

Interface micro-texturing for interlaminar toughness tailoring: a film-casting technique

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Abstract

In this work, we developed a film-casting technique to deposit thin ($\sim 13\text{ }\mu\text{m}$) layers of poly(lactic acid) (PLA) on the interface of carbon/epoxy prepregs, with the aim of increasing the interlaminar toughness. PLA patches with fractal shape were explored, based on preliminary results showing that the toughening effect increases when PLA is deposited at multiple scales simultaneously. Double Cantilever Beam (DCB) and 4-point End-Notched Flexure (4ENF) tests showed an increase in interlaminar toughness of, respectively, up to 80% for Mode I and 12% for Mode II. This is specially remarkable because the interface thickness is only $13\text{ }\mu\text{m}$. Moreover, it was demonstrated that this technique can promote interaction between neighbouring layers where PLA has been cast, thus triggering fibre bridging and leading to a further enhancement of toughness.

Keywords: Carbon fibres (A), Delamination (B), Fibre bridging (C), Fractography (D), Thermoplastic

1. Introduction

Interlaminar failure (or delamination) is among the most common failure modes in fibre-reinforced composites [1]. Its propagation is typically governed by the toughness of the interface between layers [2] rather than simply by the strength of adhesion between layers, even though the latter is more commonly measured in the literature on interface modification (particularly in Mode II [3]).

For Fibre Reinforced Polymers (FRP), various techniques have been developed to prevent interlaminar failure, such as Z-pinning [4] and stitching [5], which however may cause

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a local reduction of the mechanical properties in the laminate. In order to avoid this,
 10 a frequent approach to increase the interlaminar toughness is the use of a thermoplastic
 phase [3], either as an additive to the matrix or directly applied to the interlaminar region
 [6]. Yasaee et al. [7, 8] designed an interleaved crack-arrest feature using different ma-
 terials, including a thermoplastic film and polyamide particles, showing the capability of
 such 10 mm wide strip to locally increase the fracture toughness G_c . They concluded that
 15 the increased interlayer thickness only contributed marginally to the increase of fracture
 toughness, as most of the contribution was provided by the inserted fibres or films acting
 as a mechanical link between the fracture surfaces. Wang et al. [9] followed a similar ap-
 proach by exploring the effect of 150 μm thick patches of poly(ethylene-co-methyl acrylate)
 and poly(ethylene-co-methacrylic acid) (EMA and EMAA) on the Mode I interlaminar
 20 toughness (G_{Ic}) of carbon-fibre/epoxy laminates, showing an increase in the propagation
 value of G_{Ic} of up to 100%. This was however accompanied by a nearly 30% loss of short-
 beam shear strength and a significant reduction of flexural modulus, in line with similar
 studies on composite laminates toughened with thermoplastic film interlayers [10].

Electrospun nanofibres [11] have also been proposed to achieve thermoplastic tough-
 25 ening in carbon-fibre composites [10, 12]. Li et al. [10] used polysulfone (PSF) electro-
 spun nanofibrous membranes to toughen the epoxy matrix of unidirectional carbon-fibre
 preregs, which led to an increase in G_{Ic} of around 280% without affecting the flexural
 properties; however, they did not investigate the effect on Mode II toughness. A similar re-
 sult was obtained by Saghafi et al. [13], who studied the influence of polyvinylidene fluoride
 30 (PVDF) electrospun membrane thickness on Mode I toughness of interleaved carbon-fibre
 reinforced polymers (CFRP), showing an increase of 42% and 98% with an interleave of
 20 and 42 μm thickness respectively.

Another successful approach to increase interlaminar toughness is the patterning of
 the interlaminar region with small controlled particles, rather than with a continuous
 35 film. This was achieved by Zhang et al. [14] by inkjet printing a solution of poly(methyl
 methacrylate) (PMMA) and poly(ethylene glycol) (PEG) onto the surface of CFRP prepreg
 tape. With this technique, they achieved a 40% increase in the propagation value of G_{Ic} ,
 with a limited reduction of interlaminar shear strength, while the effect on the Mode II
 toughness (G_{IIc}) was not investigated.

Most of the approaches reported above proved successful at increasing the interlaminar toughness. However, very often this happens at the expense of the strength/stiffness of the laminate [15]; in some cases, they do not allow for a μm -level control of the thermoplastic phase thickness [9], or even more often they allow for improvements in Mode I with significant drawbacks in Mode II, and vice versa [6]. In this study, we propose a film-casting method to deposit extremely thin layers ($13\mu\text{m}$) of poly(lactic acid) (PLA, thermoplastic) directly onto the interface of carbon/epoxy prepreg, with the aim of achieving an increase of interlaminar toughness in both Modes I and II, and at the same time limit the amount of added material. The effect of $13\mu\text{m}$ -thick PLA patches on crack propagation is investigated, with regards to two specific configurations: (a) PLA patches cast only on the initial delamination plane and (b) PLA patches cast also on the two neighbouring plies (top & bottom) in order to promote ply interaction and evaluate its effect on G_{Ic} and G_{IIc} .

2. Materials and methods

2.1. Film casting of PLA

As Figure 1(a) shows, a $25\mu\text{m}$ -thick Kapton film was laser-cut (with an Oxford Lasers DPSS etcher) to produce holes of the pattern desired for the PLA patches and then applied to the surface of an uncured unidirectional **thin-ply** carbon/epoxy prepreg (SkyflexTM USN020A [16], properties in Table 1), in order to act as a mask for the subsequent casting of PLA. The Kapton film was vacuum-pressed onto the prepreg for a short time in order to provide a good adhesion to the latter. A solution prepared from PLA pellets dissolved in 1,4 dioxane (mass ratio 1:12) [17] was then cast onto the masked uncured prepreg by means of an automatic film applicator (Elcometer 4340, Elcometer Limited, UK). After casting, the masking was removed in order to allow for full evaporation of the solvent, eventually leaving a texturing of dry PLA patches on the prepreg surface, deposited in correspondence of the holes laser-cut in the masking film (Figure 1(b)). **Thin-ply prepreg was used for its low thickness ($\sim 20\mu\text{m}$), in order to promote the interaction of PLA patches deposited on consecutive plies, as will be shown in Section 2.2 and Figure 3.**

2.2. Pattern of PLA patches

Following a scoping study detailed in Appendix A, we chose a fractal shape for the PLA patches, as this combines a wide range of scales and exhibits a large perimeter-to-area

ratio, and these two features were observed to provide a considerable toughening effect in both failure modes. The selected shape was taken from the Julia set [18] (Figure 2(a)), with the following complex rational function:

$$f(z) = z^2 + 0.8i, \quad (1)$$

z being a generic point of the complex space. In order to obtain a high area fraction of PLA whilst keeping a large perimeter-to-area ratio, a cut-off series of 21 terms was chosen to select the points in the complex plane, providing a connected domain [19]. The fractal shape thus obtained from Equation 1 is plotted in Figure 2(a), while Figure 2(b) shows the actual corresponding laser-cut Kapton film.

2.3. Specimens tested

Three types of interface are tested and compared:

- 0) **Baseline**: epoxy interface of the unmodified prepreg, with no PLA (Figure 3(a));
- 1) **Fractal $\times 1$** : PLA fractal patches cast on the delamination plane only, with an area fraction $A_f = 33\%$ (Figure 3(b));
- 2) **Fractal $\times 3$** : PLA fractal patches cast on the delamination plane *and* on the two neighbouring interfaces (top & bottom), with $A_f = 33\%$ on each of the 3 plies (Figure 3(c)).

2.4. Test configuration

For Mode I, a Double Cantilever Beam (DCB) test was chosen, whereas for Mode II a 4-point End-Notched Flexure (4ENF) setup was adopted, in order to monitor the full crack propagation and characterise the R -curve of the material, instead of only measuring the initiation value. Figure 4 shows the dimensions of the DCB and 4ENF specimens.

2.5. Specimen preparation

Three unidirectional panels of 130 plies each were laid up, each panel containing six DCB and six 4ENF specimens as reported in Figure 5(a), **with a nominal thickness of 3.1 mm**. This led to 6 repetitions for each of the three patterns tested (Baseline, Fractal $\times 1$

95 and Fractal $\times 3$) in both modes. For both the DCB and 4ENF sides of the panel, a 12.5 μm -thick Teflon film was inserted as a crack starter in between the 65th and 66th ply, whose interface corresponds to the delamination plane. For specimens of type 1 (Fractal $\times 1$), PLA patches were only cast on top of ply 65, whilst for specimens of type 2 (Fractal $\times 3$), they were also cast on top of the neighbouring plies 64 and 66, i.e., below and above the
100 plane of the preimplanted insert.

The panels were then cured in an autoclave according to the supplier's recommendations. After curing, a portion of the panel was cut using a wiresaw and then polished in order to measure the thickness of the PLA film, which proved to be around 13 μm (Figure 5(b)). All specimens were subsequently labelled and waterjet-cut from the panels.
105 The edge surfaces were polished and then sprayed with a white primer paint, resulting in a thin white background to easily visualise the crack tip. A 0.3 mm pencil was used to draw a mark every 1 mm increment, starting from the tip of the Teflon insert and extending throughout the entire delamination region.

Before testing, all specimens were manually pre-cracked in Mode I (both DCB and
110 4ENF [20]), by clamping them in a vice grip and carefully sliding a 1.2 mm-diameter roller in the Teflon insert region; the crack tip advance from the Teflon film (Δa_{pc}) was monitored through a magnifying glass. Unlike pre-cracking directly with the Instron machine, which could cause an unstable crack jump of several mm, this procedure allowed for a small pre-cracking of $\Delta a_{\text{pc}} \simeq 1\text{-}2\text{ mm}$, therefore just enough to break through the resin-rich
115 pocket ahead of the Teflon film, without reducing the useful delamination length for the test. Furthermore, long pre-crack jumps have been shown to produce jagged crack fronts, unlike the straight fronts obtained from smaller pre-cracks in static conditions [21].

2.6. Test methods

2.6.1. Mode I

120 DCB tests were carried out on a 1 kN load cell Instron machine, at a displacement rate of 0.75 mm/min and with an acquisition rate of 10 samples per second. The crack length was measured optically via a travelling microscope, and such measurements were verified to be accurate, as described in Appendix B. A video strain gauge system (iMetrum) was used to record the displacement between two target points on the loading pins. The data
125 reduction follows the standard ASTM-D5528 [22], with a Modified Beam Theory (MBT)

scheme; the initiation value G_{Ic}^{init} was determined with the 5%/MAX criterion [20, 23].

2.6.2. Mode II

4ENF tests were carried out on a 50 kN load cell Instron machine, at a displacement rate of 0.25 mm/min and with an acquisition rate of 5 samples per second. As in Mode I, the crack length was measured optically by means of a travelling microscope. A video strain gauge system (iMetrum) was used to record the relative displacement between a fixed point on the support fixture and a target point on the central hinge of the loading fixture. The data reduction was carried out following the scheme proposed by Martin & Davidson [24], with a compliance calibration determined from the experimental data [25]; the initiation value G_{IIc}^{init} was determined with the 5%/MAX criterion.

3. Results

3.1. Mode I

All three types of interface tested exhibited a stable crack propagation, which proceeded smoothly without jumps after onset of failure. This is visible from the sample load-displacement curves reported in Figure 6(a), with the corresponding R -curves in Figure 6(c). The values of toughness are reported in Table 2 along with the standard deviation, as well as in Figure 6(e) in the form of a bar chart. Figure 7(a) reports the G_{Ic} values normalised to the baseline. Both patterns of PLA (fractal patches on one and on three interfaces) provided an enhancement of G_{Ic} for both initiation and propagation. The Fractal $\times 3$ specimen exhibited extensive fibre bridging (see Figure 8(a)) and the largest increase in propagation toughness (180% of the baseline).

3.2. Mode II

Crack advance was also stable in Mode II, with a nearly constant propagation value of the load (see Figure 6(b) for sample load-deflection curves and Figure 6(d) for the corresponding R -curves), as expected from this type of 4-point flexural test. Toughness values are reported in Table 2 and Figure 6(f), showing that, as in Mode I, also in Mode II the best performance is provided by the Fractal $\times 3$ specimen. With PLA patches cast on multiple layers (Fractal $\times 3$), the propagation value of the interlaminar toughness G_{IIc} reaches 112% of the baseline epoxy interface. Figure 7(b) reports the G_{IIc} values normalised

155 to the baseline. The fracture surface of the Fractal $\times 3$ specimen is shown in Figure 8(b),
with PLA patches clearly visible on both halves of the specimen once it is split open.

4. Discussion

4.1. Mode I

While the baseline (epoxy) interface exhibited a nearly constant value of toughness,
160 the PLA-textured specimens showed a significant R -curve behaviour (Figure 6(c)), with
 G_{Ic} increasing from the initiation value as a process zone developed ahead of the crack
tip. In the Fractal $\times 3$ specimen, this was accompanied by a large amount of fibre bridging
(Figure 8(a)). Because of the use of thin plies, whose thickness (nominally $\sim 24 \mu\text{m}$) can be
close or equal to zero in some spots of the prepreg, PLA patches cast on neighbouring plies
165 can locally touch and bond to each other upon curing of the laminate. This provides the
crack with a path to migrate from the initial plane to a different interface, thus increasing
the energy dissipated during propagation. The amount of fibre bridging observed proves
that the casting of PLA patches on multiple layers promoted an interaction between plies
that would have otherwise behaved independently and led to a delamination localised
170 on a single interface (i.e., on the plane of the preimplanted insert). In addition to the
thermoplastic-toughening of the delaminating interface, fibre bridging provided a further
shielding of the crack front, acting as an extrinsic toughening mechanism in the wake of the
crack [26]. The largest gain compared with the baseline toughness is a 80% increase in the
propagation value (Figure 7(a)). However, the formation of a complete bridging zone was
175 not immediate and took around 12-15 mm to fully develop, as visible in Figure 6(c). This
explains why the initiation value of G_{Ic} did not increase considerably from the baseline
value (Figure 7(a)), and it is indeed very similar for the ' $\times 1$ ' and ' $\times 3$ ' patterned specimens.

4.2. Mode II

180 The baseline interface exhibited a relatively flat R -curve, whereas the PLA-textured
specimens led to a slight increase of G_{IIc} with crack length (Figure 6(d)). As shown in
Figure 7(b), with PLA patches on multiple layers, G_{IIc} increased by around 12% in both
initiation and propagation values. Similarly to Mode I, there may have been a toughening

effect related to the crack migration, although this did not lead to extensive bridging but only to the formation of micro-cracks on the interfaces adjacent to the delamination plane.

4.3. Fractography

The fracture surfaces of the specimens were analysed via SEM microscopy. It was observed that failure of PLA is characterised by a much smoother fracture surface than the epoxy's (Figure 9, with details highlighted in Figure 10 for Mode I), which typically creates cusps with limited strain to failure. Such epoxy features seem to be more pronounced at the boundary with the PLA, suggesting a local thickening of the epoxy interlayer around the perimeter of the PLA patches (Figure 9(f)). This could be another toughening mechanism (particularly in Mode II, as pointed out by Hojo et al. [27]), in addition to the inherent ductility of PLA and the already mentioned fibre bridging. However, this effect would only contribute partially to the overall toughness, as the local increase of thickness is of the order of a few μm , while it has been demonstrated that the interply thickening effect is significant only around or above 0.1 mm for thermoset systems [28]. Finally, SEM analysis showed that failure was mainly cohesive, as suggested by the PLA residues found mirrored on both halves of the specimens (Figure 9(c)).

The SEM images in Figure 9 also provide a further explanation for why the toughness increase was much more significant in Mode I than in Mode II, in addition to fibre bridging (which is observed to be more pronounced in the first failure mode). For the epoxy matrix, Mode I failure is characterised by a very limited plastic deformation compared with Mode II, which instead leads to the accumulation of micro-cracks and shear cusps at the crack tip (Figure 9(a) vs 9(b)). Consequently, the toughening effect of PLA (which inherently has a large strain to failure) on the interface is stronger in Mode I than in Mode II.

5. Conclusions

This paper investigated the potential of film-casting a thermoplastic material onto the interface of carbon/epoxy laminates, in order to increase the interlaminar toughness in Modes I and II. It can be concluded that:

- a manufacturing technique was developed to imprint thin patches of PLA on the surface of thin-ply preregs. The current casting settings allow for a PLA thickness of 13 μm , but there is potential for further reducing the amount of added material;
- it was demonstrated that such thin texturing of PLA can increase the interlaminar toughness of uni-directional CFRP in both Mode I and Mode II;
- local bonding of PLA patches deposited on neighbouring layers can promote ply interaction and fibre bridging, which provide further interlaminar toughening;
- a significant contribution to the interface toughening is provided by the formation of resin pockets induced by the texture of PLA, leading to a non-uniform resin layer. This is observed not to occur in the presence of a continuous film of PLA with uniform thickness;
- an 80% increase in Mode I and 12% in Mode II propagation toughness has been obtained, and there is scope to believe that further benefits can be gained by extending this method to other thermoplastic materials.

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Appendix A. Scoping study

As a preliminary study to investigate the potential of textured film-casting for interlaminar toughening, five different patterns were tested (Figure 11):

1. **Square - 1 scale:** a pattern of simple square patches, with area fraction $A_f = 50\%$;
2. **Square - 3 scales:** a multi-scale version of a square (Sierpinski [29]), with $A_f = 50\%$;
3. **Fractal:** a fractal pattern (from Julia set, $f(z) = z^2 - 0.8 + 0.156i$), with $A_f = 50\%$;
4. **Full film $\times 1$:** a continuous film of PLA throughout the delamination region ($A_f = 100\%$);
5. **Full film $\times 3$:** a continuous film of PLA on the delamination plane and on the top/bottom layers (again, $A_f = 100\%$).

By comparing patterns (1)–(3), we investigated the effect of the number of scales at which PLA is deposited (at equal A_f), whereas comparing (4) and (5) allowed us to evaluate the effect of ply interaction. Mode I and II results are reported in Figure 12. Because this is a scoping study only, two specimens were tested for each of the five patterns, and only the propagation values are reported here, in terms of mean value without error bars.

From this scoping study, it was concluded that:

- PLA patches can be cast on the prepreg with remarkable precision, (Figure 13(a));
- by comparing patterns (1)–(3) ($A_f = 50\%$), the toughness (for both modes) increases with the number of scales at which PLA is cast. This means that, at equal A_f , G_c increases with the perimeter-to-area ratio of the thermoplastic patch, therefore with the epoxy/PLA transitions encountered by the crack front during propagation;
- by comparing patterns (4)–(5) (full film, $A_f = 100\%$), casting of PLA on multiple layers indeed promotes ply interaction, which can significantly increase G_c at least in Mode I via fibre bridging. This is evident from the fractography analysis in Figure 13(b)–(c), where fibre bridging is seen to be much more diffused in the ‘ $\times 3$ ’ than in the ‘ $\times 1$ ’ solution, with contribution from neighbouring plies rather than just from the two layers in contact at the delaminating surface;
- additionally, a continuous film of PLA does not improve the interlaminar toughness (unless cast on multiple layers, and only in Mode I via bridging). This is because an important toughening effect in both modes is provided by the resin pockets induced by the texture of discontinuous PLA patches, eventually with fibre waviness and nesting; features that are not present when the interface contains a continuous PLA film of uniform thickness.

The above conclusions motivated the choice of the fractal pattern studied in §2.2, in order to combine a wide range of scales (from the μm to the mm level), and to achieve a large perimeter-to-area ratio, thus increasing the probability of patches on neighbouring plies to locally touch each other and bond (hence promoting bridging).

Appendix B. Accuracy of crack length measurements

Errors in the measurement of crack length have been demonstrated to cause significant errors in the evaluation of toughness, especially in Mode II (up to 20% [25]). In this work, the accuracy of crack length measurements has been assessed in two ways:

- a) check on the compliance calibration curves, i.e., the experimental compliance C plotted against the observed crack length a . Data reduction based on MBT (§2.6) prescribes a linear relation $C^{\frac{1}{3}}(a)$ and $C(a)$ for Mode I and Mode II respectively. As visible in Figure 14(a)–(f), all compliance calibration curves showed a good degree of linearity. Observation of the crack tip is indeed more challenging in Mode II, so there is in principle a possibility that the sample curves reported in Figure 14(d)–(f) are affected by a systematic error, which would shift the curve horizontally; however, this would not affect the calculated toughness, since in the 4ENF configuration the critical energy release rate G_{IIc} is independent of the crack length [30], and therefore it only depends on the slope of the compliance calibration curve [24];
- b) observation of the fracture surface. The R -curve calculated from the experimental data was superimposed to the fracture surface of the corresponding specimen after the test, as outlined in Figure 15. This showed that the crack front observed and measured during the test compared favourably with the visible crack arrest fronts, with an error of less than ± 0.5 mm, as prescribed by the standard [22].

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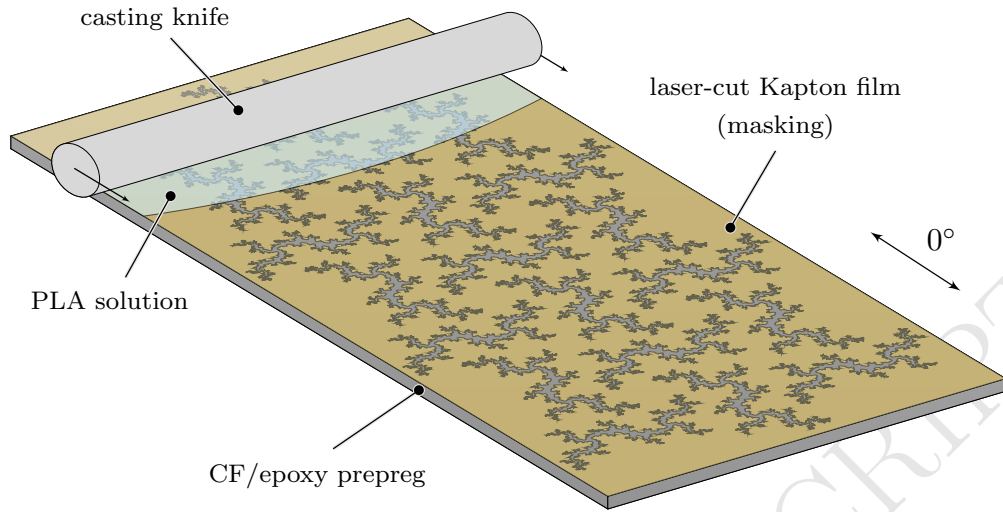
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Table 1: Mechanical properties of Skyflex™ USN020A prepreg [16, 31].

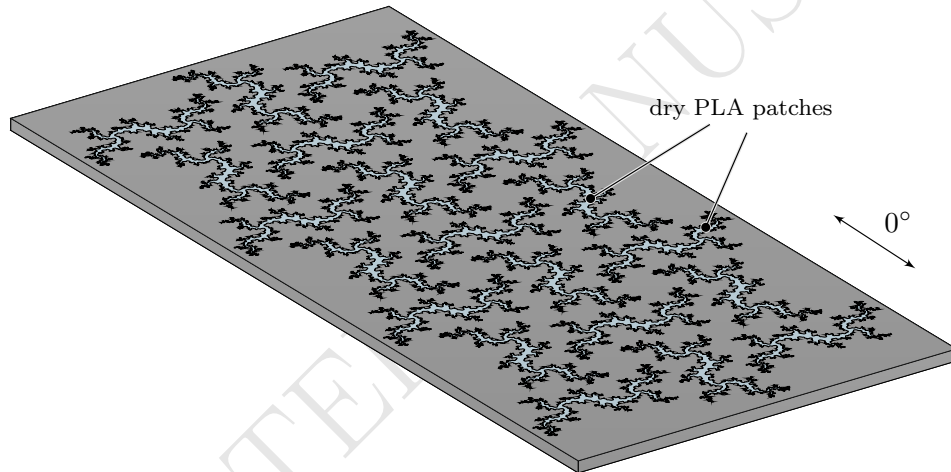
Property	Symbol	Units	Value
Fibre diameter	ϕ_f	μm	6.8
Fibre volume fraction	V_f	-	0.43
Fibre longitudinal modulus	E_f	GPa	235
Mean fibre tensile strength	$X_{f,m}$	GPa	4.4
Ply nominal cured thickness	t	mm	0.024
Ply longitudinal modulus	E_x	GPa	102
Ply transverse modulus	E_y	GPa	6
Ply major Poisson's ratio	ν_{xy}	-	0.3

Table 2: Mode I and Mode II toughness results (standard deviation in parentheses).

Pattern	G_{Ic} [kJ/m ²]		G_{IIc} [kJ/m ²]	
	Initiation	Propagation	Initiation	Propagation
Baseline	0.19 (0.02)	0.19 (0.01)	0.77 (0.09)	0.80 (0.04)
Fractal $\times 1$	0.21 (0.03)	0.22 (0.01)	0.78 (0.12)	0.86 (0.09)
Fractal $\times 3$	0.23 (0.02)	0.35 (0.07)	0.86 (0.13)	0.89 (0.07)

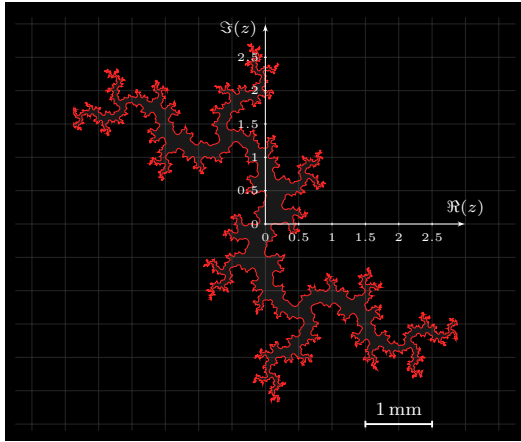


(a) Sketch of the film-casting of PLA patches on the surface of the prepreg.

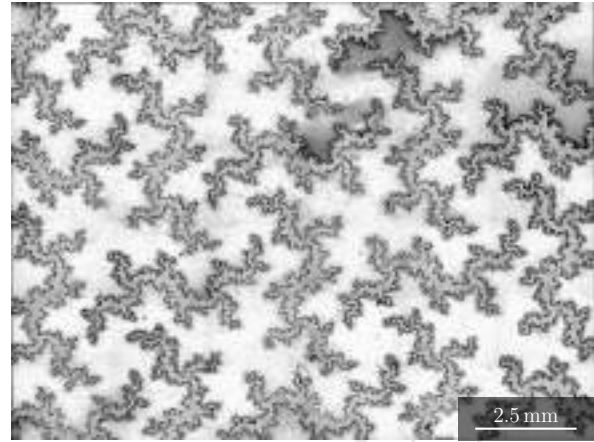


(b) Dry PLA patches deposited after removal of masking and evaporation of solvent.

Figure 1: Film-casting technique: (a) spreading of solution via film applicator and (b) resulting prepreg with cast PLA patches.

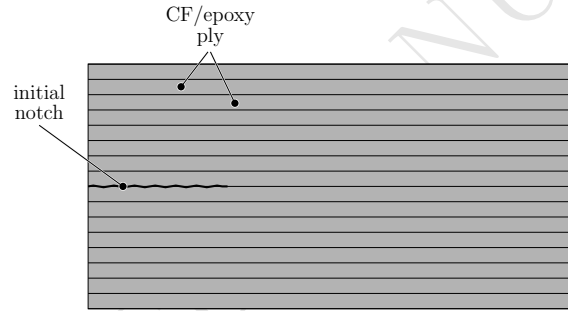


(a) Geometry of the fractal shape as per Equation 1.

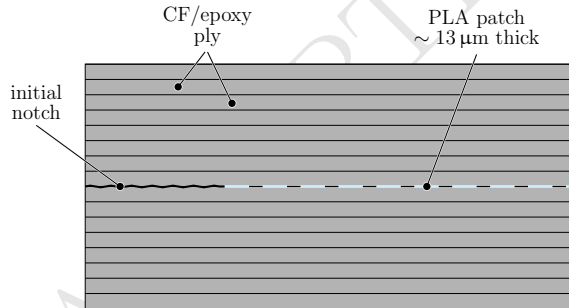


(b) Optical micrograph of the laser-cut Kapton film, leading to an array of patches with 33% area fraction.

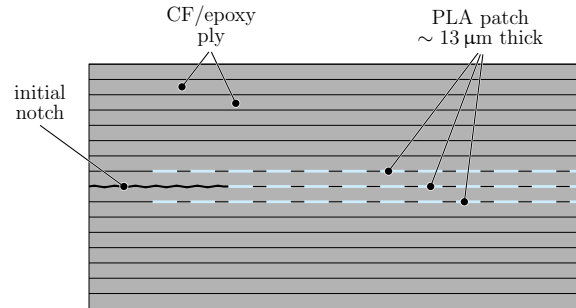
Figure 2: Design of the fractal patches: (a) Julia fractal shape in the complex space and (b) microscope image of the array of patches laser-cut in the Kapton film.



(a) Baseline specimen: epoxy interface.



(b) Fractal $\times 1$ specimen: fractal patches of PLA on delamination plane.



(c) Fractal $\times 3$ specimen: fractal patches of PLA on delamination plane and top/bottom plies.

Figure 3: Sketch of the cross-sectional view of the specimens tested, highlighting the position of PLA patches with respect to the plane of the initial delamination.

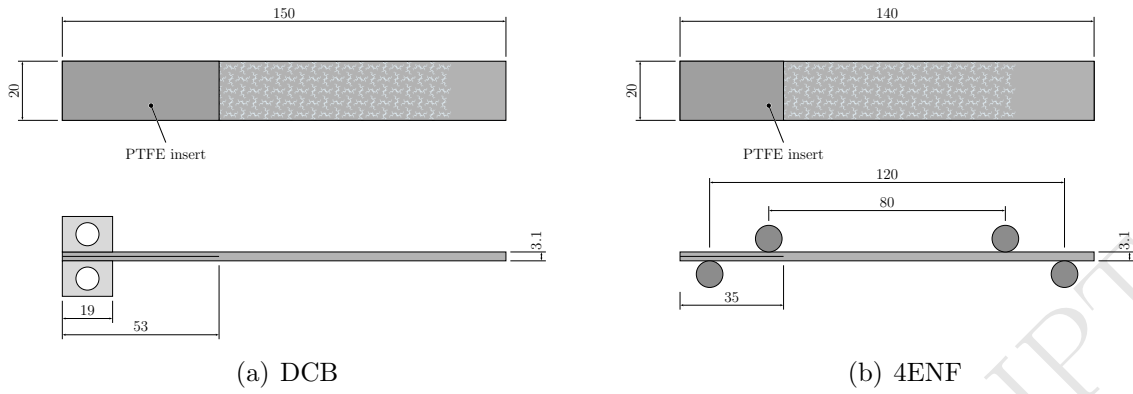


Figure 4: Geometry of the specimens used for (a) Mode I and (b) Mode II testing: in-plane (top) and side view (bottom). Dimensions are in mm.

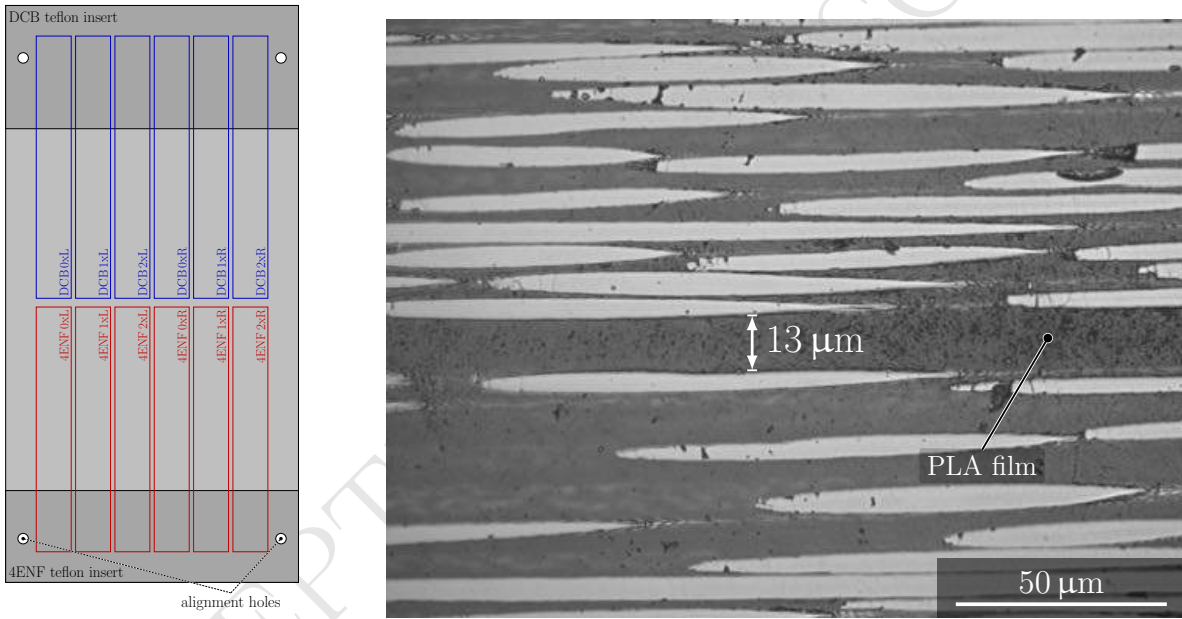
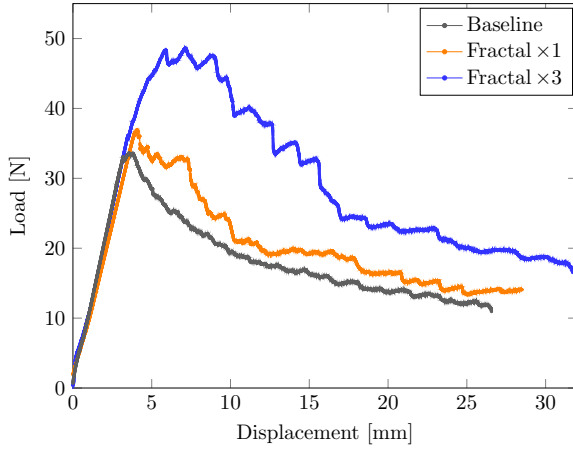
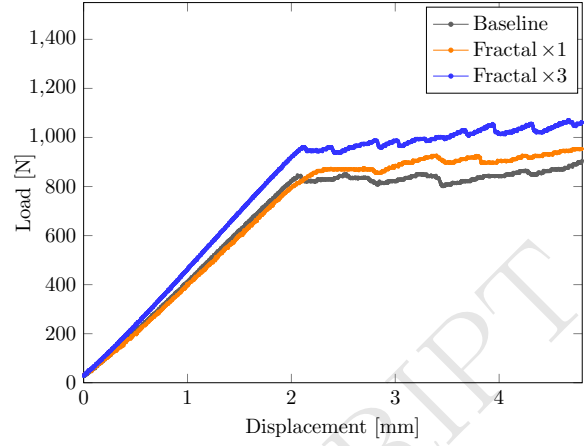


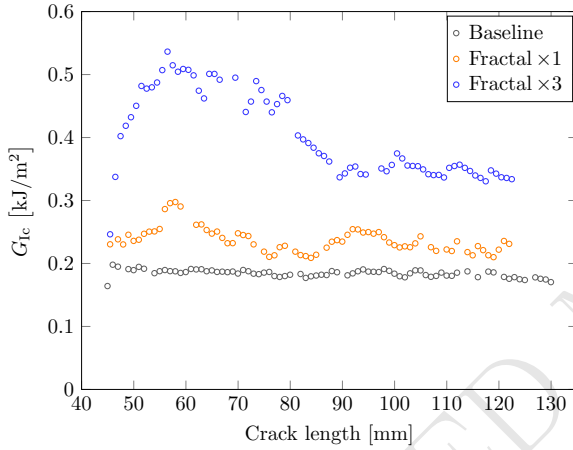
Figure 5: (a) Sketch of the specimen arrangement in the panel (as for the specimens labelling: '0', '1' and '2' denote the Baseline, Fractal $\times 1$ and Fractal $\times 3$ respectively, whereas 'x' identifies the panel. 'L' and 'R' refer to left and right part of the panel); (b) resulting PLA thickness after curing of the laminate.



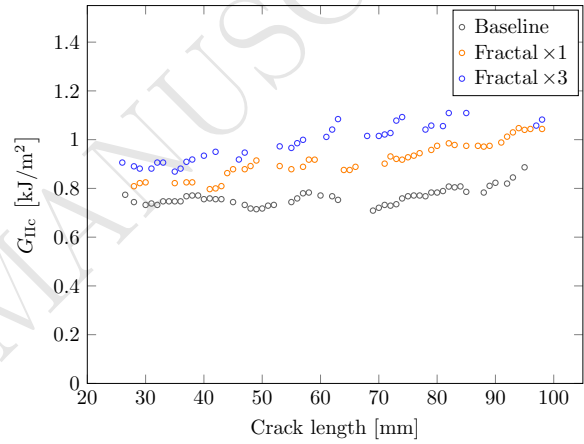
(a) Typical DCB load-displacement curves



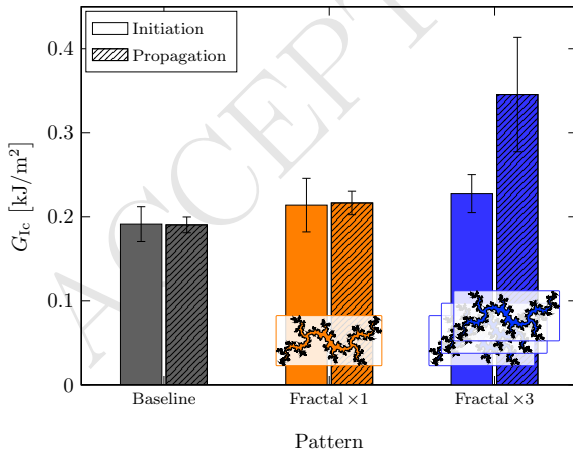
(b) Typical 4ENF load-displacement curves



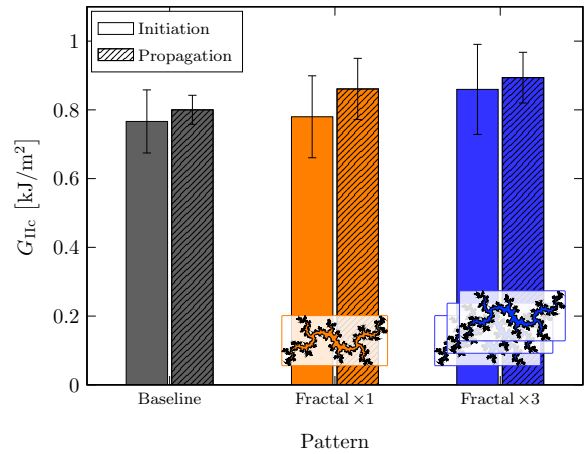
(c) Typical Mode I R -curves



(d) Typical Mode II R -curves

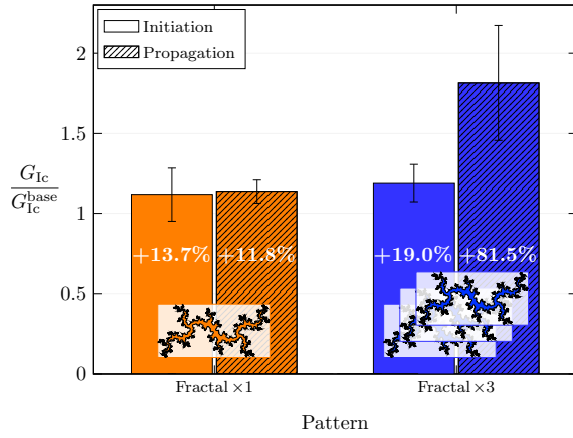


(e) Mode I toughness results

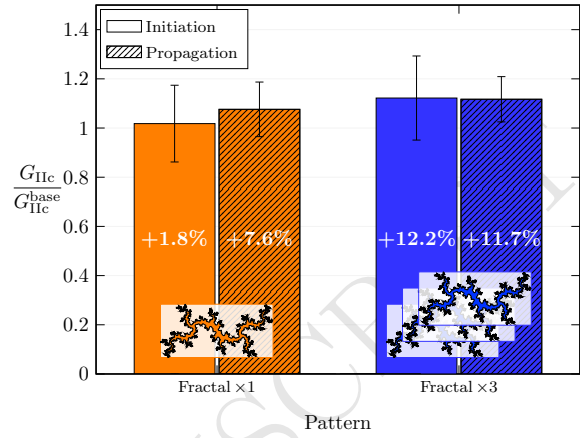


(f) Mode II toughness results

Figure 6: Results of DCB and 4ENF tests: (a),(c),(e) Mode I and (b),(d),(f) Mode II. In the load-displacement curves (a)–(b), differences in the slope of the elastic loading are related to slightly different pre-crack lengths between specimens.



(a) Mode I toughness increase

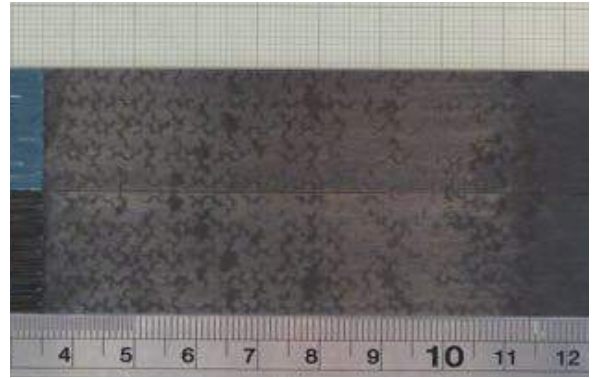


(b) Mode II toughness increase

Figure 7: Interlaminar toughness increase provided by the PLA-textured specimens, in terms of the ratio to the baseline material's toughness G_c^{base} .

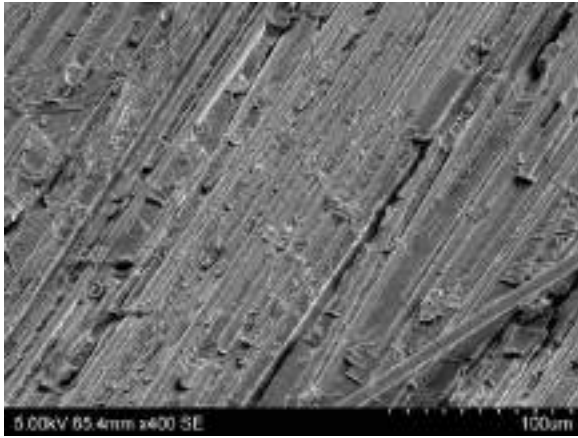


(a) DCB - Fibre bridging

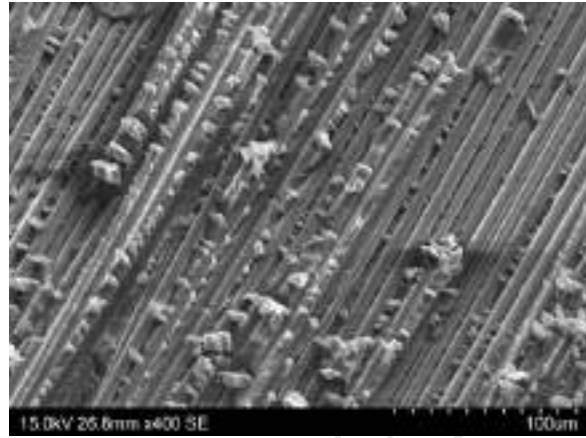


(b) 4ENF - fractured specimen (halves side by side)

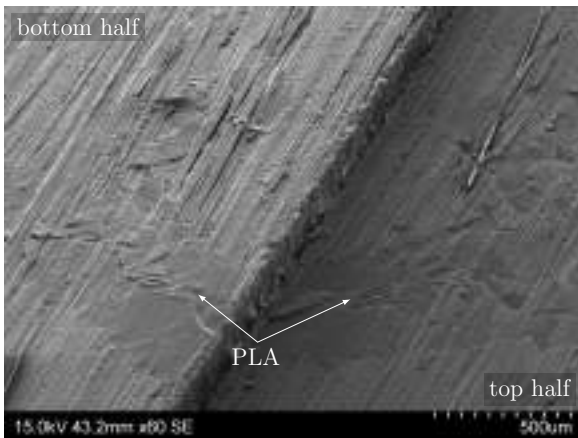
Figure 8: Fractal $\times 3$ specimen: (a) fibre bridging during DCB test and (b) fracture surface of a 4ENF specimen, with PLA patches visible on both halves.



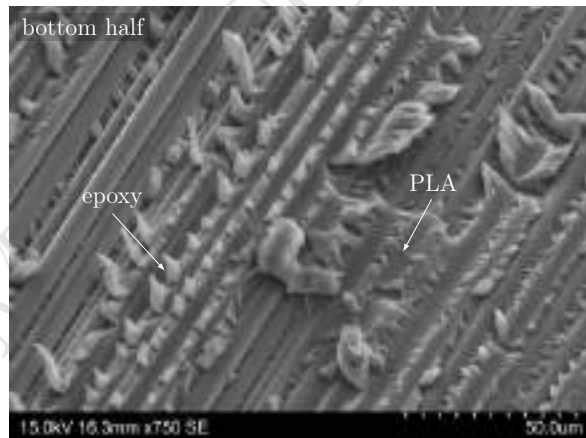
(a) Baseline, DCB



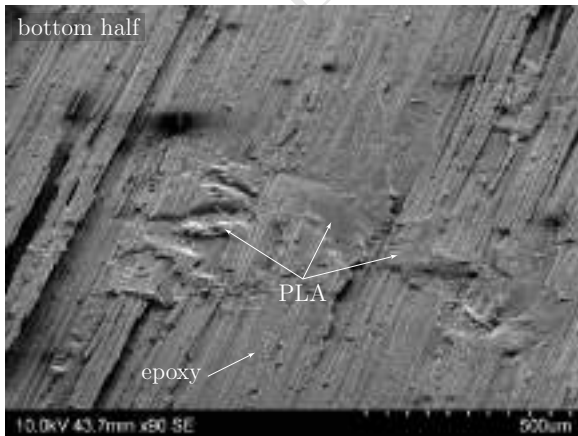
(b) Baseline, 4ENF



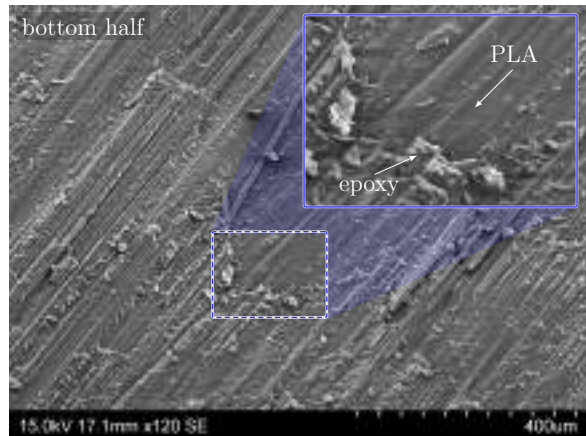
(c) Fractal $\times 1$, DCB: PLA on both halves (side-by-side) of the specimen, suggesting cohesive failure.



(d) Fractal $\times 1$, 4ENF



(e) Fractal $\times 3$, DCB



(f) Fractal $\times 3$, 4ENF: detail of the epoxy thickening at the boundary with PLA.

Figure 9: SEM images of the fractured specimens: (a),(c),(e), Mode I and (b),(d),(f) Mode II.

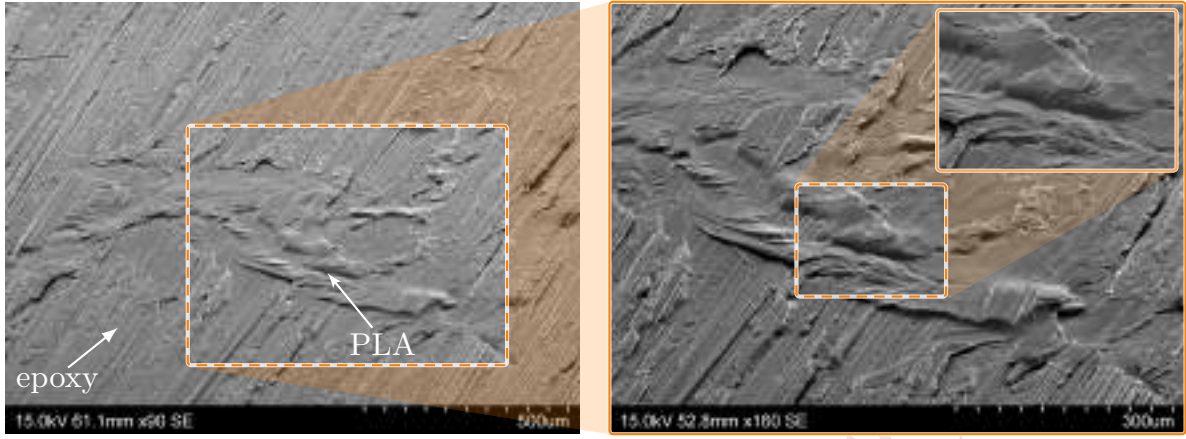


Figure 10: Fractal $\times 1$, DCB: PLA patch on fracture surface (left) and detail of Mode I failure features (right).

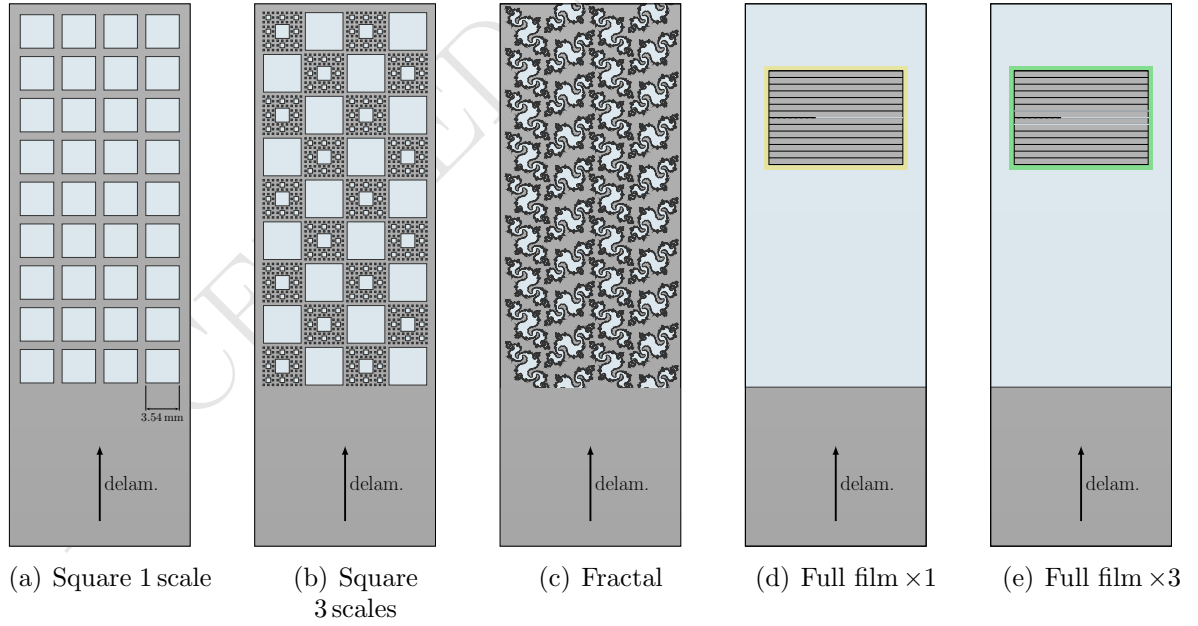
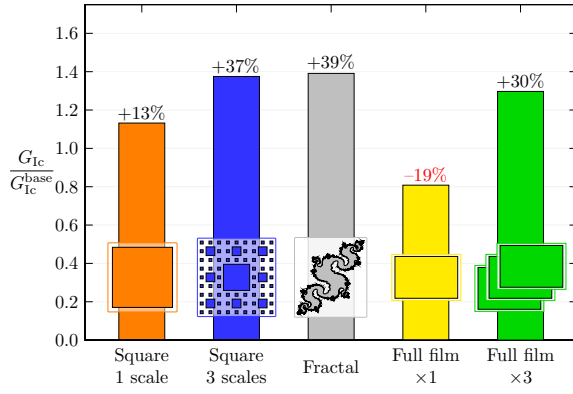
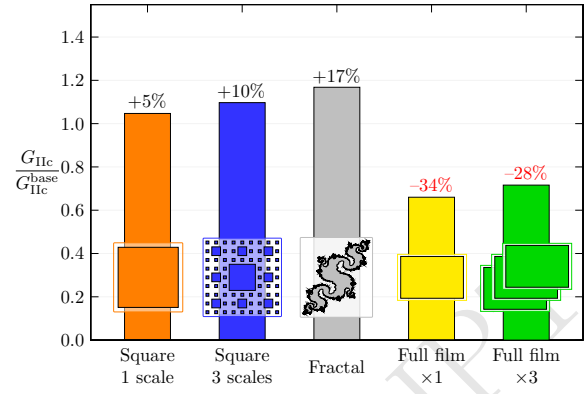


Figure 11: Sketch of the five types of PLA-textured interface tested in the scoping study.

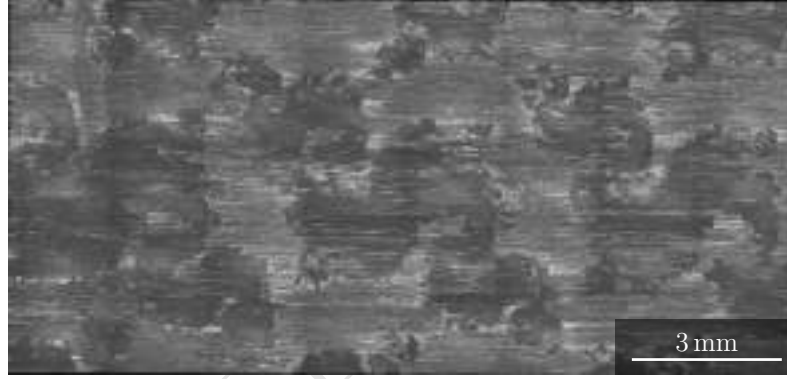


(a) Scoping study: Mode I toughness ratio

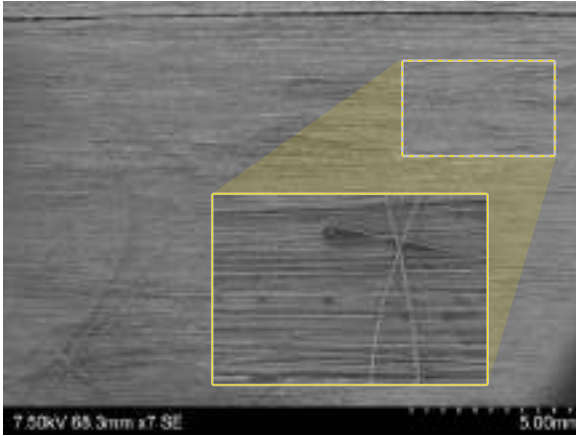


(b) Scoping study: Mode II toughness ratio

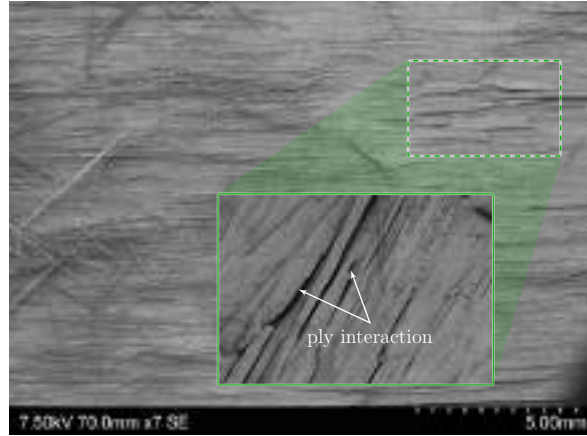
Figure 12: (a) Mode I and (b) Mode II propagation toughness ratio to the baseline (epoxy) interface, for the five different configurations tested in the scoping study.



(a) Fractal pattern (optical microscope)



(b) Full Film $\times 1$ (SEM)



(c) Full Film $\times 3$ (SEM)

Figure 13: (a) Optical micrograph of the DCB fractal specimen tested in the scoping study, and SEM images showing the DCB fracture surface of (b) Full Film $\times 1$ and (c) Full Film $\times 3$. Differences in the extent of fibre bridging are highlighted, proving that, when PLA is cast on multiple layers, entire bundles of fibres protruding from neighbouring plies can contribute to bridging the fracture surfaces.

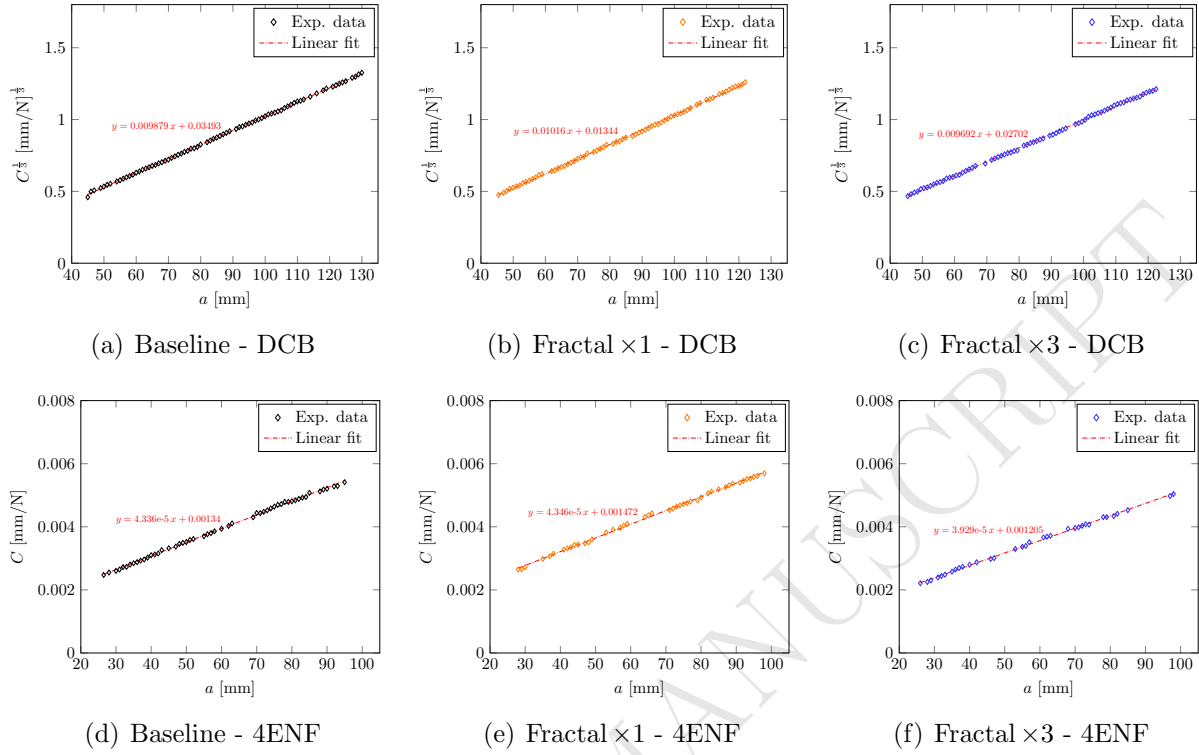


Figure 14: Typical compliance calibration curves: (a)–(c) DCB and (d)–(f) 4ENF, referred to the same specimens reported in Figure 6; C is the compliance of the specimen and a is the crack length.

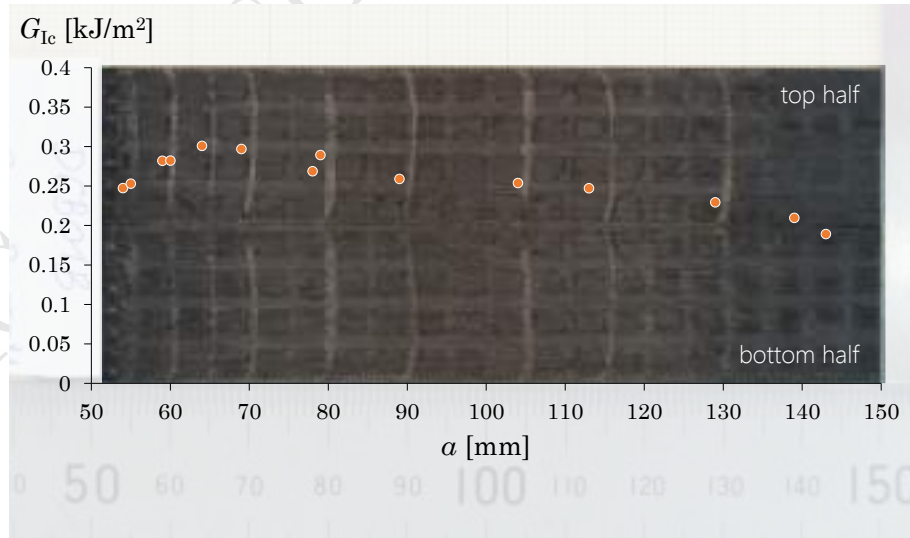


Figure 15: Fracture surface of a Square-1 scale DCB specimen, with the two halves placed side by side against a ruler. The R -curve is superimposed in order to assess the accuracy of crack length measurements.