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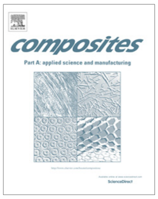

Short communication

Cure path dependency of mode I fracture toughness in thermoplastic particle interleaf toughened prepreg laminates

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ABSTRACT

The effect of cure cycle on fracture behaviour of a commercial thermoplastic particle interleaved prepreg system was investigated. Laminates were manufactured at 700 kPa in an autoclave using eight different thermal cycles that included both raising the cure temperature above the standard 180 °C cure cycle and incorporating an intermediate dwell stage between 150 and 170 °C prior to reaching the 180 °C cure temperature. Double cantilever beam tests were conducted on specimens from the cured laminates. The stick–slip crack behaviour, observed in samples manufactured using the standard cure cycle, changed to stable crack growth when processing deviated by 10 °C. The mode I fracture toughness values were reduced by 11–22% when incorporating an intermediate dwell stage before the final cure temperature. Scanning electron microscopy inspection of the fracture surfaces showed differences between samples made by standard cure cycles and those made using process deviations.

1. Introduction

Interleaving or interlayer toughening is a common technology used to improved delamination resistance of advanced composite materials. By creating a thick material interlayer between plies, interleaving may reduce limitations on plastic zone development ahead of the crack tip, allowing higher absorption of fracture energy [1–3]. One popular method for increasing interlayer thickness is the addition of thermoplastic particles [4]. As well as increasing the plastic zone ahead of the crack tip, this also may involve a number of supplementary toughening mechanisms including particle bridging, crack pinning, crack path deflection and microcracking [5,6]. The addition of an interlayer has been shown to increase the interlaminar fracture toughness of the parent laminate [2,7,8], and there are now a number of commercial material systems using thermoplastic particles in the interleaf region.

Curing reactions of thermosetting resins are exothermic, and when combined with the low through-thickness thermal conductivity of prepregs, thermal gradients and local temperature overshoots can arise within a part while curing [9]. This phenomenon is particularly prevalent in the manufacturing of thick composite parts, where high temperatures can lead to polymer degradation and the generation of residual stresses [10]. For composites containing thermoplastic interleaf particles, temperature overshoots present the additional concern of potential particle melting. Paris [11] reports that increasing part temperatures above the melting temperature of polyamide based interleaf particles resulted in a change morphology of the interlayer, increasing the measured mode I fracture toughness by up to 50%.

To reduce thermal gradients and overshoots in thick parts, the common practice is to vary the manufacturing cure cycle to feature lower ramp rates and/or additional lower temperature dwell stages [12]. It is well understood that because of the heterogeneous nature of fibre reinforced composites, such variations in the manufacturing cure cycle can affect the generation of residual stresses in a part [13,14]. However, the effect of cure cycle modifications on the delamination resistance in particle interleaf systems with heterogeneous interlayers has yet to be fully addressed. Altering the cure path has been shown to influence phase separation of dissolved thermoplastics in a particle toughened epoxy [15], the resulting effects on mechanical properties were not investigated. A study by White and Kim [16] investigated the effect of staged curing on interlaminar properties, and they found that the cure path had a negligible effect on the fracture toughness in a non-particle toughened material. The influence of the manufacturing process on interlaminar properties of an interleaf particle toughened prepreg was investigated by Zhang and Fox [17]. They found that for laminates cured using the quickstep process, mode I fracture toughness was 2.6 times higher than those cured in an autoclave.

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Any cure path influence on part properties where temperature gradients are present during the manufacturing process could result in different properties across the part. This study was undertaken to establish a link between cure path and the interlaminar performance of a thermoplastic particle interlayer toughened prepreg. Mode I opening tests were conducted on specimens manufactured using a variety of cure cycles. The cure cycles were chosen to investigate the effects of temperature overshoots and typical cure cycle modifications used to avoid temperature overshoots, including additional temperature dwell stages and lower ramp rates.

2. Laminate manufacturing

A commercially available prepreg, HexPly® M21 with IMA fibre, representative of the 'interlayer toughened' class of carbon fibre reinforced epoxy systems, was used to produce $[0]_{18}$ laminates with a nominal thickness of 3.4 mm. All laminates were cured in an autoclave using a vessel pressure of 700 kPa and a vented vacuum bag. The eight cure cycles chosen for the study are detailed in Table 1.

The standard cure cycle featured a 2 °C/min ramp to a typical 180 °C dwell temperature, denoted as cure cycle 5. In cure cycles 1–4, intermediate dwell stages were introduced to gel the resin prior to reaching the 180 °C final cure temperature. To investigate any effects of ramp rate, cure cycles 1 and 2 had identical dwell stages but had variations in temperature rise rate. The final three cure cycles (6–8) have raised curing temperatures of 185 °C, 190 °C and 200 °C, which were used to study the effect potential temperature overshoots in thicker parts or hotter sections of the autoclave.

A 20 μm thick Fluorinated Ethylene Propylene (FEP) fluoropolymer release film was inserted into the laminate mid-plane during lay-up to act as a crack initiator. After cure, piano hinges were bonded to the test specimens for load introduction using Araldite® 2014-1 epoxy paste adhesive.

During cure cycle 8, the target 200 °C autoclave set-point approached the upper limits of the bagging consumables. During the manufacturing process, the vacuum bag containing this laminate developed a leak just prior to reaching the 200 °C cure temperature. Specimens from this cure cycle had a similar total and interlaminar thickness as laminates from the other cycles (see Table 1).

3. Test procedure

Six specimens were cut from a single plate each cure cycle and tested for Mode I fracture toughness according to the ASTM D5528-13 test standard [18] using a displacement rate of 2 mm/min. The Modified Beam Theory (MBT) method, shown in Eq. (1), was used to calculate G_{IC} values [18]:

$$G_{IC} = \frac{3P\delta}{2b(a + |\Delta|)} \tag{1}$$

Some samples displayed stick–slip behaviour under loading. Where this occurred, propagation values of G_{IP} were recorded at the peak load values, using the analysis methods outlined by Hojo et al. [19].

4. Results and discussion

Stick–slip, stable crack growth, and a combination of the two were observed during the mode I testing of specimens processed by the various cure cycles. These responses are reported in Fig. 1, which shows DCB load vs load point displacement graphs for typical test specimens from each cure cycle. In Fig. 1a the samples from cure cycles 150 f and 150 s, which had the lowest temperature intermediate dwell stage, exhibited mostly stable crack propagation in the resin interlayer, with no fibre bridging. Increasing the intermediate dwell temperature to 160 °C and 170 °C (Fig. 1b) caused the specimens to show some stick–slip behaviour during testing. Fig. 1c shows the responses of samples from the 180 and 185 °C single stage cure cycles, both of which exhibited stick–slip behaviour throughout testing. Increasing the cure temperature to 190 and 200 °C (Fig. 1d) resulted in stick–slip behaviour at small crack lengths, but towards the end of the test, a transition to stable crack growth was observed. This phenomenon was common in all specimens from cycles 190 and 200 but was not observed in any other cycle. Upon visual inspection of the fracture surfaces it was found that this was caused by a transition of the crack from the particle toughened interlayer into the fibrous intralaminar region.

Delamination resistance curves (R-curves) for samples from cure cycles 150 f, 180, and 200 are displayed in Fig. 2. For cycles 150 f and 180, the G_{IP} values remain constant throughout the test. For cure cycle 200, G_{IP} is seen to start around 0.3 kJ/m^2 and then drops to 0.26 kJ/m^2. This was observed in the 190 and 200 °C temperature cure cycles and was found to correspond with the crack transitioning from the interlayer into the interlaminar region; this phenomenon has also been observed by Zhang and Fox [17].

Average fracture propagation values for each of the cure cycles are displayed in Fig. 3. Increasing the intermediate dwell temperature has shown to increase the measured mode I fracture toughness of particle interleaf specimens. In addition, laminates manufactured with a single stage cure cycle have higher G_{IP} values than laminates manufactured with an intermediate dwell stage. For the raised temperature cure cycles at 190 and 200 °C, the mode I fracture toughness was calculated for both interlayer (solid bars) and intralaminar (dashed bars) crack growth; the fracture toughness was lower in the intralaminar region than in the interlaminar region.

Table 1
Cure cycles.

Cure cycle number	ID	Ramp rate (°C)	Initial dwell stage		Final stage		Average cured interleaf thickness ± one standard deviation (μm)
			Temperature (°C)	Time (min)	Temperature (°C)	Time (min)	
1	150 f	1	150	220	180	150	31.5 ± 4.3
2	150 s	0.5	150	220	180	150	31.7 ± 3.7
3	160	1	160	180	180	150	32.6 ± 9.9
4	170	1	170	150	180	150	32.3 ± 4.2
5[a]	180	2	–	–	180	120	29.9 ± 6.1
6	185	2	–	–	185	160	31.3 ± 3.9
7	190	2	–	–	190	160	31.3 ± 6.0
8	200	2	–	–	200	160	31.4 ± 3.8

[a] Standard cure cycle.