DELAMINATION TOUGHENING AND HEALING PERFORMANCE OF 3D WOVEN COMPOSITES

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ABSTRACT

This paper presents an experimental investigation of a novel three-dimensional (3D) woven fibre-polymer composite material that has the unique combination of properties to both resist and self-repair delamination cracks. The hybrid 3D woven composite contains a combination of through-the-thickness z-binders made of carbon tows (used for high delamination toughness) and thermoplastic filaments (used for self-healing). The delamination toughness and self-healing properties of 3D woven composites reinforced separately or concurrently with z-binders made of carbon tows and thermoplastic filaments are investigated. The hybrid 3D woven composites investigated here demonstrate large improvement to the mode I interlaminar fracture toughness (~1200%) with the unique capability of self-repairing internal damage.

1 INTRODUCTION

In the absence of through-the-thickness reinforcements, fibre reinforced polymer composites are susceptible to delamination damage [1]. An effective method to improve the delamination toughnness of composites is through-the-thickness (z-binder) reinforcement using carbon, glass, aramid or other structural fibres inserted using stitching, tufting, z-anchoring, z-pinning or three-dimensional (3D) weaving [2]. However, a long standing issue with through-the-thickness reinforced composites is the high-cost associated with the repair of damage caused by impact, over-loading, fatigue loading, and environmental degradation [3]. This paper presents an experimental investigation into a novel 3D woven fibre reinforced polymer composite that combines high delamination toughness with the capacity to self-repair cracks through the use of a hybrid combination of z-binders.

2 METHODOLOGY

2.1 Composite materials and manufacturing process

The composite materials used in this study were fabricated using a 200 gsm plain woven T300 carbon fabric (AC220127 supplied by Colan Ltd). The fibre preforms consisted of 34 plies of the carbon fabric that were manually woven in an orthogonal pattern using one or two types of z-binder materials: carbon fibre tows and EMAA (poly[ethylene-co-(methacrylic acid)]) filaments. The carbon z-binder tow was 800 tex (12K) roving (Tenax® STS40) with a nominal diameter of ~0.75 mm. The carbon fibres within the z-binder have a Young's modulus of 240 GPa and average tensile strength of 4 GPa, as reported by the supplier. The EMAA z-binder was produced from fused pellets of Nucrel® (Dupont Packing and Industrial Polymer), which is an ethylene acid copolymer containing 19% by weight of methacrylic acid randomly distributed along the EMAA polymer chain. The EMAA z-binder was originally rectangular in cross-section (1.0 × 2.0 mm) with a tensile modulus and ultimate strength of 3 GPa and 16 MPa, respectively [4].

The orthogonal through-thickness weaving of the carbon tows and EMAA filaments was performed along the warp direction of the carbon fabric preforms in straight, parallel rows, as shown in Fig.1. Three types of 3D woven composite were made containing z-binders made of (i) carbon only, (ii) EMAA only, or (iii) combination of carbon and EMAA. Irrespective of the type of 3D woven composite, the spacing between the z-binders both along and between the rows was 10 mm and 5 mm, respectively. The weave density (i.e. number of z-binders per unit area) and the weave pattern (orthogonal) for the three types of 3D woven composite was similar. However, the weave areal content (i.e. percentage area of delamination crack plane occupied by z-binders) for the 3D woven composites was different due to the difference in the cross-section area of the carbon tows and EMAA filaments used for the z-binders. The weave density for the 3D woven composites containing z-binders made of carbon only and EMAA only was 0.7 vol% and 3.2 vol%, respectively. The hybrid 3D woven composite contained a 50-50 weave density of the carbon and EMAA z-binders (i.e. 0.35 vol% carbon and 1.6 vol% EMAA).

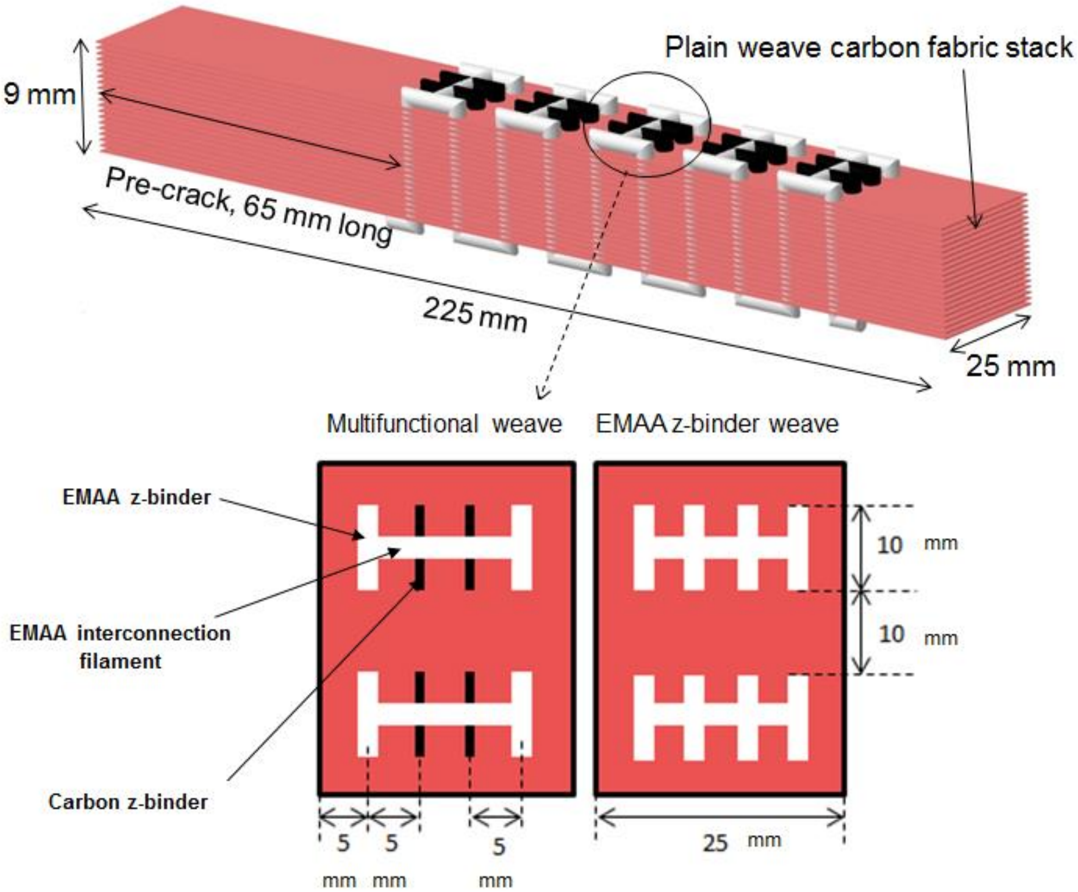

Figure 1: Schematic illustrations of the hybrid 3D woven composite in the double cantilever beam (DCB) specimen form which were used for mode I interlaminar fracture toughness testing.

An important design feature for the 3D woven composites containing EMAA z-binder (either alone or in combination with carbon) was that the neighbouring rows of the weave were interconnected by horizontal EMAA filaments placed near the surface, as shown in Fig.1. These interconnecting filaments were used to ensure sufficient supply of EMAA into delamination cracks during healing. Two additional plies of the carbon fabric were placed on both the top and bottom surfaces of the 3D woven fabrics to cover the weaves and prevent leakage of EMAA during high temperature curing and healing.

A 2D laminate without any z-binder, but with the same number of carbon fibre plies was used as the control material to bench-mark the delamination fracture properties of the 3D woven composites. The 2D and 3D fabrics were infused with epoxy resin at room temperature using the vacuum bag resin infusion (VBRI) process. The epoxy was a diglycidyl ether of bisphenol A (DEGBA) resin (SR8100) and diamine hardener (SD8824) supplied by Sicomin®. The composites were cured under pressure exerted by the vacuum bag for 24 h at room temperature, and then post-cured for 8 h at 60°C, in accordance with the epoxy supplier's recommendations.

2.2 Delamination fracture toughness testing procedure

The mode I delamination properties of the 2D and 3D woven composites were measured using the double cantilever beam (DCB) test. The DCB specimens were 225 mm long and 25 mm wide rectangular-shaped coupons. One end of the DCB specimen contained a 65 mm long and 25 μm thick polytetrafluoroethylene (PTFE) pre-crack along the mid-plane to initiate the delamination during testing. The fracture toughness tests were conducted under displacement control using a 10 kN load capacity Instron machine. The toughness was measured by applying a monotonically increasing crack opening displacement at a rate of 2 mm/min to the pre-cracked end of a DCB specimen. The delamination was grown along the specimen mid-plane in short increments, and the crack length was measured using a travelling optical microscope. The mode I interlaminar fracture toughness was calculated using the modified beam theory in accordance with ASTM Standard D5528. At least four specimens were tested for the 2D laminate and three different types of 3D woven composite.

The effect of the crack bridging traction behaviour of the z-binders on the interlaminar toughening effect for the 3D woven composites was determined by transverse tension tests. This test is often used to determine the bridging traction law for composites reinforced in through-the-thickness direction using stitches or z-pins. The transverse test specimens were manufactured using the same materials and process described earlier. The rectangular-shaped specimens were 30 mm long and 20 mm wide, and consisted of two halves of the 3D woven composite separated at the mid-plane by a thin PTFE film. Bridging the two halves were carbon or EMAA z-binders. The specimens contained 8 z-binders with a similar areal density to that used for the DCB specimen. The specimens were loaded at a constant cross-head displacement rate of 1 mm/min until all the z-binders had failed. A crack opening displacement (COD) gauge was used to measure the displacement between the two halves of the tension test specimen. Five samples from each of the 3D woven composites were tested under identical conditions.

3 RESULTS AND DISCUSSION

3.1 Fracture toughness properties

The effect of the woven z-binder material on the mode I crack growth resistance (*R*) curve of the 3D woven composites is shown in Fig.2. The *R*-curve for the 2D woven laminate is also shown, and its interlaminar fracture toughness remained low and constant with increasing crack length. By contrast the 3D woven composites showed rising *R*-curves during the initial phase of delamination crack growth and then reached a quasi-steady-state fracture toughness (G_{Ic}) value, which are given in Fig. 2b. The z-binders in the 3D woven composites did not increase the strain energy release rate for crack initiation, although they were effective at increasing the steady-state fracture toughness. Despite having the lowest areal density, the z-binders made of carbon fibre had the strongest delamination toughening effect whereas the EMAA (which was present in a higher content) was the least effective at toughening. The hybrid combination of z-binders consisting of both carbon and EMAA had an intermediate toughening effect.

The *R*-curve for the 3D woven composite containing only the carbon z-binders increased rapidly over the initial 25 mm of delamination crack growth, reaching a G_{Ic} value ~2000% higher compared to the 2D laminate. The rising *R*-curve and the improvement to G_{Ic} for this composite were due to the formation of a bridging zone by the carbon z-binders behind the crack front, as shown by cross-sectional X-ray computed tomography (CT) in Fig.3. Due to their high stiffness and strength, the carbon z-binders were highly effective at generating a high crack bridging traction load which provided the large improvement to the fracture toughness. As shown in Fig.3a-b, the carbon z-binders behind the crack front initially failed by interfacial debonding from the composite. The debonding resulted in the redistribution of bridging traction stress along the carbon z-binders, which caused them to fracture due to the localised geometric stress concentrations present at the tight bend radii of the z-binder (see Fig.3b). With further increase in crack opening displacement, the fractured ends of the carbon z-binders pulled-out of the 3D woven composite, dissipating energy through frictional sliding, and thereby increasing the interlaminar fracture toughness.

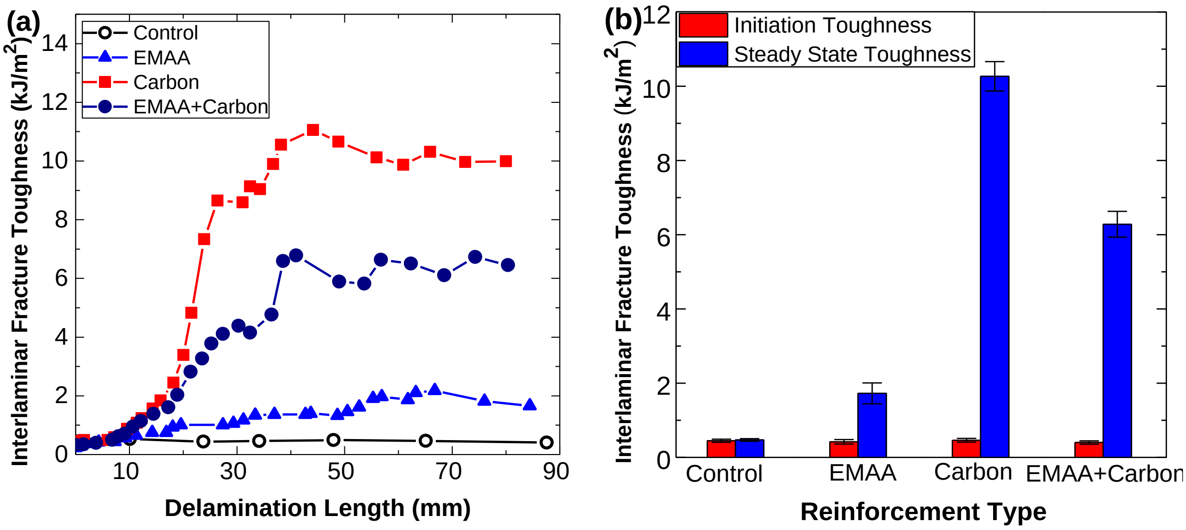

Figure 2: (a) Crack growth resistance (*R*-) curves for the 2D and 3D woven composites. (b) Mode I interlaminar fracture toughness values for the initiation and propagation of the delamination in the 3D woven composite laminates.

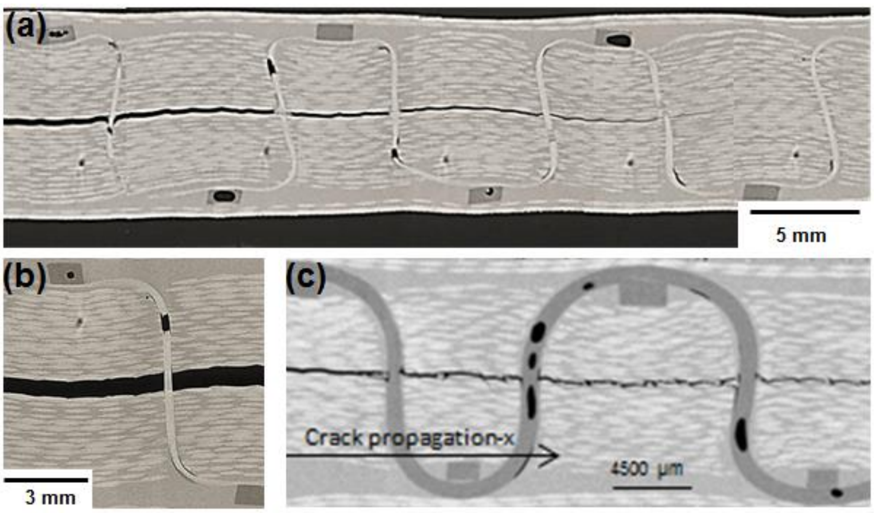

Figure 3: X-ray computed tomography images of the crack bridging mechanisms in 3D woven composites reinforced with (a-b) carbon and (c) EMAA z-binders.

The *R*-curve for the 3D woven composite containing only EMAA z-binders increased gradually with increasing delamination crack length. This toughening was due to the EMAA z-binders forming a large-scale bridging zone behind the crack front, as seen in Fig. 3c, which was similar to that formed by the carbon z-binders. The EMAA z-binders increased the G_{Ic} value of the 3D woven composite by ~250%. The improvement was primarily due to the EMAA z-binders within the crack bridging zone dissipating the stored strain energy via large-strain plastic yielding.

The *R*-curve for the hybrid 3D woven composite containing z-binders of both carbon and EMAA increased rapidly with the crack length and reached a G_{Ic} value ~1200% greater than the 2D laminate. This improvement was due to crack bridging promoted by the carbon and EMAA z-binders, as discussed earlier. The improvement of ~1200% represented an additive toughening effect as would be expected for this hybrid 3D woven composite containing half the volume fraction of each type of z-binder.

The effect of the z-binder reinforcements on the crack bridging traction behaviour was investigated further using pull-out testing, and the results are shown in Fig.4a. In this figure, the area under the load versus crack opening displacement curve represents the traction energy, *E*, dissipated during the z-binder crack bridging process. This energy can be used to estimate the improvement to the steady-state fracture toughness using the following relationship [5]:

$$G = \sum_{i=0}^{n} \frac{V_{fi} E_i}{A_i} \tag{1}$$

where A and V_f are the cross-section area and the volume fraction of the z-binders, respectively. Fig.4b shows the fracture toughness values calculated using Eq. (1), and there is agreement with the toughness values measured experimentally for the three types of 3D woven composite. Although the closed-form analytical solution in Eq. (1) can be used to calculate the mode I steady-state fracture toughness of the 3D woven composites, it cannot be used to compute the rising behaviour of the *R*-curve associated with the evolution of the z-binder bridging (although this can be done using finite element analysis).

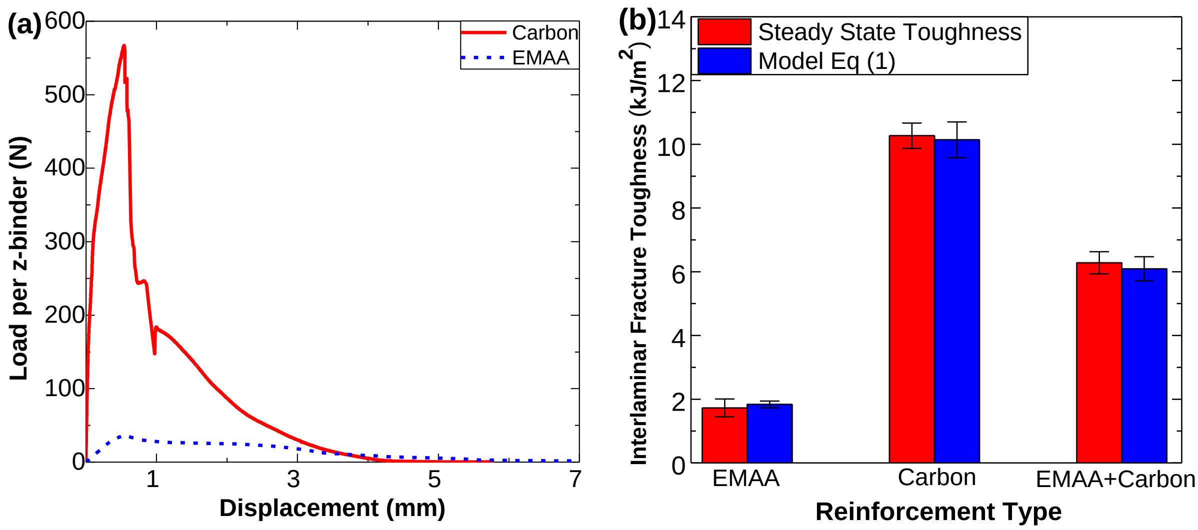

Figure 4: (a) Effect of z-binder material on the traction-load crack opening displacement of the 3D woven composites. (b) Comparison of the calculated (using Eq. (1)) and measured steady-state toughness values for the 3D woven composites.

3.2 Healing of z-binder reinforced composites

Healing of the delamination crack created by the mode I fracture toughness test was performed by heating the DCB specimens at 150°C for 30 minutes, which is reported to be the optimum healing condition for EMAA [6]. The mode I fracture toughness of the 3D woven composites was then remeasured at room temperature to determine the healing efficiency. The healing efficiency for the 3D woven composites containing z-binders of EMAA only or in combination with carbon was defined by the recovery of the fracture toughness of the healed material compared to its original toughness value. The process of delamination crack growth followed by healing was repeated three times on the 3D woven composites to evaluate the reusability of the EMAA z-binders for multiple healing operations. The recovery to the interlaminar fracture toughness values for the 3D woven composites after each healing cycle are given in Fig.5. Full recovery of the interlaminar fracture toughness was achieved for the 3D woven composite containing only EMAA z-binders. Similar findings have been reported for carbon-epoxy laminates reinforced with stitched EMAA filaments [8,9].

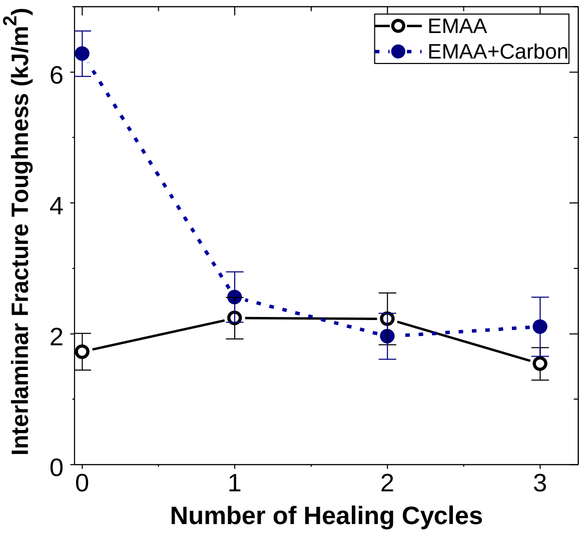

Figure 5: Effect of number of healing cycles on the fracture toughness of the 3D woven composites with EMAA z-binders only or a combination of carbon and EMAA z-binders.

Healing was enabled by the EMAA z-binders transforming from solid woven filaments into a molten state. Healing at 150^{o}C transformed the EMAA z-binders, which have a melt temperature of ~88^{o}C, into a molten state which allowed them to flow into the delamination cracks as shown in Fig.6a-b. The flow of EMAA melt into the delamination was assisted by a pressure delivery mechanism unique to this thermoplastic in the presence of epoxy resin. At the healing temperature of 150^{o}C, acid groups along the surface of the EMAA z-binders undergo a condensation reaction with amine groups in the epoxy matrix of the composite. Water is created as a by-produce of this reaction, which has a low solubility limit in EMAA [7]. Consequently, the water phase-separates within the EMAA, and due to the high temperature generates a high internal pressure onto the molten EMAA which forces some of this thermoplastic to flow into the delamination crack. The evidence of this mechanism is the micro-bubbles formed within the EMAA z-binders, as shown in Fig.6c. This high pressure delivery mechanism further improves the healing efficiency, since it forces the EMAA z-binder material that is much further away from the fracture plane to flow into the delamination crack as shown in Fig.6b. Upon cooling to room temperature, after the healing operation, the EMAA solidifies and adheres via hydrogen bonding to the composite material along the crack plane. The delamination was healed by the flow of EMAA into the crack and the 3D orthogonal weave pattern was maintained by the broken ends of the EMAA z-binders fusing together (as shown in Fig.6b).

Growing again the delamination crack in the 3D woven composites containing EMAA z-binders by repeated DCB tests demonstrated a high recovery to the fracture toughness (Fig.5). Melting and flow of EMAA created a thin adhesive layer consisting of ductile thermoplastic material spread along the site of the healed delamination. Under repeated crack growth the EMAA along the delamination formed a crack bridging zone consisting of a network of thin ductile ligaments between the z-binders (Fig. 7a). In addition, the refused EMAA z-binders also created a large-scale bridging zone along the delamination which was similar to the bridging zone in the original composites prior to healing (see

Fig.3c). The crack bridging zone consisting of the thin EMAA ligaments and refused thicker EMAA z-binders generated traction loads along the delamination which reduced the applied stress at the crack tip, thereby recovering the fracture toughness of the healed 3D woven composites. The EMAA ligaments and refused EMAA z-binders reformed within the crack bridging zone after each healing cycle, and this was responsible for the high retention of the fracture toughness with an increasing number of repairs.

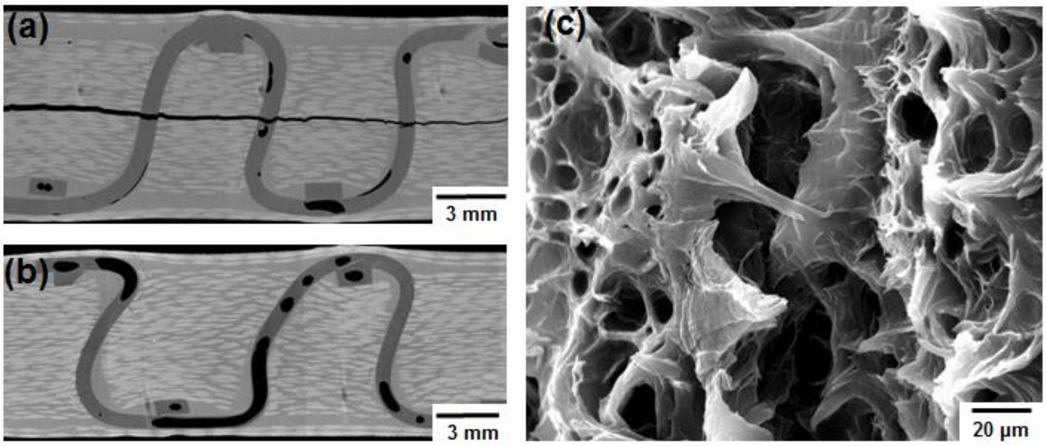

Figure 6: X-ray computed tomography of the 3D woven composites showing the EMAA weave morphology in the (a) delaminated and (b) healed laminate. (c) SEM image showing the highly porous structure of EMAA caused by bubble formation during the healing process.

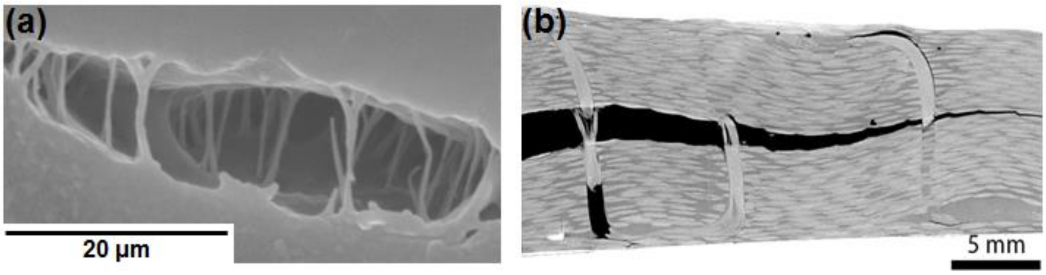

Figure 7: (a) SEM image of the side view of the delamination crack in the hybrid 3D woven composite showing crack bridging by the EMAA ligaments. (b) X-ray computed tomography of the hybrid 3D woven composite showing the damage to the carbon z-binders.

A similar healing mechanism promoted by EMAA was also observed for the hybrid 3D woven composite reinforced with z-binders consisting of EMAA and carbon fibres. However, only partial recovery of the original fracture toughness was achieved for this hybrid 3D woven composite. The fracture toughness was restored by healing to ~35 to 40% of the original toughness of the 3D woven composite. This healing efficiency was independent of the number of healing cycles. Despite this partial recovery, the fracture toughness of the hybrid 3D woven composite (with low EMAA content of 1.6%vol) after healing was similar to that for the 3D composite containing only EMAA z-binders (with high EMAA content of 3.2%vol). The recovery to the fracture toughness of the hybrid 3D woven composite was solely due to the toughening promoted by the EMAA, which is expected. Indeed, very little contribution to the crack bridging and toughening from the carbon z-binders was observed for the hybrid 3D woven composite after healing. As shown in Fig.7b, the carbon z-binders were damaged during the unloading and closing of the delamination crack prior to the healing process. The damaged carbon z-binders were incapable of generating high bridging traction loads that promote the fracture toughness of the original hybrid 3D woven composite material. Thus, in the absence of the toughening contribution from the carbon z-binders, only partial recovery of the fracture toughness of the hybrid 3D woven composite was achieved.

4 CONCLUSIONS

This study has demonstrated the effects of combining z-binder materials on the delamination resistance and self-repair functionality of hybrid 3D woven composites. Compared to the 2D woven laminate, through-the-thickness weaving using carbon or EMAA z-binder increased the mode I fracture toughness of the 3D woven composites by 2000% and 250%, respectively. In comparison, the hybrid 3D woven composite containing both carbon and EMAA z-binders increased the fracture toughness by 1200%. In this hybrid 3D woven composite, the carbon z-binders promote high delamination toughness of the original material (before healing), whereas the EMAA z-binders promote the healing of the delamination crack and partial restoration of the fracture toughness. The hybrid 3D woven composite materials investigated here has high delamination resistance found with conventional 3D woven material together with the unique capability of repairing internal damage.

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