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Viscoelastic and Drop-Weight Impact Properties of an Acrylic Resin Matrix Composite and a Conventional Thermoset Composite — a Comparative Study

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Abstract:

Novel reactive thermoplastic resin systems such as Elium® have attracted significant research attention for composite applications because they combine the low-cost processibility of thermosets and the recyclability of TPs. In this work, the viscoelastic and impact behaviours of glass fibre-reinforced composites, containing Elium® and a commercial epoxy matrix were evaluated. To complement observations from both characterisation techniques, the fracture characteristics of both materials were also investigated by scanning electron microscopy.

Keywords: Polymeric composites; Amorphous materials; Viscoelasticity; Thermoplastic matrices

Introduction

In light of recent global directives such as 2009/33/EC and 2008/98/EC, which stipulate strict targets for weight-reduction in the transport sector and cross-sectoral reduction of landfill-bound waste streams, respectively, thermoplastic (TP)-based fibre-reinforced polymer (FRP) composites are uniquely placed to future-proof FRPs as replacements for traditional materials. However, TP matrices are high viscosity melts, requiring high-cost FRP fabrication processes [1, 2]. Consequently, their low-viscosity thermoset (TS) counterparts dominate the FRP industry due to ease of processing. Recently, novel low-viscosity (~100 mPa.s; 25°C) liquid TP resin systems such as the acrylic-based Elium® by Arkema have opened up new opportunities for the fabrication of TP-FRPs using low-cost TS-FRP production methodologies.

The mechanical and thermomechanical properties of Elium®-matrix FRPs have been studied by several authors [3-6], however, there is a paucity of published comparative data (with conventional TS-FRPs). In this work, the viscoelastic and impact behaviours of glass fibre-reinforced Elium® (GF/Elium®) and glass fibre-reinforced epoxy (GF/Epoxy) FRPs have been studied comparatively. The damping characteristics of these materials provide valuable insights into internal molecular mobility and dissipation of energy [7, 8]. Fracture behaviours were also investigated to assess their associated failure processes.

Experimental

Materials and Fabrication: Elium® 180 (Arkema GRL, France) and IN2 Infusion Epoxy (Easy Composites Ltd., UK) laminates were fabricated by a room temperature (RT) vacuum infusion method. Eight plies $[45/0/-45/90]_S$ of a non-crimp E-glass fabric (Saertex GmbH & Co. KG, Germany) were used to produce 4 mm-thick laminates (0.47 fibre volume fraction). The processing schedules are as follows:

- GF/Elium®: 24 hours at RT. No post-cure.
- GF/Epoxy: 24 hours at RT. Post-cure: 40°C to 70°C; 10°C increments with a 2-hour isothermal segment at each temperature.

Mechanical and Thermomechanical Characterisation: Single-frequency (SF) and multi-frequency (MF) dynamical mechanical analyses (DMA) were performed to study the viscoelastic behaviours of both materials. Their damage resistance following drop-weight impact events at 15 J and 30 J were also assessed by measuring respective damage areas; six specimens were tested at each energy level per material. This was done by high-contrast imaging the translucent coupons using backlight illumination. Damage areas were measured using digital image processing software (Image J, National Insitutes of Health) . Relevant information on specimens and test parameters are presented in Table 1.

Table 1: DMA and Impact test information.

Test Method	Test Parameters	
DMA	Standard	BS ISO 6721 – 11
	Specimen dimensions (mm)	35 × 5
	Instrument	Triton 2000 series
	Mode	3-point bending
	Heating rate (°C/min)	3
	Amplitude (μm)	10
	Temperature range (°C)	SF: Ambient – 170 MF: Ambient – 45
	Frequency (Hz)	SF: 1 MF: 0.01 – 100
	Span (mm)	30
Impact	Standard	ASTM D7136
	Specimen dimensions (mm)	150 × 100
	Impactor mass (kg)	4.2
	Drop height (m)	15 J: 0.55 30 J: 1.05

Fractographic analyses were performed by scanning electron microscopy (SEM – Jeol JSM series microscope) to qualitatively assess the fibre-matrix interface and matrix ductility in both materials. Given that flexural stresses play a significant role in the impact damage processes, flexural testing was performed to obtain fracture surfaces, single coupons measuring 65 mm in length were flexurally loaded at 10 mm/min over a span of 30 mm. Fracture on the lower surfaces (i.e., resulting from tensile stress) were inspected for both materials. However, being a quasi-static test regime, the fracture behaviours will not be directly comparable to those observed in impact loading, where failure occurs in shorter time scales.

Results and Discussion

Glass transition (T_g) temperatures were determined as temperatures corresponding to the damping peak as shown in Fig. 1a. These were 67°C and 115°C for GF/Epoxy and GF/Elium®, respectively. GF/Elium® was also found to have a higher damping peak compared to that of GF/Epoxy.

Given their dissimilar T_g values, multi-frequency scan results were analysed at a reference temperature of 25°C to facilitate direct comparisons between the materials' viscoelastic behaviours. As is evident in Fig. 1b., GF/Elium® not only exhibited higher (8.7%) damping behaviour, but had a higher damping retention capacity than GF/Epoxy at higher frequencies (66%, from 0.01 Hz to 100 Hz, compared to 26%).

As an amorphous TP, the chains within the Elium® matrix are able to dissipate the applied energy and subsequently expend it by means of short-range motion. The rigid nature of the cross-linked chains within the GF/Epoxy reduces the extent to which it can dissipate energy at 25°C. For both materials, there is a time-dependent response because at higher frequencies, the molecules undergo limited dissipation. Both SF and MF scans showed higher energy dissipation in the case of GF/Elium®.

The observations from the viscoelastic study were not reflected in the impact testing. The damage areas for GF/Elium® and GF/Epoxy were measured as 4.4 ± 0.9% and 3.9 ± 0.3% for coupons subjected to 15 J impacts, and 7.4 ± 0.5% and 6.6 ± 0.5% for 30 J impacts, respectively. Thus, GF/Epoxy was marginally more damage resistant. Representative images for these estimations are presented in Fig. 2a.–2d.

The impact performance of FRPs involves a combination of several complex and interrelated processes, which include energy dissipation (elastic deflection and rebound) and absorption (creation of fracture surfaces); the extent of the latter controls the resultant damage processes. From a stress perspective, the inherent anisotropy of laminated FRPs gives rise to two states of stress — namely bending and interlaminar shear [10]. Their interplay results in matrix cracking and interfacial debonding, which ultimately lead to delamination and fibre fracture. As such, matrix toughness, ductility and interfacial strength play significant roles in impact performance.

To understand the lack of correlation between the viscoelastic and impact responses of GF/Elium® and GF/Epoxy, some possible influencing factors are proposed. Complementary fractographic observations on GF/Elium® (Fig. 3a.) and GF/Epoxy (Fig. 3b.) will be discussed in light of the theoretical fracture behaviours of amorphous thermoplastic and crosslinked thermoset matrices and their respective composites.

Although the fracture surfaces of both GF/Elium® (Fig. 3a.) and GF/Epoxy (Fig. 3b.) show brittle matrix failure, certain differences are observed. Brittle amorphous thermoplastics (BATPs) such as acrylics exhibit microscopic ductility and undergo ductile-to-brittle transition at high strain rates due to strain localisation [11, 12]. Microductility predominates on the GF/Elium® fracture surfaces, whereas mirror (smooth) zones are evident in GF/Epoxy. Mirror zones indicate brittle failure with unstable crack growth. The aforementioned microscopic ductility in matrices like Elium® typically does not translate into macroscopic failure behaviour because of their post-yield behaviour. This is because crazes form normal to the applied stress direction as a result of macroscopic strain softening and limited strain hardening, the latter of which gives rise to low failure strain [11, 12]. Additionally, the presence of fibres in BATPs inhibits plastic zone development [13], which induces a triaxial stress state in the matrix and limits the dissipation of energy [14]. Despite being brittle and amorphous, epoxies do not commonly exhibit this post-yield deformation behaviour because their crosslinks increase their strain hardening moduli [13].

Despite having superior dissipative characteristics, GF/Elium® contains an acrylic matrix, which is prone to strain-dependent embrittlement [11] and has low failure strain [12]. Furthermore, a high damping peak does not necessarily predict high impact strength, because the nature of chain deformation occurring within the polymer – i.e., main or side chain – determines how much of a correlation exists between both behaviours [15].

Conclusions

A comparative study was conducted to evaluate the viscoelastic and impact behaviours of GF/Elium® and GF/Epoxy. GF/Elium® was found to have higher (~9%) damping factor and superior (66%) retention of this property at the highest test frequency of 100 Hz. Improved damping characteristics typically predict impact damage resistance, however, when subjected to 15 and 30 J impact events, GF/Elium® was less damage resistant. Our observations are in agreement with those of Heijboer [15] on viscoelastic and impact responses of unreinforced acrylic polymer. These findings are important for benchmarking Elium® as a new resin system for composite applications. SEM micrographs revealed brittle matrix failure in both materials with evidence of unstable crack propagation (smoother fracture surfaces) in GF/Epoxy and microductility in the case of GF/Elium®. Further investigations are required to substantiate the tentative explanations discussed on fractographic behaviour.

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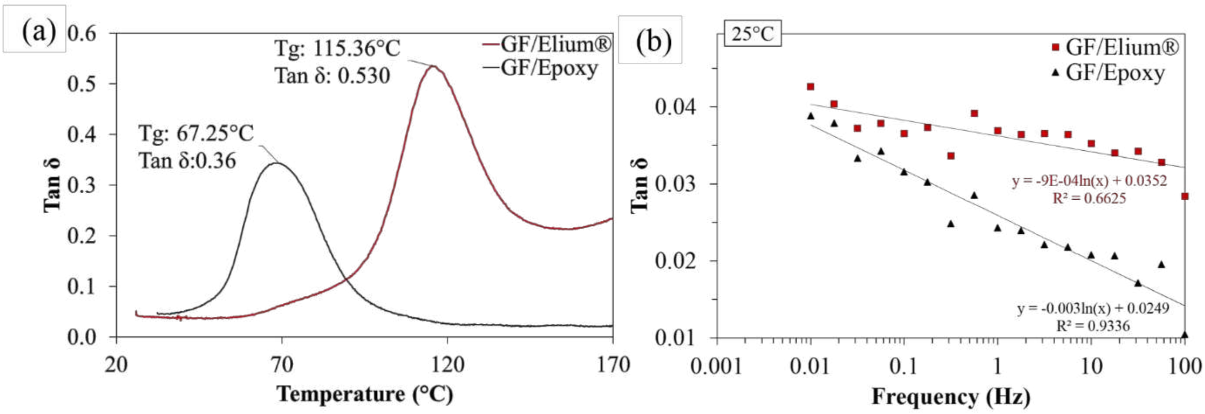
Fig. 2. Representative damage states for (a) GF/Elium® at 15 J, (b) GF/Epoxy at 15 J, (c) GF/Elium® at 30 J, and (d) GF/Epoxy at 30 J.

Fig. 3. SEM micrographs of tensile fracture surfaces from flexural specimens showing brittle matrix failure for (a) GF/Elium® and (b) GF/Epoxy.



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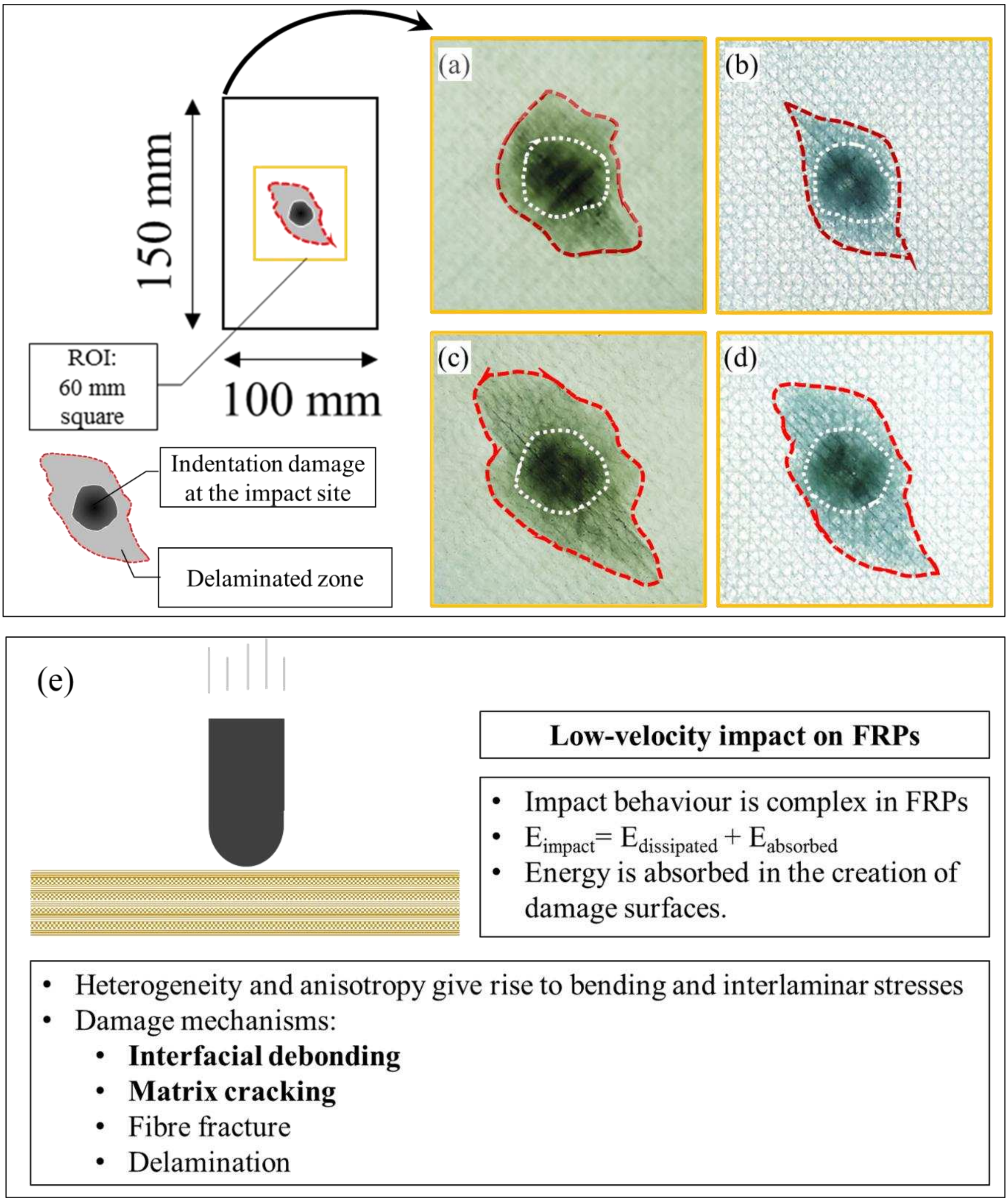
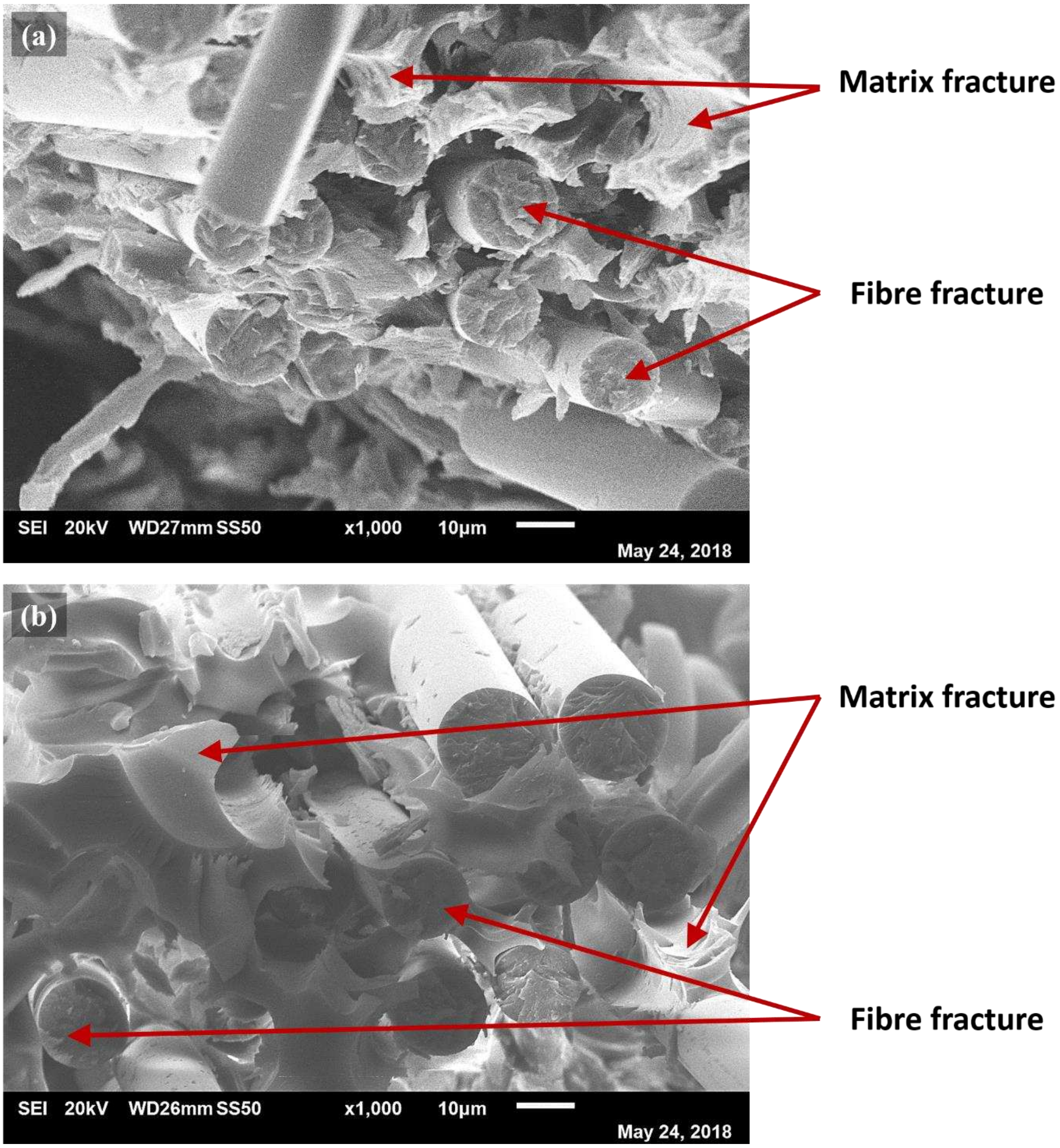


Low-velocity impact on FRPs

- Impact behaviour is complex in FRPs
- $E_{impact} = E_{dissipated} + E_{absorbed}$
- Energy is absorbed in the creation of damage surfaces.

- Heterogeneity and anisotropy give rise to bending and interlaminar stresses
- Damage mechanisms:
  - **Interfacial debonding**
  - **Matrix cracking**
  - Fibre fracture
  - Delamination

Fig. 2. Representative damage states for (a) GF/Elium® at 15 J, (b) GF/Epoxy at 15 J, (c) GF/Elium® at 30 J, and (d) GF/Epoxy at 30 J.



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