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Interlaminar Fracture Toughness of Carbon Fibre/RTM6-2 Composites Toughened with Thermoplastic-coated Fabric Reinforcement

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Abstract

Thermoplastic toughening of epoxy (RTM6-2)/ carbon fibre (CF) laminate composites without modification of the resin is reported, in which a CF fabric coated with 20 weight % of a poly (aryl ether ketone) (PAEK) was used to toughen composites. Dissolution of the PAEK during resin infusion, with subsequent reaction-induced phase separation and phase inversion during curing, formed PAEK-rich continuous layers with dispersed RTM6-rich particles within the interlaminar regions. These layers provided significant toughening during interlaminar fracture testing, in which the Mode I and Mode II fracture energies for the toughened system increased from 216 ± 7.2 to 751 ± 105 and 857 ± 99 Jm⁻² to 3316 ± 372 Jm⁻², respectively. Thus, coating of the CF fabric with PAEK avoided the significant increases in viscosity observed with resin systems containing dissolved thermoplastics; whilst generating multiphase toughening layers at the CF fabric – matrix interfaces which provide efficient interlaminar toughening.

Key Words

Polymer-matrix composites (PMCs); Fracture toughness; Interface/interphase.

1. Introduction

Advanced carbon fibre (CF) – epoxy resin composites are now used widely by the aerospace industry, due mainly to their outstanding specific mechanical properties [1]. RTM6 is a common commercial aerospace-grade epoxy system, qualified for infusion processing by the European and USA aerospace industry for more than 20 years. Due to the highly crosslinked nature of aerospace-grade epoxy resins, an RTM6 matrix is mechanically strong but also exhibits relatively low fracture resistance [2]. A common approach to increasing the fracture toughness of CF-epoxy composites is to dissolve a thermoplastic toughening modifier in the resin (such as polyether imide [3, 4], polysulfone [5], polyether sulfone [6] polyphenylene ether [7, 8], polyethylene terephthalate [9], polyphenylene oxide [10], or polyhydroxy ether

bisphenol A (phenoxy) [11]), which subsequently phase separates during curing to form a toughening dispersed phase [3]. However, the viscosity of an epoxy resin system increases significantly when a high molecular weight thermoplastic toughening component is introduced, causing severe problems in traditional infusion processes such as resin transfer moulding and resin infusion. To address this problem Cytec developed Priform™, in which the reinforcing fibres were commingled with soluble, thermoplastic fibres that dissolved in the (untoughened) epoxy resin during moulding to form a toughening dispersed phase [12]. Similarly, Wong et al reported a toughening concept based on dissolvable phenoxy fibres, and Zhang et al used phenoxy interleaves to toughen a CF/ RTM6 composite which resulted in a 3.4 fold increase in interlaminar fracture energy [13]. However, these methods were relatively complex and expensive to produce. As an alternative, Yi et al introduced the concept of “ex situ” (ES™) toughening which uses relatively simple web-coating techniques to apply a thermoplastic coating to both sides of a CF fabric. In this technique, the toughening polymer is concentrated at the matrix-reinforcement interface, giving a high local concentration of thermoplastic to increase the interlaminar toughness of laminate composites produced by dry-fabric processing techniques. Preliminary studies have shown the technique to work well with a low viscosity, untoughened epoxy system, bismaleimide and polybenzoxazine resins [14-16]. However, no study has addressed the toughening of highly-crosslinked aerospace-grade epoxy resin systems, such as RTM6, using thermoplastic-coated fabrics. In the present study, uncoated T300 CF and ES-CF fabric/RTM6 composites were manufactured using a novel resin infusion/ hot-press hybrid process, and their mode I and mode II interlaminar fracture behaviour studied.

2. Experimental

2.1 Materials

The two carbon fibre (CF) fabrics used in this study, one uncoated and the other a coated ES™ fabric, were supplied by the Beijing Institute of Aeronautical Materials (BIAM). Both were a uniweave unidirectional CF fabric with an areal density of 167±5 gsm; the warp yarn was T300 CF and the weft yarn was polyester fibre spaced at 3 mm. The ES fabric was manufactured by BIAM by coating both sides of the CF fabric with an amorphous poly(aryletherketone), PAEK, thermoplastic toughening component (Xuzhou Engineering Plastics), the structure of which is shown in Fig.1. The coating level was measured at 20.0 ±0.3 wt. % by applying resin digestion to the ES fabric (following ASTM D3171 procedure B [17]).

Fig.1. Molecular structure of the PAEK toughening coating. [14, 15]

2.2 Manufacturing

2.2.1 Processing Behaviour of Epoxy Resin Matrix System

The epoxy resin system used was RTM6-2, purchased from Hexcel. RTM6-2 is a two-component system containing the same ingredients as the original single-component RTM6 system. Samples of part A (tetraglycidyl methylene dianiline) and part B (4,4'-methylenbis(2-isopropyl-6-methylaniline and 4,4'-methylenbis(2,6-diethylaniline)) of the RTM6-2 system (here now shortened to RTM6) were preheated at 95 °C until liquid, then mixed at a weight ratio of WA:WB = 100:68.2 in a glass bottle within an oil bath set at 80 °C. A Heidolph RZR 2052 overhead stirrer was used to mix the system at 200 rpm for 30 minutes, after which the system was degassed in a vacuum oven at 80 °C for 1 hour. Samples for DSC measurements were prepared immediately after mixing and the rest of the mixed system was stored in a freezer to be used for rheology measurements and resin infusion. The dynamic mechanical properties of the composites were determined using a Perkin Elmer DMA8000 in dual cantilever bending mode. Rectangular specimens (40 x 10 x 3±1 mm) were tested at a frequency of 1 Hz from 30 °C to 300 °C at a ramp rate of 5 °C min⁻¹. The T_g of the matrix was determined using both the tan δ peak temperature for the transition and the onset of the drop in storage modulus E' at the start of the transition (sometimes termed the engineering T_g). Isothermal DSC was conducted at five temperatures, 100, 120, 140, 160 and 180 °C to simulate the thermal history of the rheometry studies. Thus, the degree of cure at any time t, α_t, during an isothermal DSC run can be calculated from the value of reaction enthalpy at time t, ΔH_t, using equation 1.

$$\alpha_t = \Delta H_t / \Delta H_{total} \quad (1)$$

Oscillatory shear rheological experiments were performed using a Thermo Scientific Haake Modular Advanced Rheometer System (MARS II) fitted with 35 mm diameter disposable aluminium parallel plates set at a 0.5 mm gap. Measurements were taken at 2.00% ±0.25%

controlled deformation auto-strain and 1 Hz frequency. Complex viscosity, η^* , was measured during two types of thermal profiles: 1) Isothermal testing at 100, 120, 140, 160 and 180 °C for 300 minutes; 2) following the manufacturer's recommended infusion cycle of 60 min at 80 °C and 120 min at 120 °C at a ramp rate of 1 °C per minute.

2.2.2 Composite Manufacturing

Resin Infusion (RI) and Hybrid RI-Hot Press Moulding

For RI, the fabrics were cut into 400 mm × 400 mm plies using a Lectra Vector TechTex FX ply cutter. Firstly, a layer of release agent (700NC, Frekote) was applied to the surface of a flat aluminium tool plate and allowed to dry. The fabric stack was then placed on the tool plate. The set up for RI is shown in Fig. 2, with spiral-split inlet and outlet tubes at two sides, peel ply (a plain woven nylon fabric) on top of the fabric stack and a distribution mesh (used to distribute the resin) placed over the top of the inlet spiral tube and over the peel-ply. Finally, the whole system was sealed with bagging film using tacky tape. For RI; the mixed RTM6 was preheated to 80 °C in an oven and the mould to 120 °C on a heated vacuum table (Elkom GmbH Vakutherm Membrane Press), then the resin was infused under vacuum and once the fabric stack was fully saturated both the inlet and outlet tubes were sealed with Hoffman clips. The lay-up was then placed in an oven and cured at 160 °C for 75 minutes. Demoulding was followed by a free standing post-cure in an oven for 120 minutes at 180 °C (ramp rate 1 °C/min). However, for composites made using RI the fibre volume fractions (V_f) of the ES composites (made using the treated ES fabric) were found to be much lower than the T300 composites (made using the untreated T300 CF fabric), at 32.4 ± 0.7 and 52.4 ± 0.3 % respectively. This was due to the vacuum-only pressure generated during RI providing insufficient compaction of the coated fabric which resulted in laminates approximately 35% thicker than those made with uncoated fabric. Reductions in compressibility have been observed previously for fabrics with surface modifications that increase the rigidity of the tows [18]. Consequently a hybrid moulding technique (Fig. 2), combining RI with hot-press moulding in a Dr Collin GmbH P300P/M press, was developed to produce laminates with higher fibre volume fractions. The layup was almost the same as RI except that two metal spacer plates were placed either side of the fabric stack in order to stop the moving platen of the hot press at the target thickness of the specimens (e.g. 4 mm for Mode I and Mode II tests). The moulding cycle proceeded in the same manner as the standard RI, but once the inlet and outlet were sealed the whole assembly was placed in the hot press (at 160 °C) and compacted under 10 bar pressure until the spacer bars were reached

and then left to cure for 75 minutes. Following demoulding, the laminates underwent the same free standing post-cure (120 minutes at 180 °C).

Fig. 2. Schematic of the RI set-up. For the hybrid RI-hot press process the spacer plates are inserted and 10 bar pressure applied in a hot press set at 160 °C.

2.3 Specimen Preparation and Composite Testing

Laminate samples for Mode I and Mode II interlaminar fracture tests had 24 layers of fabric all with a ply sequence of 0° and incorporating mid-plane inserts (Cytec ETFE release film, 13 μm thickness) of 70 mm length (which were later cut to size for the test). Specimens were cut from laminate samples using a Benetec Slida 7000 Manual Sliding Cutter: 40 mm $\times$ 10 mm for DMTA, 20 mm $\times$ 10 mm for acid digestion and 160 mm $\times$ 20 mm for Mode I and II tests. Each test was conducted on specimens from at least two panel samples. Therefore the $\pm$ standard deviations reported after the mean values in the tables and in the text reflect property variations; both panel-to-panel and specimen-to-specimen within the panels.

2.3.1 Fibre Volume Fraction

The fibre, matrix and void volume fractions of the composites were determined by removing the epoxy resin matrices by matrix digestion, following ASTM 3171 [17] procedure B which describes digestion using sulphuric acid and hydrogen peroxide.

2.3.2 Mode-I Interlaminar Fracture Toughness Test- Double Cantilever Beam (DCB)

Critical strain-energy release rate data in mode-I, G_{IC} , was determined according to ASTM D5528 [19] in which mode-I crack opening occurs due to a load applied perpendicular to the

plane of delamination using piano-hinges fitted to the end of a DCB specimen. Specimens (140 mm x 20 mm) had their side faces polished, painted white and then marked from the edge of the insert film with vertical lines every 1 mm up to 50 mm to facilitate more accurate measurement of crack length by direct visual observation using an image magnifier. Tests were carried out on an Instron 5969 universal testing machine equipped with a 10 kN load cell. G_I data was calculated by modified beam theory using equation 2.

$$G_I = \frac{3P\delta}{2b(a + I\Delta I)} \quad (2)$$

Where, P is the applied load, δ displacement, b specimen width, a crack length and $I\Delta I$ is calculated using the compliance calibration method as defined in the standard. In accordance with ASTM D5528 [12], three methods were used to determine values of G_{IC} at the initiation of delamination; visual observation, onset of non-linearity and 5% offset/maximum load (giving G_{IC-Vis} , G_{IC-NL} and G_{IC-Max} , respectively). Following initiation of delamination propagation values, $G_{IC-Prop}$, was determined as a function of crack length a .

2.3.3 Mode-II Interlaminar Fracture Test; 4-Point Bend End-Notched Flexure (4-ENF)

The 4-ENF test was used to determine Mode-II strain-energy release rate (G_{II}) data, and the procedure followed those of previous studies. [20-22] The configuration and the dimensions of a 4-ENF specimen are shown in Fig.3. The spans for the loading rollers and the supporting rollers were 60 mm and 100 mm, respectively, therefore the effective initial delamination length (a_0) was 30 mm. As with the mode-I specimens, the sides of each specimen were painted white and a scale was marked every 1 mm for 50 mm.

The 4-ENF tests were performed using an Instron 5969 universal testing machine fitted with a 10 kN load cell. The crosshead displacement speed was 5 mm/min for pre-cracking (to extend the starter crack beyond the resin pocket at the end of the insert) [23], 0.5mm/min for the remaining length and 10 mm/min for return of the crosshead after the delamination length reached around 40 mm. The G_{II} data was calculated using equation 3. Where, P is load, b specimen width, a delamination length and $\partial C/\partial a$ is the gradient from a compliance curve plotted according to modified beam theory (MBT) [19]. As with the mode-I tests, critical values of $G_{IIC-Vis}$, G_{IIC-NL} , $G_{IIC-Max}$ and $G_{IIC-Prop}$ were determined.

$$G_{II} = \frac{P^2}{2b} \frac{\partial C}{\partial a} \quad (3)$$

Fig.3. Fixture for 4-ENF mode-II testing; all dimensions in mm.

2.3.4 Scanning Electron Microscopy (SEM)

Specimens to be observed using SEM were first washed in an ultrasonic bath then dried for 4 h at 60 °C under vacuum. To improve contrast in the multiphase matrix, for selected specimens the PAEK phase was solvent etched with tetrahydrofuran (THF) for 72 hours then washed and dried. Prepared specimens were then fixed to aluminium stubs with conductive carbon paper, coated with a layer (200 Å) of AuPd alloy in an Edwards S-150-HB sputter coater and finally their edges were painted with conductive silver paint. Images were taken on a Phillips XL30 FEG SEM at 10 kV.

3. Results and Discussion

3.1 Resin Viscosity and Degree of Cure

The complex viscosity data from isothermal rheometry are shown in Fig. 4. During isothermal runs at 100, 120 or 140 °C the initial viscosities are in the range of 30-60 mPa s.

After 100 minutes, the viscosities remain in the range of 45-65 mPa s as the degrees of conversion generated (as measured by isothermal DSC) are low, ranging from 7% to 18%. This indicates that for resin infusion/injection, the processing (resin flow) time could easily be extended to more than 100 minutes without excessive increases in resin viscosity. Upon increasing the temperature to 160 or 180 °C, the resin viscosity increased rapidly within 20 and 40 minutes, respectively, due to the higher degrees of conversion attained (30% and 43% respectively). Based on these results, the mould temperature for manufacture was chosen to be 120 °C.

Fig. 4. Experimental isothermal complex shear viscosity (η^*) vs. time data and degree of cure (α data from DSC) for RTM6-2.

The effect on resin viscosity of mixing the A and B components at 80 °C for 60 minutes, followed by infusion of the resin into a RI lay-up at 120 °C is shown in the rheometry data in Fig.5, which follows the temperature profile of the standard processing cycle suggested by the RTM6-2 datasheet. Upon heating to 120 °C, the viscosity of the mixed resin drops from 160 to approximately 30 mPa s and rises only slowly to 60 mPa s over a 120 minute period at 120 °C. Thus, the system exhibits good process stability at 120 °C allowing extended infusion times.

Fig.5. Complex shear viscosity changes versus time for RTM6-2 during a standard resin injection cycle.

Table 1. Typical volume fraction results from matrix digestion of laminates produced using the hybrid RI-hot press (HP) process.

Laminate	V_f /%	V_m /%	V_v /%
T300	58.7 ± 0.6	38.9 ± 1.2	2.4 ± 0.7
ES	56.9 ± 1.1	39.7 ± 1.9	3.4 ± 0.4

3.2 Fibre Volume Fractions

Typical results from resin digestion analysis in Table 1 show both types of laminates produced by the hybrid RI-hot press (HP) process to be now of comparable quality. The HP process generated much higher levels of compaction than RI (due to application of higher pressure), and hence laminates have increased V_f .

Fig. 6. Typical DMTA storage modulus and damping data for (a) T300 and (b) ES laminates.

3.3 DMTA

The DMTA data for the T300 composite are shown in Fig 6a and Table 2 with mean values of T_g (tan δ peak) and the engineering T_g (onset of the drop in storage modulus E') of 209 and 186 °C, respectively, and a drop in E' of 1.1 orders of magnitude in the region of the glass transition between 150 and 250 °C. The addition of the PAEK coating, which is reported to have a T_g of 230 °C [14], results in significant changes in the tan δ peak of the ES composite (Fig. 6b); the peak intensity increases to 0.41 (from 0.34 for T300) and the width of the peak at half maximum increases to 29 °C, approximately 60% wider than the 18 °C of the T300 composite. In addition, a small shoulder is observed on the tan δ peak at approximately 180

°C and the ES composite exhibits much higher post- T_g damping. Thus, the $\tan \delta$ data shows that the addition of PAEK to form a multiphase interlaminar region results in a significant degree of interfacial phase mixing in the matrix of the ES composite. However, the $\tan \delta$ peak temperature decreases only slightly (by 3 °C) as is often observed for toughening of epoxy resins with a high T_g thermoplastic [24, 25]. Whereas, the increase in post- T_g damping increases the drop in E' between 150 and 250 °C for the ES composite from 1.1 to 1.35 orders of magnitude.

Table 2. Glass transition temperature (T_g) of laminates measured by DMTA.

Sample	Tan δ Peak T_g	Drop in E' T_g
	/°C	/°C
T300	209 $\pm$ 2.3	186 $\pm$ 4.4
ES	206 $\pm$ 0.5	181 $\pm$ 1.4

3.4 Interlaminar Fracture Testing

3.4.1 Mode I Fracture Energy

Typical DCB load–displacement curves of T300 and ES specimens are shown in Fig.7. The curves for all these specimens exhibited a linear region at the beginning followed by a downturn in the load. The gradient of the initial linear regions are similar indicating that the addition of PAEK results in relatively little variation in the elastic coefficient of the specimen; this is in agreement with DMTA which measured the log moduli at 30 °C of specimens from both composites to fall in the range 10.5 ± 0.2 . Crack initiation was observed to occur at higher displacements and much higher loads for the ES specimens, and for both composites a drop in load was observed as the crack began to propagate, although the load-displacement curve of the ES system is significantly less stable. Thus, the addition of PAEK both delays crack initiation and produces unstable crack propagation through this multiphase matrix, in which the secondary-phase particles can generate one or more of a number of potential additional toughening mechanisms; such as pinning or deflection of the crack, deformation of the secondary-phase particles or microcracking [10].

Fig.7. Typical DCB load–displacement curves of T300 and ES composites.

Fig. 8. Typical Mode-I R-curves from T300 and ES DCB tests.

Fig.8 shows typical R-curves of T300 and ES DCB specimens; the average G_I of the ES specimens for crack propagation is about 3 fold higher than that of the T300. The value of G_I for T300 during crack propagation is relatively stable (mean $G_I = 0.31 \pm 0.07 \text{ kJm}^{-2}$) compared to the ES (mean $G_I = 0.65 \pm 0.13 \text{ kJm}^{-2}$) which decreases by up to 25% with increasing delamination length. Despite this drop, however, the values G_{IC} for ES is still much greater. Decreasing R-curve behaviour has been observed previously for thermoplastic-toughened epoxy matrices[26, 27], when the propagating crack deviates from the interlaminar region into either the interfacial or intralaminar regions, paths which provide

easier crack propagation.

Table 3 and Fig. 8 show that the mean Mode I initiation and propagation G_I values increased significantly with the use of the PAEK-treated ES fabric, although the standard deviations also increased (to ± 13 -24%, compared to ± 3 -9%) reflecting the reduction in stability of the crack growth. Thus, the equivalent initiation values show at least a 3 fold increase (factors of ≈ 3.4 , 3.1 and 3.5 for 5%/Max to NL, respectively) and a greater than 2 fold increase (2.1) in the propagation value.

Table 3 - Mean Mode-I fracture energy values.

Sample	G_{IC-Max} /Jm ⁻²	G_{IC-Vis} /Jm ⁻²	G_{IC-NL} /Jm ⁻²	$G_{IC-prop}$ /Jm ⁻²
T300	245 $\pm$ 22	240 $\pm$ 19	216 $\pm$ 7.2	313 $\pm$ 29
ES	829 $\pm$ 103	730 $\pm$ 178	751 $\pm$ 105	647 $\pm$ 117

Fig. 9. SEM micrographs of mode I fracture surfaces: (a) onset of T300 x1k, (b) propagation ES x0.5k, (c) THF-etched ES x2k, (d) THF-etched ES x20k

The SEM image of the fracture surfaces of the T300 composite with a neat RTM6 matrix (Fig. 9 (a)) is typical of a brittle resin-fibre interfacial fracture, in which cracks initiated at the fibre matrix interface then propagated giving rise to river marks and scarps. The fibre-matrix interface also appears to be relatively weak as the fibres showed little sign of matrix adhesion. The fracture surfaces of composites toughened with the PAEK coating were much rougher and Fig.9 (b) from the propagation region of a specimen is indicative of intralaminar fracture. The image in Fig.9 (c) of a THF-etched specimen shows the complex epoxy-PAEK phase structure of the thin interlaminar regions (approximate 100 μm), whereas only single-phase RTM6 material was observed within the intralaminar regions. At a greater magnification, Fig.9 (d) shows a phase inverted particulate structure in the interlaminar region in which RTM6-rich particles are surrounded by a continuous PAEK-rich phase, similar to the morphology reported for a PAEK-toughened BMI by Cheng et al [14]. This morphology forms by reaction-induced phase separation of an initially mixed epoxy-PAEK phase in the interlaminar region followed by phase inversion to form a PAEK-rich continuous phase. The dispersed particles are quite small, with diameters 0.8-1 μm , and crack propagation has resulted in plastic deformation of these particles into prolate (elliptical) spheroids and in plastic yielding of the continuous PAEK-rich phase. The deformation of RTM6-rich particles and the plastic failure of the PAEK phase results in the significant increase in G_I values compared to the T300 composite shown in Fig. 8 and Table 4, and results in the crack deviating into the intralaminar region which provides less resistance to propagation, a contributing factor to this deviation being the lack of a toughened multiphase matrix in this region.

Fig. 10. Typical 4-ENF load-displacement curves of T300 and ES composites.

3.4.2 Mode II Fracture Energy

Typical load-displacement curves of T300 and ES specimens during the 4ENF test are shown in Fig.10. The threshold of the load drop was regarded as crack initiation, with the mean value for the ES composite being approximately 3 fold of that of the T300 and which occurred at a crosshead displacement approximately twice that of the T300, indicating that a Mode II crack propagated much earlier in the latter. In agreement with the Mode-I data (Table 4), the Mode-II data in Table 4 and Fig.11 show very significant increases in all G_{II} values with the use of the PAEK-coated ES fabric. Thus, the equivalent initiation values show essentially a 4 fold increase (factors of ≈ 4.3 , 4.2 and 3.9 fold for Max to NL, respectively) and a greater than 5 fold increase (5.3) in the mean propagation values. In the literature, a study of a similar PAEK-coated (20.2 wt.%) CF in a BMI matrix [14] reported an increase in G_{IIC} of only 1.8 fold. However Ramirez et al [28], reported equivalent 4-5 fold G_{IIC} increases for a CF composite interlaminar-toughened with 40 gsm veils of poly(phenylene sulphide) fibres.

Table 4. Mean Mode-II fracture energy values.

	$G_{IIC-Max}$	$G_{IIC-Vis}$	G_{IIC-NL}	$G_{IIC-prop}$
	$/Jm^{-2}$	$/Jm^{-2}$	$/Jm^{-2}$	$/Jm^{-2}$
T300	897 ± 116	840 ± 82	857 ± 99	733 ± 45
ES	3820 ± 325	3519 ± 480	3316 ± 372	3868 ± 329

Fig. 11. Typical Mode-II R-curves from T300 and ES 4-ENF tests.

Fig. 12. SEM micrographs of Mode-II fracture surfaces: (a) onset of T300 500 $\times$, (b) onset of ES 500 $\times$, (c) Propagation ES 500 $\times$, (d) THF-etched ES 5000 $\times$

Similar to Mode I testing, the T300 specimens in these tests showed much smoother fracture surfaces than the ES specimens. Comparing SEM micrographs of the onset region of both samples (Fig. 12 (a) and (b)) distinct cusps were observed only for the ES specimens; features which also appear in the propagation zone (Fig. 12 (c)). The THF-etched specimen in Fig. 12 (d) shows the phase separation and phase inversion in the multiphase interlaminar region which after Mode II testing shows extensive shear deformation the PAEK-rich continuous phase with little deformation of the RTM6-rich particles.

4. Conclusions

The two-component RMT6-2 shows similar rheological behaviour to that reported for RTM6, and the processing temperature was found to be in the range of 100 °C to 120 °C. The fibre volume fraction of the laminate manufactured by vacuum-only pressure resin infusion was much lower than 55% (32.4 ± 0.7), due to reduced compressibility of the coated fabrics, whereas volume fractions of $\geq 55\%$ were achievable for laminates manufactured by a hybrid

resin infusion/ hot press process which can apply significant overpressure to the fabric stack. The toughening PAEK coating on the CF resulted in only a small drop in the T_g of the ES system, down to 206 ± 2.3 °C compared to 209 ± 0.5 °C for the T300 system. Tough PAEK-rich continuous layers with dispersed RTM6-rich particles of diameter 0.8-1 μm formed within the thin interlaminar regions (approximate 100 μm) due to dissolution of the PAEK during the early stages of infusion and curing with subsequent reaction induced phase separation and phase inversion in the later stages of curing. This complex multiphase structure provided significant toughening during interlaminar fracture testing, in which the Mode I fracture energy for the toughened system increased from 216 ± 7.2 Jm^{-2} to 751 ± 105 Jm^{-2} , a 3-fold increase over the untoughened system. Similarly, the Mode II fracture energy increased from 857 ± 99 Jm^{-2} to 3316 ± 372 Jm^{-2} , a 4-fold increase. Thus coating of the CF fabric with PAEK toughening agent avoided the significant increases in resin viscosity observed with resin systems containing dissolved thermoplastics, aiding processing, whilst generating multiphase toughening layers at the CF fabric – matrix interfaces which provide efficient interlaminar toughening of the laminate composites.

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