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DEVELOPMENT OF MULTISCALE TOUGHENING TECHNIQUE TO ADDRESS THE MICRO-CRACKING PROBLEM OF COMPOSITES AT CRYOGENIC TEMPERATURE

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ABSTRACT

In the current study, the fracture properties of angle-ply ($[\pm55]_8$) carbon-epoxy (unmodified) and nanosilica-carbon-epoxy laminated (nanosilica modified) composites were investigated. Double cantilever beam (DCB) tests were carried out in order to determine the interlaminar fracture energy in terms of critical energy release rate (GIc) of the laminated composites. Both the initiation and steady-state propagation value of the interlaminar fracture energy of the nanosilica modified laminated composites generally decreased when compared to unmodified laminated composites although a slight increase in average initiation values for 8wt% nanosilica modified laminated composites was observed. Nevertheless, the toughening effects of nanosilica on the laminated composites, compared with the unmodified laminated composites, were evident at LN_2 temperature. Indeed, increased positive toughening effect of the nanosilica was seen at LN_2 temperature especially for 8wt% nanosilica modified laminated composites when compared with unmodified laminated composites.

1 INTRODUCTION

Development of lightweight composite tanks for cryogenic liquid hydrogen are utmost important for affordable launch vehicles. However, it can be seen from the available research activities [1-4] that existing fibre reinforced polymer composites suffer from premature matrix cracking, due to the high thermal stress induced by the large discrepancy between the coefficients of thermal expansion of the matrix polymer and carbon fibres. This premature micro-cracking severely degrades the strength and gas permeability of composites, posing a significant challenge to the use of fibre composite fuel tank in launch vehicles.

Typical thermoset polymers in carbon fibre reinforced composites have significantly higher coefficients of thermal expansion than carbon fibres, causing high level of tensile thermal stress when operating at the cryogenic liquid hydrogen temperature. The same microcracking also occurs in rapidly cured composites for transport applications [1].

The objective of this study is to develop and demonstrate multi-scale toughening [2] technique to address the micro-cracking problem by incorporating nano-scale materials to simultaneously improve

the fracture toughness of polymer matrix and reduce the coefficient of thermal expansion. This new technique will lower the thermal residual stress and increase resistance to matrix cracking.

Adding very small amount of carbon-based nano-fillers (< 1 wt%), such as graphene [3, 4], carbon nanofibers [5], nano-silica [6, 7] and elastomeric particulate materials [8] can significantly improve the fracture toughness of the polymer and also the coefficient of thermal expansion. Different reinforcing and toughening mechanisms have been observed for these various types of filler materials. Hybridising different filler materials can generate synergistic effects that can reduce coefficient of thermal expansion, which in turn lowers thermal stress, and increase fracture toughness to minimize micro-cracking.

The current study develop and demonstrate multi-scale toughening technique to address the micro-cracking challenge by incorporating nanosilica to improve the fracture toughness of epoxy resin to increase resistance to matrix cracking in carbon fibre reinforced laminated composites.

2 EXPERIMENTAL DETAILS

2.1 Materials

An epoxy resin with amine based curing agent was used in the current study. The shelf life of the resin is greater than 24 months at 20°C with a gel time of 1h at 25°C. The Tg can be attained up to 110-120°C with a cure schedule of 25°C for 24 h and post cure at 120°C. The mix viscosity of the resin at 25°C is 400-600 mPa s.

Unidirectional (UD) carbon fibre (205 GSM Toray) was used. Nanosilica with an average particle diameter of 20 nm was obtained from Evonic Industries.

2.2 Methods

2.2.1 Mixing of nanosilica with epoxy resin

Nanosilica was manually mixed with epoxy resin by agitating with a stick for 10 minutes. Commercial CuO was mixed with epoxy resin using probe sonication. MWCNT was mixed with epoxy resin using bath sonication.

2.2.2 Production of carbon-epoxy and nanosilica-carbon-epoxy laminated composites

Carbon-epoxy and nanosilica-epoxy laminated composites were manufactured using vacuum infusion technique. For all the tensile and DCB test specimens, a $[\pm 55^{\circ}]_8$ layup architecture was chosen. All plies were cut into 250mm x 250mm long pieces and subsequently fibres peel ply, infusion mesh, and vacuum bag were cut and fitted to the plies' size. Previously the glass table was waxed to prevent the laminate from sticking to the table.

When aiming for 4 wt% nanosilica in the epoxy resin, 20g out of 200g epoxy resin needed to be replaced with nanosilica. For making 8wt% and 12wt% 40g and 60g of epoxy resin, respectively was replaced. After polishing the lamination table and putting the release agent, resin infusion mesh was placed on the table followed by a peel ply, all 16 layers of the carbon fabric, another peel ply and another infusion mesh at the top above which the vacuum bag was placed. A 70mm wide separating Teflon film (25 µm thick) was perpendicularly inserted after the 8th layer that would later serve as pre-crack for the specimen. After placing all 16 layers in $[\pm 55^{\circ}]_8$ fibre architecture, the vacuum bag was sealed and tested for any leaks by applying the full vacuum and switched it off for at least 30 minutes. After preparing the vacuum bag the epoxy resin or epoxy resin and nanosilica mixture was mixed and degassed for 30 minutes. The epoxy resin or the nanosilica-epoxy mixture was then infused slowly using a vacuum of 1 atmospheric pressure. The laminate was then left for curing at RT for 24 hours. After 24 hours the laminate was post cured at 120°C for 2 hours. Figure 1 shows the resin infusion process and the infused composite panels.
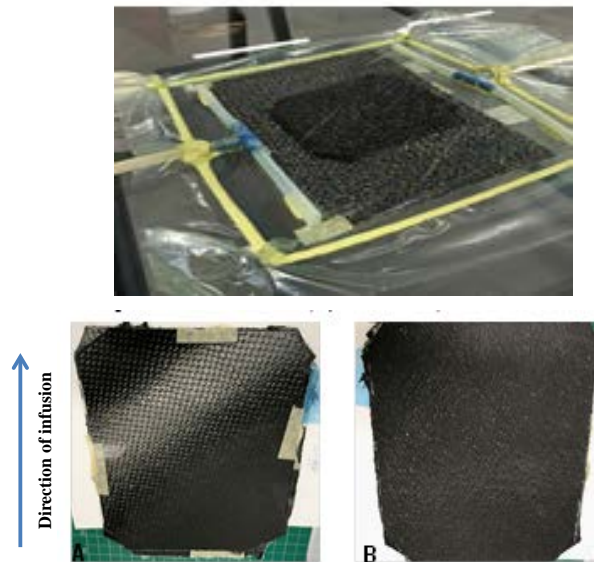
Figure 1. Resin infusion process and the infused composite panels (A) top face and (B) bottom face.

2.2.3 Cutting of the Laminated Composites

All the laminated composites were cut using a multicam CNC router. From each laminated composite, 6 specimens with a dimension of 200 mm x 20 mm were cut.

2.2.4 DCB Testing of the Laminated Composites

As shown in Figure 2, composite specimens were first attached with loading blocks as per ASTM D5528-13 and gripped the loading blocks using tensile testing grips as shown in DCB testing set up in the figure. All the DCB testing of the composites were carried out using a crosshead speed of 1 mm/min.

Figure 2. Composite DCB specimens attached with loading blocks and the loading blocks gripped through pins with the Instron tensile tester.

2.2.5 Cryogenic test arrangements of the laminated composites

Figure 3 shows a manual container built out of stainless steel for cryogenic testing. There are four containers which are arranged in way that two small containers (160 mm x 100 mm x 100 mm) seat on two large containers (200 mm x 150 mm x 150 mm). The gap between the small containers and large containers are filled using polyurethane insulation. The free volume after sitting the small containers

inside the large containers is filled with liquid nitrogen during cryogenic testing. The bottom grip of Instron tensile testing machine is attached through the containers into the top small container where the DCB specimen is attached and during testing this open area of the small container chamber is poured with liquid nitrogen up to about 5 mm above the surface of the DCB specimen so that the crack propagation can be visualised and pictured.

Figure 3. The cryogenic test setup for DCB testing.

3 RESULTS AND DISCUSSION

DCB testing was performed to see whether addition of nanosilica has any effect on the interlaminar fracture energy. Figure 4 shows the typical load displacement curve obtained from DCB testing at RT and cryogenic LN_2 temperature. From the curves, it can be seen that LN_2 temperature makes the composite less tough.
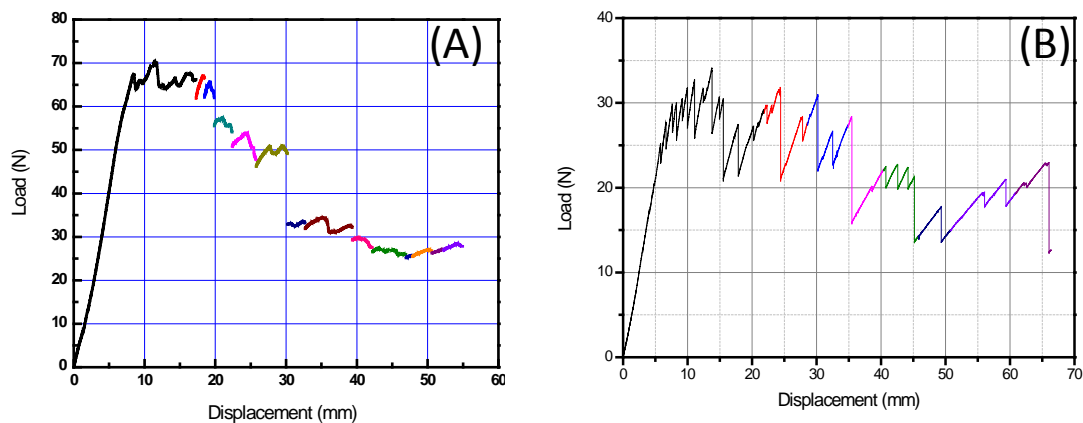

Figure 4 Typical load displacement curve obtained from DCB testing at (A) RT and (B) LN_2 temperature.

Figure 5 shows the R curves for carbon-epoxy and nanosilica-carbon-epoxy laminated composites at RT and LN_2 temperature. It can be seen from the results that the addition of nanosilica does not improve the G_{Ic} values of the laminated composites compared to carbon-epoxy (unmodified) laminated composites at RT. It can also be seen that LN_2 temperature reduces the G_{Ic} values of the unmodified composites more than the nanosilica modified laminated composites. The best retention of G_{Ic} values was seen for 8wt% nanosilica modified laminated composites.
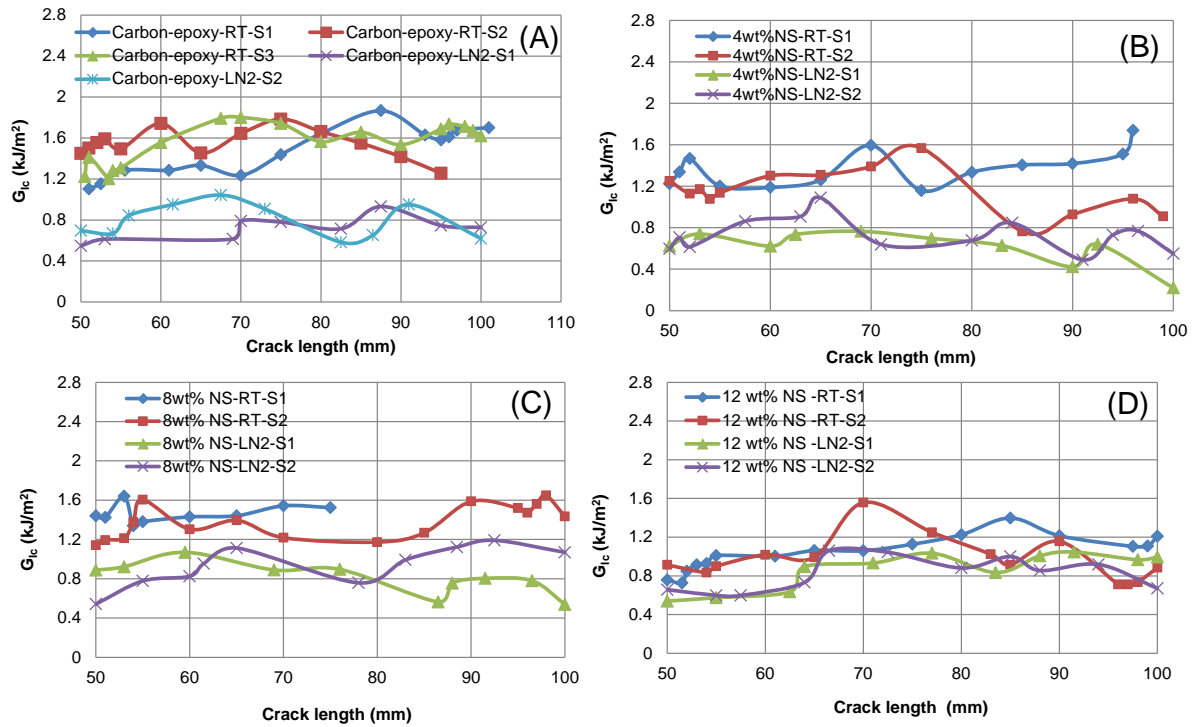
Figure 5. R curves for (A) carbon-epoxy and (B-C) nanosilica-carbon-epoxy laminated composites at RT and LN$_2$ temperature where (B) 4 wt% nanosilica (B) 8 wt% nanosilica and (C) 12 wt% nanosilica.

From Figure 6 initiation and steady state critical energy release rate (G_{Ic}) of the unmodified and nanosilica modified laminated composites at RT and LN$_2$ temperature can be seen. At RT, the initiation G_{Ic} values generally decreased for nanosilica modified laminated composites when compared to unmodified laminated composites. However, 8wt% nanosilica modified laminated composites had a slight increase in initiation values. On the other hand, the steady state G_{Ic} values decreased for nanosilica modified laminated composites when compared to unmodified laminated composites and the decrease was lowest for 8 wt% nanosilica modified laminated composites. At LN$_2$ temperature, for 8 wt% nanosilica modified laminated composites the G_{Ic} values increased about 14% of both initiation and propagation state when compared to unmodified laminated composites.
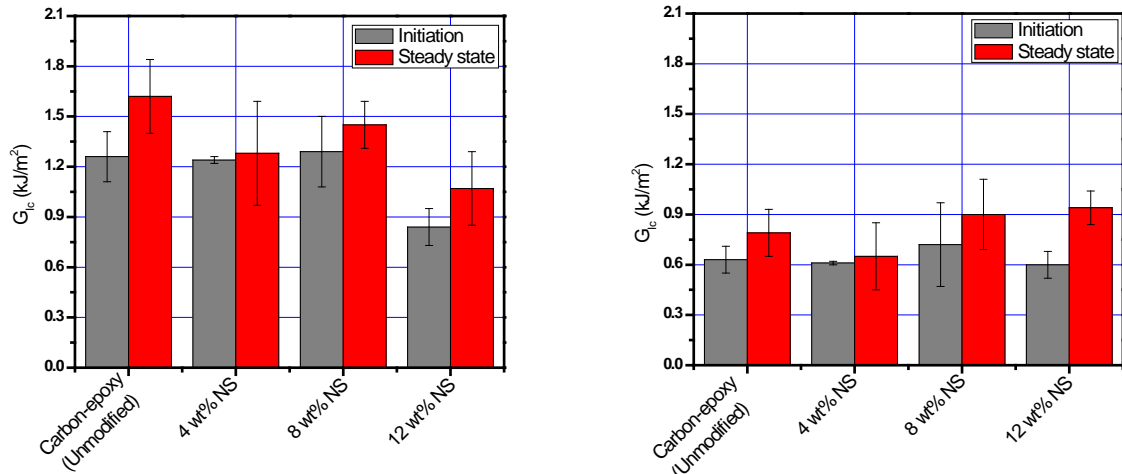
Figure 6. Initiation and steady state critical energy release rate (G_{Ic}) of the laminated composites at (A) RT and (B) LN$_2$ temperature.

4 CONCLUSIONS

DCB testing was carried out to measure the interlaminar fracture energy (G_{Ic}) of angle-ply ($[\pm55]_8$) unmodified and nanosilica modified laminated composites. From this study, it was observed that the initiation and steady-state propagation value of the interlaminar fracture energy of the nanosilica modified laminated composites generally decreased in comparison to unmodified laminated composites. However, the toughening effect was evident at LN$_2$ temperature for nanosilica modified laminated composites compared to unmodified laminated composites. About 14% increase of G_{Ic} values both initiation and steady state propagation state was seen at LN$_2$ temperature for 8wt% nanosilica modified laminated composites when compared with unmodified laminated composites.

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