Interlaminar Fracture Micro-Mechanisms in Toughened Carbon Fibre Reinforced Plastics Investigated via Synchrotron Radiation Computed Tomography and Laminography

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Abstract

Synchrotron Radiation Computed Tomography (SRCT) and Synchrotron Radiation Computed Laminography (SRCL) permit 3D non-destructive evaluation of fracture micro-mechanisms at high spatial resolutions. Two types of particle-toughened Carbon Fibre Reinforced Polymer (CFRP) composites were loaded to allow crack growth in Modes I and II to be isolated and observed in standard and non-standard specimen geometries. Both materials failed in complex and distinct failure modes, showing that interlaminar fracture in these materials involves a process zone rather than a singular crack tip. The work indicates that incorporating particle/resin, fibre/interlayer and neat resin failure is essential within models for material response, since the competition between these mechanisms to provide the energetically favourable crack path influences the macro-scale toughness. The work uniquely combines the strengths of SRCT and SRCL to compare failure micro-mechanisms between two specimen geometries, whilst assessing any edge effects and providing powerful insight into the complex micro-mechanical behaviour of these materials.

Keywords: A - Polymer-matrix composites (PMCs); A - Particle-reinforcement; B - Delamination; D - Non-destructive testing

1 General Introduction

The high specific stiffness and strength of CFRPs has led to their use in aerospace applications, where a reduction in weight has a direct impact on the payload and range of the aircraft. However, composites may suffer from low velocity impact damage whilst in service that can have a significant effect on the residual mechanical properties. Such events may cause significant internal damage in the form of delaminations, which are difficult to identify from surface inspections and may reduce compressive properties by up to 60% [1]. The need to resist impact damage, and the lack of reliable predictive models to account for impact and post-impact performance, contributes to the over-engineering of composite structures and a failure to achieve the desired weight-saving and consequent performance improvement. Given that Mode I and Mode II dominated loading conditions have been identified to occur under low velocity impacts [2], modelling such Mode I and II fracture is a key first step in developing models for impact damage resistance and post impact damage tolerance.

Incorporation of secondary phase particles into the polymer matrices has been identified as an effective way of increasing the matrix toughness. Key toughening mechanisms have been identified as; crack pinning [3], crack path deflection [3,4], particle/matrix de-bonding and

subsequent void growth[5], localized shear yielding [6] and bridging of the crack surfaces by the particles [7]. However, the relevance of such mechanisms varies between specific matrix/particle combinations, where, for example, it has been shown that the size and stiffness of the particles significantly influences the ability of the crack to be pinned or to deflect the crack path appreciably [4,8]. Toughness studies have been conducted, by systematically varying particle volume fractions, and particle stiffness and size, in which orders of magnitude of improvement in bulk toughness were shown to be possible by controlling these parameters [3,9,10]. The incorporation of such matrices between the plies of a composite, called interlayering, has been shown by several authors to increase the interlaminar toughness[11]–[17]. However, it has been established that an improvement in bulk resin properties does not directly translate to an improvement in interlaminar toughness, and this has been assumed to be due to the constraining action from the neighbouring plies [11,12]. Furthermore, increasing interlayer toughness can be ineffective if the cracks propagate by avoiding the toughened interlayer [18,19], which was observed in one of the systems investigated in this work.

Damage inside a composite material can be made visible via x-ray computed tomography (CT) using laboratory [20,21] and synchrotron sources [22–25] with specimen geometries adapted to the CT scanning technique. This permits *in situ* observations of toughening mechanisms to be made using non-invasive techniques away from free-edges under representative stress states. This has recently become feasible due to the development of Synchrotron Radiation Computed Laminography (SRCL)[21,26–29]. The experiments documented in this paper represent the first to capture *in situ* toughening mechanisms operating in Mode I interlaminar cracks in particle-toughened composite laminates, tested in both standard and non-standard geometries. They permit the direct identification of micro-mechanical features and mechanisms that affect the global Mode I fracture toughness. Mode II identification of micro-mechanisms was conducted *ex situ*, but the SRCL and SRCT techniques still provided invaluable information without changing the stress state on the sectioning plane that may introduce out-of-plane displacements. The controlled loading conditions and image quality permitted the identification of the sequence of local damage evolution, emphasizing the influence of local microstructural irregularity on the location and geometry of damage initiation and growth.

2 Methodology

2.1 Materials

CFRP test coupons, provided by Cytec Engineered Materials, were manufactured from developmental particle-toughened material systems. The toughening was confined to a $\sim 30\text{ }\mu\text{ m}$ thick particle-toughened interlayer in each system. The primary reinforcement was a proprietary intermediate modulus carbon fibre ($\sim 5.4\text{ }\mu\text{ m}$ in diameter). A 16 ply (3mm thick) uni-directional layup was prepared from pre-preg with a $40\text{ }\mu\text{ m}$ thick Polytetrafluoroethylene (PTFE) insert in the mid-plane of the sample in order to control the initiation of fracture. Materials were laid up by hand and cured by the manufacturer according to a standard aerospace autoclave cure cycle. Two

different particle types were investigated; with the fibre type, sizing, base resin and particle volume fraction remaining constant between the material systems. Material A (Mat. A) can be identified with the visible particles in the figures, and Material B (Mat. B) in which the constituent material density of the particles is too similar to the resin to be identifiable via CT. The composition of the materials is proprietary and is not important to the key observations and conclusions of this paper. The two systems demonstrated significantly different fracture micro-mechanisms and fracture toughness, highlighting the importance of understanding micro-mechanical behaviour to develop an effective interlayer.

2.2 Specimen Geometries

To take full advantage of SRCT and to avoid artefacts, a cross-sectional geometry of 3x1mm (Figure 1(a)) was chosen to maximise the transmission of low energy X-rays and to provide a relatively uniform X-ray path through all angles of rotation. These specimens were 120mm long and had a 10mm long PTFE insert. Fracture toughness values for the narrow samples could not be obtained due to difficulties associated with their 1mm wide geometry. British Standard geometries (BS ISO 15024:2001) were employed in the fracture toughness testing and SRCL imaging, which permits the use of laterally extended specimens that are closer to practical component and structural length-scales. The specimens (Figure 1(b)) were 20mm wide, 3mm thick and 150mm long. There was a 50mm long PTFE insert placed at the mid-plane. In contrast to SRCT, the sides of the specimen do not pass through the field of view during imaging, allowing the sides of the specimen to be painted with white brittle paint to discern the approximate location of the crack tip.

Figure 1: Specimen geometries used (a) for SRCT and (b) for SRCL

2.3 Mode I testing

A wedge-loaded *in situ* double cantilever beam arrangement was used for both specimen geometries. The wedge was driven into the mid-plane in a displacement-controlled manner as shown in Figure 2(a). The specimens were lightly loaded in Mode II in order to release the insert from the resin, since this was found to help the insertion of the wedge into the interlaminar region. An initial loading step was applied to create approximately 10mm of Mode I crack extension prior to SRCT or SRCL testing so as to reduce any effects from the initial Mode II loading.

Figure 2: Schematic of the scan locations with (a) the wedge-loaded Mode I experimental set-up and (b) the three point bend arrangement for Mode II

2.4 Mode II testing

Mode II loading was conducted using a three-point bend arrangement with a 100mm span for both the SRCT and SRCL specimens as shown in Figure 2(b). The crack was propagated 10mm following an initial Mode II pre-crack. The narrower specimens were clamped at the support furthest from the crack to maintain sample stability under loading due to the narrow width. The approximate crack tip location was identified using a microscope, and more accurately via a radiograph immediately prior to scanning. For the wider specimens imaged via SRCL, the desired locations for imaging were established by reference to the crack tip location, revealed via the brittle painted edges.

2.5 Fracture Toughness Testing

British Standard geometries (BS ISO 15024:2001) were used for both the Mode I and Mode II tests, with five specimens tested in each mode at a cross head displacement rate of 2.5mm/min. Aluminium blocks were attached to the Mode I specimens, which were subsequently pre-cracked in Mode I, and reloaded to propagate the crack for 50mm in a double cantilever beam arrangement. The fracture toughness was calculated using the area method to give the average propagation toughness over a certain area. Mode II end-notched flexure tests were conducted using a span of

100mm, following a Mode II pre-crack, with the specimen placed such that the delamination crack tip was 35mm from the support. The fracture toughness was calculated from the peak load and deflection according to [30]:

$$G_{IIc} = \frac{9Pa^2\delta}{2B(1/4L^3 + 3a^3)} \cdot 1000, \quad (1)$$

where P is the peak load (N), a is the delamination length (mm), δ is the crosshead displacement at crack delamination onset (mm), B is the specimen width (mm), and L is the span length (mm). The factor 1000 is included for unit conversion to obtain J/m².

2.6 Synchrotron Radiation Computed Tomography

SRCT was conducted at the Swiss Light Source (SLS) on the TOMCAT beamline at the Paul Scherrer Institut, Villigen, Switzerland. Scans were conducted at a voxel resolution of 0.69 μ m, with a detector size of 2560x2160 pixels. 1501 projections were taken in each 180-degree rotation, with an exposure time of 150ms and beam energy of 19kV (with more details described in previously published work [20]). A propagation distance of 22mm was used to take advantage of phase contrast, a technique used to enhance contrast between materials of similar attenuations (highlighted in [31]). Reconstructions were completed at the SLS via the in-house GRIDREC method [32] and subsequently analysed using ImageJTM [33].

2.7 Synchrotron Radiation Computed Laminography

SRCL [34] differs from SRCT in that the sample is rotated about an axis inclined with respect to the X-ray beam normal (and with the specimen plane normal approximately oriented parallel to this rotation axis), as described in [20]. This permits imaging of laterally extended objects (e.g the wider geometry shown in Figure 1 (b)) that would otherwise result in full attenuation of the beam at certain angles of rotation in the CT method, with details described in previously published work [34,35]. In comparison to such a limited-angle CT (i.e. not acquiring the full angular range of CT due to exceeding absorption), the SRCL scanning scheme maximises the coverage in the Fourier domain, which often leads to more completely reconstructed images than limited-angle CT [29,35].

The SRCL scans were completed at beamline ID19 of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. 2400 projections were taken with an exposure time of 100ms at an X-ray energy of 19kV, and using edge-enhancing phase contrast [36]. The scans were conducted at a pixel size of 1.4 μ m, with a detector size of 2040x2040 pixels. The reconstructions were performed using an algorithm based on filtered back-projection [37], with an in-built ring correction to minimise such artefacts in the reconstructed volumes.

3 Results and discussion

This section discusses the SRCT and SRCL results, highlighting important micro-mechanical features that should be included in physically representative micro-scale models. The difference in the quality of the scans, SRCT vs. SRCL, highlights the advantages of using narrower samples due to the increased detail achievable in SRCT, whilst SRCL scans were primarily useful to validate that the micro-mechanisms are consistent across the specimen geometries and scales. Key micro-mechanisms are described in Tables 1 and 2 following the discussion sections, and the fracture toughness values of both material systems are given in Figure 3.

Figure 3: Mean fracture toughness values and standard deviation for both materials

Table 1: Key Mode I micro-mechanisms and normalised fracture toughness values

	Mat A	Mat B
Micro-mechanisms	<ul style="list-style-type: none"> • Propagation through damage evolution initiating with particle debonding • Crack deflection from the ply interface back into the interlayer • Bridging ligaments formed from resin and particles • Tortuous crack path following particles with regions of bifurcation • Brittle crack appearance at 45° in particle depleted regions 	<ul style="list-style-type: none"> • Limited particle interactions due to suspected high particle/resin interface strength driving crack into the ply • Fibre bridging occurring with a discrete crack tip • Crack propagation within the ply rather than at the ply/interlayer interface • Instances of ligamented resin failure at the ply/interface
Normalised Toughness	1	0.49

Table 2: Key Mode II micro-mechanisms and normalised fracture toughness values

	Mat A	Mat B
Micro-mechanisms	<ul style="list-style-type: none"> • No clearly identifiable crack tip with crack propagation via evolution of damage initiating with particle debonding • Large echelon bridging formations across the interlayer consisting of particles and resin • Crack path preferentially following particle rich regions • Thin/brittle cusps forming in particle depleted regions 	<ul style="list-style-type: none"> • Crack path predominantly in the ply, featuring a smooth path with matrix shearing of fibres, and broken fibres • Macro-and micro- cusp formations indicative of brittle failure, with propagation along the ply/interlayer interface • Limited particle de-bonding events present in interlayer • Specimen width influencing the proportion of interlaminar vs. intralaminar failure
Normalised Toughness	1	0.43

3.1 Mode I failure in Mat. A

Figure 4(a) shows a CT image “slice” of a Mode I interlaminar crack growing from left to right in Mat. A whilst under load. Note that this “slice” is near the middle of the specimen, away from free edges. There is a clear variation in microstructure, particularly the interlayer thickness (from about $40\text{ }\mu\text{m}$ to $20\text{ }\mu\text{m}$) and in local particle concentrations. The crack opening displacement (COD) is increasing towards the left hand side of the image, with early (i) particle/resin de-bonds occurring. Through the discontinuous coalescence of these de-bonding events, (ii) bridging ligaments can be seen to occur at a number of locations. Across the width of the sample, particle/resin de-bonding can be observed to occur at significant distances ahead of a continuous crack tip, whilst particle-depleted regions exhibit no such damage. In particle-depleted regions, such as at (iv), straight, smooth crack segments are generally observed as the crack path propagates towards and then along the fibre/resin interface. In particle-rich regions, such as at (iii), the crack path appears to be deflected back into the interlayer, towards particle de-bonds that offer a lower energy crack path. It is believed that the resulting, more tortuous crack path, together with the formation of bridging ligaments increases the overall toughness. This single image illustrates three important components that must be considered in a physically representative model; the particle/resin interface, the fibre/resin interface, and neat resin behaviour. Each of these is in constant competition to offer the lowest energy crack path and it is clear that this is highly dependant on local microstructure.

Figure 4(b) shows a second “slice”, which reveals some other microstructural features and also illustrates the increase in COD behind the Mode I *in situ* crack tip. In this case, the microstructure is much more consistent across the region, and features such as neat resin cracking and fibre/resin interface cracks (Figure 4(a)(iv)) are not visible. Early (i) particle/resin de-bonds are observed more clearly in this image, and there is a significant region of (ii) bifurcation, where these de-bonding events have overlapped. As a clear crack path appears at the left of the image,

there are some de-bonds that do not coalesce, forming secondary cracks. Creation of these additional crack surfaces directly increases energy absorption, and increases the extent of bridging ligaments (e.g. at (iii)) in the crack wake, which in turn result in an increased overall toughness.

Three-dimensional analysis was conducted on 30 consecutive CT ‘slices’ from which the crack could be segmented (in blue) from the surrounding volume following the application of thresholding tools and a median filter. Figure 4(c) shows the segmented crack, which shows locations of (i) bridging ligaments clearly. Such tools can be used to quantify the sizes of ligaments in the crack wake, for application in models, whereby the traction forces between the crack faces could be estimated. The figure also shows a substantial amount of (ii) fibre/resin interface failure with a thin crack in this particle-depleted region. At (iii), the segmentation technique at this large crack opening could require improvement, but at smaller crack openings (at the crack tip), the straightforward thresholding technique was sufficient since the grey-scale value remained low (i.e.: black).

Figure 4: Mode I crack path in Mat. A captured via SRCT illustrating; (a) variations in microstructure and the effect on local crack path, (b) particle/resin de-bonding and bifurcation of the crack, (c) a 3D crack segmentation showing bridging ligaments and regions of fibre/resin interface failure, and (d) a lower resolution slice captured via SRCL showing the increase of COD behind the crack tip

Figure 4(d) shows a SRCL scan of a Mode I crack in a standard-sized specimen. The larger field of view illustrates the process zone length of such a crack, but the lower resolution and characteristic SRCL artefacts reduce the level of detail that can be observed. However, the quality of the scan limits the ability to distinguish microstructural effects on the crack path easily. The

Fresnel fringes around the crack openings make distinguishing the true crack surface difficult. As the COD increases, the attenuation of air within the open crack produces a similar grey-scale value as that associated with the surrounding plies which complicates the quantification of crack opening. Nevertheless, the overall characteristics are consistent with the SRCT observations. Again, the figure shows (i) particle/resin de-bonding events preceding (ii) crack coalescence, and the subsequent formation of (iii) bridging ligaments and their decay in the wake of the crack, as seen at (iv). Throughout the reconstructed volume, the crack tip appears to be consistent across the 2.8mm field of view in the centre of the standard-sized specimen.

3.2 Mode I failure in Mat. B

The particles in Mat. B produced significantly different Mode I failure micro-mechanisms compared to the evolutionary damage observed in Mat A, and resulted in a 51% lower mean value of the toughness. Investigating the micro-mechanisms, Figure 5(a) shows that under Mode I loading, the material fails within the intralaminar region. Compared to Mat. A, there is no clear indication of particle/resin de-bonding within the resin rich region. Instead, the crack predominantly propagates at least one fibre deep within the ply, with less frequent occurrences of fibre/matrix interface failure at the interlayer. The plan view (Figure 5(a)) indicates non-uniform crack propagation, whereby the crack front across the width of the sample is not perpendicular to the direction of crack propagation. The figure shows an un-cracked region at (i), and a significant increase in COD on the left hand side at (ii), indicating that the crack has progressed significantly further on one side of the specimen. In Figure 5(b), there appears to be matrix that has separated from a fibre ('peeling') at (i), since the size of this feature is smaller than the nominal fibre diameter. At (ii), there is a bridging fibre, and this bridging behaviour was observed to occur throughout the volume of the specimen. The relative proportion of damage deeper within the ply and not at the ply/interlayer interface suggests that the ratio of the effective overall ply/interlayer strength to single fibre/matrix strength is higher in Mat. B than in Mat. A, for which only instances of ply/interlayer, and not single fibre/matrix interface failure, are present (Figure 4).

Figure 5(c) shows an interesting region of two overlapping crack segments, one on each ply interface. In this image, several (i) bridging ligaments are present, due to fibres "peeling off" the surfaces of the plies, along with regions of clearer fibre/matrix interface failure that appear to show some (ii) smaller scale ligamented behaviour on the bottom surface. Examining across the volume of the specimen, this bifurcation occurs when the crack jumps from one side of the interlayer to the other, and is observed at three separate locations in Figure 5(a). It is unclear why the crack path changes sides, but fracture mechanics states that the crack path will follow the most energetically favourable route. Looking at the microstructure of this sample, there are regions where there are stray fibres within resin rich regions and regions where the interlayer thickness drops from its nominal thickness to less than a fibre's diameter, which may promote this behaviour.

Figure 5: Mode I crack path in Mat. B captured via SRCT showing; (a) a top down view of the crack showing a Mode I crack propagating on either side of the interlayer, (b) a side view of a representative slice featuring a fibre bridge and fibre/matrix 'peeling', (c) a region of an overlapping crack with fibre bridging and fibre/matrix interface failure, and (d) an SRCL slice identifying bridging and broken fibres in the crack wake

Figure 5(d) shows an SRCL image of intralaminar failure in Mat. B under Mode I loading, supporting the contention that the micro-mechanisms observed in the narrower specimens reflect those observed within standard-sized specimens. However, the figure does show the drawback of conducting SRCL scans at a larger voxel size and inherently poorer resolution, whereby the location of the crack is impossible to distinguish from the fibres or resin. At (i), bridging fibres can be distinguished by the bright lines that connect the top of the crack surface to the bottom. However, the lack of contrast at the surfaces makes quantification of even simple CODs difficult, whereby a trainable segmentation technique cannot be accurately applied consistently across the volume. In the image, there are bright lines whose abrupt endings signify an end of a (ii) broken fibre. Such data could be used to estimate at what COD (and fibre strain) the bridging fibres are expected to break, and, therefore, provide an estimate of the effective bridging zone length.

3.3 Mode II failure in Mat. A

Figure 6 shows Mode II cracks propagating through an irregular microstructure in Mat. A. As in Figure 4(a), there are particle-depleted and particle-rich regions in which significantly different fracture paths result. Figure 6(a), illustrates that (i) particle/resin de-bonding occurs further ahead of significant (iii) crack coalescence under Mode II loading than in Mode I (Figure 4), and that again there is no distinct crack tip in this material system. On the left hand side of the

figure, the crack path follows the particle rich regions and then cracks through a neat resin region with an (iv) “echelon” of thin and brittle crack segments at 45° to the fibre direction. These correspond to “hackles” that have been previously documented within interleaved laminates under Mode II loading [38]. Looking at the evidence in this figure, where there is absence of damage formation within (ii) particle-depleted regions, it can be stated that particles facilitate initial damage formation within the interlayer, and hence act as preferential de-bonding sites.

Figure 6: Mode II crack path in Mat A captured via SRCT showing; (a) a variation in microstructure with particle depleted regions surrounded by highly concentrated particle clusters, (b) a preferential crack path through particle rich regions and the formation of echelon bridging arrangements, and (c) a 3D crack segmentation showing bridging ligaments and substantial amount of secondary micro-cracks

The notion that particles act as preferential de-bonding sites is supported in Figure 6(b), in which the crack path is clearly following particle-rich regions on the left hand side of the image (at (ii)). In the central region, the segments of the initial echelon crack geometry, coalesce leaving what appear to be (i) large bridging ligaments. This coalescence is clearly different from the Mode I observations and was observed in numerous regions throughout the volume. Such ligaments contain a distribution of particle vs. resin concentrations, representing a range of properties. In addition the ligaments vary in size, which represents further variation in the bridging separation-traction response. Figure 6(c) reinforces this, where the segmented crack shows regions of echelon bridging locations, where (i) predominantly consists of resin and (ii) is almost entirely a particle. There is further evidence for crack path preferentiality at (iii) and the 3D segmentation also highlights the amount of secondary micro-cracking occurring under Mode II conditions when compared to Mode I (Figure 4(c)). Understanding the formation of these bridging ligaments and their mechanical properties will be an important feature to consider when developing a predictive

model. The exact constitutive response of the bridging ligaments is not yet known, but optimising material components to promote such behaviour would undoubtedly increase the traction forces between the plies and consequently enhance the toughness [19].

Figure 7: Mode II crack in Mat. A captured via SRCL showing; (a) a slice emphasizing the discontinuous behaviour behind the crack tip, and (b) a 3D crack segmentation illustrating the length of the Mode II process zone

Figure 7(a) shows an SRCL ‘slice’ of the Mode II crack, where (i) particle/resin de-bonding can be identified, along with the significantly more (ii) discontinuous cracking behaviour when compared to Mode I (Figure 4(d)). This figure illustrates that SRCL scans with a larger field of view complement the higher resolution SRCT scans, which depicted the more detailed microstructural effects on the geometry of the crack coalescence. Figure 7(b) shows an SRCL scan of the damage zone behind the crack tip, with the crack segmented in grey. The 3D view shows the extent of the discontinuous cracking behind the crack tip and that there are still intact resin segments about 2mm behind the crack tip (i.e. initial de-bonds). The uneven geometry through the coalescence region illustrates the additional friction forces that would be expected during the shear loading across the interlayer. Figure 7 illustrates the problem that has faced researchers in standardising Mode II fracture toughness tests, since there is no clear location where the crack tip can be identified. This causes particular difficulties when an initiation toughness value is specified; it is less problematic if a propagation value is required.

3.4 Mode II failure in Mat. B

Under Mode II loading, Mat. B had a 57% lower mean value of the toughness than Mat. A, with failure occurring predominantly in the intralaminar region. A full volume analysis showed that about 30% of the crack region had significant damage within the interlayer, which was absent under Mode I conditions. Figure 8(a), again reveals that the crack front is not uniform in this material, with no cracking at (i), and that as for Mode I, the majority of the crack is at least one fibre deep within the ply (e.g. (ii)). The region indicated at (iii), shows the crack propagating along the ply/resin interface and not within the ply itself; a behaviour that has been highlighted previously [39]. It is in these regions where damage forms within the interlayer (Figure 8(c)). Firstly, a representative region was analysed that featured the crack propagating within the ply. Figure 8(b) shows the typical cracking behaviour observed in Mat. B. There were more (i) isolated damage events within the interlayer compared to Mode I loading, and again the geometry of such damage suggests particle/resin interactions. On the left hand side of the image, the sharp change in direction suggests the location of a (ii) broken fibre. The crack path then appears to propagate along a fibre deep into the intralaminar region, which provides further evidence of the critical role of the relative fibre-interface and particle-interface strength/toughness in determining the crack path and overall toughness.

Figure 8: Mode II crack path (via SRCT) in Mat. B showing (a) a top down view of the crack showing the crack propagating on either side of the interlayer that has a significant variation in interlayer thickness, (b) a side view of suspected fibre damage on the left and isolated damage events in the interlayer, and (c) a macro- and micro- hackling events and suspected particle-resin interaction

Figure 8(c) shows an atypical region (<30% of the volume) in which the crack path extends into the interlaminar region at several locations with some (i) suspected particle interactions. There appear to be (ii) micro- and (iii) macro-45° echelon cracks occurring in the interlayer, which is consistent with previously reported Mode II behaviour and observations made within particle

depleted regions in Mat. A. The circular (spherical in 3-D) feature at (iv) suggests a particle/resin de-bonding event, but again, the extent to which such debonding was observed was much less than in Mat. A. The lack of such events suggests that the interface between the particles and matrix is relatively strong, such that de-bonding is largely prevented. Crack propagation via the coalescence of particle-de-bond initiated cracks is, therefore, suppressed leaving propagation at the ply/resin interface or within the ply as the lowest resistance path. However, this path results in less ligament formation and generally smoother crack surfaces. This may explain 57% lower mean toughness value of Mat. B against Mat. A.

Figure 9: Mode II interlaminar crack in Mat. B captured with SRCL featuring; (a) echelon crack arrangements and the crack path swapping between the top and bottom of the interlayer, and (b) a 3D crack segmentation emphasizing the length of the process zone in Mode II and showing significant damage occurring within the interlayer across almost the entire volume

Figure 9(a), shows an SRCL slice through a standard-sized specimen. The failure in this case was predominantly in the interlayer, contradicting the predominantly intralaminar failure observed in the narrower specimens. About 90% of the scanned volume at the centre of the standard sized specimen contained significant damage in the interlayer. This may suggest a specimen geometry effect on the fracture micro-mechanisms observed, whereby the crack has initiated from the edges in the ply and stayed in that region in the smaller specimens. There is evidence of (i) particle interactions, whereby the crack propagation proceeds via the coalescence of (ii) “echelon” crack segments along the ply/resin interface, whose smoother appearance may

contribute to the lower toughness when compared to Mat. A. The figure also shows the crack changing sides between the top and bottom of the interlayer interface (e.g at (iii)), which was observed to occur across the 3D volume. The global view (Figure 9(b)) shows that Mat. B fails with distinct failure modes, the relatively smooth fibre-interface failure, and micro- and macro-damage events that occur within the interlayer. Although the appearance of the failure modes between the specimen geometries is similar, the proportion of these failure modes is inconsistent. Additional surface observations of the sides of the narrow and the standard specimens indicated that intralaminar failure has occurred at the free surface for both geometries. This discrepancy between the free surface behaviour and constrained behaviour was also observed in standard sized Mat. A specimens, but requires further investigation. Nonetheless, the fact that the atypical micro-mechanical behaviour in one specimen geometry is actually the representative behaviour in another emphasizes the importance of validating the SRCT against the SRCL observations since the behaviour in this case is not completely representative.

4 Conclusions

The results presented in this work are the first of a series of experiments targeted at gaining a better understanding of the key mechanisms operating in interlayer-toughened CFRPs. The use of high resolution SRCT and SRCL provides a very powerful approach for identifying and quantifying these mechanisms and comparing between materials and fracture modes. The constraining action of the plies and the variations in local particle distributions are all contributing factors to the fracture micro-mechanisms observed in this work. The work indicates that particle/resin, fibre/interlayer and neat resin failure need to be included in models for material response, as there is a clear competition between these mechanisms to provide the energetically favourable crack path and thereby determining its consequent effect on the macro-scale toughness. Although the nominal interlayer thickness of Mat. A and Mat. B are similar, the properties of the interlayer particles have produced significantly different failure mechanisms and toughness values. This complex and quite distinct behaviour of the cracks in the two materials and between Mode I and Mode II fracture indicates that interlaminar failure in current toughened composite materials involves much more of a process zone than an easily identifiable sharp crack tip. By inspecting the chronology of the processes occurring in Mat. A, which has the higher fracture toughness in both Mode I and II, the mechanisms that deflect the crack from the fibre/resin interfaces back into a tortuous path within the particle-rich regions is key. The current work leads to the suspicion that the presence of prior de-bonding events facilitates a lower energy crack path and permits the formation of bridging ligaments, which are key contributors to macro-scale fracture toughness.

Through the use of SRCT and SRCL facilities, the Mode I fracture micro-mechanisms have been assessed non-destructively under a representative stress state. The unique ability of SRCL to scan laterally extended objects proved invaluable in validating the observations made in the narrower specimen geometries via SRCT, and has indeed highlighted that in some materials and loading conditions the micro-mechanisms can be significantly different. Therefore, all work on narrow CFRP specimens should always be validated with standard geometries.

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