THE INTERLAMINAR FRACTURE PROPERTIES OF FIBRE REINFORCED THERMOPLASTIC COMPOSITES:THE EFFECT OF PROCESSING TEMPERATURE AND TIME

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ABSTRACT

An experimental study has been conducted to study the effect of processing temperature and dwell time on the critical energy release rate, Gc of unidirectional carbon fibre-reinforced poly ether imide (CF/PEI) under Mode II and Mixed-Mode I/II loading. Under Mode II loading, the value of GIIc, increased as a function of both processing temperature as well as with dwell time at the required temperature. A linear relationship was observed between the value of GIIc and the logarithm of the dwell time, t. Under Mixed-Mode loading conditions, the R-curves showed a continuous increase in the value of GI/IIC as a function of crack length, possibly due to the effect of fibre bridging. A comparison between the fracture toughness under Mode II shear loading and Mixed-Mode I/II loading indicated that Mode II loading yielded higher fracture energies for the same temperature and dwell time, possibly due to the occurrence of additional toughening mechanisms. The results suggest that the optimum processing temperature and dwell time for this material are 300°C and 60 minutes respectively. It is planned that these processing parameters will be employed in the repair of impact-damaged panels based on this thermoplastic material.

***KEYWORDS**: Interlaminar fracture, damage, thermoplastic composites*

1.0 INTRODUCTION

With the continuous increase in the use of polymers and structural composites in a wide range of applications, such as the automotive, aerospace, defence and construction industries, appropriate techniques are required for repairing damage that may occur during the operational lifetime of the structure. However, according to Wu *et al*. (2008), many of the existing repair methods are costly, time-consuming and require reliable detection techniques as well as a skilled workforce. In addition, these procedures are mainly applied to external repairs and accessible damage, instead of internal and invisible microcracks. In contrast, self-healing polymeric materials offer an attractive alternative for repairing damage. In principle, self-healing polymeric materials offer the potential to substantially recover their load-carrying ability after repair. Such recovery can occur autonomously (self-sufficiently) or be activated through the application of a specific stimulus, such as heat or radiation.

According to the comprehensive review on repair by Wu *et al.* (2008), the conventional methods for the repair of advanced composites include welding, patching and in-situ curing of new resins. Techniques for the repair of thermoplastics include (i) fusion bonding through resistance heating, infrared welding, dielectric and microwave welding, ultrasonic welding, vibration welding, induction welding and thermobond interlayer bonding, (ii) adhesive bonding and mechanical fastening such as riveting. However, as previously mentioned, there are some limitations to these traditional methods, including their cost and the fact that they can be time-consuming. In addition, there is a need for reliable detection techniques and a skilled workforce. It is also worth noting that existing methods are limited to the repair of external and accessible damage instead of internal and visible microcracks. In an attempt to fill this gap, a number of self-healing polymers have been recently introduced.

Jud *et.al.* (1981) investigated the mechanisms of crack-healing in glassy polymers (PMMA-PMMA, SAN-SAN and PMMA-SAN) using the compact tension test specimen (CT). In this study, two batches of specimens were considered; these being (i) specimens that were not completely broken and were exposed to a crack-healing treatment at elevated temperatures and (ii) samples in which the fracture surfaces of the broken CT specimens were polished and then subjected to a welding treatment at elevated temperatures. From the experimental results, it was found that at temperatures above the glass transition temperature, T_g, the fracture toughness, K_{Ii} varied from very low values, of the order of the surface free energy of the polymer , γ, to $K_{Io,}$ *i.e.* the fracture toughness of the original material, as a function of healing time, t_p, and temperature, T_p. Based on a diffusion model, a relationship in the form of $K_{Ii} \propto t^{1/4}$ was developed and applied.

In a subsequent study by Davies *et.al* (1989), the process of crack healing in carbon fibre-reinforced PEEK composites was investigated. Double cantilever beam (DCB) specimens were repaired at temperatures between 320°C and 380°C and the degree of recovery in the delamination resistance was assessed. This initial study suggested that the Mode I delamination resistance can be fully recovered in these carbon fibre reinforced thermoplastic composites.

Reyes and Sharma (2010) studied the reparability of impact-damaged woven glass fibre reinforced polypropylene composites. A simple compression moulding process was employed to repair the damaged panels. Following four-point-bend tests, on the repaired samples, a significant recovery in the flexural strength and modulus of the thermoplastic matrix composites was reported.

Wu *et al.* (2008) reviewed the literature on self-healing polymers and reported that one of the commonly-observed healing techniques include molecular inter-diffusion via thermal action. In the current research programme, a similar healing technique is used to join a fibre-reinforced thermoplastic at different temperatures and dwell times.

2.0 EXPERIMENTAL PROCEDURE

2.1 Materials

In this study, a carbon fibre-reinforced polyether imide (CF/PEI) from Ten Cate Advanced Composites has been investigated. The material was a unidirectional composite supplied in prepreg form with a nominal thickness of approximately 0.25 mm. **TABLE 1** presents some of the fundamental properties of this material.

TABLE 1 Fundamental properties of the unidirectional carbon fibre reinforced PEI

Properties	**Value**
Density (kg/m^3)	1510
T_g (°C)	217
Flexural strength (MPa)	870
Flexural Modulus (GPa)	50

2.2 Experimental Test Methods

Interlaminar Fracture Testing

Interlaminar fracture test specimens were prepared by stacking twelve plies of unidirectional carbon fibre-reinforced polyether imide (CF/PEI) in a picture frame mould with an opening size of 200 x 240 mm. To introduce a pre-crack, a folded aluminium alloys with a nominal thickness of 20 μm was inserted at the mid-plane of the stacked prepreg. The stacked prepreg was then heated to different processing temperatures (220, 240, 260, 280 and 300 °C). Two different dwell times were considered at each processing temperatures; these being 5 and 60 minutes. A fixed pressure of 6.5 bar was employed during processing. This was followed by cold pressing to room temperature. After consolidation, the moulded panels had a thickness of approximately 3 mm.

Mode II End Notched Flexure (ENF) Testing

Mode II End Notched Flexure (ENF) testing was carried out in accordance with European Structural Integrity Society (ESIS) protocol. The nominal specimen thickness width (B) was 20 mm, the initial crack length (a_o) was 30 mm and the total specimen length was 120 mm as illustrated in **FIGURE 1.**

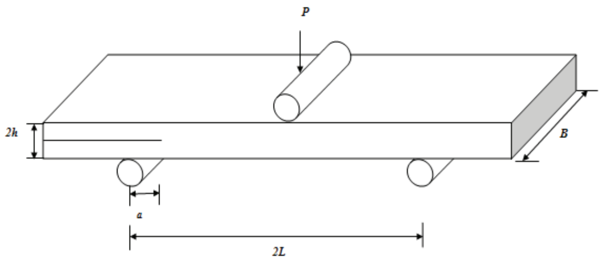

FIGURE 1 Schematic of the test configuration for the Mode II ENF Test.

Based on modified beam theory, the critical energy release rate, G_{IIC} was calculated using

$$G_{IIC} = \frac{9P\delta a^2}{2B(2L^3 + 3a^3)} \tag{1}$$

whereby G_{IIC} = the critical strain energy release rate, P = load, δ = displacement of the cross-head, a = effective crack length, B = width of the specimen and L = half distance between the supports.

The Mode II fracture tests were performed at a crosshead displacement rate of 1 mm/min using an Instron 4505 universal testing machine. The load-displacement data were measured using a 5 kN load-cell.

Mixed-Mode Interlaminar Fracture Testing

The mixed-mode I/II Interlaminar fracture properties of the composites was characterised using the mixed-mode flexure geometry (MMF) shown in **FIGURE 2.** This test is similar to the ENF test configuration, with the difference being that the load is applied to only one arm. The ratio of the length of the starter defect (a_o) to the half span (L) was fixed at 0.5. In this test, the load is applied at the mid-span, yielding a ratio of Mode I (G_I) to Mode II strain energy release rate (G_{IIC}) of 4/3.

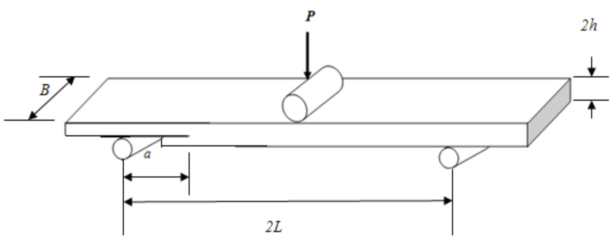

FIGURE 2 Schematic of test configuration for Mixed Mode Flexure Testing

The test was conducted at a cross-head displacement rate of 1 mm/min using an Instron 4505 screw-driven universal testing machine.

The mixed-mode critical strain energy release rate, $G_{I/IIC}$ was calculated via:

$$G_{I/IIC} = \frac{21P^2 a^2 C}{2B(7a^3 + 2L^3)} \quad (2)$$

where P is the applied load and C is the specimen compliance.

3.0 RESULTS AND DISCUSSION

FIGURE 3 shows representative Mode II load-displacement traces following tests at temperatures between 220°C and 300°C. The load increases in a linear fashion until a maximum value is reached, associated with crack propagation from the starter defect. From the plots, it is clear that the maximum load increases as a function of processing temperature, reflecting a gradual increase in the Mode II critical energy release rate with increasing processing temperature.

Over the temperature range of 220°C to 300°C, there is a continuous increase in the critical strain energy release rate, as shown in **FIGURE 4**. It is also evident that the value of G_{IIC} reaches a maximum at 320°C. Also, it is also clear that, for a given processing temperature, there is an increase in the critical strain energy release rate as the dwell time is increased, as is also shown in FIGURE 4. TABLE 2 summarizes the critical energy release rate for different temperatures and dwell. It is clear that the Mode II fracture toughness of the fully- consolidated composite is very high, being in excess of 3000 J/m^2. These high value are significantly above values for other thermoplastics, such as PEEK, and all thermosets, such as epoxy resins.

A plot of G_{IIC} as a function of the logarithm of dwell time is presented in **FIGURE 5**. From the FIGURE, there is evidence that there is a linear relationship between the critical energy release rate and the logarithm of log time. Similar observations have been reported by Jud *et.al* (1981) in the study of crack healing in PMMA and SAN polymers where it was observed that the fracture toughness, K_{Ii} increased with contact time, according to $K_{\mathrm{Ii}} \propto t^{1/4}$, at temperatures above the glass transition temperature, T_g , of the polymer.

In general, under Mode II loading conditions, this unidirectional carbon fibre reinforced poly ether imide (CF/PEI) exhibited an increase in G_{IIC} with increasing processing temperature and dwell time.

In **FIGURE 6**, it is evident that there is an increase in the value of $G_{I/IIc}$ with processing temperature, from 220°C up to 300°C for a constant dwell time of 5 minutes. It is interesting to note that the values of $G_{I/IIC}$ are much lower than the Mode II values, with the maximum value reaching only 1200 J/m^2. This suggests that this form of loading is more critical for these composites.

To understand the effect of dwell time on $G_{I/IIC}$, typical load-displacement traces are plotted for specimen processed at two temperatures (240°C and 300°C) at dwell times of 5 minutes and 60 minutes, in FIGURE 7. From the FIGURE, there is a noticeable increase in the maximum load at the temperature is increased and as the dwell time is increased from 5 to 60 minutes at a given temperature. A summary of the experimental results under Mixed-Mode I/II tests is given in TABLE 3.

FIGURE 8 shows typical R-curves for different temperatures at a dwell time of 5 minutes. From the plots, it is clear that there is an increase in the value of $G_{I/IIC}$ as a function of crack length. This is likely due to the presence of fibre bridging, along the crack path, as depicted in FIGURE 9.

FIGURE 10 presents a comparison of the critical strain energy release rate under Mode II shear loading and the results of Mixed-Mode I/II loading conditions. As reported above, it is clear that Mode II loading yielded higher values of G_c at the same temperature and dwell time, suggesting that there is an additional toughening mechanism under Mode II shear loading. These findings are in agreement with the work by Kim *et al.* (2004)

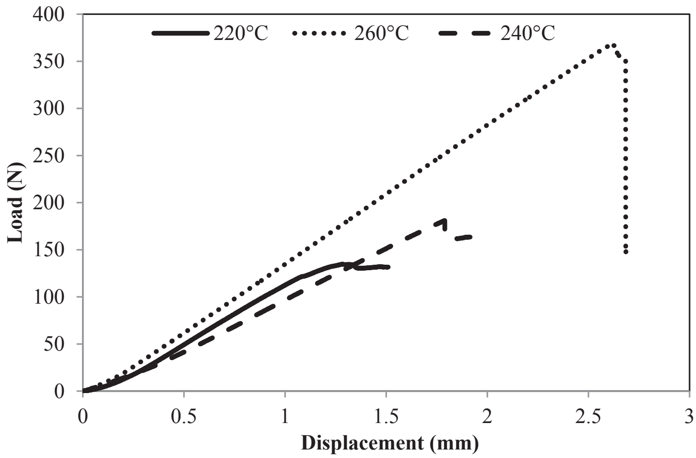

FIGURE 3 (a) Typical load-displacement traces for Mode II samples, processed at 220°C, 240°C and 260°C respectively with 5 minutes of dwell time.

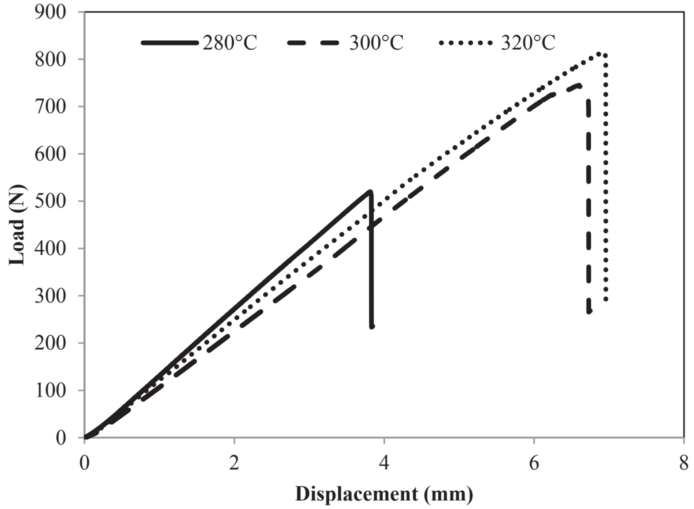

FIGURE 3 (b) Typical load-displacement traces, for Mode II samples processed at 280°C, 300°C and 320°C respectively with 5 minutes of dwell time.

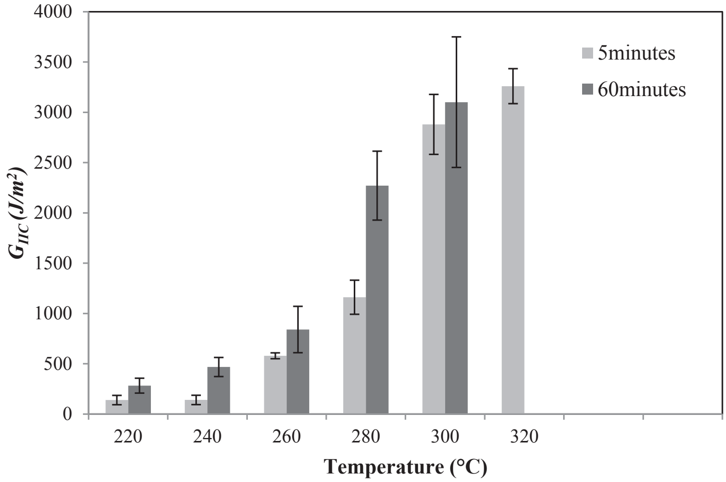

FIGURE 4 Plot of G_{IIC} as a function of processing temperature and dwell time.

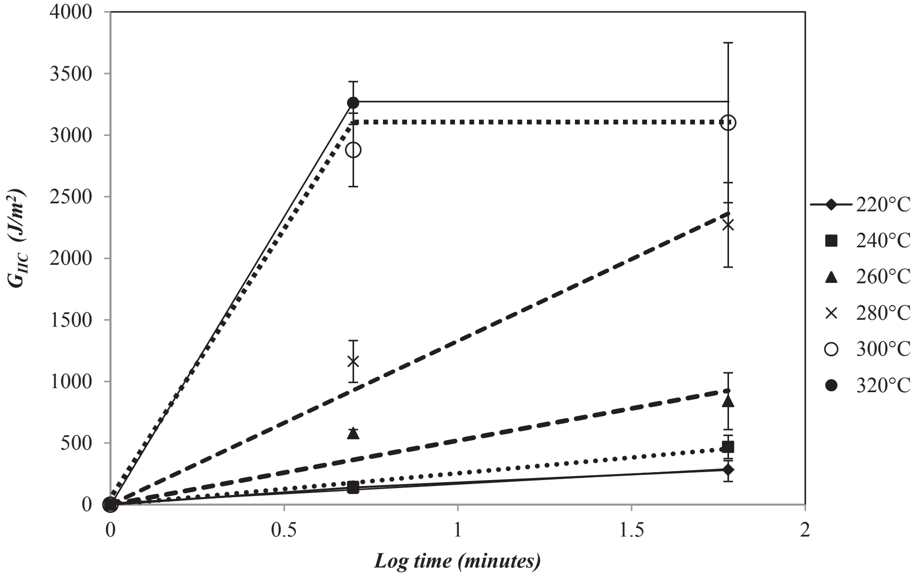

FIGURE 5 Plot of G_{IIC} versus Log time

TABLE 2 Average values of G_{IIc} for different processing temperatures and dwell times.

Temperature (°C)	Dwell time (mins)	G_{IIC} (J/m^2)
220	0	0
	5	139(46)
	60	283(75)
240	0	0
	5	14(46)
	60	468(95)
260	0	0
	5	580(30)
	60	840(231)
280	0	0
	5	1162(170)
	60	2271(343)
300	0	0
	5	2880(298)
	60	3101(649)
320	0	0
	5	3260(174)

Note:-
**The values in the bracket correspond to the standard deviations*

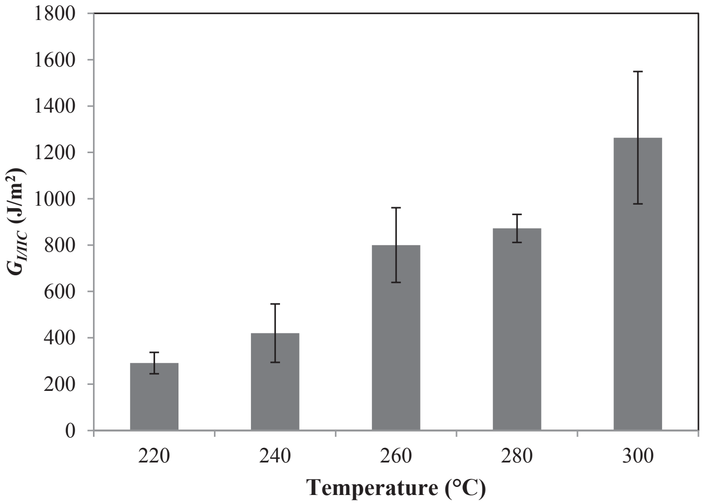

FIGURE 6 Plot of $G_{I/IIC}$ at different processing temperatures for the Mixed-Mode fracture specimens manufactured using a dwell time of 5 minutes

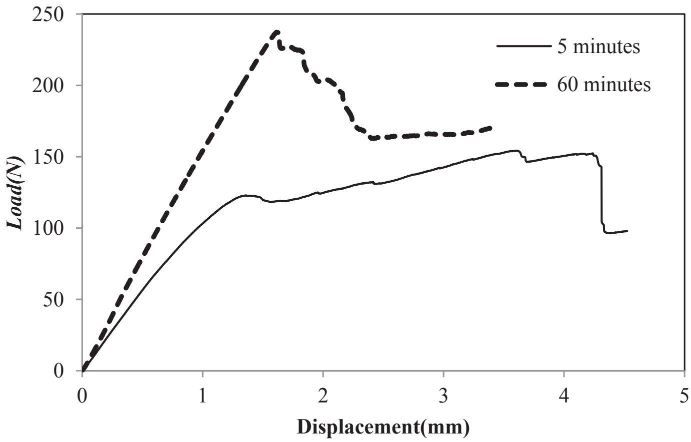

FIGURE 7 (a) Typical Mixed-Mode load-displacement traces following processing at 240°C with dwell times of 5 and 60 minutes.

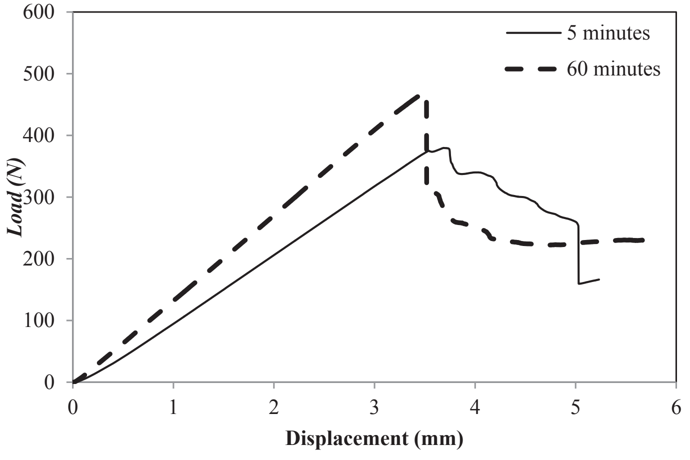

FIGURE 7 (b) Typical MMF load-displacement traces following processing at 300° with dwell times of 5 and 60 minutes.

TABLE 3 Average values of $G_{I/IIC}$ for Mixed Mode Interlaminar Fracture Tests at different processing temperatures and dwell times.

Processing temperature (°C)	Dwell time (minutes)	$G_{I/IIC}$ (J/m^2)
220	5	291 (46)
	60	509 (57)
240	5	420 (126)
	60	1191 (168)
260	5	800(161)
	60	686(69)
280	5	873(61)
	60	1088(211)
300	5	1264(310)
	60	1256(255)

Note:-
**The values in the bracket corresponds to the standard deviations*

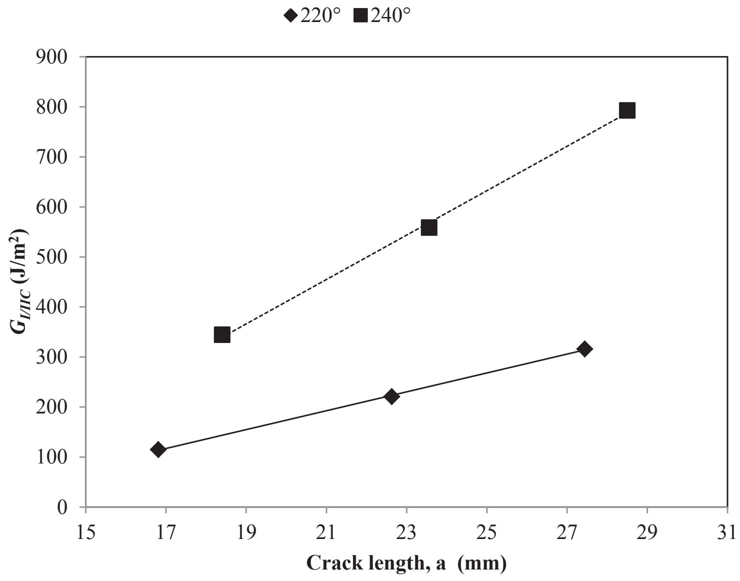

FIGURE 8 Representative R-curves for MMF test specimens processed at different processing temperatures, manufactured using a dwell time of 5 minutes.

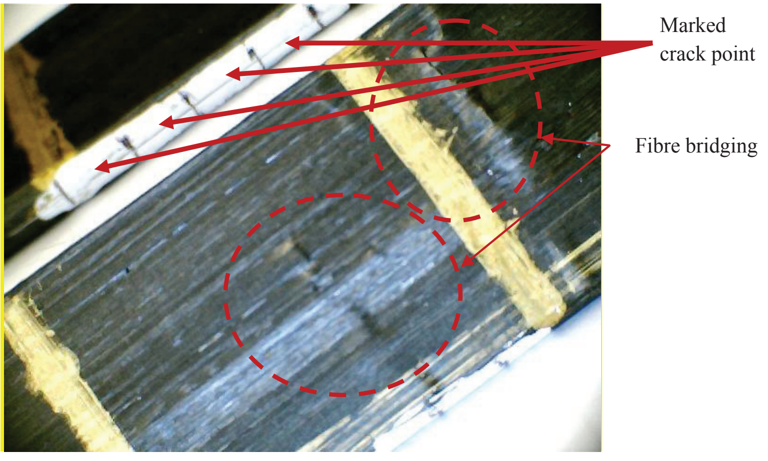

FIGURE 9 Photograph of a mixed-mode test specimen showing fibre bridging as the crack propagates from the initial crack to the mid span position.

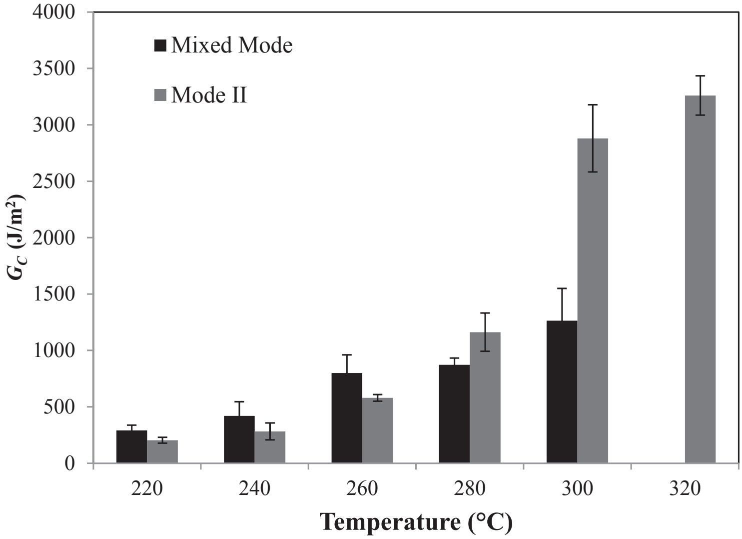

FIGURE 10 G_c versus processing temperature following both Mode II and Mixed-Mode I/II Interlaminar Fracture tests on specimens manufactured using a dwell time of 5 minutes.

4.0 CONCLUSIONS

An experimental study has been conducted to investigate the effect of varying the processing temperature on the interlaminar fracture properties of a carbon fibre reinforced polyether imide (CF/PEI). The tests have been conducted at four different repair temperatures above the glass transition temperature of the poly ether imide, PEI matrix (T_g of 217°C); these being 220°C, 240°C, 260°C and 300°C, all of which are.

Data from both end-notched flexure (ENF) as well as mixed-mode interlaminar fracture testing suggest that the optimum processing temperature in terms of achieving the required critical energy release rate, G_c is 300°C, with a dwell time of 60 minutes.

Further work will investigate the reparability of damaged CF/PEI following low-velocity impact, in which the optimum processing parameters identified in this current work will be considered. The test laminates will be repaired using a one-step compression moulding procedure using the hot press machine. The laminate stiffness values after impact as well as an effect of repair temperatures and time will be assessed via indentation tests. A detailed analysis of the repaired sites will be carried out using optical microscopy, looking at impact sites repaired at different temperatures, to investigate the presence of any residual damage after repair.

Overall, the findings of this research work suggest that it is possible to repair damage in fibre-reinforced thermoplastics, such as carbon fibre-reinforced poly ether imide (CF/PEI).

5.0 ACKNOWLEDGEMENTS

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