ARE FLAX-EPOXY COMPOSITES TOUGH?

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Abstract
The purpose of this study was to determine the influence of fibre architectures on the interlaminar fracture toughness of flax fibre epoxy composites. The fracture toughness was investigated for both Mode I (G_{IC}) and Mode II (G_{IIC}) for seven flax-epoxy architectures: one plain weave, two twill 2x2 weaves, a quasi-unidirectional and a unidirectional composite, in both [0,90] and [90,0] lay-ups. The results of the mode I and Mode II showed promising results of the flax-epoxy composite performance. The addition of flax fibre increases the G_{IC} and G_{IIC} of the composites over that of the unreinforced brittle polymer by at least 2-3 times. Further improvements are made with the use of woven textiles.

1. Introduction

Delamination is one of the main concerns while designing composite structures. Therefore, it is essential to understand how a composite material resists to interlaminar fracture following an impact for example. The resistance against unstable interlaminar crack growth is called the interlaminar fracture toughness. This resistance can be determined for three loading modes: tension (mode I, G_{Ic}) in-plane shear (mode II, G_{IIC}) and out-of-plane shear (mode III). For composite laminates, the loading in Mode I is usually obtained with the double cantilever beam (DCB) test, the second mode with the end notched flexural (ENF) test and the third with a mixed-mode bending method for delamination testing (MMB) [1]. Interlaminar fracture toughness properties can be influenced by many factors such as: matrix type, matrix ductility, fibre type, fibre architecture, fibre orientations, fibre-matrix interface quality, laminate thickness and laminate stacking sequence.

A matrix is considered brittle when the G_{IC} value is lower than 200 J/m^2 and once combined to a fibre reinforcement, the composite interlaminar fracture toughness in Mode I will typically be double or triple [2]. Dhakal et al. [3] found that the weave architecture with the most cross-over points (higher picks and ends counts) leads to the highest resistance to impact damage and increased toughness properties in the case of plain weave jute/methacrylate soybean oil composites. Liu et al. [4] found similar results, where the type of woven flax-epoxy fabrics with varying stacking sequences and textile features resulted in an increase of the G_{IC} compared to the pure epoxy resin. To increase the toughness of the composite, other structural designing approaches could be applied such as z-pinning [5], stitching [6], 3D-weaving [7] which are known to improve the G_{IC} and G_{IIC}. The introduction of such features leads to a more tortuous in-plane crack propagation path, partly responsible for the increase of crack initation and propagations values.

In this study, the fracture toughness and tensile toughness are investigated in order to understand how the damage spreads in the impacted samples from previous in-house studies [8, 9], and how the damage area would relate to the damage tolerance capacity of each composite system. Mode I and Mode II interlaminar fracture toughness is studied for 7 flax fibre textile architectures (plain weave, twill 2x2, [0,90] and [90,0] cross-ply from quasi-UD and UD) combined to a brittle epoxy matrix.

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2. Experimental

2.1 Materials and production

In this study a total of 7 flax fibre architectures were investigated. Table 1 presents the flax preform features. It has to be noted that the quasi-UD fabric has 90% of the fibres in longitudinal direction while 10% are along the transverse direction and are used as binding yarns to keep the UD fibres together. All the textiles were used untreated. An *Epikote 828LVEL* epoxy combined with 15.2 phr of *Dytek DCH-99* hardener was used to produce the thermoset (Th) composites. The composites were manufactured using the Resin Transfer Moulding technique (RTM) as described in [10]. A spacer was used to ensure that a proper thickness would be reached. The targeted fibre volume fraction (V_f) was 40%. Before composite manufacturing, all the textiles were dried for at least 24 hours at 60°C to prevent moisture problems.

Table 1: Description of the used flax preforms and matrix.

Flax fibre preform architectures	Areal density ρ_{sur} (g/m^2)	Lay-up*
Plain weave	285	$[0]_4$
Twill 2x2	400	$[0]_4$
	150	$[0]_8$
Quasi-UD	300	Quasi-UD $[0,90]_s$
		Quasi-UD $[90,0]_s$
UD2	200	UD2$[0,90]_{2s}$
		UD2$[90,0]_{2s}$

* The 0° direction is along the fibre direction for unidirectional fabrics and along the warp direction for woven fabrics. More details on the fabrics are found in [8]

2.2 Interlaminar fracture toughness testing

Fracture toughness testing in mode I was done following the Double Beam Cantilever (DCB) test as defined in ASTM D5528 standard. The mode II fracture toughness was investigated using the End-Notched Flexure (ENF) test as described by Carlsson et al. [11] and the Japanese standard JIS K7086. Four to eight flax-epoxy samples were tested for both DCB and ENF. The samples were reinforced at the outside with 4 layers of quasi-UD glass fabric on each side to reinforce the sample and avoid a mixed-mode crack growth due to heavy bending of the beams.

The interlaminar pre-crack (a_0) was made by inserting a high temperature polyimide film with a 12 µm thickness and a length between 50 and 55 mm. The DCB tests were carried out on an Instron 4505 tensile machine with a crosshead speed of 1 mm/min, to produce stable crack growth, and a load cell of 1 kN. For the ENF, the tests were performed on an Instron 4467 with a 100mm span length and a crosshead speed of 1mm/min.

Mode I interlaminar fracture toughness calculation was based on the modified beam theory method. The modified equation takes into account the relation between the compliance of the specimen and the detected crack length using Eq. 1. In Mode I interlaminar fracture toughness determination, the initiation of fracture was defined using the three methods as decribed in [12]: the non-linearity of the load-displacement curves (NL), the 5% offset and the visual observation (VIS). The steady-state propagation value of G_{IC} is derived from the average propagation points when the G_{IC} value does not change with increasing crack length. For the G_{IIC} strain energy release rate, the classical elastic beam theory is used (Eq. 2),

$$G_{IC} = \frac{3P\delta}{2b(a-\Delta)}\frac{F}{N} \qquad \text{Eq.1}$$

$$G_{IIC} = \frac{9P\delta a^2}{2b(2L^3+3a^3)} \qquad \text{Eq.2}$$

where P is the load (N), a is the delamination length (mm), b is the specimen width, δ is the load point displacement (mm), L is the span length (mm), the Δ value is calculated from a linear regression analysis of $(C/N)^{1/3}$ – delamination length curve, *C* is the compliance (*δ/P*), *F* and *N* are the correction factors for the large displacement and finite displacement of the loading block respectively.

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3. Results and discussion - Interlaminar fracture toughness

3.1 Mode I Fracture toughness

The initiation fracture toughness value was determined using three methods: NL, 5% offset and VIS. It was found that visual determination of the initiation point (VIS) does not reliably represent the real value of the critical point for crack onset as it was closer to the propagation values in most cases. This means that the crack already started to grow before one can visually observe it and therefore, the initiation value was then considered to coincide with the non-linearity of the load-displacement curve (NL), as it has been found to be the lowest value of all three methods.

In this study, an epoxy matrix has been used with an average fracture toughness of 300 J/m^2, as tested by Aravand et al. [13]. An increase in fracture toughness over the unreinforced polymer was observed in all cases as seen in Figure 1. The interlaminar fracture toughness for initiation varies from 457 J/m^2 for the plain woven composite to 777 J/m^2 for the Quasi-UD [90,0]. The propagation value varies from 663 J/m^2 for the UD [90,0] to 1597 J/m^2 for the Low twist. Compared to glass and carbon fibre composites with non-toughened epoxy matrix (G_{Ic}= 150-500J/m^2) in literature [2, 14, 15], the mode I fracture toughness values of flax fibre composites are 3-5 times higher than for glass or carbon epoxy composites. It was also observed that the values are rather close to carbon-PES and carbon-toughened epoxy values [16-18].

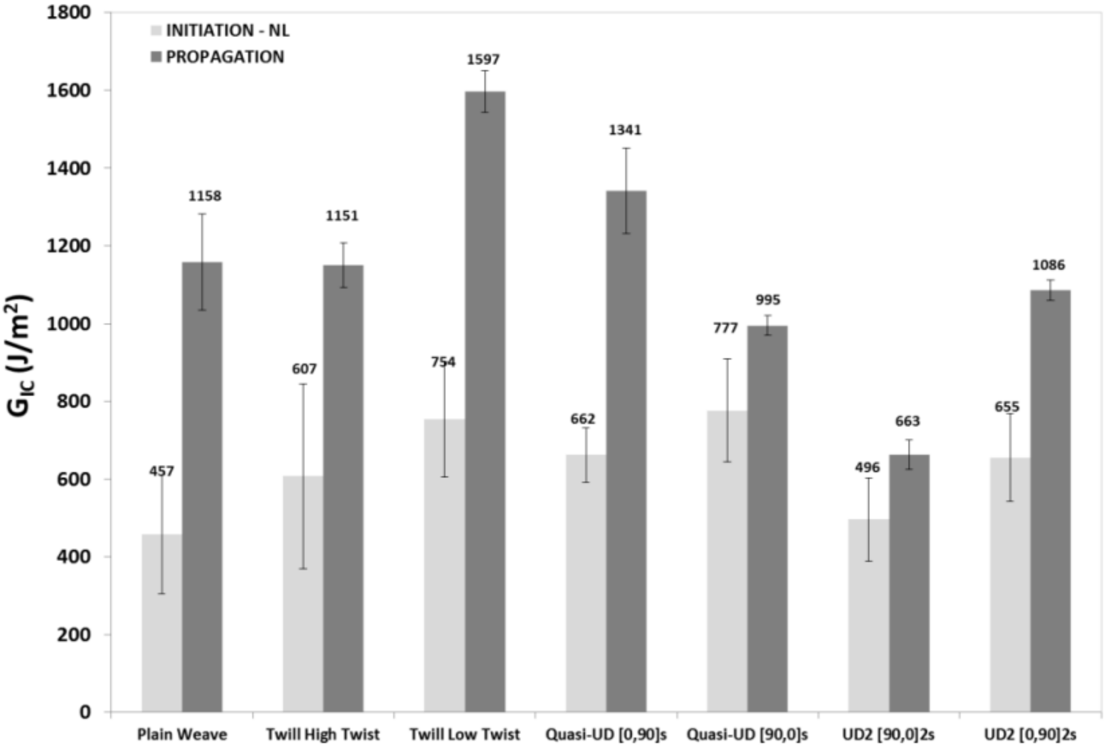

Figure 1: Double Cantilever Beam test - Fracture Toughness Mode I for flax-epoxy composites.

The woven fabrics and [0,90] laminates have an increased delamination resistance by a factor 1.5-2 compared to UD [90,0] reinforcements with propagation values varying from 1158 to 1597 J/m^2 compared to 663 J/m^2 as seen in Figure 1. This additional resistance may be due to the irregular path of the crack growth, caused by the yarn twist and crimp. In the case of cross-ply laminates, an increase in propagation interlaminar fracture toughness is observed, becausethe crack propagates into the 90^o layers instead of staying inbetween the 0^o layers for both UD and quasi-UD. Furthermore, crack jumping between two neighbouring 0°/90° interfaces of UD [0,90] was observed. This caused an increase in fracture toughness due to this deviation from the expected in plane crack path while for the UD [90,0] specimen, the crack propagated mid-plane, inbetween the 0° layers.

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Several mechanisms contribute to the composites' interlaminar fracture toughness. The dominant toughening mechanism, crack deflection, occurs when the crack deviates to follow the yarn or fibre undulations [19]. This creates a new and/or larger crack area resulting in higher fracture energy. The second mechanism is the pull-out of the fibres from the matrix, dissipating the energy by friction. The third mechanism is related to fibre bridging: the fibres debond from the surface without breaking, and link both surfaces together; additional energy is then consumed when the bridging fibres eventually fracture.

3.2 Mode II interlaminar fracture toughness

The main difference between mode I and II is that the first is a tensile mode where the two half beams (vertical opening of the crack surfaces) move apart from each other. The shear loading condition in mode II leads, for the ENF specimen configuration, to an unstable crack growth allowing only the measurement of the crack initiation value presented in Figure 2.

It was observed that the high twist twill and the quasi-UD laminates have similar mode II fracture toughness values, which can be related to the fineness of the yarns used in these preforms. The UD2 [90,0] laminate displays lower properties as the crack easily travels in between the 0 degrees layers. As for the UD2 [0,90] laminate, more resistance to crack growth was seen as the crack grows perpendicular to the 90°-fibres, and hence it can deviate around the fibres and even "jump" to the neighbouring 0° layers, in the same fashion as in mode I. The plain woven fabric seems to have the highest resistance to mode II crack growth, although it had an average performance for damage resistance [8, 9].

It can be said that the more complex the composite internal weave structure, the more the G_{IIc} increases as there is a higher resistance to crack growth in shear mode. The energy absorbing mechanisms observed in mode II have similarities to the ones of the mode I such as matrix cracking, fibre breakage, the effect of internal structure of the composite (textile geometry) and crack path deflection previously discussed.

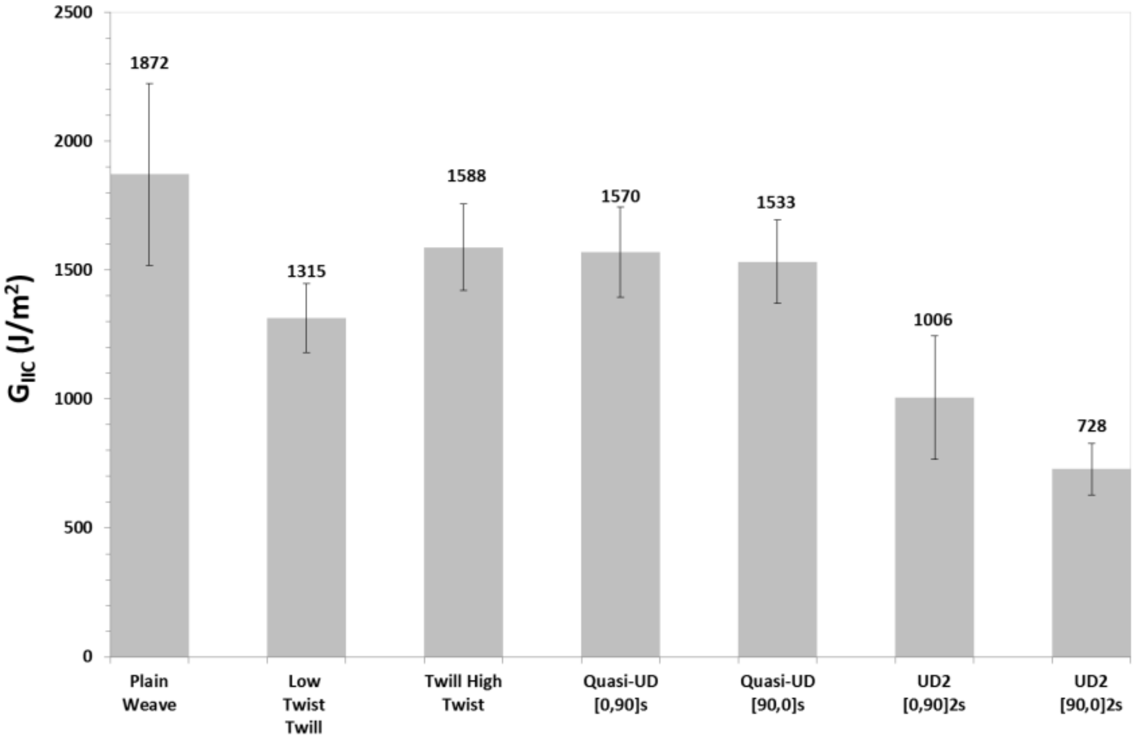

Figure 2: End Notched test – Fracture toughness Mode II.

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4. Conclusions

The objective of this work was to determine the influence of fibre architectures and choice of matrix on the impact behaviour and the interlaminar fracture toughness of flax fibre composites. As in most composites, it could be expected that the damage area after impact is directly related to its capacity to resist delamination growth, hence it was considered essential to evaluate the interlaminar fracture toughness. However, in a later publication, it will be shown that the correlation between interlaminar fracture toughness and impact resistance is not as straightforward in flax fibre composites as it is in glass and carbon fibre composites, because other (in-plane fibre fracture dominated) damage mechanisms seem to play a bigger role.

It has been found that the addition of flax fibre increases both the G_{IC} and G_{IIc} of the materials over that of the unreinforced polymers and those of glass and carbon fibre composites. Additional improvements are made with the use of woven textiles compared to the [0, 90] cross ply lay-ups. For mode II, the highest resistance to interlaminar shear (Mode II) is recorded for the plain weave fabrics. This might be due to the shape of the yarns (high twist) used for the preform weaving, as the crack has to travel around the highly twisted yarns making it harder for the crack to initiate leading to an increase in shear delamination properties.

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