Mode I and II interlaminar critical energy release rates in all-carbon interlayer unidirectional fibre-hybrids based on ultrahigh-modulus and high-strength fibres

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Abstract

Delamination resistance governs the damage mode in interlayer hybrid composites. The interlaminar critical energy release rates (CERR) G_{Ic} and G_{IIc} are for the first time reported for the different interfaces in hybrids composed of ultrahigh-modulus, low-elongation (LE) carbon fibre layers and high-strength, high-elongation (HE) carbon fibre layers in an epoxy matrix. The G_{Ic} and G_{IIc} values at the LE/LE interface were several times lower than for the HE/HE. The G_{Ic} of the LE/HE interface was close to that of the HE/HE interface, whereas its G_{IIc} was significantly higher than the average of those of the LE/LE and HE/HE interfaces, and 1.5 times higher than LE/LE value. This synergy should be taken into account for the design of LE/HE carbon fibre UD hybrids featuring stable pseudo-ductile behaviour.

Keywords: hybrid laminates, all-carbon hybrids, fracture toughness, hybrid effect

1 Introduction

Traditional fibre-reinforced polymer composites suffer from low failure strains and sudden failure. Fibre-hybrid composites can successfully deal with these disadvantages if they are designed to be pseudo-ductile. Fibre-hybrid composites typically consist of low-elongation (LE) and high-elongation (HE) components and can be created in three different configurations: intrayarn, intralayer or interlayer. The most technologically simple and versatile configuration is the interlayer one, which is composed of LE and HE layers [1, 2]. The pseudo-ductile plateau on the stress-strain diagram is typically achieved by using thin unidirectional layers with a certain proportion of the LE and HE components, and keeping the mode II energy release rate G_{II} at first LE fracture lower than the critical energy release rate G_{IIc} of the applied materials; both factors prevents unstable delamination formation [3]. For a given layer thickness, stacking sequence, mechanical properties and mode II critical energy release rate (CERR) G_{IIc} , a damage mode map can be produced [4].

Despite its importance for hybrid composite design, CERR for such hybrid composites has not been investigated thoroughly nor consistently. As noted in a recent review [2], published results for carbon/glass and flax/basalt hybrids all suffer from the same drawback: only one of the two reference composites was tested instead of both, preventing a convincing discussion on synergetic effects. Few observations, scattered in the existing literature, are worth noting.

Jalalvand et al. [4] used the value of $G_{IIc} = 1000 \text{ J/m}^2$ for the interface between glass and carbon fibre reinforced epoxy UD prepregs, determined using the technique with a central cut in the carbon layers [5]. This is higher than typical values of $800 \dots 1000 \text{ J/m}^2$ of G_{IIc} for glass/epoxy and carbon/epoxy UD laminates (see, for example, [5]). This points to the possibility that there might be a positive hybrid effect on CERR. Hwang et al. [6] studied mode II interlaminar CERR using the ENF test and compared the CERR values for a UD carbon/glass fibre-hybrid laminate and a UD carbon fibre laminate. They measured G_{IIc} values in the range of 400 to 600 J/m^2 for the hybrid laminates, in comparison with $\sim 400 \text{ J/m}^2$ for the carbon fibre laminate. The values for glass fibre laminates were not measured. The difference was explained by “the

existence of glass fibres on the surfaces of the pure carbon fibre side of the [ENF] specimen". These two cases suggest that there may be a positive hybrid effect for mode II CERR in interlayer UD composites. This hypothesis remains unproven in the literature.

Analysis of mode II CERR is hampered by the fact that the toughness values determined according to the standard [7] can depend on factors such as specimen thickness, through-the-thickness normal stress, and the planarity of the crack [5]. The authors [5] argued that "the fracture energy is not a material [or a hybrid material system] constant". This statement, put forward for UD laminates in [5], was further elaborated in [8] for glass/carbon hybrid interfaces.

The effect of pseudo-ductility is also present in all-carbon hybrids combining layers reinforced with ultrahigh-modulus pitch-based and high-strength PAN-based carbon fibres, based on thin ply preregs for UD and quasi-isotropic (QI) laminates [9, 10]. In [9], the value of 500 J/m² was quoted as "the estimated fracture toughness G_{IIc} of the tested materials", which was compared with the theoretically calculated threshold to avoid catastrophic delamination. Czél et al. [10] studied QI laminates, which are composed of UD hybrid sub-laminates. The fracture toughness for these sub-laminates is also investigated. G_{IIc} for the UD sub-laminates was measured using hybrid specimens with a cut central layer, following the methodology of Cui et al. [5]. The measured values for G_{IIc} were 650 J/m² and 1053 J/m² for the two studied types of all-carbon hybrid laminates: [T1000₈/XN80₂]_s and [MR60₁₀/XN80₂]_s respectively (T1000 refers to Torayca T1000GB (12k), MR60 – to Pyrofil MR60H 24P (12k) and XN80 – to Granoc XN-80 (12k)). No analysis of the difference or similarity of the CERR for the hybrid and non-hybrid interfaces was given in [9, 10].

In the present paper, we report CERR values G_{Ic} and G_{IIc} for carbon-carbon unidirectional hybrid composites based on ultrahigh-modulus and high-strength carbon fibres as well as, for the first time, for both of its reference, non-hybrid composites. We employ the standardised testing techniques and treat the determined fracture toughness as a material characteristic. This characteristic will be applicable in practice for laminate thicknesses of the same range as studied here, for example, in cohesive law definition or creation of damage mode map. These measured values can be used to design pseudo-ductile interlayer hybrids and better understand their delamination resistance.

2 Materials and specimens manufacturing

Two carbon/epoxy preregs were provided by *Composite Materials Italy* (CIT) - Toray Group. For convenience, we introduce the following notation: T for the prepreg based on TORAYCA® T800 fibres and D for the prepreg based on DIALEAD® K63720 fibres. The properties of the fibres and the preregs, according to the manufacturer, are shown in Table 1. SEM images (see Figure 7 and Figure 10, discussed in the Results section) show that the diameter of T800 fibres is reasonably consistent, whereas the DIALEAD fibre diameters vary significantly, in the range of 7...11 µm. The ER450 epoxy, which was used in both preregs, is a toughened system [11]. For laminate production, the preregs were placed in a steel frame with inner dimensions 260x260 mm, and hot-pressed for 2 hours at a pressure of 0.5-0.6 MPa and a temperature of 135°C. The pressure variation reflects precision of the pressure control; the large relative variability is caused by the thermal inertia of the thick plates and by the fact that the pressure of 0.5-0.6 MPa is at the low end of the pressure scale of the device. The frame prevents lateral flow of the resin with the fibres under compaction. The thickness of frame was 5 mm. Prepreg packs were always thinner than the frame; the pressure was acting on the fibres via a paper stack to avoid the contact of loading plates to the steel frame during the compaction and the curing process.

Table 1 Properties of the fibres and the prepregs

Prepreg fibres	Abbreviation	Resin	Fibres				Fibre volume fraction	Prepreg nominal thickness, mm
			Young's modulus, GPa	Strength, MPa	Failure strain	Diameter, μm		
TORAYCA® T800	T	ER450	294	5880	2%	5	63-65%	0.10
DIALEAD® K63720	D	ER450	630	2650	0.4%	11	63-65%	0.25

An aluminium (A) foil, with a thickness of 12 μm , was coated with a release agent PMR-90EZ and was located in the middle of the stack. After curing, the panels were cut into strips with a diamond coated, and water-cooled high-speed disk saw. The specimen length was 200 mm, width 20 ± 0.2 mm. The aluminum foil helped to create a weak surface on the contact T/T, D/D or T/D. However, the weak surface is not a real crack. To get the initial crack tip in the needed location of the pristine interface, we use “precracking” by opening the weak surface in mode I with the use of thin wedge. To create an initial crack of a given length in the range of $a_0 = 35 \dots 45$ mm, a 0.5 mm thick steel knife with a wedge end was pushed into the pre-crack, while the specimen was clamped in a vice at the desired position of the crack tip [12]. The pre-crack position was determined by observation of the specimen edge and marked with a narrow white mark. The measurement accuracy of this initial position was estimated to be about 0.5 mm.

Three general lay-up configurations were tested: $T_m/A/T_m$, $D_m/A/D_m$ and $T_m/A/D/T_m$. These lay-ups were labelled T/T, D/D and T/D, respectively, with m being the number of T- and D-layers. The lay-up of the specimens used in DCB and ENF tests and their thickness is given in Table 2. The hot press displayed a pressure variation of 5...8% during the process of curing, which contributes to the thickness differences for the same lay-up.

Table 2 Lay-up and thickness of the tested DCB and ENF specimens.

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	$T_{20}/A/T_{20}$	4.31 ± 0.02	20.0 ± 0.33
		D/D	$D_{10}/A/D_{10}$	4.67 ± 0.04	19.9 ± 0.06
		T/D	$T_{20}/D/A/T_{20}$	4.26 ± 0.04	19.8 ± 0.07
ENF	200	T/T	$T_{20}/A/T_{20}$	4.38 ± 0.03	25.5 ± 0.05
		D/D	$D_{10}/A/D_{10}$	4.27 ± 0.03	25.5 ± 0.09
		T/D	$T_{14}/D/A/T_{14}$	3.37 ± 0.01	25.6 ± 0.01

Note: “ $\pm$ ” denotes standard deviation for five specimens

The T/D specimens were slightly unsymmetrical because of non-symmetrical positioning of the aluminium foil. The resulting offset of the pre-crack from the neutral line was about 0.1 mm (half thickness of D layer). According to [13], an offset of comparable size led to deviations of about 10% in the G_{Ic} values from a DCB test and had no significant influence on the G_{IIc} from an ENF test.

In [14], a criterion is described for an acceptable degree of non-symmetry, formulated in terms of flexural rigidity ratios: $D_c = \frac{D_{12}^2}{(D_{11}D_{22})} < 0.25$ and $B_t = \left| \frac{D_{16}}{D_{11}} \right|$ as low as possible. In these formulae D_{ij} are the components of the bending stiffness matrix from classical lamination theory. i and j in this notation can be 1, 2 or 6 referring to, respectively, the longitudinal,

transverse, and shear directions (the notation used in [14]). Conformity to these two conditions ensure that the test is free from crack jumping and non-uniformity of the strain energy release rate across the specimen width; the reader is referred to [14] for the details. Evaluation of these criteria for the studied laminates give $D_c \cong 0.005$ and $B_I = 0$ for all the layups.

3 Test methods

3.1 Imaging of the internal geometry and fracture surfaces

The internal structure of the T/D specimens was characterised with X-ray computed tomography (micro-CT) using a Nanotom GE micro-CT device. The parameters of the acquisition were as follows: tube voltage 60 kV, current 430 μ A, no filter, 360 projections, frame averaging 2. A molybdenum target was used. The resulting pixel size of the image was 0.8 μ m. The fracture surface was imaged with SEM (JEOL JSM 7001F, voltage 20 kV, working distance 10 mm). No conductive coating was applied to the specimens. The images were taken in the region about 5 mm away from the pre-crack (“initiation area”) and in the middle of the final cracked zone (“propagation area”).

3.2 Double-cantilever-beam (DCB) tests

Double-cantilever-beam (DCB) tests were used in the mode I experiment according to ASTM D5528-14 standard [12]. Figure 1 shows the test configuration and the used nomenclature, and Table 2 specifies the tested laminate lay-up and dimensions. The initial crack length was 35...45 mm. The tests were performed at a displacement rate of 5 mm/min.

Figure 1 Configuration and parameters of the DCB test.

The data processing of the test results was done with Modified Beam Theory method, recommended by the standard [12]. Compliance C of the specimen was calculated as $C = \delta/P$, where δ is the displacement of the machine, P is the load (see Figure 1 for the notations). The dependency of the compliance on the crack length was taken in the form

$$C^{1/3} = A \cdot (a + \Delta) \quad (1)$$

where a is the crack length, A , Δ are fitting parameters. To determine these parameters, linear regression was applied to a set of points $((C_{lin}^i)^{1/3}, a_i^0)$, where i is the specimen index, C_{lin} is the compliance in the original linear region of the load-displacement curve (see Figure 5 further in section 4.2 for an example of the calibration). This procedure mixes the data of the specimens of the same lay-up, which in principle have slightly different thicknesses. Since the thickness variation was below 1% (see Table 2), the influence of this variability could be safely neglected.

Once the calibration was performed, the crack length during loading was calculated as

$$a = \frac{1}{A} C^{1/3} + \Delta, \quad (2)$$

with compliance $C(\delta) = \delta/P(\delta)$. According to the standard [12], the delamination initiation moment is defined by the condition $C(\delta) = C_{+5\%} = 1.05 \cdot C_{lin}$. The initiation CERR value was then calculated as

$$G_{Ic} = \frac{3P^* \delta^*}{2b(a + |\Delta|)}, \quad (3)$$

where P^* and δ^* are the load and displacement corresponding to $C_{+5\%}$, b is the specimen width. The same formula was used to calculate propagation G_{Ic} values.

3.3 End-notch flexure (ENF) tests

A three-point flexure scheme for a beam with an end notch (ENF) was used for the mode II experiments according to the ASTM D7905M-14 standard [7]. Figure 2 shows the test configuration. Table 2 specifies the tested laminate lay-up and thickness. The nominal width b of all the specimens was 25.6 mm, whereas the span length $2L$ (see Figure 2) was 120 mm, and the initial crack length a_0 was 35...45 mm. The tests were performed at a displacement rate of 5 mm/min. The loading was stopped when the crack propagated to the length of 50...60 mm. To perform compliance calibration over a wide range of the crack length, some specimens were unloaded and loaded again, with the formed crack serving as an initial crack in the next loading cycle.

Figure 2 Configuration and parameters of the ENF test.

The data processing of the test results was done with a compliance calibration procedure, recommended by the standard [7]. Compliance C of the specimen was calculated as $C = \delta/P$, where δ is the displacement of the machine and P is the load (see Figure 2 for the notations). The dependency of the compliance on the crack length was assumed to be

$$C = A + ma^3 \quad (4)$$

where a is the crack length, A and m are fitting parameters. The dependency (4) was fitted using the values of the compliance C_{lin} in the linear part of load-displacement curves (constant part of compliance-displacement curves) for all the specimens and all loadings of each specimen for each lay-up. This procedure mixes the data of the specimens of the same lay-up, which in

principle have a slightly different thickness. Since the thickness variation was below 1% (see Table 2), the influence of this variability could be safely neglected.

Once the calibration was performed, the crack length during loading was calculated as

$$a = \left(\frac{C-A}{m} \right)^{1/3}, \quad (5)$$

with compliance $C(\delta) = \delta/P(\delta)$. According to the standard [7], the delamination initiation moment is defined by the condition $C(\delta) = C_{+5\%} = 1.05 \cdot C_{lin}$. The initiation CERR value was then calculated as

$$G_{IIc,init} = \frac{3m(P^*a^*)^2}{2b}, \quad (6)$$

where the load P^* was taken as the load corresponding to $C_{+5\%}$, and the crack length a^* was calculated with (5) for this load.

The standard [7] warns that the crack propagation in ENF test is likely to be unstable and does not recommend calculation of the propagation values of G_{IIc} . This recommendation was followed in the present work.

4 Results and discussion

4.1 Microstructure of the hybrid interface

The microstructure near the lamina interface can affect the interlaminar CERR. Figure 3a illustrates the microstructure of the hybrid laminate near the interface of T- and D-layers. There are no visible signs of the intermingling of the two fibre types at the interface: the layers are separated. At the same time, no significant resin interlayer is observed between T- and D-ply in the laminate.

Using VoxTex software [15], the distribution of fibre directions in the T and D layers near the interface was determined according to the procedure described in [16]. Figure 3b shows the distributions of the fibre orientation angles in- and out-of-plane, and Figure 3c – the corresponding standard deviations. The misalignment of D-fibres is in the typical range for prepreg-based laminates [16]. T-fibres have considerable in- and out-of-plane deviations from the nominal direction, with a standard deviation of the out-of-plane orientation angle as high as 15°. The fibre orientation distributions for the full volume of the corresponding T- and D-ply in both the hybrid and the reference laminates are similar to the distributions shown in Figure 3b.

Figure 3 Internal structure of the hybrid laminate near the interface: (a) three orthogonal slices of a micro-CT image; (b) fibre misalignment in T and D plies for a volume of 50 μm thickness: distribution of in-plane and out-of-plane fibre orientation angles; (c) standard deviations of the misalignment angles.

The high out-of-plane orientation of the fibres in T-plyies suggests the expected high difference between the initiation and propagation values of G_{Ic} for T-fibre laminate. This difference can be created by several phenomena, outlined and discussed in [17]: deflection of the crack front as the crack propagated along the fibre/matrix interface, change of the dimensions of the damage zone and fibre bridging. As shown in the next sub-section, the propagation value of G_{Ic} for T-fibre laminates is indeed much higher than the initiation one, in contrast with D- and hybrid laminates.

4.2 Mode I interlaminar critical energy release rate G_{Ic}

Figure 4 shows the typical results of a DCB test: load-displacement (see Figure 4a) and compliance-displacement (see Figure 4b) dependencies and images of the tests (see Figure 4c). These curves depend on three factors: lay-up, thickness and initial crack length. For the sake of a clear comparison, Figure 4 presents tests on different lay-ups (T/T, D/D and T/D) but with similar initial crack lengths (38 ± 4 mm). The layups for the DCB tests (see Table 2) were chosen in such a way that the specimens had similar initial compliance for the same crack length (see Figure 4a and Figure 4b).

The different specimens (

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	T ₂₀ /A/T ₂₀	4.31 ± 0.02	20.0 ± 0.33
		D/D	D ₁₀ /A/D ₁₀	4.67 ± 0.04	19.9 ± 0.06
		T/D	T ₂₀ /D/A/T ₂₀	4.26 ± 0.04	19.8 ± 0.07
ENF	200	T/T	T ₂₀ /A/T ₂₀	4.38 ± 0.03	25.5 ± 0.05
		D/D	D ₁₀ /A/D ₁₀	4.27 ± 0.03	25.5 ± 0.09
		T/D	T ₁₄ /D/A/T ₁₄	3.37 ± 0.01	25.6 ± 0.01

Note: “±” denotes standard deviation for five specimens

) had considerably different initial cracks length; hence, the differences in the load-displacement curves for individual specimens are large and statistically significant. Figure 4 therefore shows typical raw test results; the scatter of the data is characterized by the G_{Ic} values resulting from these four tests with different initial crack length (

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	T ₂₀ /A/T ₂₀	4.31 ± 0.02	20.0 ± 0.33
		D/D	D ₁₀ /A/D ₁₀	4.67 ± 0.04	19.9 ± 0.06
		T/D	T ₂₀ /D/A/T ₂₀	4.26 ± 0.04	19.8 ± 0.07
ENF	200	T/T	T ₂₀ /A/T ₂₀	4.38 ± 0.03	25.5 ± 0.05
		D/D	D ₁₀ /A/D ₁₀	4.27 ± 0.03	25.5 ± 0.09
		T/D	T ₁₄ /D/A/T ₁₄	3.37 ± 0.01	25.6 ± 0.01

Note: “±” denotes standard deviation for five specimens

Figure 4 Raw data of the DCB tests, typical test results: (a) load-displacement diagrams for T/T, D/D and T/D specimens for similar values of the initial crack length; (b) corresponding compliance-displacement diagrams; (c) photos of the T/T, D/D and T/D tests.

Table 3 Results of DCB tests, with * indicating parameters corresponding to the compliance $C_{+5\%}$.

Sample type	Δ , mm	Initiation							Propagation		
		a_0 , mm	C_{lin} , mm/N	$C_{+5\%}$, mm/N	a^* , mm	δ^* , mm	P^* , N	G_{Ic} , J/m ²	G_{Ic} , J/m ² average, std. dev.(CoV)	averaging range, a, mm	G_{Ic} , J/m ² average, std. dev. (CoV)
T/T	- 3.159	34.6	0.0201	0.0212	35.3	1.725	81.4	266	235 ± 23 (9.7%)	25 ... 50	636
		33.2	0.0180	0.0189	33.8	1.518	80.3	231			726
		33.9	0.0190	0.0200	34.5	1.312	63.1	209			680
		32.4	0.0169	0.0177	33.0	1.279	72.2	235			831
D/D	- 0.637	46.1	0.0226	0.0237	46.8	1.300	54.9	118	105 ± 10 (9.5%)	5 ... 50	140
		50.0	0.0300	0.0315	50.8	1.001	31.8	105			103
		42.6	0.0186	0.0195	43.0	1.956	50.0	107			114
		42.2	0.0181	0.0190	42.9	0.853	44.9	93			126
T/D	- 1.290	38.0	0.0153	0.0161	38.7	0.897	55.6	102	106 ± 6 (5.6%)	5 ... 50	101
		39.0	0.0167	0.0175	39.7	0.950	54.9	104			140
		44.0	0.0233	0.0245	44.7	1.262	51.4	113			125

		42.1	0.0207	0.0217	42.8	1.224	56.5	111			141	
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Significant fibre bridging was observed for the T/T interface, but not for D/D and T/D (see Figure 4c). The fibre bridging in the T/T interface and its absence for D/D can be linked to the high out-of-plane fibre misorientation in T-ply, whereas it is low in D-ply (see section “4.1 Microstructure of the hybrid interface”).

The compliance-displacement curves (see Figure 4b) show a clear initial region with a constant compliance (C_{lin}). Since the delamination onset is at the end of this region, the C_{lin} values were used for compliance calibration.

The load-displacement diagrams were processed, according to equations (2-3), to yield the initiation value of CERR G_{Ic} and propagation values: dependence of G_{Ic} on the crack length a (R-curves). The values are shown in

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	T ₂₀ /A/T ₂₀	4.31 ± 0.02	20.0 ± 0.33
		D/D	D ₁₀ /A/D ₁₀	4.67 ± 0.04	19.9 ± 0.06
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Note: “±” denotes standard deviation for five specimens

, and the R-curves are presented in Figure 6. For the delamination onset,

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	T ₂₀ /A/T ₂₀	4.31 ± 0.02	20.0 ± 0.33
		D/D	D ₁₀ /A/D ₁₀	4.67 ± 0.04	19.9 ± 0.06
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Note: “±” denotes standard deviation for five specimens

lists the raw data for each test: initial crack length a_0 , the constant compliance C_{lin} , the threshold compliance $C_{+5\%}$, the crack length a^* , the displacement δ^* and the load P^* , corresponding to the threshold compliance. The value of initiation CERR G_{Ic} was calculated with equation (3), using the Δ values shown in

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	T ₂₀ /A/T ₂₀	4.31 ± 0.02	20.0 ± 0.33
		D/D	D ₁₀ /A/D ₁₀	4.67 ± 0.04	19.9 ± 0.06
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ENF	200	T/T	T ₂₀ /A/T ₂₀	4.38 ± 0.03	25.5 ± 0.05

		D/D	D ₁₀ /A/D ₁₀	4.27 ± 0.03	25.5 ± 0.09
		T/D	T ₁₄ /D/A/T ₁₄	3.37 ± 0.01	25.6 ± 0.01

Note: “±” denotes standard deviation for five specimens

. The latter were derived from the compliance calibration equations, shown in Figure 5. The propagation CERR value was calculated by averaging G_{Ic} values in the constant region of the R-curves (see Figure 6). These intervals are listed in

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	T ₂₀ /A/T ₂₀	4.31 ± 0.02	20.0 ± 0.33
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		T/D	T ₁₄ /D/A/T ₁₄	3.37 ± 0.01	25.6 ± 0.01

Note: “±” denotes standard deviation for five specimens

. The results confirm the consistency of the measurements. The coefficients of variation of G_{Ic} values are within 10% for initiation and within 15% for propagation. The G_{Ic} values for T800/ER450 laminates provided by the manufacturer are in the range 500-600 J/m² [11], which is similar to the present measurements for propagation.

Figure 5 DCB compliance-calibration diagrams for all three lay-ups.

The G_{Ic} initiation value for the T/T interface is three times lower than those for propagation, and there is a long rising region in the R-curves before the propagation value stabilises (see Figure 6). This may be caused by fibre bridging (see Figure 4c). SEM fractography (see Figure 7) also suggests that fibre bridging is the major energy absorption mechanism: it shows intensive contamination of the crack surface by short (about the fibre diameter) pieces of

broken fibres and matrix “dust”. The number of such pieces seems larger in the crack propagation zone than in the initiation zone (compare Figure 7a and b for the T/T specimen).

Figure 6 R-curves from the DCB test.

Figure 7 Typical SEM images of the fracture surface of the DCB specimens: (a) area of the crack onset (b) area of the crack propagation. The lay-up is indicated on the images.

The G_{Ic} values for the D/D laminate are 2.2 times lower than for the T/T interface for the initiation, and about six times lower for propagation (see

Test type	Specimen nominal length, mm	Interface type	Lay-up	Specimen thickness, mm	Specimen width, mm
DCB	200	T/T	T20/A/T20	4.31 ± 0.02	20.0 ± 0.33
		D/D	D10/A/D10	4.67 ± 0.04	19.9 ± 0.06
		T/D	T20/D/A/T20	4.26 ± 0.04	19.8 ± 0.07
ENF	200	T/T	T20/A/T20	4.38 ± 0.03	25.5 ± 0.05
		D/D	D10/A/D10	4.27 ± 0.03	25.5 ± 0.09
		T/D	T14/D/A/T14	3.37 ± 0.01	25.6 ± 0.01

Note: “ $\pm$ ” denotes standard deviation for five specimens

). The initiation and propagation G_{Ic} values for D/D laminate differ by just 15%. This small difference is attributed to the absence of fibre bridging in D/D case (see Figure 4c). This absence may, in turn, be caused by the low out-of-plane misalignment of the D-fibres (see section “4.1 Microstructure of the hybrid interface”). SEM fractography for D/D laminate (see Figure 7) indeed reveals a clean fibre surface with occasional fibre breaks. This suggests interface debonding as the main mechanism for the crack propagation and energy absorption for the studied D/D laminate.

The mode I CERR of the hybrid T/D interface is almost the same as the mode I CERR of the D/D interface, both for initiation and propagation. Fibre bridging was not observed (see Figure 4c). The fracture surface in the crack initiation region (see Figure 7a) shows clean fibre surfaces on both the T and D sides of the crack. The G_{Ic} values were hence determined by the same mechanism as in D/D interface. In the propagation region, matrix “dust” is present (see Figure 7), suggesting an additional mechanism of energy absorption. However, this is not reflected in the G_{Ic} propagation values: no statistically significant increase was measured for T/D laminates in comparison with D/D.

4.3 Mode II interlaminar critical energy release rate G_{IIc}

Figure 8 shows the typical results of an ENF test: load-displacement (see

Figure 8a) and compliance-displacement (see

Figure 8b) dependencies. These curves depend on three factors: lay-up, thickness and initial crack length. For the sake of a clear comparison,

Figure 8 presents tests on different lay-ups (T/T, D/D and T/D) but with similar initial crack length (36.0 ± 0.4 mm). The slope of the load-displacement diagrams are ordered as $T/T > D/D > T/D$. This is caused by the difference in the T and D fibre stiffness and the difference in the laminate thicknesses (see Table 2). Compliance-displacement curves (see

Figure 8b) show a clear region with constant compliance (C_{lin}), corresponding to the linear load-displacement relation. The C_{lin} values were used for the compliance calibration.

Figure 8 ENF tests, raw data: (a) Load-displacement diagrams for T/T, D/D and T/D specimens with the close values of the initial crack length; (b) the corresponding compliance-displacement diagrams.

Figure 9 displays the compliance calibration curves. All three layups demonstrate the proportionality of the compliance to the cube of the crack length. The order of the compliance values, which is $T/T < D/D < T/D$, is also (as the order of the slopes of the load vs displacement) explained by the difference in T and D fibre stiffness and the difference in the laminate thicknesses (see Table 2). The regression equations, shown in Figure 9, and their inversions according to equation (5) were used to calculate the crack length a^* values, corresponding to the delamination initiation compliances $C_{+5\%}$ (see section 2.4).

Figure 9 ENF compliance calibration: dependencies $C = A + ma^3$ for the three lay-ups.

Table 4 summarises the results of all ENF tests, including the initial crack length a_0 , linear compliance C_{lin} , threshold compliance $C_{+5\%}$, corresponding load P^* , crack length a^* , and the G_{IIc} values calculated using equation (6). The T/T and D/D results displayed consistent results with coefficients of variation of 8.2% (T/T) and 7.2% (D/D). The T/D results displayed a larger scatter: the coefficient of variation of G_{IIc} values was 17.0%. Despite the larger scatter, the G_{IIc}

values passed the Student's t-test for difference of the means (D vs T, D vs T/D and T vs T/D) with a confidence of 99% or higher.

Table 4 Results of the ENF tests.

Sample type	a_0 , mm	C_{lin} , mm/N	$C_{+5\%}$, mm/N	a^* , mm	P^* , N	G_{IIc} , J/m ²	G_{IIc} , J/m ² average, std. dev., (CoV)
T/T	34.3	0.00184	0.00193	36.4	899	890	934 ± 77 (8.2%)
	36.1	0.00193	0.00203	37.6	903	965	
	37.4	0.00200	0.00210	38.9	866	944	
	43.1	0.00240	0.00252	44.5	676	971	
	44.9	0.00255	0.00268	46.4	591	807	
	41.8	0.00230	0.00241	43.3	810	1031	
D/D	27.4	0.00250	0.00262	30.2	677	424	469 ± 34 (7.2%)
	32.2	0.00273	0.00287	34.7	632	488	
	33.6	0.00280	0.00294	35.9	597	465	
	38.1	0.00310	0.00325	40.0	555	502	
T/D	34.2	0.00390	0.00409	35.2	579	1208	1376 ± 234 (17.0%)
	45.1	0.00648	0.00680	46.2	452	1264	
	35.7	0.00418	0.00439	36.9	544	1166	
	34.3	0.00391	0.00411	35.2	690	1705	
	33.6	0.00381	0.00400	34.7	664	1537	

As expected, the G_{IIc} values significantly exceed the corresponding G_{Ic} values for mode I. The D/D interface, as for mode I proved to be about two times weaker than T/T interface.

In contrast with mode I, the G_{IIc} initiation and propagation values for the T/D interface are higher than those for T/T and D/D, being roughly close to the sum of G_{IIc} values of the components of the hybrid. This is an unexpected and very large hybrid effect of +96% in comparison with the average of the component values. This is one of the largest synergies ever reported for a mechanical property in fibre-hybrid composites [1, 2].

SEM fractography (see Figure 10) revealed different fracture features in the T/T and D/D cases compared to the hybrid T/D case. In both the T/T and D/D interfaces, the crack initiation (see Figure 10a) and propagation (see Figure 10b) zones revealed a smooth fibre surface with occasional fibre breakage, but without full fibre removal from its position in the unidirectional ply. In the hybrid case, the morphology of the fracture surface both in the T and D side of the crack is different. Several phenomena can contribute to the increase of energy absorption during the crack formation. Firstly, significantly more matrix is seen on the crack surface than for T and D specimens. This indicates much more irregular debonding than in the non-hybrid cases, leading to an increase in the effective CERR. Secondly, fibre breakage is more prevalent than for T and D specimens. Finally, on both sides of the T/D cracks, fibres of both the T and D type are noticeable thanks to their difference in diameters. This means that the T and D layers were intermingled despite their apparently good separation in Figure 3. This could help to explain the additional energy absorption for detachment of the fibres crossing the crack plane, and corresponds to the similar observations in [6] for glass/carbon hybrid UD laminates.

Figure 10 Typical SEM images of the fracture surface of the ENF specimens: (a) area of the crack onset (b) area of the crack propagation. The lay-up is indicated on the images.

5 Conclusion

The mode I and II interlaminar CERRs in carbon-carbon interlayer unidirectional fibre-hybrids have been measured and for the first time compared to the CERR of the reference composites. The reported values (see

<i>Test type</i>	<i>Specimen nominal length, mm</i>	<i>Interface type</i>	<i>Lay-up</i>	<i>Specimen thickness, mm</i>	<i>Specimen width, mm</i>
DCB	200	T/T	T20/A/T20	4.31 ± 0.02	20.0 ± 0.33
		D/D	D10/A/D10	4.67 ± 0.04	19.9 ± 0.06
		T/D	T20/D/A/T20	4.26 ± 0.04	19.8 ± 0.07
ENF	200	T/T	T20/A/T20	4.38 ± 0.03	25.5 ± 0.05
		D/D	D10/A/D10	4.27 ± 0.03	25.5 ± 0.09
		T/D	T14/D/A/T14	3.37 ± 0.01	25.6 ± 0.01

Note: “±” denotes standard deviation for five specimens

and Table 4) can be further used for the simulation of mechanical behaviour of hybrid all-carbon laminates.

The comparison of the CERR of the studied hybrids and their components allowed estimation of the “hybrid effect”, which is the difference between the CERR value of the hybrid interface and the average of the values for the reference composites. For mode I CERR, the hybrid effect was observed to be negative. The CERR of the hybrid corresponded to the lowest of the values for the two components (laminates with ultrahigh-modulus fibres). For mode II CERR, a strong positive hybrid effect was observed: the CERR value for the hybrid is close to the sum of the values for the components. The mechanism of the high hybrid effect in mode II is only partially explained by SEM fractography and merits detailed investigation in future work.

These findings reveal a previously almost unexplored research direction of hybrid effect in interfaces of fibre-hybrid composites and could contribute to explaining hybrid effects for other properties, such as impact resistance.

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Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time due to technical or time limitations; the data can be provided on demand.

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