lead to an enhanced viscous dissipation, thus increasing the damping parameter of the samples. SEM investigations revealed that the bonding agents uniformly wetted the metal surface during manufacturing and no clean, non-wetted metal surfaces were evident in the failed samples. All bonding layers exhibited a cohesive failure, which is indicative of an effective adhesion both with the metal and the composite layer. The mixed bonding agents clearly exhibited their superior bond strength as larger composite remnants were evident in these cases only.

The reactive functional groups within the methacrylate adhesives and the epoxy acrylate resins facilitated in situ polymerization with the acrylic matrix, which was key to attaining desirable and effective composite-metal bonding, a prerequisite for enhanced mechanical properties. The low-cost TP-FML technology demonstrated herein can readily be upscaled for industrial production to deliver products with a unique combination of properties.

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