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MECHANICAL AND IMPACT CHARACTERIZATION OF HYBRID COMPOSITE LAMINATES WITH CARBON, BASALT AND FLAX FIBRES

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ABSTRACT

Ternary hybrids including carbon, basalt and flax fibres in an epoxy matrix have been fabricated by hand lay-up, then consolidated by vacuum bagging using two different stacking sequences. Both configurations involved the use of carbon fibres on the outside, whilst basalt and flax fibres were disposed internally either in a sandwich or in an intercalated sequence. They were subjected to tensile, flexural and interlaminar shear strength test, then to falling weight impact with three different energies, 12.8, 25.6 and 38.4 Joules, studying damage morphology and impact hysteresis cycles. Intercalation of basalt with flax layers proved beneficial for flexural and interlaminar strength. As regards impact performance, the differences between the two laminates were quite limited: however, the presence of a compact core of flax fibre laminate or else its intercalation with basalt fibre layers had a predominant effect on impact damage features, with intercalation increasing their complexity.

KEYWORDS

B. Impact behaviour; B. Mechanical properties; A. Hybrid; Flax

INTRODUCTION

During last decades, hybridisation, hence the introduction of layers of different fibre materials in composites has been often attempted. The first attempts involved glass/carbon hybrid composites, where it was found that performance is likely to exceed what would be expected from consideration of the rule of mixtures, which can be defined as a positive hybridisation effect [1]. In the specific case of traditional thermosetting composites, such as unsaturated polyester and epoxy, where the environmental advantage of hybridisation is not always obvious, this procedure may be perceived in different ways. A possibility is to consider hybridisation as an intermediate step for the substitution of traditional fibres, such as E-glass ones, with vegetable fibres, such as flax, hemp or jute, in composites, possibly with limited degradation in properties: this was mainly the approach of early studies [2-4]. Dealing with carbon fibre composites, the above mentioned approach comes somehow short, since the difference in properties with plant fibres is very considerable. On the other side, it has been suggested that the introduction of fibres with distinctly different properties would lead to a composite with properties more tailored on the requirements from service. This would compensate for some degree of complication involved in having different types of fibres. More specifically, carbon fibre composites, though outstanding in terms e.g., of tensile properties, suffer from limited toughness, which can be improved by their hybridisation with plant fibre composites [5], a characteristic found of interest for biomedical applications [6].
More recently, basalt fibres have been often considered as a suitable replacement for glass fibres, since they offer a lower environmental impact, in that their production does not involve the use of sizing agents, and they can effectively compete with glass in sectors, such as automotive, for example for the improved resistance to acid environment [7]. As a matter of fact, also hybrid laminates including carbon and basalt fibres have been produced, in which case the effect of the

stacking sequence proved determinant in the achievement of a higher performance, although positive hybridisation effect was always obtained for all laminate configurations produced [8]. On the other side, the introduction of vegetable (lignocellulosic) fibres e.g., flax, in the laminate would reduce its weight and being not excessively detrimental in terms of properties, such as flexural, which can benefit from the fact that lignocellulosic fibres have a hollow structure for the presence of an internal lumen [9].
All these considerations need nonetheless to be inserted in more global observations and experiences of the suitability of these hybrid composites to service. It is therefore important in this regard to carry out testing that are more representative of the real, even if accidental, events occurring during composites use in structures, such as automotive components. This is the case with falling weight impact testing, which has normally been performed on these materials to obtain information on absorbed energy, on the morphology of damage and on its evolution. Studies on the respective relevance of the elastic, plastic and damping energy during the impact event have proved suggestive of the mode of energy absorption the laminate can perform [10-11]. However, the higher complexity of hybrid laminates with more than two different fibres make it quite cumbersome the assessment of a positive hybridisation effect especially, because the relation between the fibres in the laminates, expressed by the stacking sequence, may equally play a role in it [12].

In this work, composite laminates based on epoxy resin and reinforced with three types of fibres, carbon, basalt and flax, were produced by hand lay-up then consolidated by vacuum bagging, with two different stacking sequences. More specifically, carbon fibres, as the stiffest amongst the three, has always been used in the external layers, while as for internal layers basalt and flax fibres have been changed their mutual positions in the laminate. The performance of the laminates with the two different stacking sequences has been compared by carrying out static mechanical tests, namely tensile, flexural and interlaminar shear strength and falling weight impact tests, particularly concentrating on the characteristics of energy absorption during the impact event.

EXPERIMENTAL

MATERIAL CONFIGURATIONS

Composites were obtained by using an epoxy resin CR83 by Syka with amine hardener CH83-2, a system with glass transition temperature exceeding 80°C. Three different types of reinforcement were used in the composites production, in particular basalt fibres fabric, atlas weave (areal weight 350 g/m²) by Basaltex, unidirectional non crimp flax fabric (areal weight 380 g/m²) with textured polyester as stitching thread, 1/cm, commercial name Fidflax, by Fidia, and plain woven carbon fabric (areal weight 200 g/m²) by Toray. Laminates were produced by hand lay-up, ensuring that flax fibre fabric was well dried before use by putting them in oven at 60°C for a time of around 15 minutes. Vacuum bagging at 0.88 bar at a controlled temperature of 28°C was applied, followed by a post-cure at 80°C for 14 hours, in a frame with dimensions 600x400 mm.
The amount of fibres introduced was in the order of 53 wt.% total, of which 27 wt.% flax (F), 12 wt.% carbon (C) and 14 wt.% basalt (B). The layers reinforced with the three different fibres were stacked together in two different configurations, so to form two hybrid laminates, referred to as laminate N.1 and laminate N.2, respectively. The relevant stacking sequences are reported in Figure 1.
All the obtained laminates had thickness equal to 4 ± 0.2 mm.

MECHANICAL AND IMPACT TESTING

Details on the tests carried out are reported in Table 1. Per each of the laminates and of the static tests (tensile, three-point flexural and ILSS) ten samples were tested. In the case of impact tests by falling weight (IFW), the samples were impacted from a height of 3 meters to measure the maximum energy absorbed. Once performed this measurement, other samples were impacted from two lower heights, 1 and 2 meters respectively, to evaluate damage produced by impact at energies not sufficient to result in laminate penetration. For each impact height and laminate, three samples were impacted. Since, as reported in Table 1, an impacting mass of 1.3 kg is used, the nominal energies of impact applied are equal to 12.8, 25.6 and 38.4 Joules, the last one excessive with respect to the impact resistance of the laminates, hence leading to penetration.

RESULTS

The starting point from these considerations over the results obtained would be the assessment of whether a positive hybridisation effect is obtained or not, and of possible differences between the two stacking sequences. As from tensile properties, reported in Table 2, it can be noticed that the performance of the two laminates is very similar. On the other side, tensile properties can be used for the aforementioned evaluation of the hybridisation effect.

In very general terms, one can start from the consideration of an epoxy composite with approximately 50 wt.% fibres, half of which are flax fibres, whereas the remaining half is equally divided between carbon and basalt fibres. As for fibre orientation, studies on quasi-isotropic or randomly oriented fibres were selected. From literature some data over tensile properties of the originating composites, hence containing only carbon, basalt or epoxy fibres, concentrating on those which have around 50 wt.% reinforcement, are reported in [13-15]. In particular, tensile strength values obtained for carbon/epoxy are between 483 and 609 MPa in [13], for basalt/epoxy are between 140 and 160 MPa in [14] and for flax/epoxy are between 125 and 155 MPa in [15]. From these considerations, it would be expected that a minimal target for the hybrid composite is to exceed at least the maximum tensile properties of both flax and basalt fibres, which would offer some justification to the addition of carbon fibre layers to the laminate.

In practice, this is achieved: however, tensile strength values are only around 15-20% above what could be expected from a pure basalt/flax fibre hybrid with no added carbon fibres. This can be ascribed to problems in consolidation of the composite and possibly to the non negligible presence of voids. As for comparison between the two laminates, from tensile properties, reported in Table 2, it can be suggested that for a strength that is basically very similar, intercalation of basalt fibre layers with flax fibre ones results in improved rigidity. On the other side, it may be reasonable to suppose that the action of both carbon and basalt during flexure is showing tensile mode of fracture (evidence is given on basalt composites in [16] and more recently on carbon/flax/carbon hybrid fibre composites in [17]), while it is likely that flax fibre composite is more prone to the onset of compression damage, due to its lower rigidity. As regards flexural properties and interlaminar strength, laminate n.2 shows a substantially higher flexural strength, with basically the same ultimate strain and higher ILSS, as depicted in Table 3. This rigidity appears particularly beneficial in terms of flexural properties, where intercalation substantially hinders the bending of flax fibres, which is likely to be enhanced by the presence of a core of flax layers, tending to interact complexly during flexure. In other words, as it was previously noticed, intercalated layers enable more gradual damage propagation during loading, whereas in the case of the presence of a compact plant fibre composite in the core of the laminate, its collapse dominate the whole degradation process [18]. This has been recently been observed on basalt/flax/basalt hybrid laminates, in the presence of two basalt layers on each side of the laminate, suggesting that the main a benefit of introducing basalt fibre layers would be experimented as regards flexural properties [19]. A similar positive effect is encountered in the case of ILSS results, as can be observed from the curves reported in Figure 2, where the introduction of intercalated layers appears to delay the collapse of flax core.

As regards falling weight impact though, the higher flexural rigidity obtained with intercalation leads to no particular benefits in terms of penetration energy: in particular, impact penetration energy was equal to 29.45 ± 0.1 J for laminate n.1 and to 29.25 ± 0.33 J for laminate n.2, respectively. On the other side, intercalation slightly increases the variability of the performance, as might be expected every time the complexity in a composite becomes higher. As regards flax fibre composites, it has been previously noticed that toughness is dominated by the fibre volume effect rather than by reinforcement architecture: this would suggest that the different sequence of flax layers might possibly not have a strong influence on crack propagation in the laminate [20].

Further considerations concern the analysis of hysteresis curves obtained during impact by falling weight tests. As a first indication, which is essential to start the analysis, the area included in the closed loop of the curve, corresponds to energy absorbed by the laminate [21]. Examples of force vs. deflection impact hysteresis cycles for loading at the three impact energies considered, namely 12.8 J, 25.6 J and 38.4 J, are represented in Figure 4. Considering that the thickness of the laminates is around 4 mm, it may be expected that impact at the highest energy results in severe forming and penetration, since the final deflection, when load comes back to zero, is around the order of the laminate thickness. The analysis of impact hysteresis cycles, whose interest to evaluate the modes of damage propagation in composites has been proposed in a number of instances [22-23], allowed obtaining more information on the two different configurations. In particular, a number of parameters were calculated, which are reported in Table 4a and 4b for laminate n.1 and n.2, respectively. The linear stiffness has been defined as the average slope of the impact curve until the first load drop takes place, which is intended as the final point of quasi-elastic behaviour of the laminate during impact. Another parameter which is important to evaluate impact resistance of a laminate is the so called "damage degree", defined as the ratio between the absorbed energy E_a, as described by the area under the hysteresis cycle and the incident energy E_i, which is the area inside the hysteresis cycle. The difference between E_a and E_i is given by the rebound energy of the sample, and damage degree increases with the presence of more severe damage, as the laminate has an increased difficulty in allowing the impactor to rebound [24-25]. From comparing Table 4a with Table 4b, it can be clear that the differences in impact loading properties between the two laminates are very limited, if not negligible at all.
This type of analysis can be refined by trying to divide the impact hysteresis cycle, hence E_a, into different parts, as illustrated in Figure 5. As it is suggested from the comparison of Figure 4 and Figure 5, the plastic energy area is of very limited extension for impact at 12.8 and 25.6 J. This is reflected by the values of the damage degree, which are basically very similar in the two cases, whilst they are much higher in the case of impact at 38.4 J. In contrast, the maximum deflection and the residual deflection grow considerably with impact energy. In contrast, the variation of linear stiffness is very limited : the value of linear stiffness has proved elsewhere to be particularly related to the overall rigidity under impact performance of the flax fibre reinforced laminates [26]. It might be suggested therefore that even for impact at energies causing penetration both laminates does not undergo a substantial loss of rigidity resulting in uncontrolled deformation and usually in the end in loss of material at rear, usually defined as "spalling" [27]. This is a typical feature of ballistic impact on natural fibre composites, but diffused also in low velocity impact on them [28]. This suggests that the elastic part of impact loading is virtually unaffected by the rise in impact energy, on the other hand, the amount of plastic energy is very variable with higher impact energy, since it is due to a not easily predictable combination of viscous behaviour causing inelastic deformation and creation of fracture surfaces.

In Figure 6a-d both surfaces (impacted and rear surface) of the two laminates impacted at 25.6 and 38.4 J are represented. For all energies, the rear surface of the laminate is subjected to tearing even if the hysteresis cycles indicate that residual deformation at the end of loading is limited. In particular, tearing becomes more complex and elongated at 25.6 J, where it is accompanied by some indentation on the impacted surface, more evident in laminate n.2, most likely due to the presence of flax fibre layer closer to the surface than for laminate n.1. Finally, penetration is clear at 38.4 J, although on the rear surface "branched" crack structures are evident rather than the typical cross-like damage features observed elsewhere for plant fibre composites, for example in a recent study on ramie fibre composites [29]. In other cases, where damage is more concentrated around the impact point as the result of an increased hindrance to delamination, or else an increased tendency of the composite layers to be "locked" together as an effect of impact. This is for example the case with hemp fibre reinforced composites, where typically diamond-shaped penetration areas are observed [30-31].

To better clarify the difference in impact failure mode of the two laminate configurations, in Figure 7a-c side views of the laminates fractured at different impact energies are offered. The mode of failure appears quite different for the two configurations; in particular, the critical point where delamination is likely to occur is at the interface between basalt and flax layers, the latter being visible by the characteristic waviness of the fibre structure. This is highlighted by red circles in particular on 12.8 J laminates, where it is less evident and frequent, yet already present. Passing to higher impact energies, delaminations in this part of the section are more frequent, as visible at 25.6 J especially on laminate n.1, less so on laminate n.2, and in particular at 38.4 J, which is an energy exceeding the one that induces penetration, it is clear that the two configurations present considerably dissimilar penetration features. More precisely, it can be noticed that, although in general terms the value of maximum deflection of the laminates is similar, the presence of more interfaces between layers including different fibres in laminate n.2, brings to a more complex mode of failure, hence to impact diffusing on a larger area, as shown by the wider crack propagation, whereas for laminate n.1 damage appears more concentrated. However, as suggested above by the substantial preservation of the values of linear stiffness, commenting about results in Table 4, Figure 7c confirms there is no evident spalling from impact at 38.4 J for neither of the laminates. On the other side, the different complexity in the mode of damage shown by two laminates with not very different impact performance suggests the possibility to tailor damage creation in a laminate to the needs of the structure it is included in: this process would be further refined by the use of hybrid fabrics for reinforcement (e.g., flax/basalt hybrid leno weave [32]).

CONCLUSIONS

The fabrication of two configurations of ternary hybrids including carbon, basalt and flax fibres in an epoxy matrix, in both cases placing carbon fibres on the outside, whilst disposing basalt and flax fibres internally either in a sandwich or in an intercalated sequence, indicated the criticality of the introduction of flax fibre laminates in this context, in particular as regards impact resistance. As a matter of fact, falling weight impact demonstrated a large presence of delaminations at the interface between flax and basalt layers. This could be the cause of the quite deceiving performance of the hybrids, which just slightly exceeds that of pure flax or pure basalt fibre reinforced laminates. It was found that intercalation with basalt layers does reduce the proneness of flax fibre laminates to bending, enhancing their rigidity, which is beneficial as regards flexural and interlaminar strength: however, impact performance is not substantially increased with respect to the other configuration. On the other side, trying different stacking sequences was demonstrated to sensibly modifying the mode of fracture under impact loading, an effect that could be possibly exploited by designing the stacking sequence to obtain a given distribution of damage absorption across the laminate. Damage features appeared in any case quite complex and quite far e.g., from the typical cross-like or diamond-like crack arrangements normally observed on plant fibre composites on the rear surface when undergoing penetration. This could suggest the possibility to tailor the mode of fracture of these hybrids under impact by modifying their stacking sequence, while leaving basically unchanged the overall impact performance, as appearing from impact hysteresis curves.

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Tests	ASTM standard	Samples dimensions (mm)	Testing apparatus	Other information
Tensile	D3039	250x25x4	Instron 3382 (100 kN load cell)	Crosshead speed 2 mm/minute
Three-point flexural	D790	90x18x4	Lloyd Instruments 30K (30 kN load cell)	Crosshead speed 1 mm/minute Span 72.5 mm Supports diameter 5 mm Loading nose diameter 5 mm
Interlaminar shear strength (ILSS)	D2344	40x10x4	Lloyd Instruments 30K (30 kN load cell)	Span 18 mm Crosshead speed 1 mm/minute Supports diameter 6 mm Loading nose diameter 3 mm
Falling weight impact	D7136	150x100x4	In-home built IFW tower	Impacting mass 1.3 kg 12.7 mm impactor

Table 1 Experimental set-up of mechanical and impact tests

Laminate	Tensile strength (MPa)	Tensile strain	Tensile modulus (GPa)
n. 1	189.23 ± 3.75	0.0253 ± 0.002	16.20 ± 0.52
n. 2	185.24 ± 5.66	0.0218 ± 0.0005	16.89 ± 0.31

Table 2 Tensile properties of the two different laminates

Laminate	Flexural strength (MPa)	Flexural strain	Flexural modulus (GPa)	ILSS (MPa)
n. 1	256.08 ± 9.79	0.0185 ± 0.0008	16.42 ± 0.55	24.33 ± 0,88
n. 2	286.67 ± 15.26	0.0195 ± 0.001	17.08 ± 1.00	25.35 ± 0.98

Table 3 Flexural and ILSS properties of the two different laminates

Impact energy (J)	Max. load (kN)	Max. deflection (mm)	Residual deflection (mm)	Linear stiffness (N/mm)	Damage degree
12.8	4.02±0.10	4.44±0.30	0.70±0.17	1265±84	0.484±0.024
25.6	4.77±0.12	6.21±0.08	1.24±0.05	1258±48	0.545±0.036
38.4	4.98±0.19	8.61±0.32	4.65±0.44	1425±138	0.883±0.063

Table 4a IFW properties of laminate n.1 impacted at different energies

Impact energy (J)	Max. load (kN)	Max. deflection (mm)	Residual deflection (mm)	Linear stiffness (N/mm)	Damage degree
12.8	3.66±0.05	4.32±0.03	0.70±0.02	1237±51	0.497±0.005
25.6	4.63±0.03	6.36±0.02	1.21±0.05	1224±6	0.540±0.041
38.4	5.09±0.11	8.84±0.08	4.17±0.26	1451±161	0.870±0.062

Table 4b IFW properties of laminate n.2 impacted at different energies

LAMINATE N.1	LAMINATE N.2
C 0/90	C 0/90
C 45/-45	C 45/-45
B 0/90	F 90
F 0	B 0/90
F 90	F 0
F 0	F 90
F 90	B 0/90
B 0/90	F 0
C 45/-45	C 45/-45
C 0/90	C 0/90

Figure 1 Stacking sequences of the two laminates (C=carbon, B=basalt, F=flax)

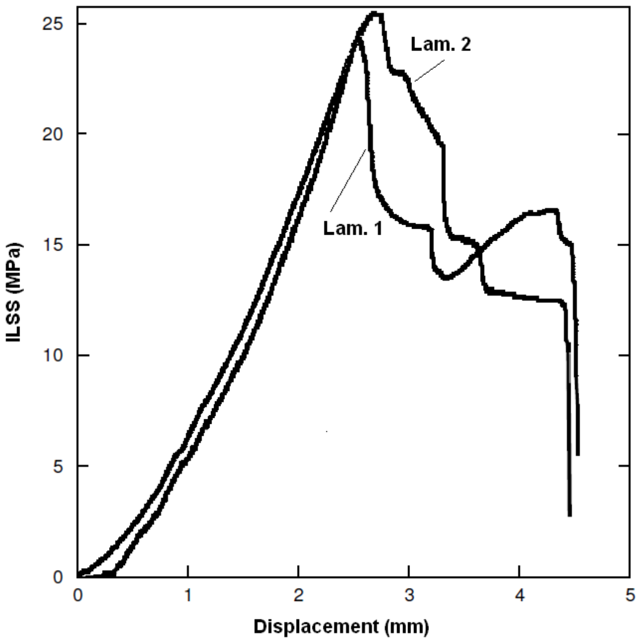


Figure 2 Typical ILSS vs. displacement curves for the two laminates

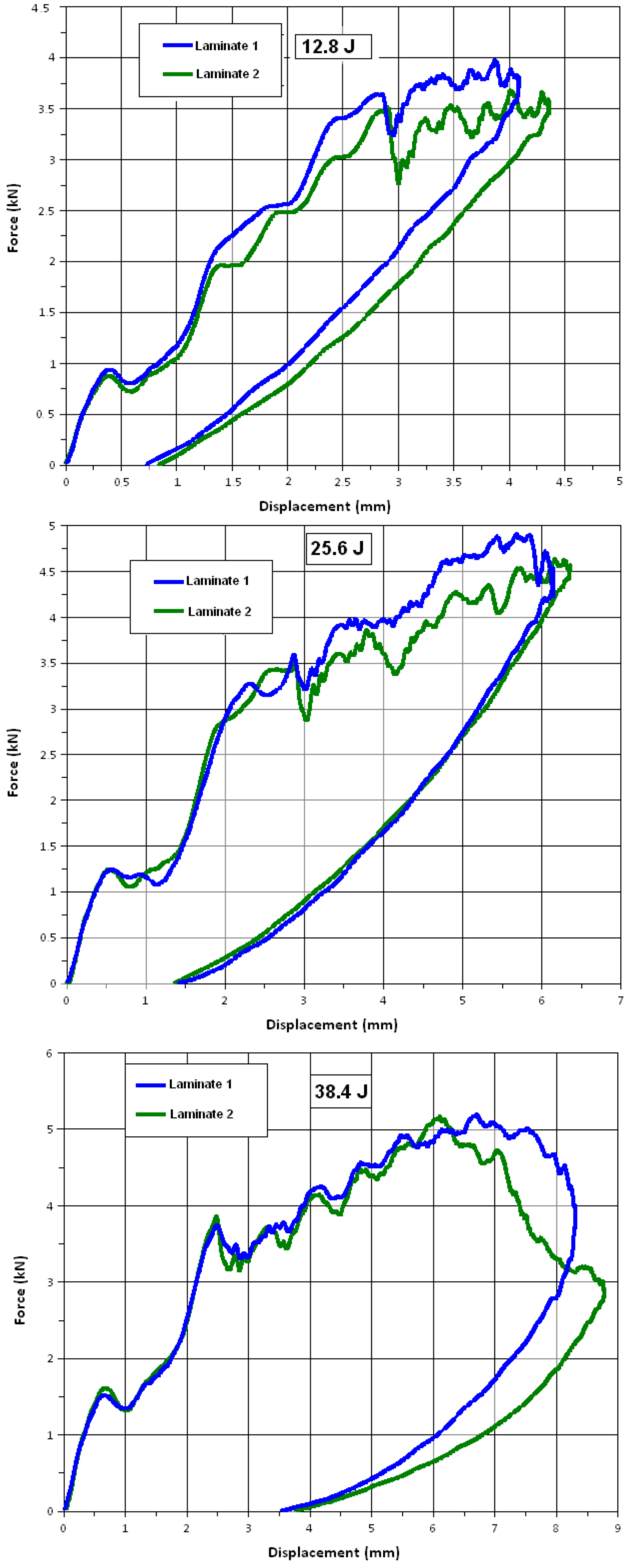


Figure 4 Impact hysteresis curves for the two laminates at different energies

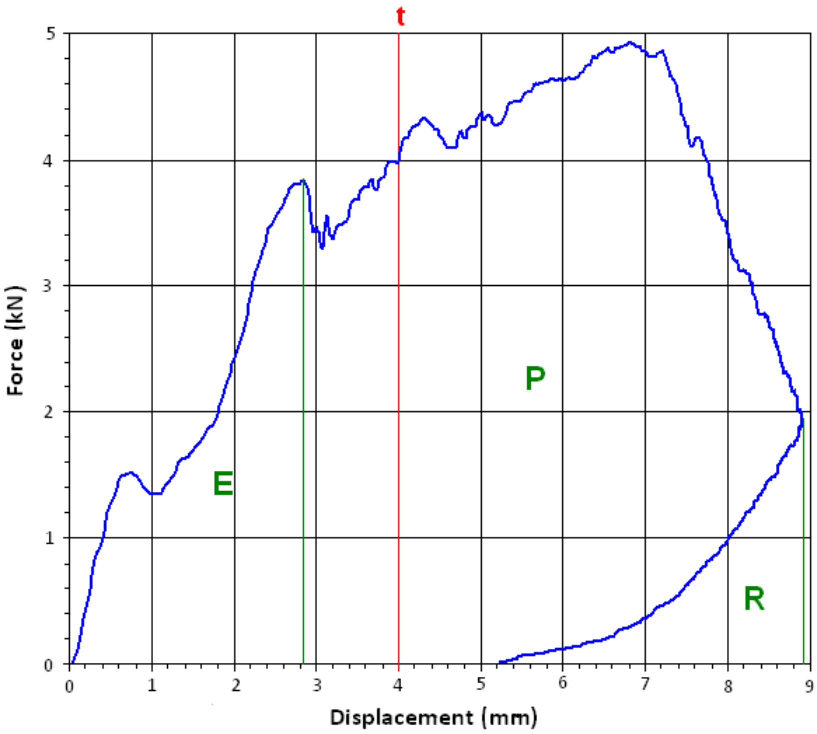


Figure 5 Example of partition of the impact hysteresis cycle
(E= Elastic energy; P=Plastic energy; R=Rebound energy).
The red line corresponds to the impactor reaching the back face of the laminate (t=thickness).

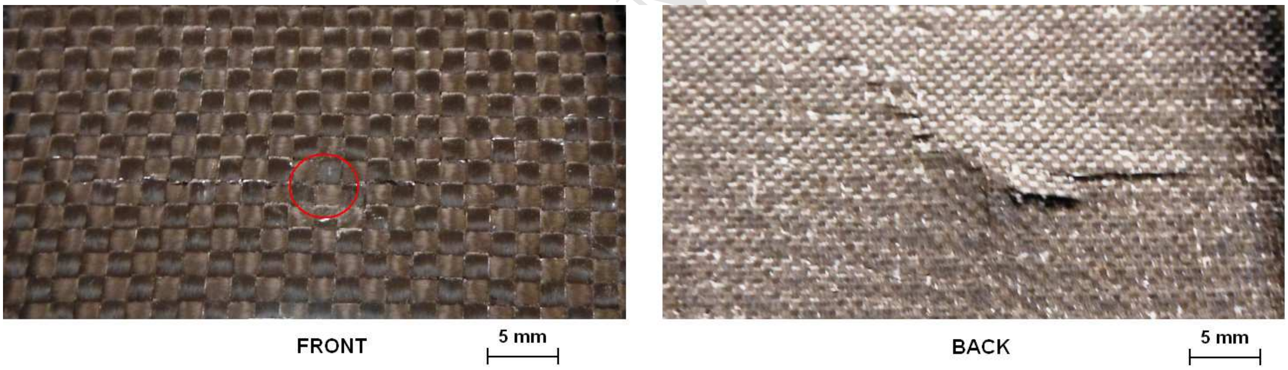


Figure 6a Surfaces of laminate n.1 impacted at 25.6 J

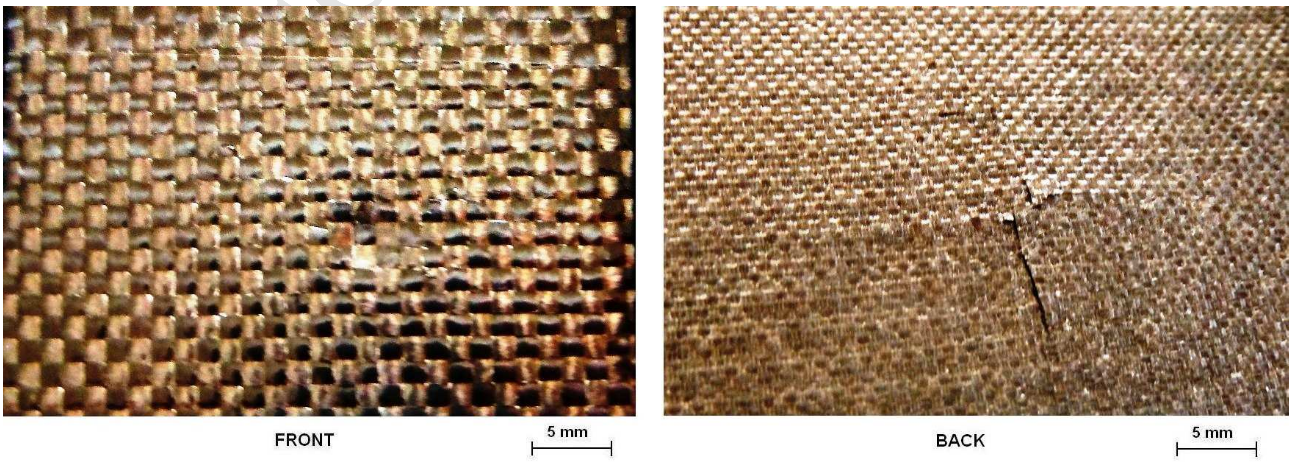


Figure 6b Surfaces of laminate n.2 impacted at 25.6 J

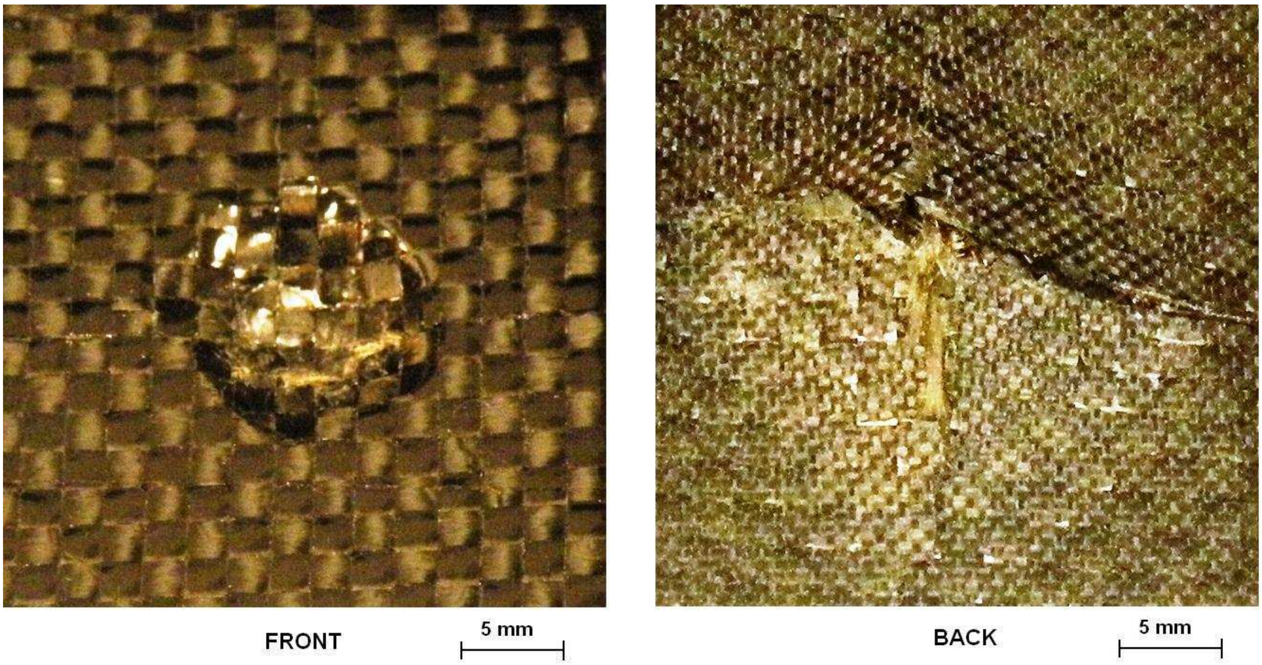


Figure 6c Surfaces of laminate n.1 impacted at 38.4 J

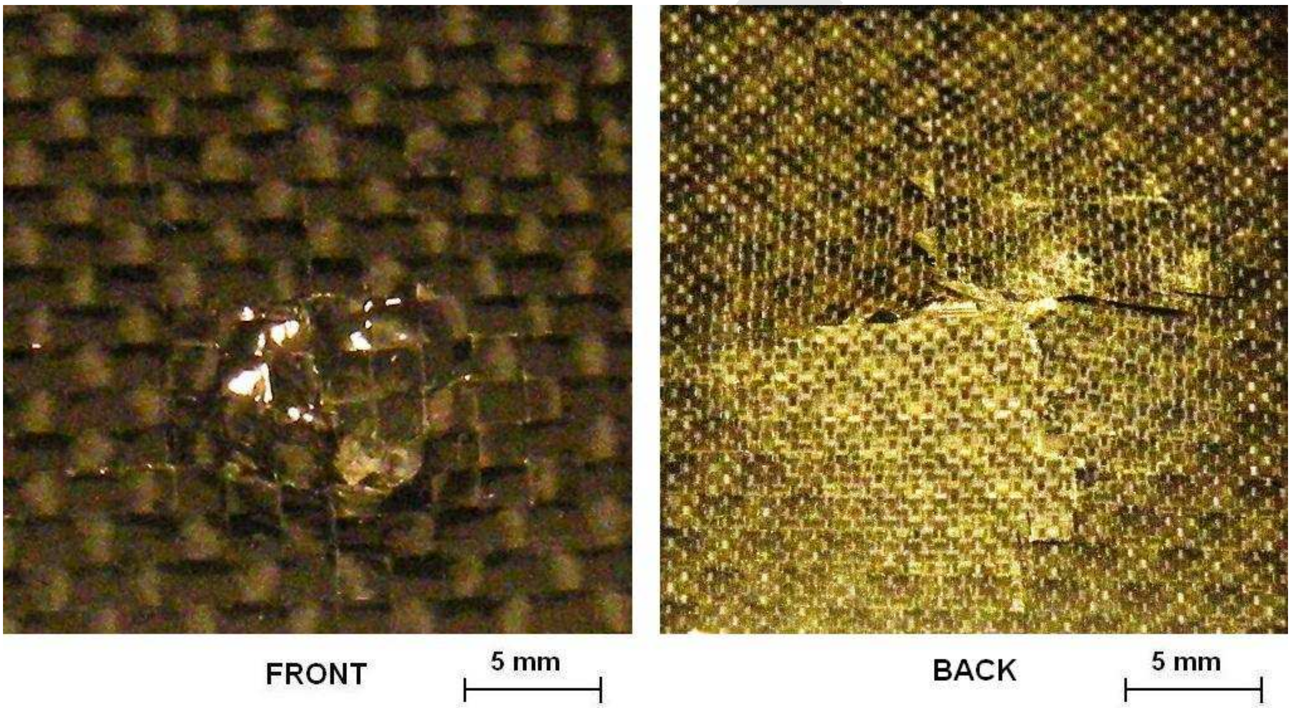
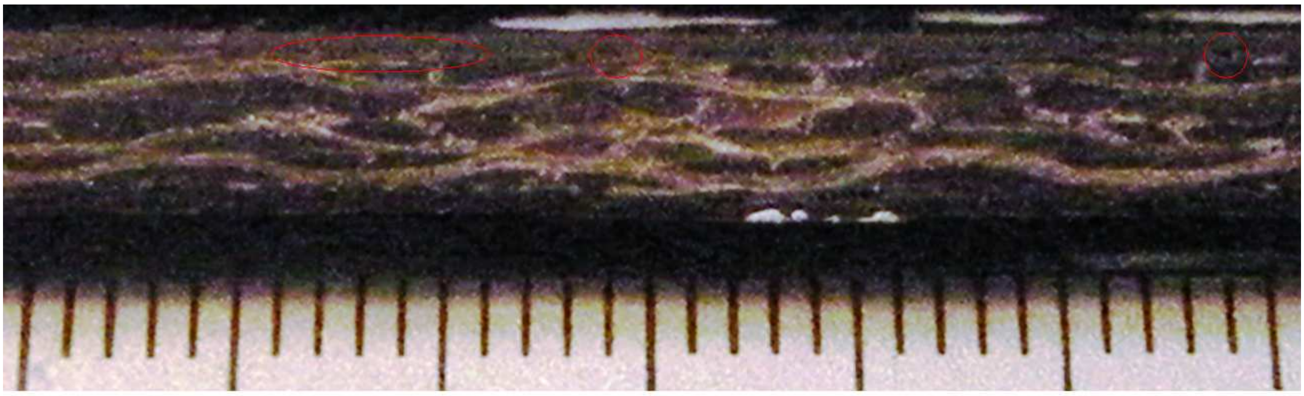


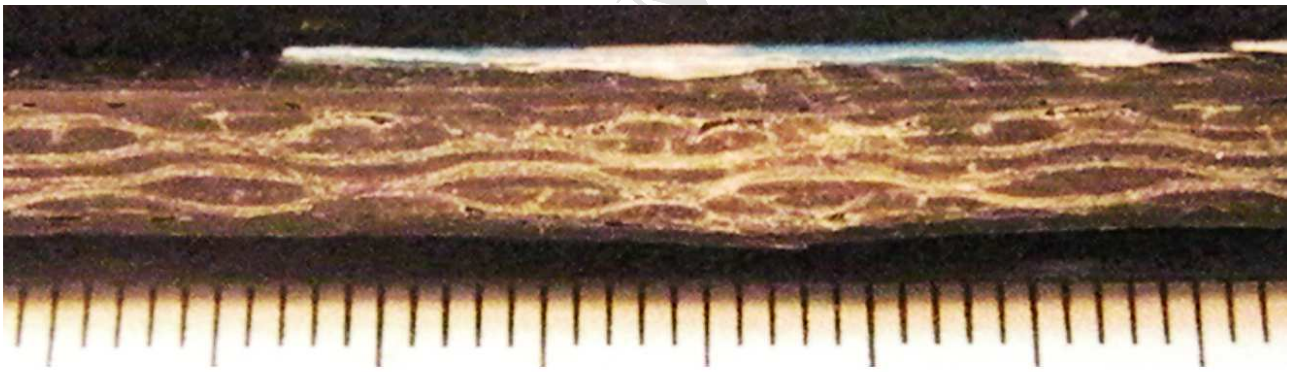
Figure 6d Surfaces of laminate n.2 impacted at 38.4 J



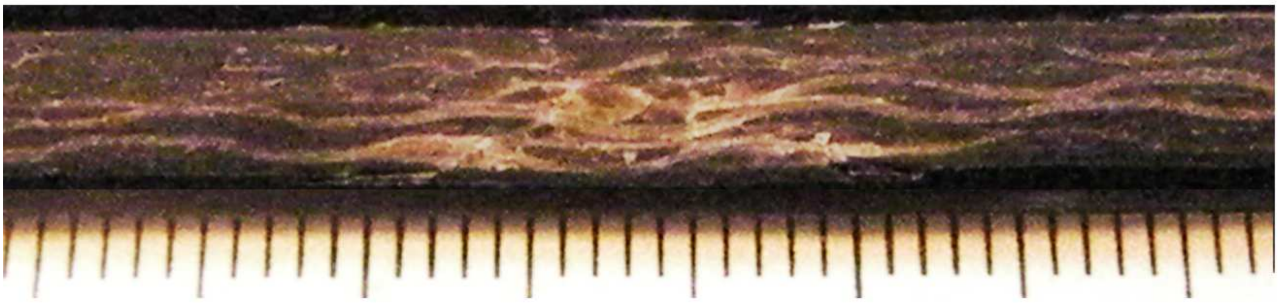
LAMINATE N.1

LAMINATE N.2

Figure 7a Side view of laminates impacted at 12.8 J

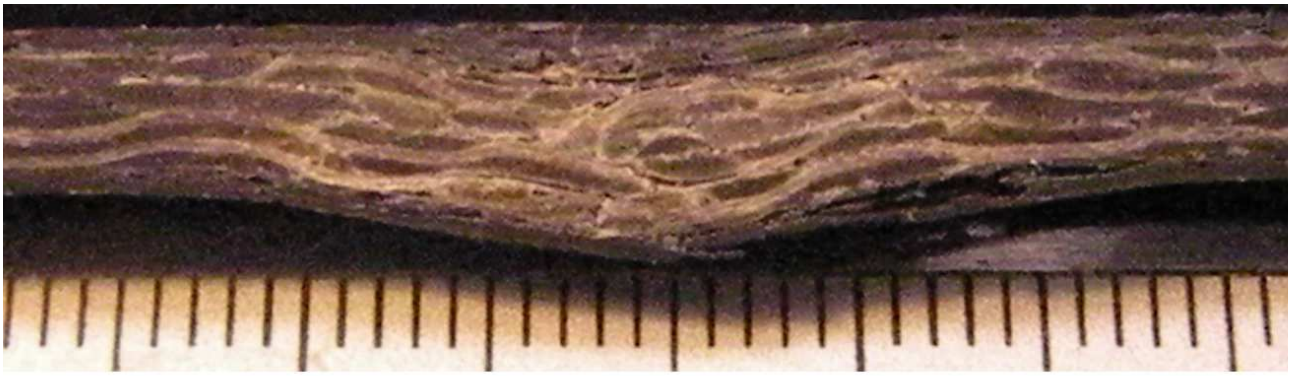


LAMINATE N.1

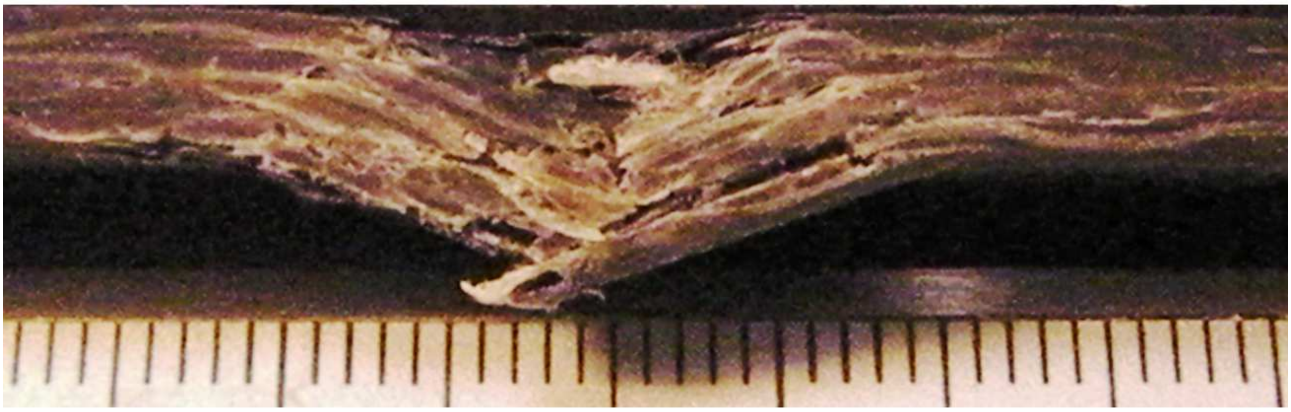


LAMINATE N.2

Figure 7b Side view of laminates impacted at 25.6 J



LAMINATE N.1



LAMINATE N.2

Figure 7c Side view of laminates impacted at 38.4 J