Mode I delamination behavior of flax-epoxy laminates made with UD and UD-mat reinforcements

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1. Abstract

Natural fibers are attracting increasing interest due to their low cost and low environmental impact, while offering intrinsic mechanical properties comparable to glass/epoxy composites. However, laminates remain susceptible to delamination, a critical failure mode that can significantly reduce the strength of the composite. While synthetic materials are well-documented, the delamination behavior of unidirectional flax-epoxy remains poorly understood. This study aims to evaluate the mode I delamination properties of unidirectional (UD) and UD-mat flax fiber composite laminates, made of unconditioned and preconditioned reinforcements, to assess their potential as durable structural materials. UD glass laminates are also evaluated for comparison. The laminates were manufactured using the RTM process to get 8 layers laminates at a fiber volume fraction around 40%. First, laminates made with different pre-crack lengths, made with a 20 µm thick wax paper, were tested. This was done to develop a repeatable testing method while reducing the initial nonlinearities to conform to the ASTM D5528 requirements. Second, the mode I delamination behavior of laminates made of four different types of flax reinforcement has been evaluated, namely a UD, UD-mat, UD-mat made of fibrillated short fibers and a pre-compacted UD-mat made of fibrillated short fibers. The results show that reducing the initial pre-crack length from 40 to 25 mm is required to meet the linear behavior requirement of the ASTM standard, considering that a highly non-linear behavior with large hysteresis loops was observed at a_0 = 40 mm. For the UD flax laminate, a dispersion of less than 5% and G_{IC} values perfectly aligned with those reported in the literature were obtained. The results also show that adding a mat binder to the UD reinforcement (comparing UD and UD-mat laminates) has virtually no effect on the G_{IC} value. However, the laminate with UD-mat reinforcement made of fibrillated fibers shows an increase of 11.4 % and 21.2 % in G_{IC} compared to the UD and UD-glass laminates, while pre-compaction of UD-mat reinforcements (no fibrillation) reduces G_{IC} by 38,8 % and 33,4 %. Surface fibrillation of short flax fibers enhances interaction with UD fibers in adjacent plies, contributing to improve G_{Ic}. Conversely, the compaction of UD-mat reinforcement affects the distribution of fibers through the

thickness of the laminate and promotes the creation of a resin-rich mat binder between the UD phases within the laminate, making it more susceptible to delamination.

2. Introduction

Natural fibers, particularly flax, are attracting increasing interest due to their low cost and minimal environmental impact, while offering intrinsic mechanical properties comparable to glass/epoxy composites. Flax-based composites are therefore increasingly considered for lightweight structural applications. However, laminates remain susceptible to delamination, a critical failure mode that can reduce a composite's strength by up to 60% [1, 2]. Mode I, the most severe in terms of crack opening, allows for the characterization of interlaminar toughness via the G_{IC} parameter. While synthetic materials are well-documented, the delamination behavior of natural fiber composites remains poorly understood. Unlike conventional glass/epoxy composites, natural fiber composites exhibit more complex damage mechanisms associated with the intrinsic variability of plant fibers as well as fiber/matrix interactions, particularly the moisture state prior to manufacturing, which affects resin impregnation and the resulting interfacial properties of flax/epoxy laminates [3, 4]. In addition, recent studies have shown that the interlaminar fracture behavior of flax/epoxy composites depends significantly on the reinforcement architecture, leading to distinct crack initiation and propagation mechanisms compared to conventional glass-based laminates [5, 6]. Flax/epoxy composites exhibit relatively stable crack propagation behavior together with more progressive failure mechanisms compared to conventional glass/epoxy laminates [1, 6]. The condition of flax fibers prior to manufacturing directly affects resin impregnation and fiber/matrix interfacial properties, which can consequently influence the mechanical performance and delamination behavior of the resulting composites [3, 4]. More generally, several studies have highlighted that the energy dissipation mechanisms in natural fiber composites remain strongly dependent on the internal organization of reinforcement and on the interactions developed within the laminate [1, 5 ,6].

This study aims to measure and analyze the mode I delamination properties of unidirectional (UD) and unidirectional-mat (UD-mat) flax-epoxy composite laminates to assess their potential as a durable structural material. More specifically, it aims to evaluate the influence of the flax mat binder on G_{IC} properties for a laminate made with a UD-mat reinforcement manufactured using a papermaking process. At the same time, modifications to the reinforcement, such as surface fibrillation of short fibers and dry compaction before molding known to improve the laminate tensile properties [7, 8], are also evaluated for delamination. The results are also compared with those obtained with a UD glass-epoxy laminate.

3. Experimental

3.1. Materials

Flax fibers from Safilin Inc. (Szczytno, Poland) and SikaBiresin® epoxy 820 resin are used as base materials. Two formats of fibers have been used: Tex 400 yarns for the unidirectional (UD) layer and short fibers cut from a Tex 5000 ribbon for the mat layer. The resin was chosen for its low viscosity to ease wetting in the resin transfer molding (RTM) process. This system includes a base epoxy resin (SikaBiresin® CR72) and a hardener (SikaBiresin® CH72-3), mixed at an 18%

mass ratio. For the UD-mat reinforcement, the fabrication process uses a dynamic former from ALIMAND Inc. The protocol used for manufacturing the reinforcement is described in detail in previous works [7, 8] and will therefore not be included here. For comparison with the flax reinforcements, a stitched unidirectional glass reinforcement from Texonic Inc. (Saint-Jean-sur-Richelieu, Canada), the TG09U50 of 301 g/m^2, was also used to mold unidirectional glass-epoxy laminates.

3.2. *Molding and specimen preparation*

Flat panels were molded from a stack of 8 reinforcement layers. The unidirectional (UD) and unidirectional with a mat binder (UD-mat) reinforcements have a surface density of 245 g/m² and 300 g/m², respectively. The UD reinforcement is made of the same Tex 400 flax yarns placed side by side. All panels have been molded with the UD fibers of each ply oriented at 0° to obtain a $[0]_8$ stacking sequence. Before molding, the reinforcements were dried at 60°C for 2 hours. Four aluminum shims located between the mold halves were used to control the mold cavity thickness, which was adjusted to obtain a target volume fraction of 40%. The values obtained after manufacturing was 39.85 ± 0.14% for flax-epoxy and 40 ± 0.14% for glass-epoxy plates. Impregnation was carried out at room temperature, with a gradual, controlled pressure increase from 0.5 to 4 bar. After injection, the panels were oven-cured for 4 hours at 80°C. For the mode I tests, a pre-crack was integrated at mid-thickness of the plate using waxed paper approximately 20 µm thick, conforming to ASTM D5528.

3.3. Experimental testing

Figure 1a shows the schematic of a double cantilever beam (DCB) specimen and a photograph (Figure 1b) of a typical sample. The molded plates were waterjet-cut to achieve clean edges and dimensions conforming to ASTM D5528. The samples have a total length L = 125 mm, a width b = 25 mm, and a thickness h adjusted according to the reinforcement used. Three initial effective lengths of the pre-crack (measured from loading axis) of 35, 30 and 25 mm were tested for flax laminates. For the glass/epoxy laminate and given its higher stiffness, a_o was set at 35 mm. After cutting, the edges were cleaned, and metal hinges were bonded to allow for the application of mode I opening load. Figure 2 shows the experimental setup. The tests were performed on a tensile testing machine (from Test Resources Inc.) equipped with a 500 N load cell. Hinges ensure symmetrical loading, while a video camera measures the crack propagation and specimen opening during the test. The force P and the opening (displacement δ) are continuously recorded for G_{IC} calculation, while the correlation between the applied load P, opening (δ) and the crack length (a) is made using the elapsed time recorded by the camera during testing. The mode I energy release rate (G_{IC}) is calculated using equation (1) from the P–δ curves and the crack length a, following the Modified Beam Theory (MBT).

$$G_{IC} = \frac{3P\delta}{2b(a+\Delta)} \quad (1)$$

In this equation, *b* the specimen width and *Δ* a correction factor for the crack length obtained by calibrating the specimen's compliance. The G_{IC} values at initiation and propagation are then calculated.

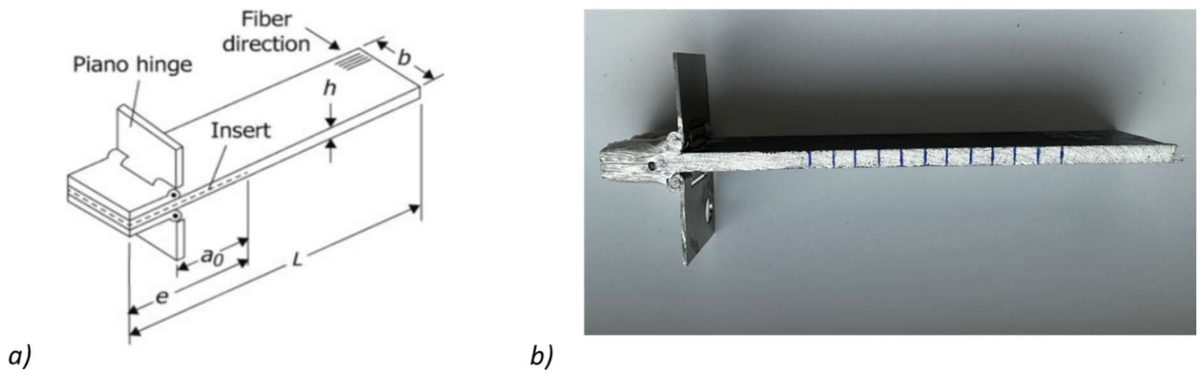

Figure 1. a) Schematic of the specimens following ASTM D5528 and b) photograph of a typical test specimen.

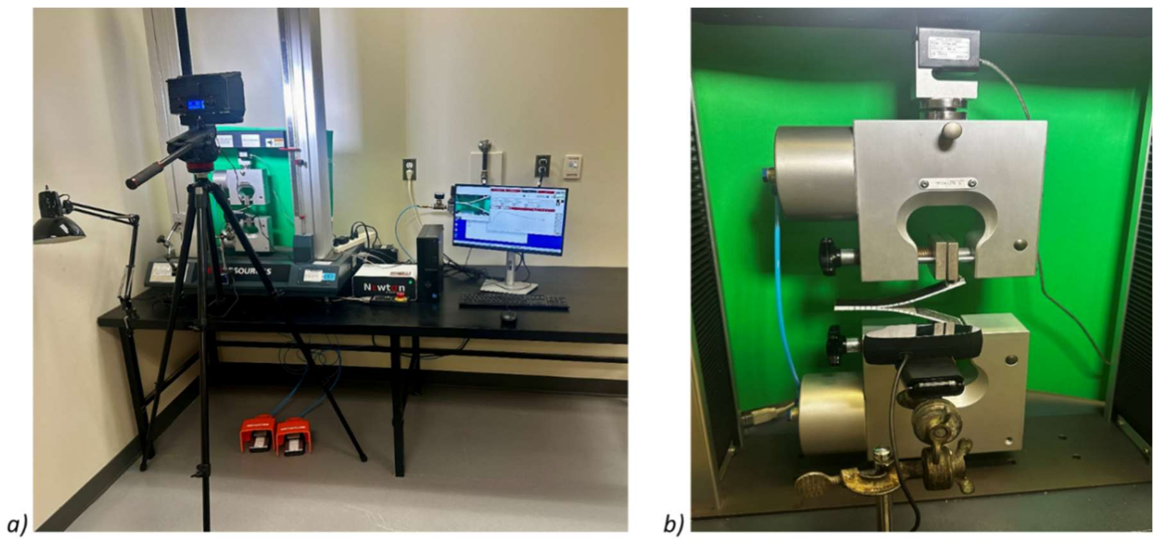

Figure 2. a) Testing setup and b) photograph of a specimen during testing.

3.4. *Test performed*

Table 1 shows the test performed. Except for UD, all other tests have been performed on coupons made of UD-mat (UDM) flax reinforcements. The C letter refers to reinforcements that have been individually pre-compacted at 3.6 bars for 20 minutes under a 50 T press (at ambient temperature) prior molding, while the F letter refers to the UD-mat reinforcement made of fibrillated short flax fibers for the mat binder. Fibrillation was made using a laboratory refining Paper and Fibre Research Institute (PFI) mill. Again, experimental details of the refining protocol are described in previous works [7].

Table 1. Test performed for mode I tests.

Test ID	Reinforcement type	Conditioning
UD	Unidirectional flax	---
UDM	UD-mat flax	---
UDMC	UD-mat flax	Compaction
UDMF	UD-mat flax	Fibrillation
UDMFC	UD-mat flax	Fibrillation and compaction
UDG	UD glass	---

4. Results

4.1. Typical P-δ curves

Figure 3 shows typical load-unload curves obtained during cycling of a UD flax laminate for a pre-crack a_0 of 40 mm. The cycling exhibits a nonlinear behavior, a strong energy dissipation due to hysteresis, and a significant residual displacement at the end of the test. These effects indicate a substantial internal rearrangement of the material, which completely disrupts the energy dissipation measurement related to the propagation of delamination. It also precludes the use of the compliance method to measure G_{IC} in the propagation phase of the delamination. To avoid these effects and obtain a linear behavior until delamination initiation (during initial loading), different pre-crack lengths were tested (Figure 4). A shorter a_0 increases stiffness, reduces nonlinearity, and improves measurement accuracy to meet the requirements of the ASTM test standard. A length of a_0 = 25 mm was ultimately selected for all flax-epoxy tests. Typical P–δ curves for UDM, UDMF, UDMC and UDG are shown in Figure 5. For UDM, the NL, 5%, and MAX points for the calculation of G_{IC} are also shown. As specified in ASTM D5528, these points correspond to the linearity deviation (NL), to a load corresponding to a 5% reduction from the initial stiffness (5% offset) and to the maximum force (MAX) measured during the test. For the glass laminate (UDG), a discontinuous propagation of delamination is observed in the curve as sudden load jumps. This behavior corresponds to the sudden crack propagation in resin-rich zones of the laminate, which are created by the presence of polyester thread binder in the UD reinforcement. The presence of a mat binder in flax laminates avoids this behavior, which is advantageous for ensuring a more gradual propagation of delamination as shown in Figure 5.

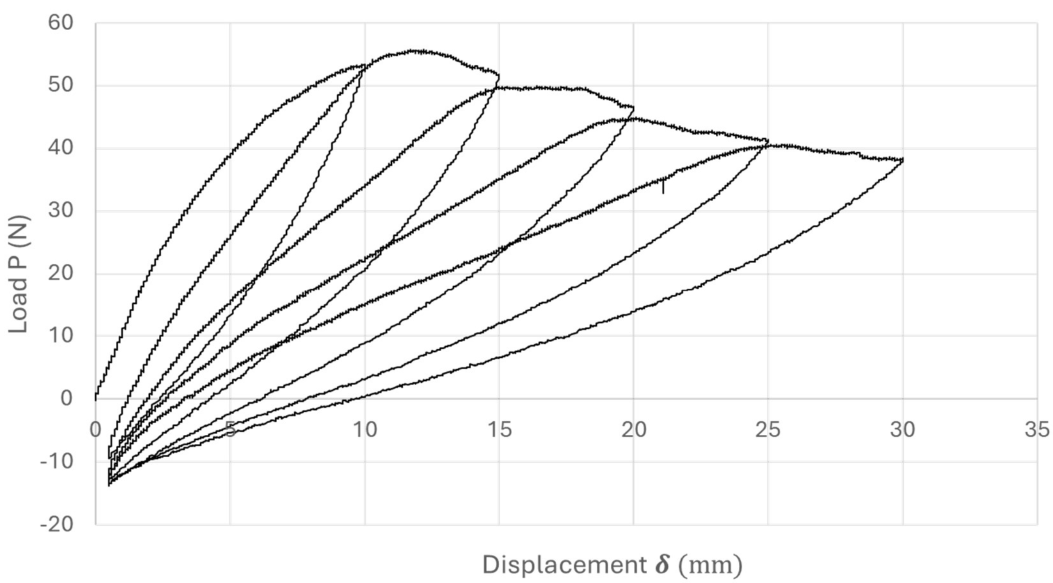

Figure 3. Typical P-δ curves for the UD laminate showing large hysteresis loops and residual displacements after testing.

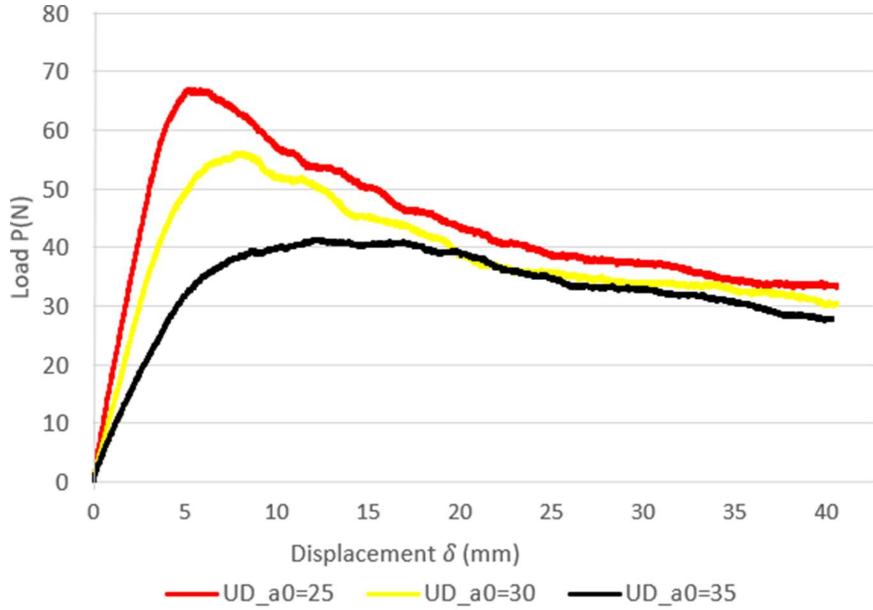

Figure 4. Effect of the pre-crack length on the P-δ curves for the UD laminate.

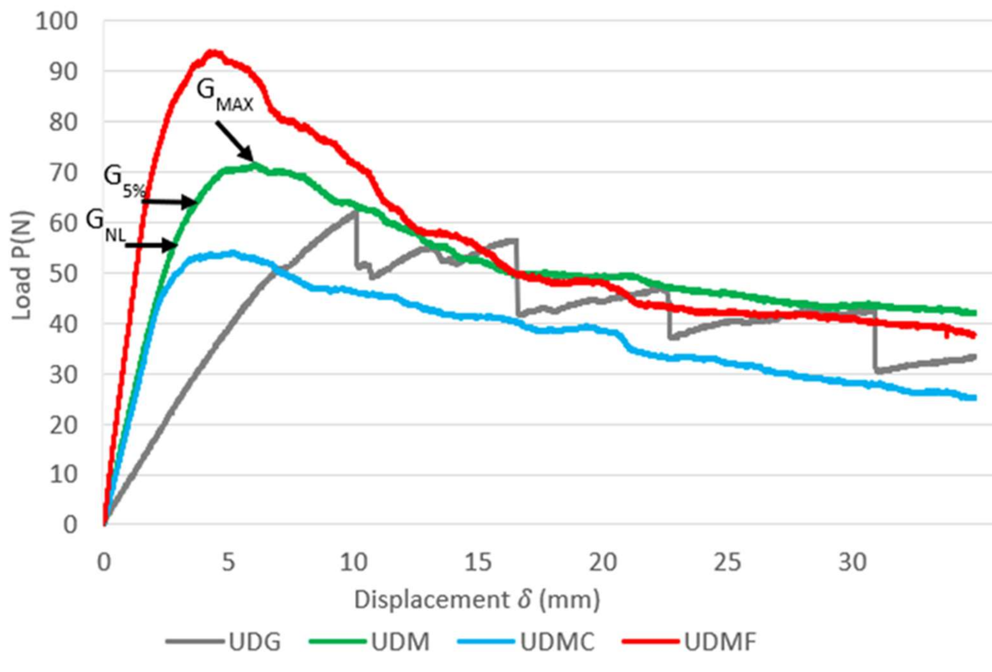

Figure 5. P-δ curves for the 5 tests of UD flax laminates showing the NL, 5% and MAX locations for G_{Ic} calculations.

4.2. G_{IC} results and analysis

The histogram in Figure 6 shows the G_{IC} results obtained for all tests in Table 1. G_{IC} values calculated at NL (GNL), 5% (G5%), and MAX points (GMAX) are shown. First, for the UD-flax tests, values between 575 and 867 J/m² are obtained with a dispersion of less than 5%. These results are very close to those obtained by Saadati et al. [1] who obtained values ranging from 574 to 903 J/m² for UD-flax laminates at $V_f = 40\%$. Second and for all groups of results (NL, 5% and MAX),

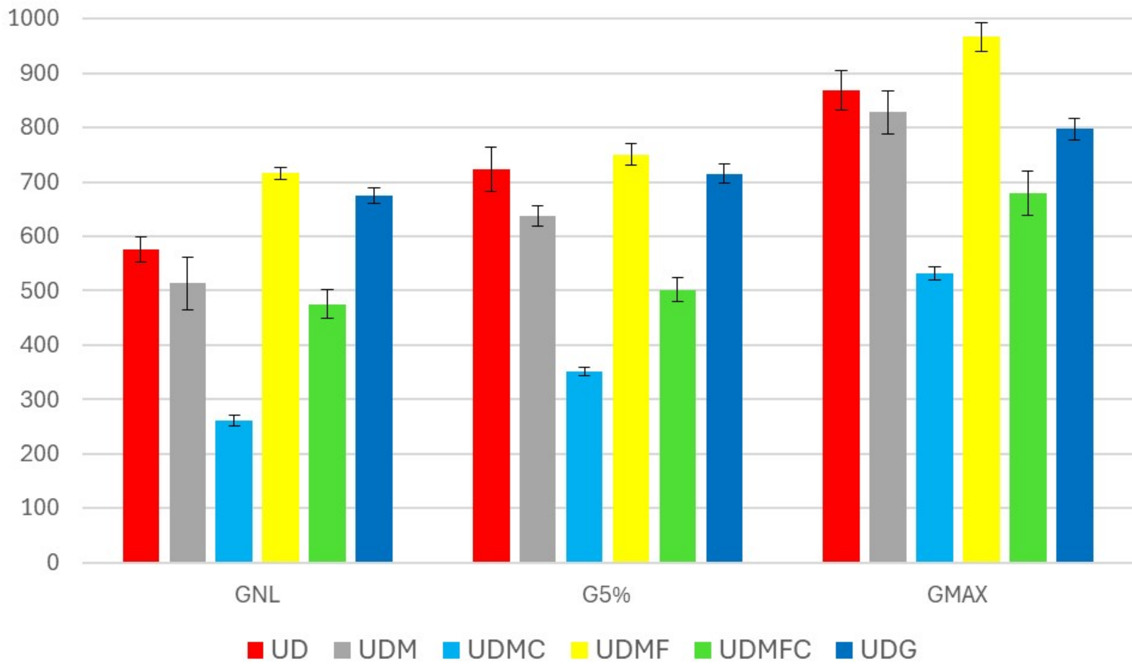

Figure 6. G_{IC} results at NL, 5% and maximum loads for all tests.

the best results are obtained for the UD-mat made of fibrillated short fibers (UDMF). Overall, surface fibrillation increases G_{IC} while compaction of reinforcements (before molding) has the opposite effect. This is clearly observed by comparing the UDM, UDMC, UDMF and UDMFC results (grey, light blue, yellow and green bars respectively). An increase of G_{IC} is obtained when passing from UDM to UDMF, followed by an important decrease for UDMFC with final values even lower than those of UDM. The same compaction effect is observed when comparing UDM and UDMC. Surface fibrillation of short flax fibers enhances the interaction between plies through the detached nanofibers of the mat binder penetrating the adjacent UD phases, thereby increasing the fiber bridging during crack propagation. This is observed in Figure 7 showing the fiber bridging of typical UDM, UDMF and UDMFC specimen. Fiber bridging was more important in UDMF compared to UDMFC specimen. Conversely, compacting reinforcements before molding, although it has proven beneficial for the tensile properties of similar laminates [8], produces more compact UD phases in the laminate and consequently some separation of plies in the mat phase, resulting in the creation of a resin-rich mat binder between the UD phases, a situation more prone to matrix fracture and delamination. This is supported by the UDMC result (reinforcement without surface fibrillation but compacted) showing the lowest G_{IC} results of all tests. Compared to UD glass-epoxy (UDG) laminate, G_{IC} of the UD and UDM are very close to that of UDG, especially for Gmax for which the results are statistically the same at around 800 J/m^2. This means the introduction of a mat binder in the UD flax reinforcement do not ameliorate nor deteriorate the mode I delamination toughness, but to ameliorate it, surface fibrillation of short flax fibers seems efficient, at least for the fiber content evaluated in this work. The influence of fibrillation and reinforcement compaction becomes evident when comparing the micrographs in Figure 8 showing mesoscopic observations of fracture surfaces taken near the tip of the initial defect for flax

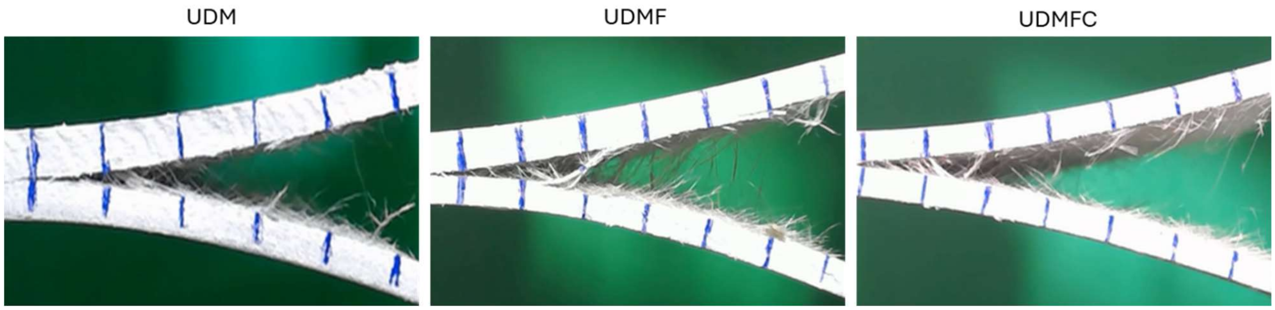

Figure 7. Side views of DCB test during testing of typical UDM, UDMF and UDMFC specimens.

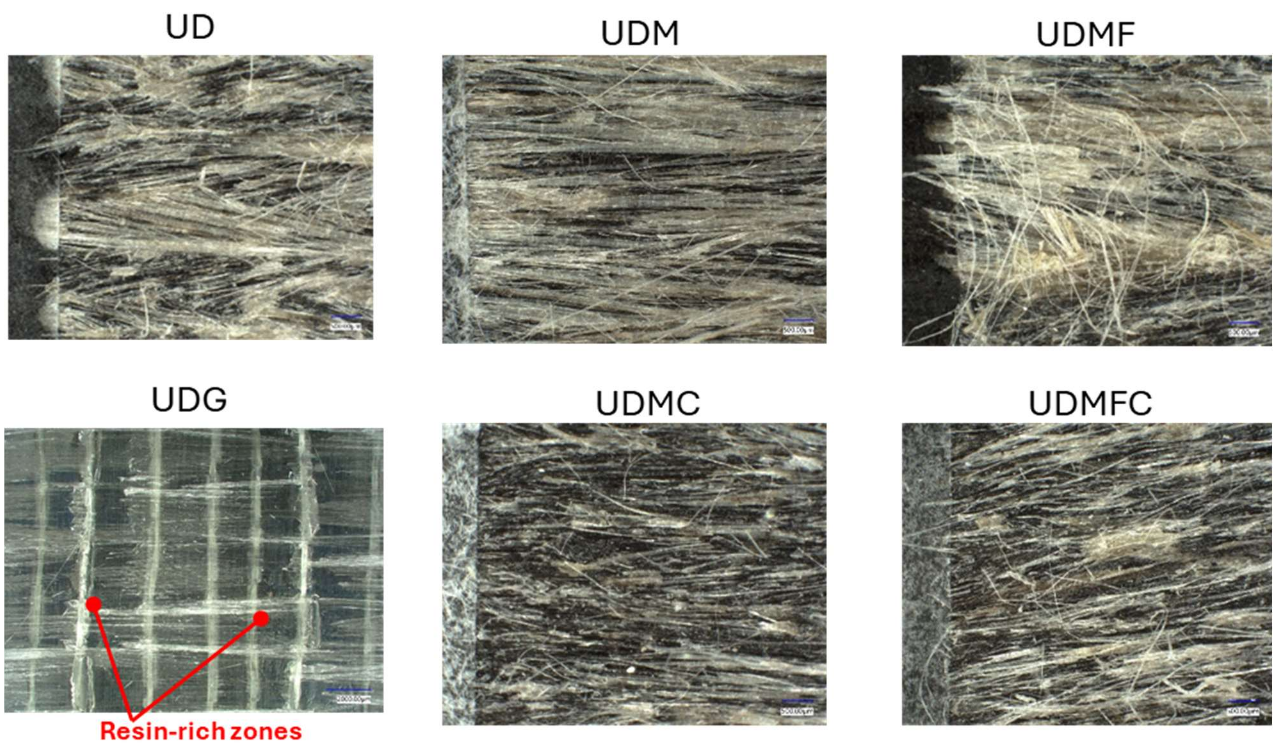

Figure 8. Fracture surface of UD, UDM, UDMC, UDMF and UDMFC specimens taken near the tip of initial pre-crack where the values in Figure 6 were calculated.

laminates (placed to the left of each image) where the G_{IC} values were calculated. For the UD laminate, the absence of the mat binder leads to a direct contribution from the unidirectional fibers located on either side of the crack, with fibers debonded from the matrix (light beige color in the images). The addition of mat in the UDM laminate induces a combined contribution from the latter and the UD fibers, with the global effect of maintaining the G_{IC} values to a relatively constant level compared to UD (Figure 6). The introduction of surface fibrillation of short flax fibers (UDMF case) leads to a significant increase in fiber-matrix and mat-UD fiber interactions, due to the presence of microfibrils, which considerably increases the G_{IC} values. As soon as compaction is introduced (UDMC and UDMFC cases), the opposite effect takes place (a decrease in G_{IC}), and this is more pronounced in the UDMC case than in the UDMFC case, as the fibrillation of the latter helps mitigate the negative effect of compaction. The darker appearance of the fracture surfaces of the UDMC and UMMFC samples clearly reflects this effect with less fiber debonding, particularly for the UDMC sample. Finally, the resin-rich areas in glass-epoxy laminates, which caused the force jumps in Figure 5, are clearly visible as dark areas in Figure 8 adjacent to light grey areas showing fiber debonding. Table 2 shows the relative variation of the maximum G_{IC} (GMAX) of all laminates compared to the references UD-flax (UD) and UD-glass (UDG) laminates. UDMF is improved by 11,4 % and 21.2 % compared to UD and UDG respectively while the worst case is clearly UDMC with reductions of 38,8 % and 33,4 %.

Table 2. Variation of G_{IC} (GMAX) of each laminate with respect to UD flax and UDG laminates.

Test ID	G_{Ic} variation vs UD (%)	G_{Ic} variation vs UDG (%)
UD	0.0	+8.8
UDM	-4.5	+3.9
UDMC	-38.8	-33.4
UDMF	+11.4	+21.2
UDMFC	-21.7	-14.8

5. Conclusion

Mode I tests have been realized on unidirectional (UD), UD-mat flax (with and without fiber fibrillation and reinforcement pre-compaction) and UD glass epoxy laminates all at V_f around 40%. After adjusting the initial crack length to 25 mm in flax laminates, the tests showed good stability and repeatability in the mode I test results. The G_{IC} obtained, with a dispersion of less than 5%, demonstrates reproducible toughness perfectly aligned with values reported in the literature for similar tests [1]. These results confirm the reliability of the experimental protocol, despite the use of a natural fiber composite that is inherently highly nonlinear and susceptible to energy dissipation through hysteresis. The presence of a mat binder in the flax laminate contributes to stabilize the crack propagation while maintaining a G_{IC} value similar to that of UD flax (UD) and UD glass (UDG) laminates. It has been shown that surface fibrillation of short flax fibers in the mat binder improves resistance to delamination, while compaction of the reinforcement produces the opposite effect. Fibrillation promotes the interaction of UD fibers in adjacent plies while compaction of UD fibers reduces the through-thickness homogeneity of fiber distribution, with the creation of a resin-rich mat binder between the UD phases, thus reducing the delamination strength. The maximum G_{IC} increases by 21,2 % with surface fibrillation for UDMF while it decreases by 33,4 % for UDMC compared to the UDG laminate.

6. References

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