

# Failure mechanisms of biological crossed-lamellar microstructures applied to synthetic high-performance fibre-reinforced composites

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## Abstract

This paper investigates whether the toughening mechanisms of a biological crossed-lamellar microstructure can be reproduced in a synthetic high-performance carbon fibre/epoxy matrix composite. The mechanics of the failure process in synthetic crossed-lamellar microstructures was investigated using the Finite Element Method. This enabled the design of a high-performance carbon-fibre reinforced polymer (CFRP) with such microstructure. Two different procedures were then developed to synthesise the first crossed-lamellar microstructures in CFRP in the literature. Test specimens were subsequently manufactured. Three-point bend tests were carried out in an SEM environment, showcasing the damage diffusion capability of the microstructure under stable conditions. The results show that the crossed-lamellar microstructure can be synthesised in CFRP with good accuracy, and that the mechanical toughening mechanisms associated with the natural crossed-lamellar microstructures can be reproduced in this synthetic material.

**Keywords:** Microstructures (A), Fibre-reinforced composite material (B), Mechanical testing (C), Fractography (C), Biomimetics

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## 1. Introduction

Microstructures found in natural composites, such as bone, mollusc shells and wood, allow for energy dissipation through various strategies, making these composites significantly more damage tolerant than their brittle and relatively weak main constituents [1–3]. This has inspired researchers to apply similar principles in synthetic composites in order to improve their damage tolerance [4–9].

One example of such microstructure found in biological composites is the crossed-lamellar microstructure, the most common microarchitecture within molluscs [10]. The crossed-lamellar microstructure varies slightly between different species and it has been experimentally characterised in the literature [10–18], with a vast amount of research focusing on the *Strombus gigas* shell (Figure 1(a)) [19–27]. The microstructure of the *Strombus gigas* shell will therefore be used as an example of such microstructure in this paper.

The crossed-lamellar microstructure is highly organised and consists mainly of brittle aragonite, a form of calcium carbonate, which in the *Strombus gigas* shell represents 99.9 w% of the composite [24]. Despite consisting almost solely of the brittle ceramic phase, the crossed-lamellar microstructure is the toughest among the molluscs, reaching toughness values that are up to four orders of magnitude higher than the toughness of monolithic aragonite [21]. In comparison, one of the strongest natural microstructures with analogous constituents, the brick-and-mortar type nacreous microstructure with 95 w% ceramic phase, has a work of fracture that is seven times lower than that of the *Strombus gigas* shell [21].

The outstanding toughness of the shell arises from its microstructure (seen on the fracture surface of the shell in Figure 1(b) and schematically illustrated in Figure 1(c)) which, on the coarsest length scale, comprises three macroscopic layers with  $0^\circ/90^\circ/0^\circ$  orientation (identified as inner, middle and outer layers and marked I, M and O, respectively, in Figures 1(b) and 1(c)) [21]. Each of these macroscopic layers further consists of 1<sup>st</sup> order lamellae with  $\pm 35^\circ$  to  $\pm 45^\circ$  orientation with respect to the thickness

of the shell. These 1<sup>st</sup> order lamellae further consist of 2<sup>nd</sup> and 3<sup>rd</sup> order lamellae (not  
 30 illustrated in Figure 1(c)) on smaller length scales. It has been found that the microstructural features illustrated in Figure 1(c) are the most important contributing to the high toughness of the shell [21].

The most important toughening mechanisms associated with the crossed-lamellar microstructure are (see Figure 2) [29]:

- 35 • parallel tunnel cracking between the 1<sup>st</sup> order lamellae on the tensile side (i.e. inner layer) and subsequent crack arrest at the interface between the inner and

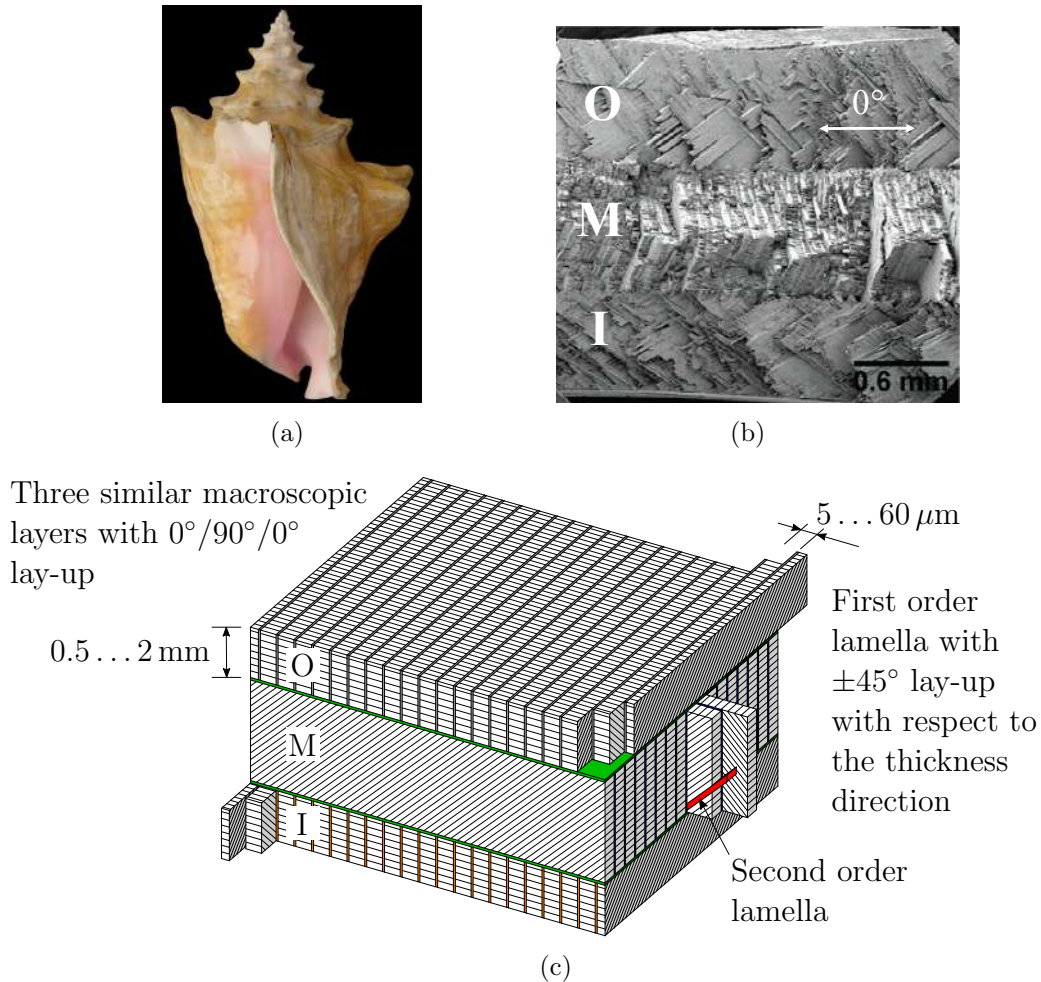


Figure 1: (a) *Strombus gigas* shell [28]; (b) fracture surface of the shell showing a crossed-lamellar microstructure (adapted with permission from Su et al. [20]. Copyright 2004 American Chemical Society.); (c) schematic illustration of the microstructure (not to scale).

middle layers;

- crack deflection as the cracks advance to the middle layer; and
- crack bridging, debonding and frictional sliding of the lamellae in the middle layer.

40 When the shell is loaded in bending with the inner layer in tension (see Figure 2), multiple parallel tunnel cracks form and extend on the tension side along the depth of the specimen, as shown in Figure 2 [21]. These cracks rely on the weak protein interfaces between the 1<sup>st</sup> order lamellae, and as they grow through the thickness of the inner layer, they arrest at the macroscopic interface between the inner and middle layers  
45 due to the change in the layer orientation. At a later stage, secondary tunnel cracks are observed to form by growing from the macroscopic inner/middle layer interface towards the tensile surface (see Figure 2).

Gradually, the cracks advance to the middle layer and grow along the  $\pm 45^\circ$  orienta-

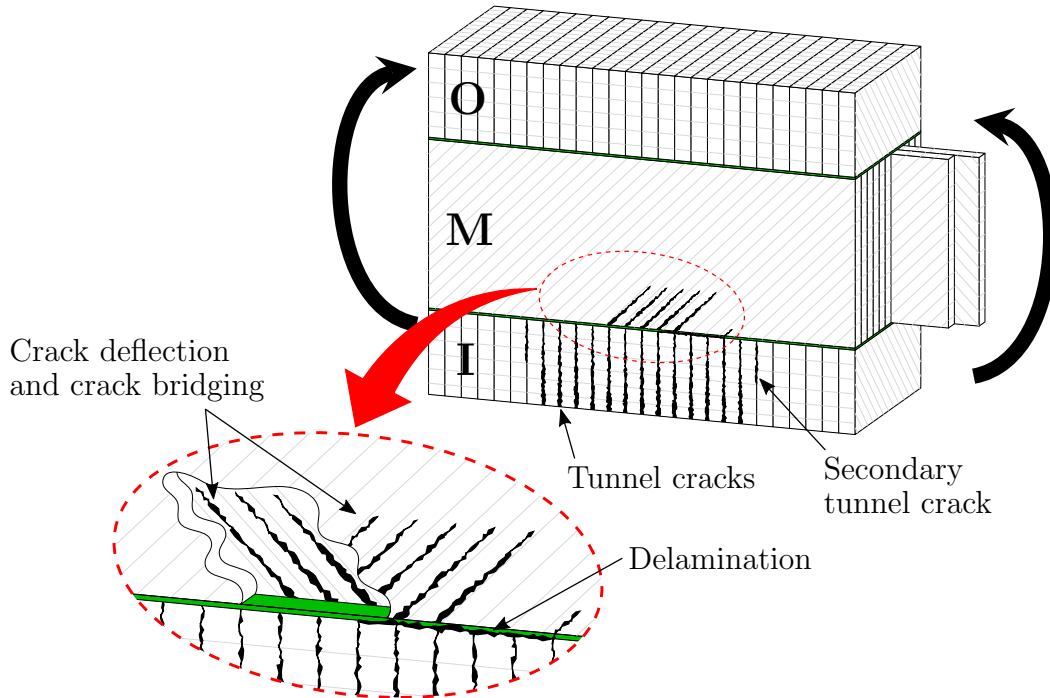


Figure 2: The main toughening mechanisms associated with the crossed-lamellar microstructure are tunnel cracking in the inner layer, crack deflection in the middle layer, and crack bridging, debonding and frictional sliding of the lamellae in the middle layer.

tion of the lamellae so that they deflect from their original direction of propagation (see  
50 Figure 2). As the lamellae have an alternating orientation with respect to the neighbouring lamellae, the cracks propagate along a  $+45^\circ$  direction in every other lamella, while the neighbouring lamellae remain intact at that location and act as bridging lamellae. Likewise, when the cracks advance in a  $-45^\circ$  lamella, the neighbouring  $+45^\circ$  lamella act to bridge that crack.

55 The crack bridging involves debonding and frictional sliding of the lamellae, and lamellae pull-out [30]. The adjacent lamellae debond and slide against each other, creating large fracture surfaces and dissipating energy through friction. In order for the crack to propagate through the thickness of the shell, it does not only have to propagate along the weak  $\pm 45^\circ$  interfaces but also to break half of the lamellae (in a direction  
60 parallel to the crack in the other half) or to fully pull them out. The latter tends to occur with very significant and stable energy dissipation in the process.

In addition to the toughening mechanisms discussed above, delamination between the macroscopic layers (see Figure 2) and additional bridging of ligaments have been suggested to occur and to improve the damage tolerance of the shell [21, 30]. Upon  
65 delamination between the macroscopic interfaces, the crack advances to the middle layer after travelling a short distance along the macroscopic interface.

Despite its outstanding toughness, few attempts have been made to mimic the toughening mechanisms of the crossed-lamellar microstructure in synthetic composites, and none using CFRP. Hou et al. [31] used bamboo lamellae with a polymer adhesive  
70 to study the effect of the fibre orientation angle for a crossed-lamellar microstructure in this material system. Chen et al. [32] manufactured and tested specimens made of silicon and photoresist in a proof-of-concept study, and were able to reproduce one tunnel crack, delamination, and crack bridging.

Salinas [18] used dry biaxial stitched glass fibre and epoxy resin to manufacture  
75 glued two-layer specimens with in-plane and into-plane fibre orientations (omitting the 1<sup>st</sup> order interfaces due to stitching). They tested the specimens in three-point bending and concluded that a configuration with in-plane fibres in the loading direction is the

stiffest and strongest. The dimensions of the specimens and their failure mechanisms were not reported.

80 Kaul and Faber [33] produced a crossed-lamellar ceramic laminate of mullite using tape casting, oriented lamination and templated grain growth, and reported achieving a microstructure that produced a torturous crack path. Karambelas et al. [34] used silicon nitride and boron nitride to prototype a crossed-lamellar microstructure using a method based on co-extrusion, cutting and re-bonding the materials. Although they  
85 succeeded in incorporating some of the toughening mechanisms of the natural crossed-lamellar microstructure, including tunnel cracking, crack deflection, crack bridging and frictional sliding, the failure of the specimens was unstable.

Gu et al. [35] designed and prototyped a *Strombus gigas* -inspired microstructure using bi-material 3D printing with a stiff polymer as the bulk material and a soft  
90 polymer at the interfaces. They investigated the effect of the level of hierarchy on the impact resistance of the composite and found that the crossed-lamellar microstructure can significantly improve the impact resistance by creating large amounts of diffuse damage.

It can be summarised from the literature that there have been to date no single  
95 work published (to the knowledge of the authors) that explores bio-inspired crossed-lamellar microstructures in high-performance engineering fibre-reinforced composites. In this work, we design, prototype and test a crossed-lamellar CFRP microstructure. We demonstrate that it is possible to replicate the damage mechanisms of the biological crossed-lamellar microstructure. This work hence opens up a completely new field for  
100 the design of damage tolerant CFRP structures.

## 2. Methods

### 2.1. Microstructure definition

A parametric study using a unit cell FE modelling approach was carried out to study the feasibility of the crossed-lamellar microstructure in a carbon/epoxy material

105 system, and to obtain values for the geometric parameters that define the microstructure. A tessellated unit cell is shown in Figure 3, and the full details of the model and the parametric study are included in Appendix A. The FE model was loaded in displacement-controlled pure bending through periodic boundary conditions, and the parameters of the study included (see Figure 3):

- 110 • the height  $h_i$  of each macroscopic layer  $i \in \{I, M, O\}$ ;
- the thickness of the prepreg plies comprising the macroscopic layers,  $t_{\text{ply}}$ ;
- the fibre orientation angle of the prepreg with respect to the thickness  $\pm\theta$ ;
- the macroscopic interfaces, with epoxy matrix or thermoplastic polyethersulfone (PES) film with a parametric thickness  $t_{\text{MI}}$  (which increases the resistance to
- 115 delamination and facilitates the accumulation of damage near the interface).

Based on the results of the parametric study, fully detailed in Appendix A, it was concluded that a microstructure with a configuration specified in Table 1 is suitable for investigating experimentally whether the toughening mechanisms of the crossed-lamellar microstructure can be reproduced in a prototyped CFRP microstructure. The

120 material system associated with the chosen configuration is standard-thickness UD prepreg Hexcel IM7/8552 and PES film SU301050 supplied by Goodfellow Cambridge Ltd.

## 2.2. Prototyping

### 2.2.1. Co-curing prototyping route

125 Based on the predictions of the FE analysis, as well as the manufacturing feasibility considerations, and the aim to achieve a high level of energy dissipation, a composite

Table 1: The values of the geometric parameters of the microstructure obtained from a parametric study.

| $h_I$ [mm] | $h_M$ [mm] | $h_O$ [mm] | $t_{\text{ply}}$ [mm] | $\pm\theta$ [°] | $t_{\text{MI}}$ [ $\mu\text{m}$ ] |
|------------|------------|------------|-----------------------|-----------------|-----------------------------------|
| 1          | 2          | 1          | 0.125                 | 45              | 50                                |

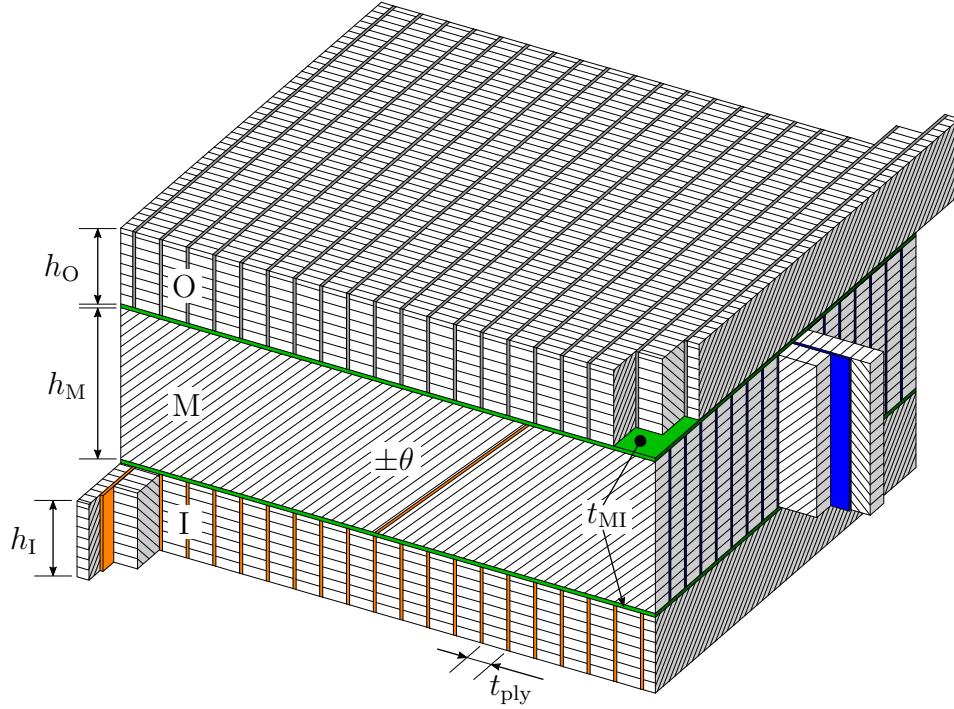


Figure 3: A tessellated unit cell model showing the variables used in the parametric study.

with crossed-lamellar microstructure was prototyped according to the specifications outlined in the previous section (Table 1 and Figure 3). To this end, a prototyping procedure with multiple steps, as schematically illustrated in Figure 4(a), was developed progressively over multiple iterations. This procedure is based on constructing the  
130 microstructure of uncured prepreg and curing it directly to its final form.

First, sub-laminates with  $\pm 45^\circ$  architecture were sequentially laid up and cut into 2 mm wide partial strips using a ply cutter, which in initial investigations was found to provide the required level of accuracy and repeatability. An experimental parametric  
135 study on the process parameters (i.e., the thickness of the sub-laminate and cutting speed) was performed, and it revealed that cutting speeds that are too high or too low, as well as sub-laminates that are too thick, will lead to fibre fray. Therefore, the sub-laminate thickness was chosen to be 0.5 mm and the cutting speed was chosen to be 50 mm/s. In addition, alignment holes were cut at each corner of the sub-laminates  
140 using the ply cutter.

The sub-laminates were subsequently stacked on an alignment plate that had pins

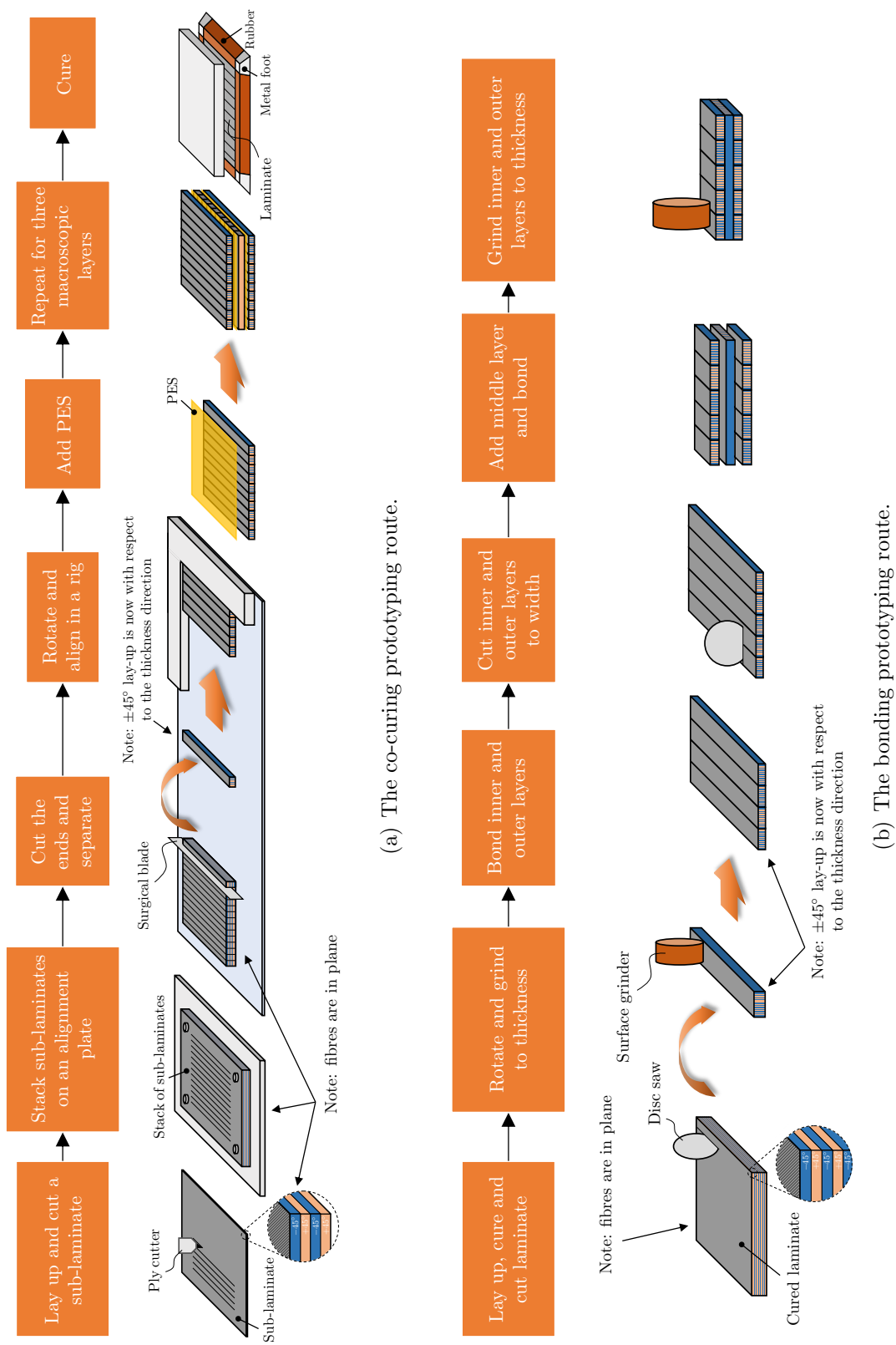


Figure 4: Two prototyping procedures were developed to manufacture the crossed-lamellar microstructure in CFRP.

matching the alignment holes in order to ensure accurate positioning of the partial cuts. The cuts on each sub-laminate were manually extended to run across the whole length of the laminate before adding a new sub-laminate onto the stack. When a stack height  
145 of 5 mm was reached, the stack was removed from the alignment plate and the strips were separated along the cut lines using a thin surgical blade, and rotated  $90^\circ$  to their side so that, after the rotation, the  $\pm 45^\circ$  lay-up was with respect to the thickness. The rotated strips were aligned and joined using alignment tools until a nominal layer size of 91 mm x 91 mm was reached. The operations were repeated for the three macroscopic  
150 layers of the crossed-lamellar microstructure.

All three manufactured layers were debulked separately under vacuum for 24 hours. The top surface of the layers was protected by a plate resting on feet at the corners of the laminate to prevent the fibre and ply orientations from changing due to the applied vacuum. After debulking, the nominal in-plane dimensions of the layers were  
155 87 mm x 87 mm and the thickness was 2.3 mm. The three-layer microarchitecture was assembled from these layers, with a  $50\text{ }\mu\text{m}$  PES film between each layer.

A lateral support of RTV-101 silicone rubber was cast around the prototyped laminate to accommodate for unevenness of the edges of the laminate and to give it structural stability during curing. Four steel feet were placed at the corners of the  
160 laminate, each 6 mm in height, and their height was adjusted to correspond to the thickness of the uncured laminate by adding metal shims on top of them (three pieces on each foot, each 0.5 mm thick). A 20 mm thick steel plate wrapped in release film was placed on the feet to obtain a flat top surface. The laminate was cured in an autoclave according to the prepreg manufacturer's instructions (curing pressure of 7 bar and cu-  
165 ring temperature of  $110^\circ\text{C}$  for one hour, followed by  $180^\circ\text{C}$  for two hours), except that the heating and cooling rates were set to a low value ( $1^\circ\text{C}/\text{min}$ ).

Figure 5(a) shows the microstructure in CFRP achieved with the co-curing procedure (obtained by reconstructing microscopic images). The  $\pm 45^\circ$  blocks (Figure 5(c)) are highlighted. The thickness of the cured laminate was 6.8 mm and the configura-  
170 tion with 1 mm thick inner and outer layers was manufactured from this laminate by

grinding down the thickness of the inner and outer layers with a surface grinder.

### 2.2.2. Bonding prototyping route

In order to improve the alignment and consolidation of the microstructure, an alternative prototyping procedure was investigated. This procedure was based on curing a laminate, and subsequently cutting, rotating and re-bonding it to achieve the crossed-lamellar microstructure, as schematically illustrated in Figure 4(b). Apart from the macroscopic interfaces, the parameters used with this procedure are the same as those used with the co-curing procedure (Table 1). In the bonding prototyping route, the macroscopic interfaces consisted of 3M Scotch-Weld 9323 B/A epoxy adhesive.

175 Firstly, a 175 mm x 175 mm laminate with the lay-up sequence  $[+45^\circ/-45^\circ]_{20S}$  was laid up using Hexcel IM7/8552 prepreg. Peel ply was added on top and bottom of the laminate, and two layers of glass cloth and a 10 mm thick aluminium caul plate were placed on top of the laminate. The edges of the laminate were dammed with cork and the laminate was cured in an autoclave according to the manufacturer's recommended  
185 cure cycle. The thickness of the cured laminate was 9.8 mm.

The cured laminate was subsequently cut into six 3 mm wide and 96 mm long blocks using a disc saw. The blocks were then rotated  $90^\circ$  to their side so that the width of the block, and the fibre orientation angle, were now in the thickness direction (Figure 4(b)). The thickness was then further ground from 3 mm to 2 mm using a surface grinder, and  
190 the long sides of the block were ground to remove the imprint of the peel plies so that the final width of the blocks was 9.6 mm.

At this stage, five blocks were bonded together along their long sides in order to manufacture the inner and outer layers of the microstructure. Prior to bonding, the long sides were roughened with sand paper and cleaned, and subsequently glued using  
195 3M Scotch-Weld 9323 B/A epoxy adhesive. The laminate was left to cure clamped at room temperature for 24 hours. The width of the cured laminate was 48 mm and, after curing, two pieces corresponding to the width of the original blocks were cut from it across its width to make the inner and outer layers of the microstructure.

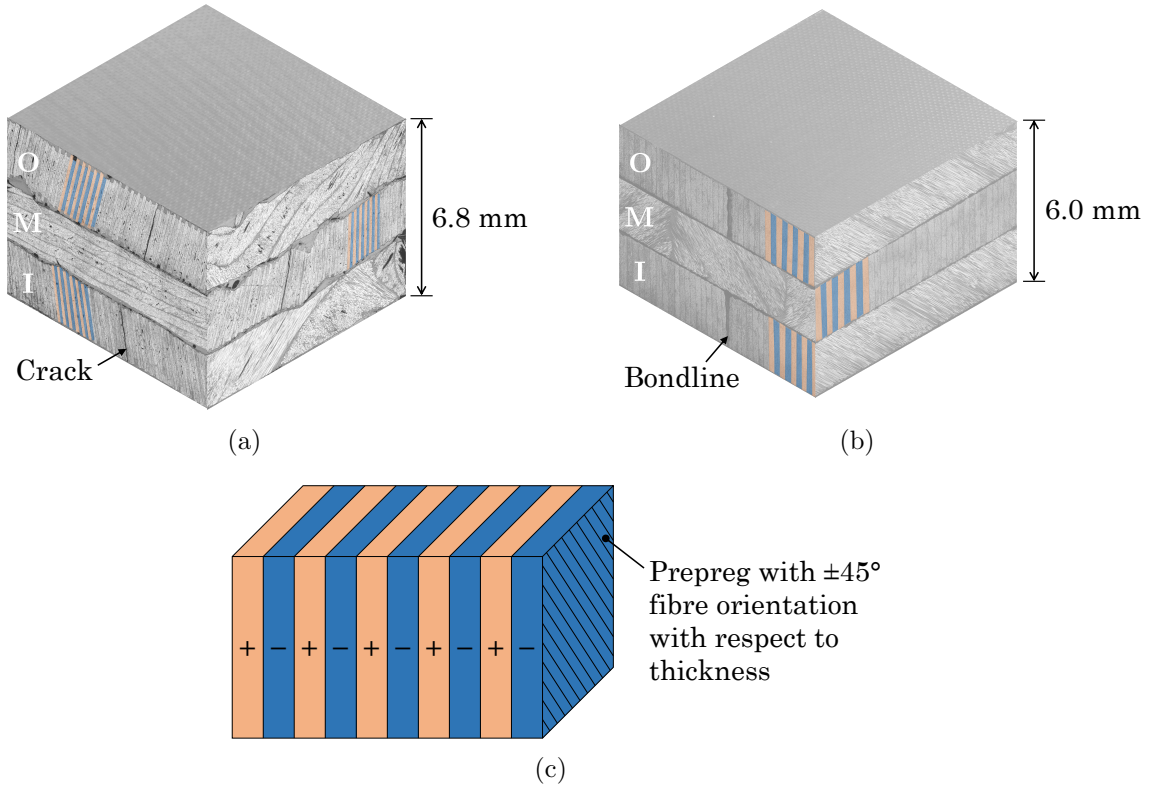


Figure 5: The prototyped crossed-lamellar microstructure in CFRP achieved with (a) the co-curing procedure and (b) the bonding procedure, with (c) the highlighted  $\pm 45^\circ$  blocks.

The middle layer was manufactured by cutting a 48 mm long piece (the width of the  
 200 glued laminate) from the remaining sixth block. The three layers were then bonded to-  
 gether using 3M Scotch-Weld 9323 B/A epoxy adhesive to compose the crossed-lamellar  
 microstructure, with the bonded surfaces roughened with sand paper prior to bonding.  
 The microstructure was clamped and cured at room temperature for 5 hours to reach  
 handling strength and further cured in mild heat ( $65^\circ\text{C}$ ) for two hours to accelerate  
 205 the strength build-up according to the adhesive manufacturer's instructions. After fully  
 cured, 1 mm was ground off from the top and bottom surfaces of the manufactured la-  
 minate to achieve a microstructure with 1 mm thick inner and outer layers. Figure 5(b)  
 shows the achieved microstructure prior to the final grind.

### 2.3. Testing

Specimens from both prototyping routes (hence two different microstructures with dimensions listed in Table 2) were tested in a three-point bending (3PB) configuration (Figure 6) in order to assess the toughening mechanisms of the crossed-lamellar microstructure in CFRP. The 3PB tests were carried out in an SEM environment using a Deben Microtest Module with a 5 kN load cell. In order to enhance the quality of the SEM images, the specimens were polished and gold-sputtered on the side surface prior to testing. Furthermore, pre-existing cracks on the co-cured microstructure (see Figure 5(a)) were filled with Araldite 2011 epoxy adhesive prior to testing.

The specimens were loaded at a displacement rate of 0.2 mm/min, and the loads and the displacements were recorded with an acquisition rate of 200 ms. The displacements were read directly from the built-in extensometer and corrected after the test to account for the stiffness of the testing rig.

The tests were regularly paused in order to take SEM images. In the beginning,

Table 2: The dimensions of the tested specimens with a crossed-lamellar microstructure.

| Specimen | $d$ [mm] | $t$ [mm] | $w$ [mm] |
|----------|----------|----------|----------|
| Co-cured | 36       | 4.8      | 7.8      |
| Bonded   | 36       | 3.9      | 9.8      |

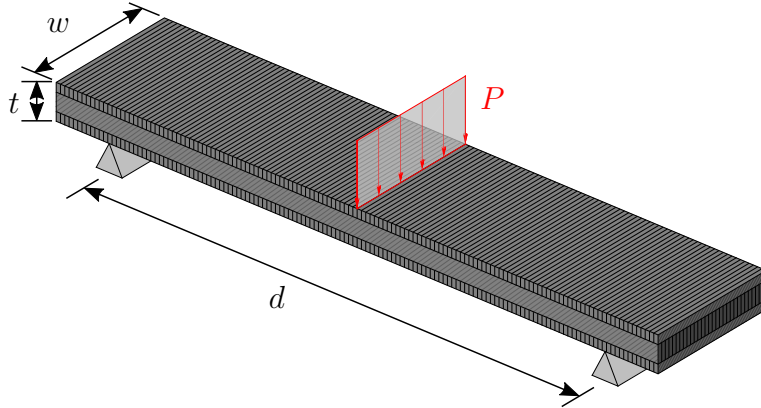


Figure 6: Sketch of the set-up of the three-point bending test showing the dimensions and the testing orientation of the specimen.

before major damage occurred in the specimens, the displacement was held at load intervals of 50 N. At a later stage, when the damage was growing, the tests were paused  
225 at regular displacement intervals or when other interesting features were observed.

### 3. Results

Figures 7 and 8 show selected SEM images obtained during the testing of the co-cured microstructure (Figure 7) and the bonded microstructure (Figure 8). Figures 7(a) and 8(a) show intact specimens with the macroscopic layers highlighted prior  
230 to testing, while the progressive failure of the specimens is illustrated in Figures 7(b)-(f) and 8(b)-(f) for the co-cured and bonded microstructures, respectively.

In both microstructures, the damage initiated by tunnel cracking in the inner layer, and subsequently arrested at the macroscopic interface (Figures 7(b) and 8(b)). Cracks then deflected to grow along the fibre direction in the middle layer at various locations  
235 along the interface (Figures 7(c) and 8(c)) but they did not propagate far into the middle layer, as seen in Figures 9(a) and 9(b) showing a more detailed view of the deflected cracks.

Secondary tunnel cracks (i.e. cracks growing from the macroscopic interface towards the tensile surface) were observed in the co-cured microstructure at a relatively early  
240 stage (Figure 7(c)), while in the bonded microstructure they occurred at a later stage (Figure 8(d)).

In the co-cured microstructure, cracks appeared in the outer layer while the middle layer was still carrying load (Figure 7(d)), and eventually the plies in the middle layer began to debond (Figure 7(e)), leading to the final failure of the specimen (Figure 7(f)).  
245 Figure 9(c) shows the debonded plies in the middle layer after the final failure.

In the bonded microstructure, the damage propagated in the form of delamination at the inner/middle layer interface (Figure 8(d)). The deflected cracks propagated more gradually, some eventually growing across the middle layer (Figure 8(e)), while the middle layer retained its load-carrying capacity. The test was stopped prior to the  
250 final failure of the specimen due to the testing machine reaching its maximum extension.

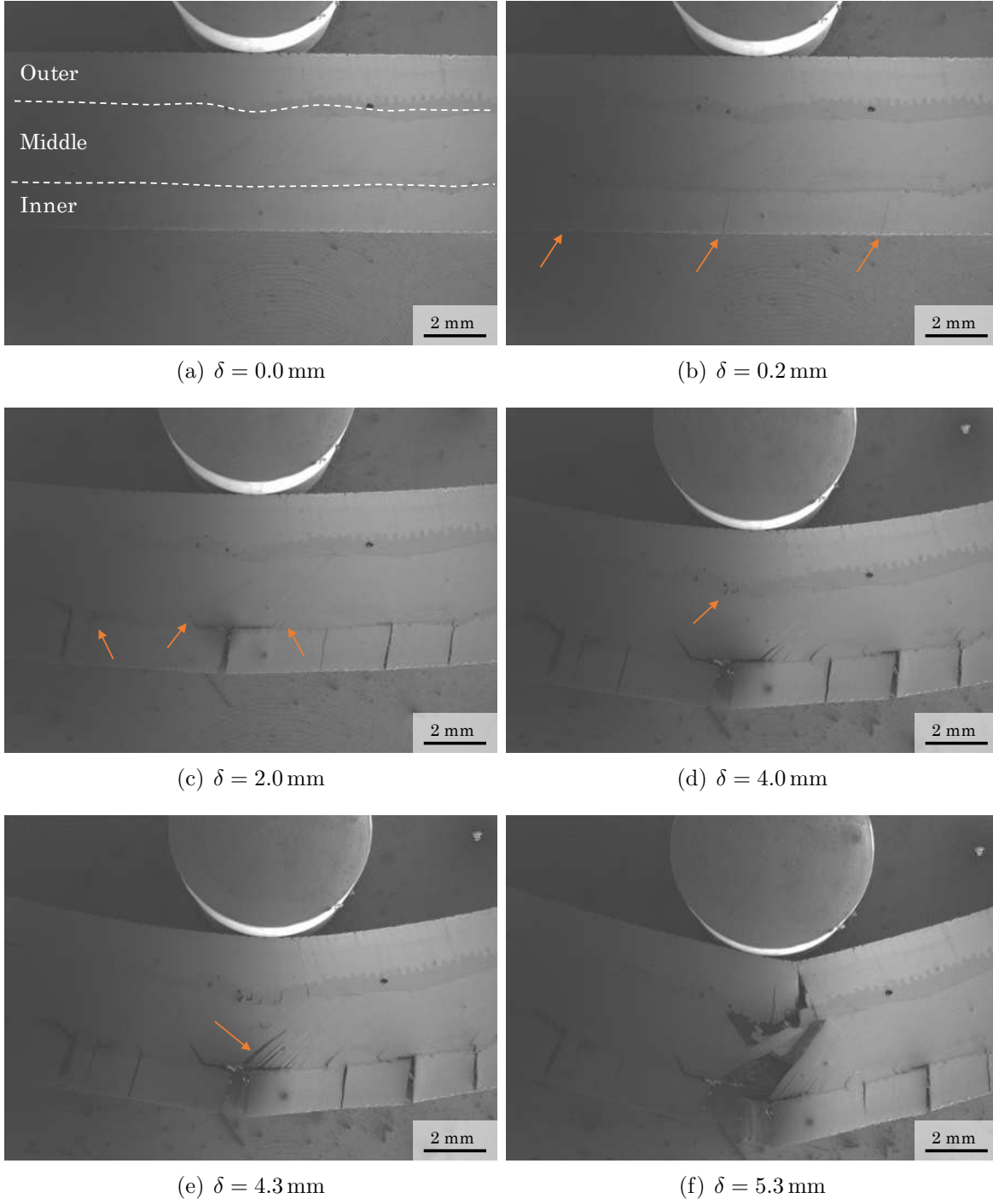


Figure 7: (a) Co-cured microstructure in the SEM highlighting the macroscopic layers and (b)-(f) SEM images showing the progressive failure of the specimen.

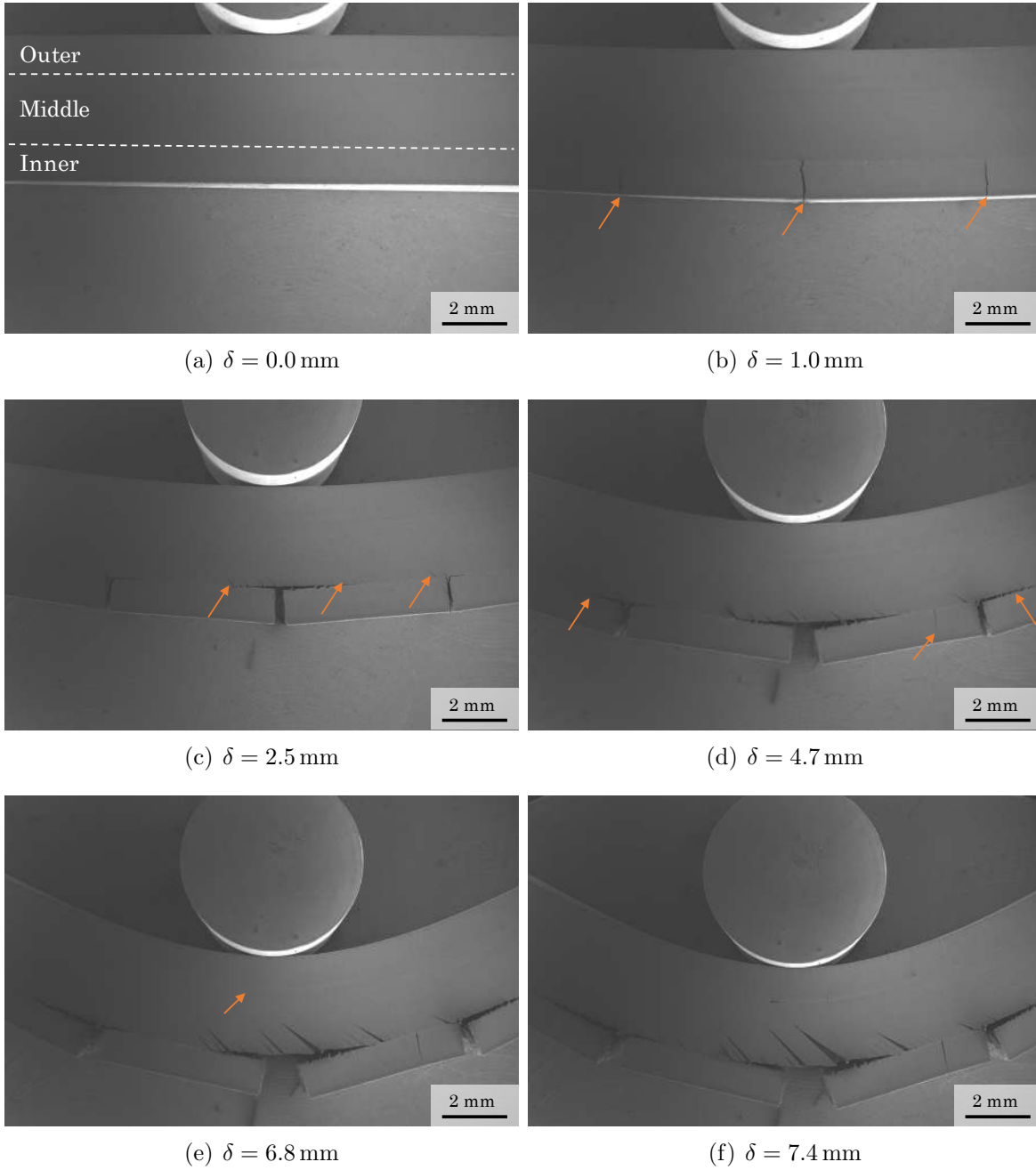


Figure 8: (a) Bonded microstructure in the SEM highlighting the macroscopic layers and (b)-(f) SEM images showing the progressive failure of the specimen.

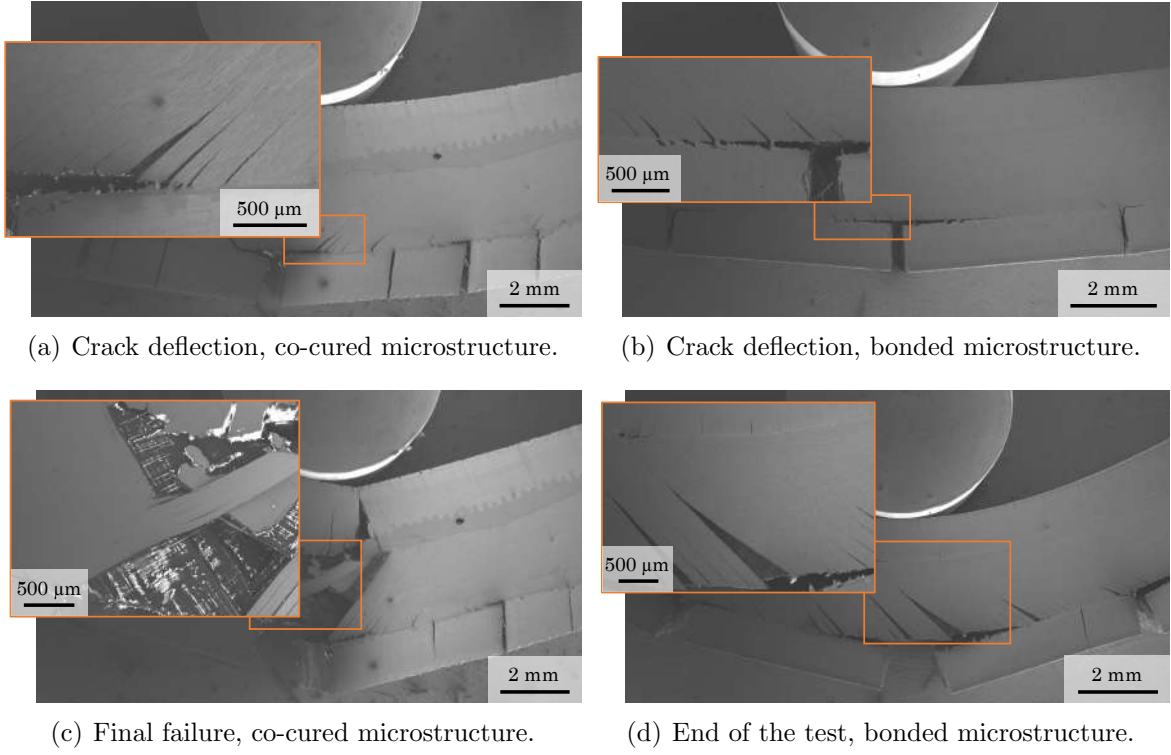


Figure 9: Details of the 3PB tests of the CFRP specimens with a crossed-lamellar microstructure.

Figure 8(f) shows the specimen at the end of the test, and a more detailed view is given in Figure 9(d).

Both microstructures dissipated energy in a stable manner and under increasing load, as seen in the load,  $P$ , vs displacement,  $\delta$ , curves given in Figure 10. The load drops observed in the curves are associated with specimen relaxation when the displacement was held to take SEM images. The annotations (b)-(f) in the curves correspond to the SEM images in Figures 7 and 8.

After testing, the specimens were manually broken into two to take further SEM images of the fracture surfaces (Figure 11). Figures 11(a) and (b) show a perpendicular view of the fracture surfaces of the co-cured and bonded microstructures, respectively. The cracks followed the  $\pm 45^\circ$  fibre orientation in the middle layer without breaking the fibres, the adjacent plies debonding from each other and creating large fracture surfaces with a regular pattern (Figures 11(c) and (d)). A detailed view (Figures 11(e) and 11(f)) reveals diffuse damage as multiple cracks extend into the middle layer along the fibre

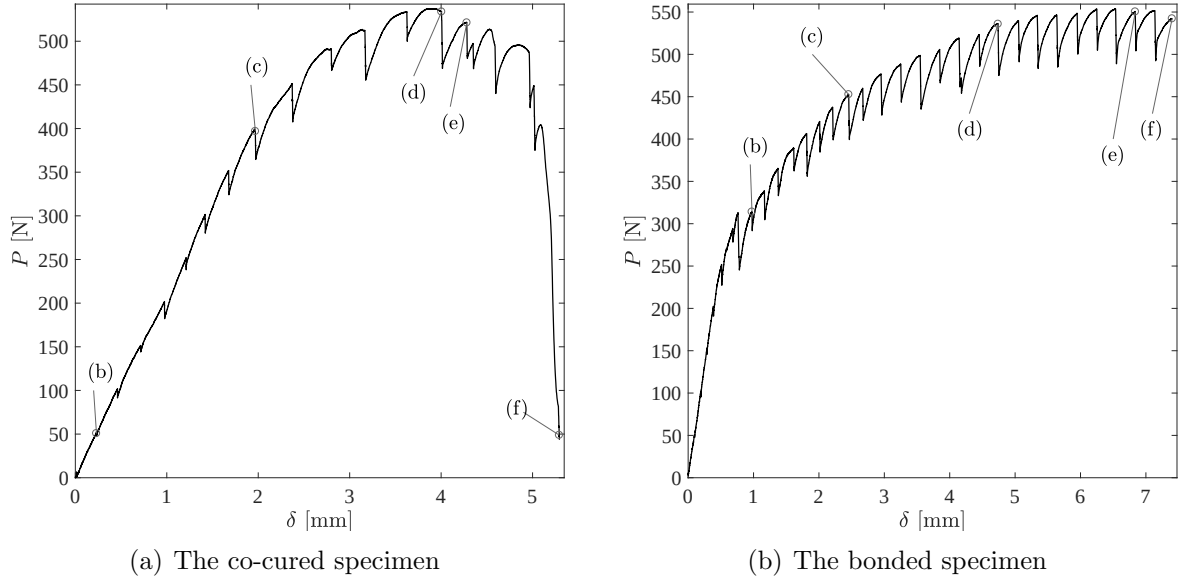


Figure 10: The load vs displacement curves of the test specimens. The annotations correspond to the images in Figures 7 and 8, and the load drops are associated with specimen relaxation while the tests were paused for taking SEM images.

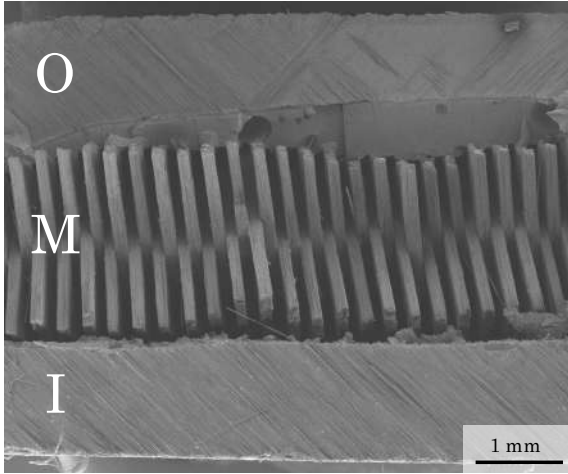
265 direction. Some fibre failure caused by manually breaking the specimen is observed on the fracture surface of the bonded specimen (Figure 11(b)).

## 4. Discussion

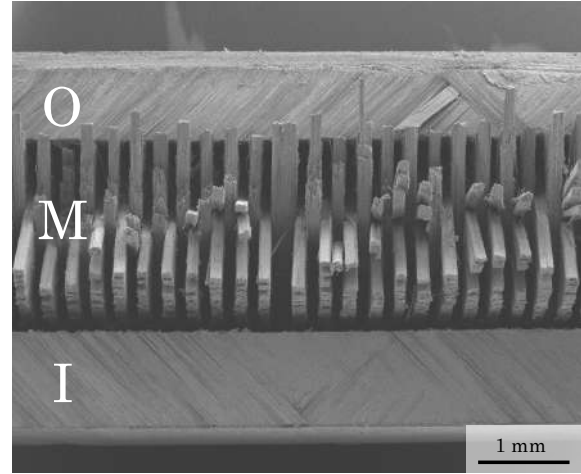
### 4.1. Prototyping procedure

Two different prototyping procedures were investigated, and the crossed-lamellar microstructure was prototyped successfully and with relatively good accuracy with both  
 270 procedures (Figure 5). In the co-cured microstructure, the prepreg did not maintain exactly its vertical orientation during curing, and some cracks formed along ply interfaces due to thermal stresses (Figure 5(a)). Furthermore, it is possible that the PES film may partially dissolve in the epoxy during curing and undergo reaction induced phase  
 275 separation forming a gradient interphase [36].

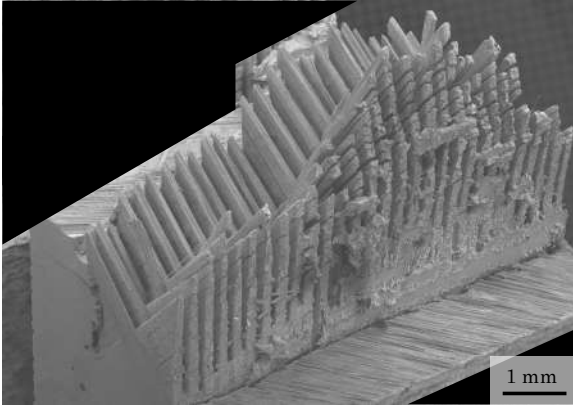
The bonding procedure can be seen to allow for better control over the dimensions and orientations of the microstructure (Figure 5(b)). The bonded microstructure has even layer heights, as also seen when comparing the fracture surfaces in Figu-



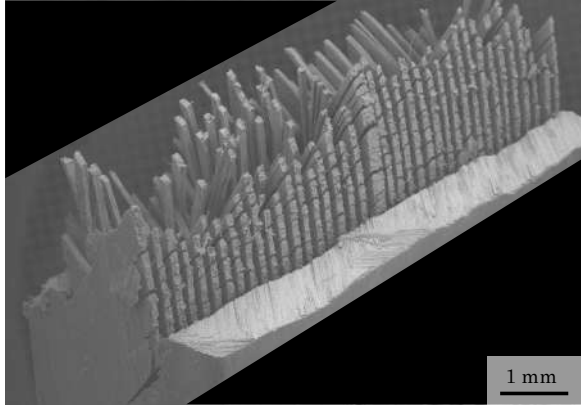
(a) Perpendicular view of the fracture surface, co-cured specimen.



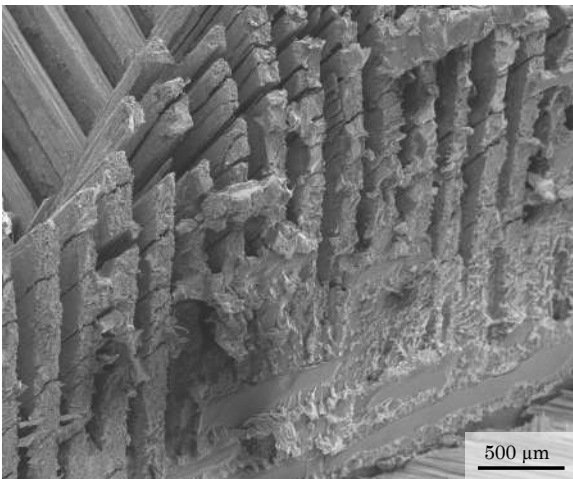
(b) Perpendicular view of the fracture surface, bonded specimen.



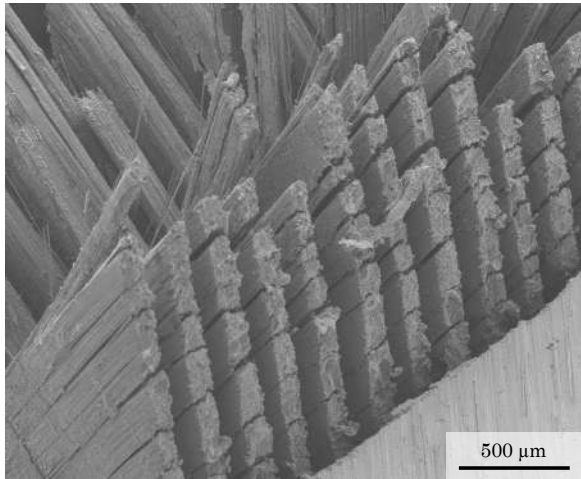
(c) Lamella pull-out and splits along the fibre direction, co-cured specimen.



(d) Lamella pull-out and splits along the fibre direction, bonded specimen.



(e) Detail of the splits along the fibre direction, co-cured specimen.



(f) Detail of the splits along the fibre direction, bonded specimen.

Figure 11: SEM micrographs of the tested specimens.

res 11(a) and (b). Furthermore, the consolidation of the bonded microstructure is  
280 better, with no voids visible in the microstructure. In addition, the bonding procedure  
introduces additional interfaces to the inner and outer layers that have approximately  
the thickness of one prepreg ply, as seen in Figure 5(b).

#### 4.2. Toughening mechanisms

Toughening mechanisms associated with the crossed-lamellar microstructure were  
285 successfully reproduced in both tested microstructures. Figures 7(b) and 8(b) show  
primary tunnel cracking where cracks initiate from the tensile surface and grow towards  
the macroscopic interface. Secondary tunnel cracks that initiate from the macroscopic  
interface and grow towards the tensile surface are observed in Figures 7(c) and 8(d).  
The co-cured microstructure had a higher tunnel crack density, (Figure 7), whereas the  
290 tunnel cracks in the bonded microstructure initiated at the weak bondlines with only  
one additional primary tunnel crack forming under the load pin (Figure 8).

Figures 9(a) and 9(b) show crack deflection along the fibre direction in the middle  
layer. The crack deflection led to a large amount of diffuse damage by forming a regular  
pattern of splits along the length and across the width of the specimens (Figure 11).  
295 The propagation of the deflected cracks was slow and stable, and in fact, many cracks  
were arrested without them growing far into the middle layer due to the bridging effect  
of the  $\pm 45^\circ$  lay-up. In the middle layer, the favourable crack propagation direction  
changes from  $+45^\circ$  to  $-45^\circ$  (or vice versa) at each ply interface (Figure 11). As a result  
of this mismatch along the width of the specimen, the crack smears and ‘struggles’ to  
300 propagate in the middle layer.

Both microstructures also exhibited some degree of delamination, which in the co-  
cured microstructure was eventually arrested by the tough PES interface and migrated  
to the middle layer as deflected cracks (Figure 7). In the bonded microstructure, the  
interface was not sufficiently tough to arrest the delamination which grew around each  
305 primary tunnel crack (Figure 8(d)).

The post-mortem investigation revealed large ‘triangular’ fracture surfaces and la-

mella pull-out (Figure 11), which are associated with large amounts of energy dissipation in the natural crossed-lamellar microstructure. The debonding was readily observed in the co-cured specimen during the test (Figure 9(c)), while the bonded specimen was  
310 better able to preserve its structural integrity up to large curvatures (Figure 9(d)).

Figure 10 shows that the toughening mechanisms of the crossed-lamellar microstructure led to stable energy dissipation, reaching large displacements under increasing or constant load. The majority of the energy dissipation arises from the toughening mechanisms associated with the middle layer, i.e., the extensive crack deflection and crack  
315 bridging, and the delamination and lamella pull-out with large fracture surfaces.

In both microstructures, after extensive distributed damage, the damage eventually localised in the area under the load pin and the damage growth transitioned to the middle layer as the deflected cracks opened and grew upwards in the middle layer. During this damage propagation, the load continued to grow, but at a decreasing rate  
320 as seen in Figures 10(a) and (b) after the instance (c) marked in the graphs. When the mechanical response was dominated by the damage propagation in the middle layer, both microstructures yielded qualitatively similar responses.

In the case of the co-cured microstructure, the load eventually decreased as the damage propagated through the whole middle layer, leading to the final failure of the  
325 specimen. In the bonded microstructure, however, the final failure of the middle layer was not reached and the load continued to increase due to superior integrity of the microstructure prior to testing, compared with the co-cured microstructure, as seen in Figures 5(b) and (a), respectively.

## 5. Conclusions

330 This paper established that the mechanics of failure of natural crossed-lamellar microstructures can be replicated in synthetic high-performance engineering composites. It can be further concluded that:

- the mechanical toughening mechanisms of the crossed-lamellar microstructure

335 were successfully reproduced in CFRP. These mechanisms include parallel tunnel cracking in the inner layer, crack deflection when the cracks advance to the middle layer, and debonding and frictional sliding of the lamellae in the middle layer;

- the first ever prototyping routes to synthesise the crossed-lamellar microstructure in CFRP were conceived. Two routes were developed – the first route involves co-curing the whole microstructure at once while the latter relies on curing the layers separately and bonding them after curing;

340

- the co-cured microstructure has tougher macroscopic interfaces than the bonded microstructure, while the structural integrity and alignment are better in the bonded microstructure;

- the crossed-lamellar microstructure dissipates energy in a stable manner upon breaking, while preserving structural integrity up to large curvatures.

345

In summary, this paper contains the first ever synthesised crossed-lamellar CFRP microstructures, and their respective mechanical analysis. It demonstrates their energy dissipation and diffusion capabilities, and establishes their significance for light-weight damage tolerant design.

## 350 Acknowledgments

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## Appendix A. Parametric FE study for identifying suitable microstructure parameters

### Appendix A.1. The model geometry

The feasibility of the crossed-lamellar microstructure in a carbon/epoxy material system was studied using a parametric unit cell FE model shown schematically in Figure A.1 with variables in Table A.1. The objective of the model was to identify suitable configurations, i.e., configurations where the damage transitions to the middle layer under increasing load, after creating diffuse damage in the inner layer.

The unit cell model (Figure A.1) contains, in all macroscopic layers, several plies with a  $\pm\theta$  arrangement with respect to the thickness. Additionally:

- in the inner and outer macroscopic layers, the leftmost and rightmost plies have only half the thickness so that periodic boundary conditions can be applied;
- in the middle macroscopic layer, only half the thickness of the  $+\theta$  and  $-\theta$  plies are contained in the model due to its periodicity.

Table A.1: Variables considered in the parametric study.

| Variable                     |             | Baseline value                    | Other values investigated                                           |
|------------------------------|-------------|-----------------------------------|---------------------------------------------------------------------|
| Inner and outer layer height | $h_I, h_O$  | 1 mm                              | 2 mm                                                                |
| Middle layer height          | $h_M$       | 2 mm                              | 1 mm                                                                |
| Macroscopic interface        |             | PES,<br>$t_{MI} = 50 \mu\text{m}$ | PES, $t_{MI} = 25 \mu\text{m}$ ;<br>Epoxy, $t_{MI} = 1 \mu\text{m}$ |
| Fibre orientation angle      | $\pm\theta$ | $\pm 45^\circ$                    | $\pm 30^\circ$ ; $\pm 60^\circ$                                     |
| Prepreg thickness            | $t_{ply}$   | $125 \mu\text{m}$                 | $25 \mu\text{m}$                                                    |

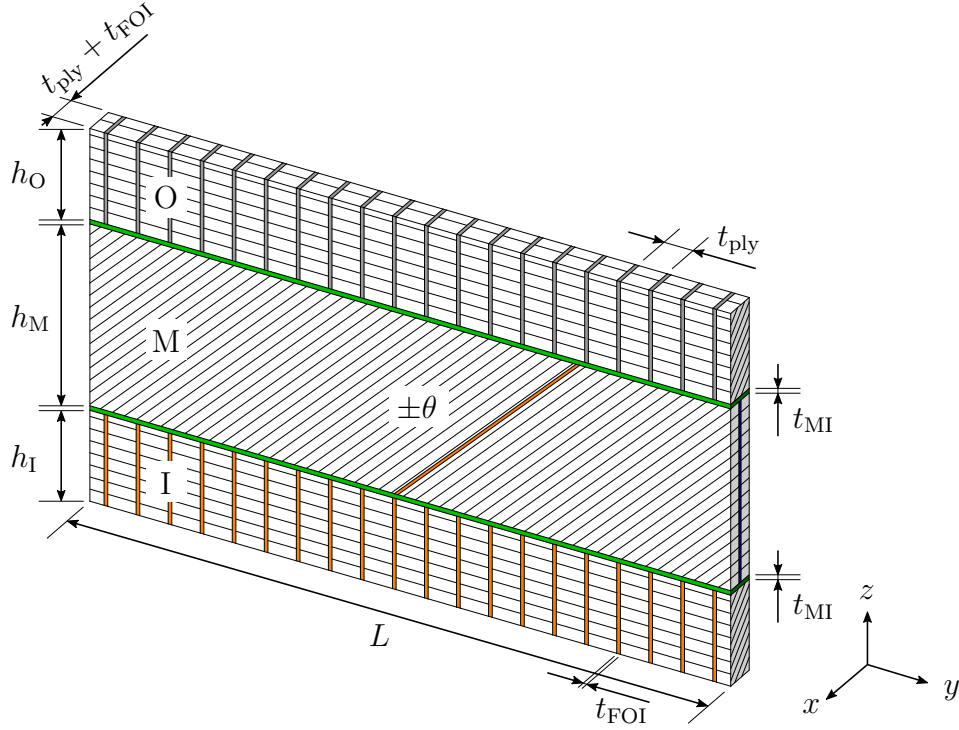


Figure A.1: The geometric parameters of the unit cell model used in the parametric study.

The unit cell model allows for multiple cracking of the 1<sup>st</sup> order interfaces in the inner layer and it is capable of simulating the delamination between the macroscopic layers, the debonding between the 1<sup>st</sup> order lamellae in the middle layer, and the splits  
500 along the 2<sup>nd</sup> order lamellae (i.e. along the fibres) at a prescribed crack density defined by  $L$  in the middle layer.

A baseline configuration is defined (see Table A.1), from which several other configurations are analysed by changing one parameter at the time as indicated in Table A.1. Configurations with different layer heights will be referred in the rest of this text using  
505 the syntax  $[h_I \ h_M \ h_O]$ . For instance,  $[1 \ 2 \ 1]$  refers to the layer heights in the baseline configuration.

The material properties associated with the model and its parameters are given in Table A.2. Depending on the ply thickness,  $t_{ply}$ , two sets of material properties for the prepreg are considered: the material properties of standard-thickness prepreg Hexcel  
510 IM7/8552 (Table A.2) and the material properties of thin-ply prepreg Skyflex USN020A

(Table A.2). Furthermore, the geometric parameters that are not treated as variables in the study are given in Table A.3.

#### *Appendix A.2. Boundary conditions*

The model is loaded in displacement-controlled pure bending about the model  $x$ -  
 515 axis (Figure A.2) using periodic boundary conditions. According to Figure A.3 and  
 assuming a small angle  $\phi$ , the displacements in the  $y$ -direction on the left and right  
 boundaries,  $u_2^L$  and  $u_2^R$ , can be written as

$$u_2^L(x, z) = \bar{u}_2^L + \phi \cdot z + \tilde{u}_2(x, z) \quad (\text{A.1})$$

$$u_2^R(x, z) = \bar{u}_2^R - \phi \cdot z + \tilde{u}_2(x, z),$$

where  $\bar{u}_2^L$  and  $\bar{u}_2^R$  are displacements due to eventual stretching,  $\phi$  is the angle of rotation  
 from the vertical plane, and  $\tilde{u}_2$  is the warping field (fluctuation terms). The periodic  
 520 boundary conditions can be expressed by combining the equations as

$$u_2^R(x, z) - u_2^L(x, z) = \bar{u}_2^R - \bar{u}_2^L - 2z\phi. \quad (\text{A.2})$$

In an FE environment, this equation can be implemented using a master node  $M_1$ :

$$u_2^R(x, z) - u_2^L(x, z) = u_2^{M_1} - 2zu_4^{M_1}, \quad (\text{A.3})$$

where  $u_i^{M_1}$  is the displacement  $i$  of the master node  $M_1$ . Bending can be applied  
 by giving  $u_4^{M_1}$  a displacement corresponding to a desired curvature while the degree  
 of freedom 2 is left free to ensure no overall forces are applied in direction 2. The  
 525 displacement in the  $x$  and  $z$ -directions on the left and right surfaces are also periodic  
 according to the equation

$$u_i^R(x, z) - u_i^L(x, z) = u_i^{M_1}, \quad i = 1, 3, \quad (\text{A.4})$$

Table A.2: Material parameters associated with the FE model. ST stands for standard-thickness prepreg and TP stands for thin-ply prepreg. In the prepreg properties, the subscript 1 denotes the fibre direction and subscripts 2 and 3 denote the transverse directions.  $E_{ij}$  is the elastic modulus,  $G_{ij}$  is the shear modulus and  $\nu_{ij}$  is the Poisson's ratio.  $k$  is the elastic stiffness of the cohesive law and is assumed to be the same for all Modes.  $\tau_I$ ,  $\tau_{II}$  and  $\tau_{III}$  are the damage initiation tractions of the cohesive interfaces in Mode I, II and III, respectively, and  $G_{Ic}$ ,  $G_{IIc}$  and  $G_{IIIc}$  are the corresponding critical energy release rates.  $\alpha$  is the exponent of the mixed-mode power law,  $Y^c$  is the transverse compressive strength of the prepreg (assumed to be the same for both materials),  $\sigma^y$  is the yield strength and  $\epsilon^f$  is the strain to failure.

| Prepreg    | ST                             | TP                                   | Prepreg                              | ST&TP        | Epoxy             | ST                    | TP              |     |                      |                      |
|------------|--------------------------------|--------------------------------------|--------------------------------------|--------------|-------------------|-----------------------|-----------------|-----|----------------------|----------------------|
| $E_{11}$   | GPa                            | 164.0 <sup>[37]</sup>                | 101.7 <sup>[38]</sup>                | $\tau_I$     | MPa               | 50 <sup>[39]</sup>    | $E$             | GPa | 4.67 <sup>[37]</sup> | 3.35 <sup>[40]</sup> |
| $E_{22}$   | GPa                            | 12.0 <sup>[37]</sup>                 | 6.0 <sup>[40]</sup>                  | $\tau_{II}$  | MPa               | 80 <sup>[39]</sup>    | $\nu$           | -   | 0.3 <sup>[41]</sup>  | 0.38 <sup>[40]</sup> |
| $E_{33}$   | GPa                            | 12.0 <sup>†</sup>                    | 6.0 <sup>†</sup>                     | $\tau_{III}$ | MPa               | 80 <sup>†</sup>       |                 |     |                      |                      |
| $G_{12}$   | GPa                            | 4.6 <sup>†</sup>                     | 2.4 <sup>[40]</sup>                  | $G_{Ic}$     | kJ/m <sup>2</sup> | 0.3 <sup>‡</sup>      | <b>PES</b>      |     |                      |                      |
| $G_{13}$   | GPa                            | 4.6 <sup>†</sup>                     | 2.4 <sup>†</sup>                     | $G_{IIc}$    | kJ/m <sup>2</sup> | 1.0 <sup>[42]</sup>   | $E$             | GPa | 2.6 <sup>[43]</sup>  |                      |
| $\nu_{12}$ | -                              | 0.3 <sup>[41]</sup>                  | 0.2 <sup>[40]</sup>                  | $G_{IIIc}$   | kJ/m <sup>2</sup> | 1.0 <sup>†</sup>      | $\nu$           | -   | 0.4 <sup>[43]</sup>  |                      |
| $\nu_{13}$ | -                              | 0.3 <sup>†</sup>                     | 0.2 <sup>†</sup>                     | $\tau_\mu$   | MPa               | 10 <sup>[42]</sup>    | $\sigma^y$      | MPa | 85 <sup>[43]</sup>   |                      |
| $\nu_{23}$ | -                              | 0.4 <sup>†</sup>                     | 0.4 <sup>†</sup>                     | $\alpha$     | -                 | 1.0 <sup>†</sup>      | $\varepsilon^f$ | -   | 0.65 <sup>[43]</sup> |                      |
| $k$        | $\frac{\text{MPa}}{\text{mm}}$ | 23.4·10 <sup>5</sup> <sup>[44]</sup> | 16.8·10 <sup>5</sup> <sup>[44]</sup> | $Y^c$        | MPa               | 304.7 <sup>[41]</sup> |                 |     |                      |                      |

<sup>†</sup> Assumed

<sup>‡</sup> Assumed based on [45]

where  $u_i^R$  and  $u_i^L$  are the displacements  $i$  on the right and left boundaries, and  $u_i^{M_1}$  is the degree of freedom  $i$  of the master node  $M_1$ , which is left free.

The front and back surfaces of the inner and outer macroscopic layers are assigned  
530 periodic boundary conditions according to

$$u_i^F(y, z) - u_i^B(y, z) = u_i^{M_2}, \quad i = 1, 2, 3, \quad (\text{A.5})$$

where  $u_i^F$  and  $u_i^B$  are the displacements  $i$  on the front and back surfaces, and  $u_i^{M_2}$  is the degree of freedom  $i$  of the master node  $M_2$ , which is left free in all directions.

The macroscopic middle layer has symmetry boundary conditions on the front and back surfaces. On the front face, the symmetry is imposed through setting the nodal  
535 displacement in the  $x$ -direction,  $u_1^F$ , equal to zero and leaving the displacements in the  $y$ - and  $z$ -directions,  $u_2^F$  and  $u_3^F$ , free. On the back face, the displacement  $u_1^B$  is set equal

| Table A.3: The geometric parameters of the FE model |                           |                         |                                |
|-----------------------------------------------------|---------------------------|-------------------------|--------------------------------|
|                                                     | $b^\dagger [\mu\text{m}]$ | $L [\text{mm}]$         | $t_{\text{FOI}} [\mu\text{m}]$ |
| ST                                                  | 126                       | 6.3 (7.3 <sup>‡</sup> ) | 1                              |
| TP                                                  | 26                        | 6.3                     | 1                              |

<sup>†</sup> Note  $b$  is defined by  $t_{\text{tply}}$  and  $t_{\text{FOI}}$

<sup>‡</sup> For configuration  $\theta = \pm 30^\circ$

to the displacement  $u_1^{\text{M}_2}$  to allow for Poisson's effect, while the displacements  $u_2^{\text{B}}$  and  $u_3^{\text{B}}$  are left free.

### Appendix A.3. Damage mechanisms and material models

540 The types of damage allowed in the FE model are schematically illustrated in Figure A.2. They include the brittle failure of the 1<sup>st</sup> order interfaces in the inner layer and at a selected location along the fibre direction in the middle layer, the plastic deformation of the thermoplastic PES at the macroscopic layer interfaces, and the debonding and frictional sliding of the lamellae in the middle layer.

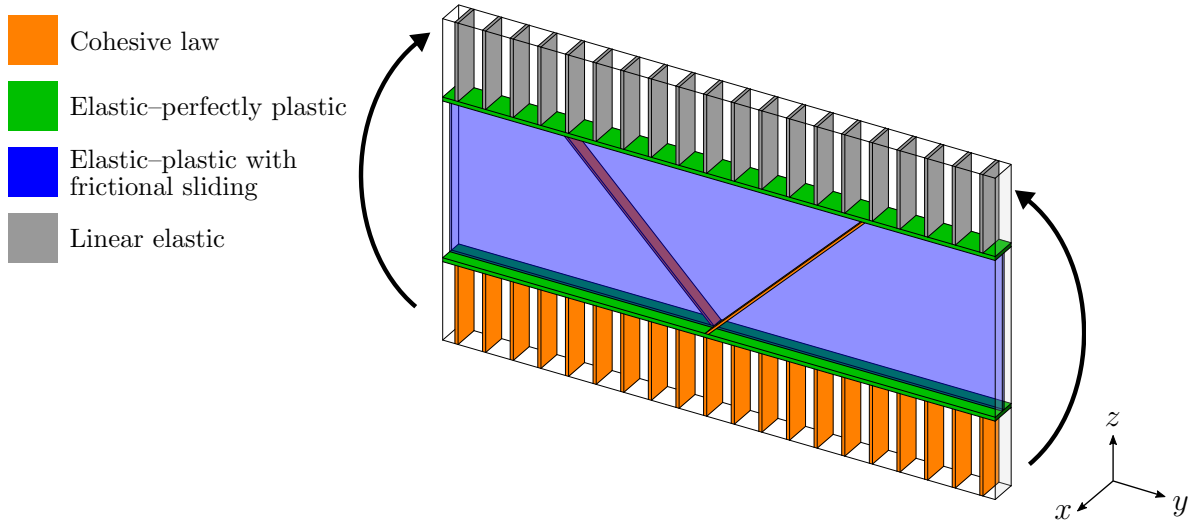


Figure A.2: The failure mechanisms that can be simulated with the parametric model include cracking in the inner layer and at a selected location in the fibre direction in the middle layer, delamination at the macroscopic layers and debonding and frictional sliding of the lamellae in the middle layer. Failure of the top layer is not included in the model. The arrows indicate the direction of applied bending.

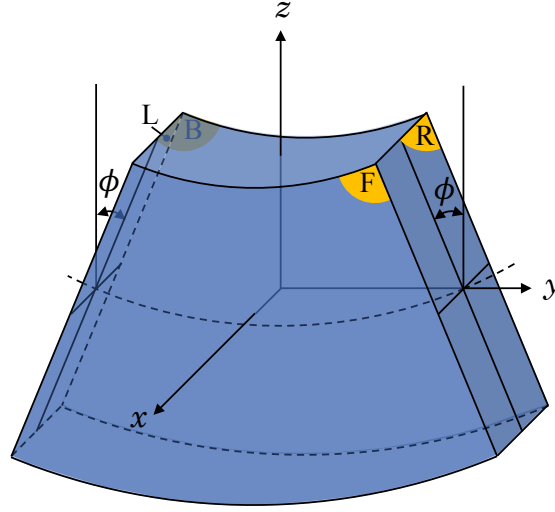


Figure A.3: The displacement in the  $y$ -direction of a beam in pure bending depends on the distance from the neutral axis,  $z$ , and the angle of distortion from neutral axis,  $\phi$ .

545 The failure of the 1<sup>st</sup> order interfaces in the inner layer and at a selected location along the fibre direction in the middle layer is modelled using cohesive elements with thickness  $t_{\text{FOI}}$  (see Table A.3). The cohesive elements follow a traction-separation law with a quadratic stress criterion for damage initiation and linear softening for damage propagation. The damage evolution is energy-based, and the mixed-mode behaviour is  
550 governed by the power-law criterion with exponent  $\alpha$ . The 1<sup>st</sup> order interface directly below the possible crack location in the middle layer is made 0.05% weaker than the rest so that the damage would propagate to the middle layer at this prescribed location.

The plastic deformation of the macroscopic PES interfaces is modelled using elastic-perfectly plastic material response with von Mises yield criterion and isotropic hardening. The constitutive law of the material in shear is schematically illustrated in Fi-  
555 gure A.4(a). The material starts yielding when it reaches its strength,  $\tau^y$ , and exhibits perfectly plastic behaviour until its strain to failure,  $\gamma^f$ . After this, the material exhibits constant frictional shear stress  $\tau_\mu$ . In the configuration with epoxy in the macroscopic interface, the interface is modelled using cohesive elements with the same behaviour as  
560 the cohesive elements at the 1<sup>st</sup> order interfaces in the inner layer.

The debonding and subsequent frictional sliding of the plies in the middle layer are

modelled as an epoxy interface of thickness  $t_{\text{FOI}}$  (see Table A.3) with an elasto-plastic constitutive law. von Mises yield criterion is used with isotropic hardening, with the yield stress adapted for the shear strength of the prepreg. After the material reaches its strain to failure, it exhibits constant frictional shear stress. The failure shear strain  
565 depends on the element thickness,  $t_{\text{FOI}}$ , according to the energy equivalence

$$U = G_{\text{IIc}} \cdot A = \frac{1}{2} \tau_{\text{II}} \cdot \gamma^{\text{f}} \cdot V = \frac{1}{2} \tau_{\text{II}} \cdot \gamma^{\text{f}} \cdot A \cdot t_{\text{FOI}} \quad \Rightarrow \quad \gamma^{\text{f}} = \frac{2G_{\text{IIc}}}{\tau_{\text{II}} \cdot t_{\text{FOI}}}, \quad (\text{A.6})$$

where  $U$  is the internal energy of the element,  $G_{\text{IIc}}$  is the Mode II critical energy release rate,  $A$  is the surface area of the element,  $\tau_{\text{II}}$  is the shear strength of the material and  $V$  is the element volume. The frictional sliding subsequent to the debonding of the  
570 interface is modelled by introducing a constant frictional shear stress  $\tau_{\mu}$  as schematically illustrated in Figure A.4(b).

The failure of the outer layer is not included in the model because that is the last part to fail in the sequential failure of the crossed-lamellar microstructure, and it does not largely contribute to the overall toughness of the microstructure. Therefore, in the  
575 outer layer, the interfaces have a thickness  $t_{\text{FOI}}$  and their response is linear-elastic with

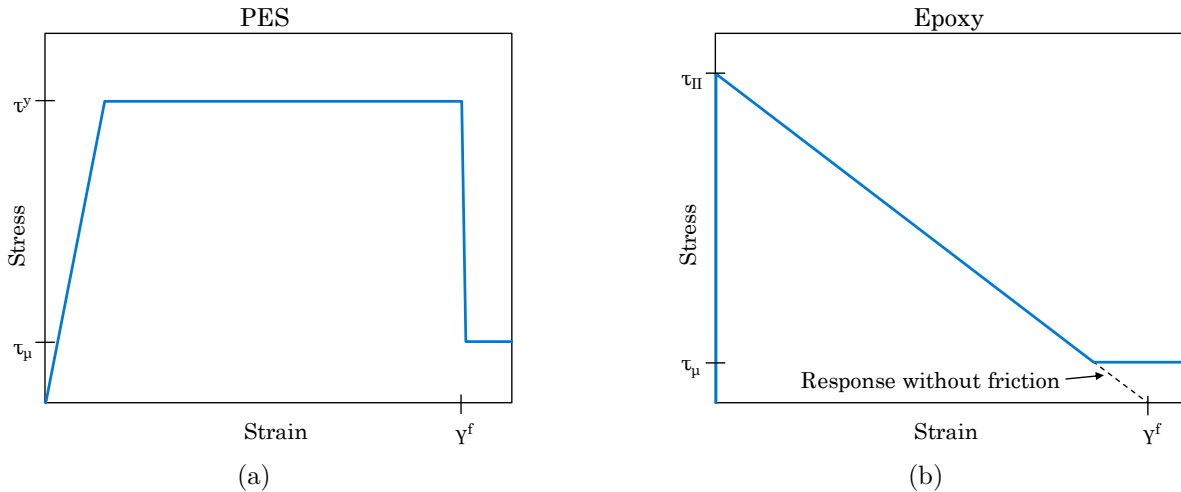


Figure A.4: Assumed constitutive laws (in shear) of a) PES and b) epoxy.  $\tau^y$  is the yield strength of the material in shear,  $\tau_{\mu}$  is the frictional shear stress,  $\gamma^{\text{f}}$  is the failure strain and  $\tau_{\text{II}}$  is the Mode II critical stress of the material. Note that the elastic part of the curve in Figure A.4(b) is very stiff but not vertical.

the material properties of the epoxy given in Table A.2.

The bulk of the CFRP prepreg is modelled as transversely isotropic linear-elastic material (Table A.2). The fibre orientation angle  $\pm\theta$  is taken into account by assigning appropriate material orientations.

#### 580 *Appendix A.4. Element and analysis properties*

The FE model described above was created and run in Abaqus/Standard (version 6.14-3). Figure A.5 illustrates the meshed FE model. The linear-elastic and elastoplastic areas of the model were modelled using 8-node solid elements with reduced integration and enhanced hourglass control. The areas with traction-separation material response were assigned 8-node cohesive elements. The Abaqus built-in automatic  
585 stabilisation scheme with an adaptive damping factor was used in order to make the solution more stable.

#### *Appendix A.5. Post-processing*

The mechanical response of the model was monitored by recording the displacement  
590 and reaction force of the master node that is used for applying the bending in the mo-

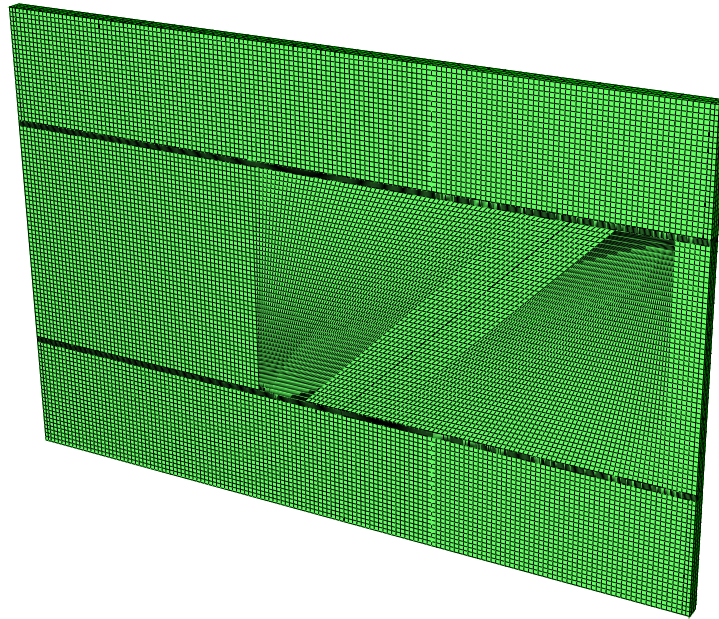


Figure A.5: Parametric FE model and the mesh that were used in the analysis.

del. The displacement and reaction force were converted to curvature,  $\kappa$ , and moment normalised by the second moment of area,  $M/I$ , according to

$$\kappa = \frac{2\phi}{L} \quad \text{and} \quad M/I = \frac{R}{2I}, \quad (\text{A.7})$$

where  $\phi$  is the applied rotation,  $L$  is the length of the model,  $R$  is the reaction force and  $I$  is the second moment of area.

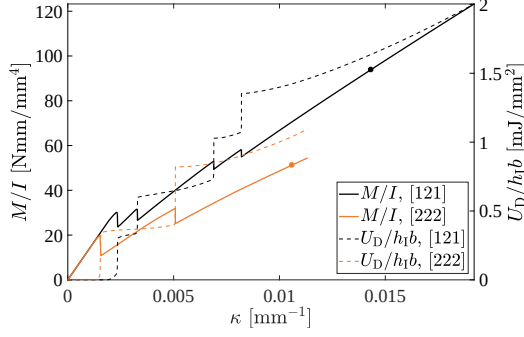
595 The initiation of compressive failure of the outer layer was evaluated by recording the instant the stress component  $\sigma_y$  exceeded the transverse compressive strength of the prepreg  $Y^c$ . As the failure of the outer layer was not modelled explicitly, the simulations were continued slightly beyond this point to monitor what would have happened to the mechanical response.

600 The damage in the inner layer was quantified by extracting the dissipated damage energy,  $U_D$ , and normalising it by the cross-sectional area of the inner layer  $h_I b$ .

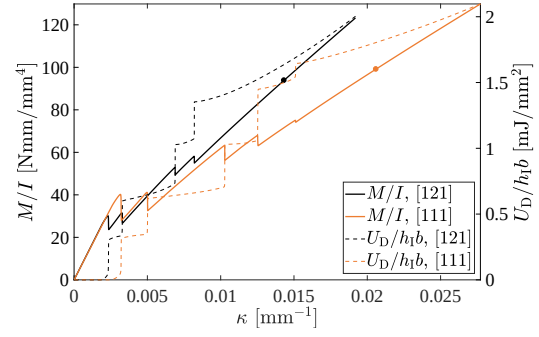
### *Appendix A.6. Results*

Figure A.6 shows the normalised moment and dissipated damage energy as functions of the applied curvature for each studied parameter. In each figure, the baseline configuration outlined in Table A.1 is shown in black. The dot on the curves indicates  
605 the instant the compressive stress in the outer layer exceeds the transverse compressive strength of the material. The load drops in the curve are associated with tunnel cracks growing in the inner layer.

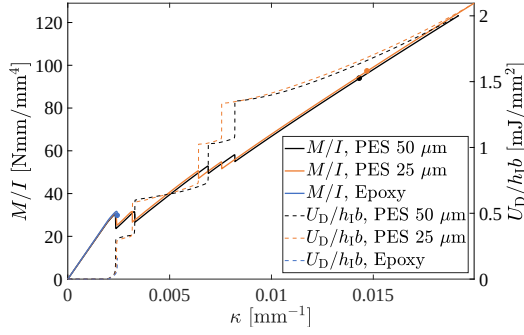
Due to the unstable failure of the model with epoxy at the macroscopic interfaces,  
610 no data for this model is available after the initiation of the first macroscopic crack (Figure A.6(c)). Figure A.7 shows the damage variable of this model at the 1<sup>st</sup> order interfaces, at the macroscopic interface and in the crack location in the middle layer at the first load drop.



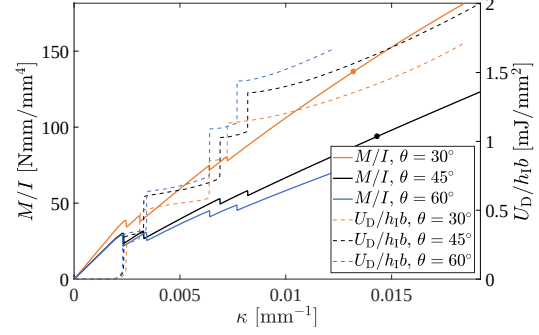
(a) Effect of inner/outer layer height.



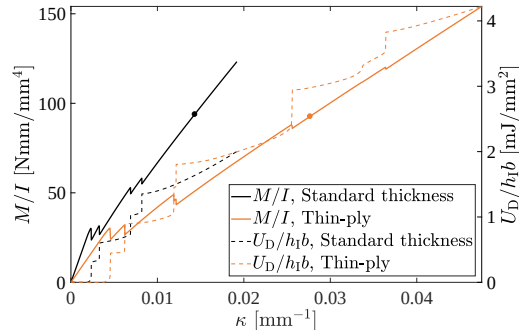
(b) Effect of middle layer height.



(c) Effect of macroscopic interface.



(d) Effect of fibre orientation angle.



(e) Effect of prepreg type.

Figure A.6: Normalised moment and energy dissipation as functions of curvature. The parameters are given in the subcaption and the dot on the curves indicates the instant the compressive stress in the outer layer exceeds the transverse compressive strength of the prepreg. The baseline configuration in each plot is shown in black.

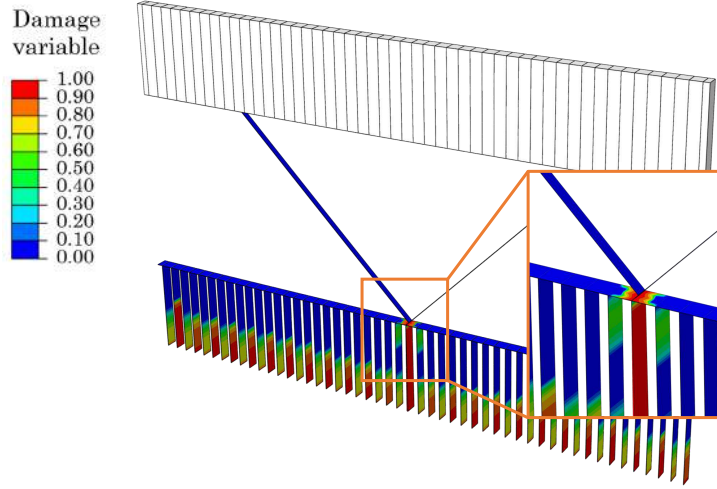


Figure A.7: The damage variable of the model with epoxy interface at the first order interfaces, at the macroscopic interface and in the crack location in the middle layer.

#### Appendix A.7. Discussion

The results indicate that increasing the inner/outer layer height leads to earlier damage initiation and lower relative energy dissipation (Figure A.6(a)) and to lower crack density in the inner layer. Furthermore, the effective stiffness of the  $[2\ 2\ 2]$  configuration after tunnel cracking is lower than in the baseline due to the lower inner/outer to middle layer ratio. Increasing the inner/outer layer heights thus has a negative overall effect on the microstructure.

Decreasing the middle layer height delays the damage initiation and leads to slightly more damage dissipation in the inner layer (Figure A.6(b)). However, due to the lower proportion of middle to inner/outer layer height than in the baseline, the effective stiffness after tunnel cracking is lower than in the baseline. Despite performing better in terms of damage initiation and energy dissipation than the baseline, prototyping the  $[1\ 1\ 1]$  configuration with the co-curing procedure was not feasible.

A configuration with epoxy at the macroscopic interfaces fails by delamination at the macroscopic interfaces (as opposed to damage propagation to the middle layer). Figure A.7 shows that the damage is growing into the macroscopic interface immediately after the first tunnel crack has formed. This suggests that a tougher interface is required in order to promote damage propagation to the middle layer.

The thickness of the PES has a small effect on the mechanical response and damage energy dissipation of the microstructure (Figures A.6(c)). The thicker interface is tougher and therefore less prone to delamination, making it the preferred choice for  
635 the interface.

The fibre orientation angle has the most pronounced effect on the stiffness of the microstructure, with the model with the smallest angle exhibiting, as expected, the stiffest behaviour (Figure A.6(d)). In this configuration, the fibres in the middle layer are more parallel to the loading direction, giving the structure a stiffer response. Despite  
640 being stiffer, the configuration with the  $\pm 30^\circ$  fibre orientation angle dissipates slightly less energy in the inner layer compared with the baseline.

The simulation with the  $\pm 60^\circ$  fibre orientation angle was terminated when the cracks in the middle layer started fully opening and the plies started debonding, as the results can only be generalised for the failure of the inner layer due to the prescribed  
645 crack density in the middle layer. The fibres in this configuration are more parallel to the thickness direction than in the baseline, facilitating the damage propagation to the middle layer, and leading to failure of the configuration at a significantly smaller curvature compared with the other fibre orientation angles.

Using thin-ply prepreg delays the onset of damage and leads to twice as much energy  
650 dissipation in the inner layer than for the baseline (Figure A.6(e)). In addition, the thin-ply solution has more interfaces per width to debond in the middle layer and is therefore likely to outperform the standard-thickness prepreg in terms of energy dissipation in the middle layer.

According to the parametric study, all the studied configurations apart from the  
655 configuration with an epoxy interface are suitable for reproducing the damage mechanisms of the crossed-lamellar microstructure in the inner layer (with varying degrees of energy dissipation) with subsequent damage propagation to the middle layer under increasing load. Therefore, taking also into account considerations of feasibility and manufacturability, the values listed in Table 1 for the various parameters were chosen  
660 for the experiments.