EFFECT OF THE SHORT FLAX FIBER MAT BINDER ON IMPACT PROPERTIES OF UNIDIRECTIONAL FLAX COMPOSITES MADE OF UD-MAT REINFORCEMENTS

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1 INTRODUCTION

In recent days, transport industries are focusing on lightweight materials having good mechanical properties and a lower environmental impact. Synthetic fiber reinforced composites are used in many application areas but the manufacturing of synthetic fibers and their end of life disposal remain major problems to the environment. Natural fibers have lower environmental impact and major advantages over glass fibers, such as a higher specific stiffness, making them very competitive for transport applications. Continuous un-twisted flax fiber yarns have shown good potential as reinforcement architecture for composites [1-3]. Impact loads are important in composite materials because they are prone to delamination and fiber-matrix debonding, failure modes having a large influence on the residual properties of laminates. A popular method used to characterize a composite to a low velocity impact is that using a drop tower [4]. The initiation of cracks in the matrix and at fiber-matrix interface lead to the formation of stresses at the fiber-matrix interface. Moreover, the development of the cracks depends on the thickness and architecture of the layers in the laminate. When an energy threshold is reached, these cracks cause delamination [5].

Most of the works devoted to impact properties in natural fiber composites involved woven fabric reinforced laminates. Panciroli and Giannini [6] investigated the effect of low-velocity impacts on quasi-isotropic flax-epoxy and glass-epoxy made of similar lamination sequences consisting in the stacking of 0, 90 and ±45 plies. Layers made of fabrics were used for the flax laminate while a unidirectional reinforcement was used for the glass laminate. For the glass laminate, the energy absorbed represented approximately 25% of the impact energy at lower energy levels (up to 20 J). Matrix cracking and delamination were dominant at these low impact energies. For the flax laminate, fiber breakage was obtained at all impact energies, resulting in a larger portion of the impact energy absorbed by the laminate compare to the glass laminate for similar energy levels. From these considerations, the impact resistance of flax fiber composites was drastically lower than their glass counterparts, behavior mainly due to the lower flax fiber strength. Ramakrishnan et al. [7] investigate the impact behaviour of flax-epoxy and commingled flax-polypropylene (PP) composite plates manufactured using the compression moulding process. Dry flax woven fabrics used with epoxy resin and balanced 2x2 twill fabric made of comingled flax-PP fibres are used to produce laminates with similar fibre volume fraction (V_f = 40 %) and consolidated thickness. For impact energies ranging from 3 J to 15 J, a large percentage of the total energy was transformed into absorbed energy, which increased almost linearly with increasing initial impact energy, and the energy absorbed by the two materials was similar. The critical energy for penetration failure was approximately 9 J for both composites. Scarponi et al. [8] address the damage resistance and post-impact damage tolerance of plain-weave hemp fabric laminates, with a V_f of 42% under the quasi-isotropic configuration $[(\pm45)/(0/90)]_{2S}$, made of bio-based epoxy and traditional epoxy matrix subjected to low-velocity impact ranging from 5 J to 40 J. For both types of resin and from 10 to 20 J, the absorbed energy represented over 80% of the impact energy, representative of large internal damage in the laminate. The authors mention that the composites investigated "exhibited a quite high compliant behaviour which enabled the specimens to absorb energy through high overall deformation and interface failures such as delamination and pull-out". Vasconcellos [4] study the resistance to low velocity impact of plain-woven hemp/epoxy composites and the influence of impact damage on their residual quasi-static tensile and cyclic fatigue strengths. For a mean V_f of 31% and similar to Scarponi et al. [8], the percentage of absorbed energy for 5 and 10 J impact energies was relatively high (about 50% for 5 J up to 80% for 10 J). For these higher energies and on the back coupon face, the crack propagated along the direction of fabric yarns while the internal damages grow from the top towards the back face in the form of a conical shape, inside which two types of internal damage were observed: interface and matrix cracks. No yarn or fibre breakage was found.

Very few works have been dedicated to laminates made of unidirectional (UD) plis as concern the impact properties of natural fiber composites. Using a pendulum-type impact apparatus, Sy and al. [9] evaluate two configurations of flax-epoxy laminates made of UD plies, a unidirectional $[0]_{8S}$ and a $[0/90]_{4S}$ cross-ply laminate. The cross-ply flax/epoxy laminate exhibited higher energy absorbing capacity at high energy levels (10 to 30 J) due to the presence of the 90° layers that helped stop the matrix crack from propagating through the laminate thickness. At low energy levels however (5 to 10 J), both laminates absorbed approximately 60–70% of the impact energy. For the $[0]_{8S}$ laminate, matrix crack parallel to fibers originated at the centre of the back face where the tensile stress is the highest. New unidirectional flax fiber reinforcements fabricated using a laboratory papermaking process have shown interesting properties for a use in natural fiber composite laminates. These reinforcements are made by projection of a solution of short flax fibers, in suspension in water, over a unidirectional (UD) layer of flax fibers (made of low-twist flax yarns) to obtain a unidirectional reinforcement backed with a thin layer of a mat binder, commonly named UD flax-mat reinforcement. During fabrication of the reinforcement, the short fibers adhere to the aligned UD flax yarns to form a mixed layer with a good cohesion for composites manufacturing. Such reinforcement showed interesting stiffness and strength results in the fiber and transverse directions [2]. During the preforming of complex double curvature part, the mat binder contributes in obtaining a good fiber preform geometry for liquid molding processes like RTM, but also it maintains the UD flax fibers well aligned and parallel when preforming is done under warm and wet conditions [3]. Using conical impactors made of different cone angles, Habibi et al. [10] study the relationship between the impact energy, the impactor angle (a conical impactor was used) and the induced damage for unidirectional flax-epoxy composites (V_f = 40%) made of UD-mat reinforcements similar to those in the actual study. At an impact energy of 2J using a 15° impactor, crack propagating parallel to fibers are observed on the face opposed to impactor face, due to tensile failure of the matrix perpendicular to fibers. For the same 2J impact, a 30° impactor showed larger damage with shear matrix cracking and fiber breakages. Larger damage zones were observed with an increase of the impact energy. The damage were characterized by a main crack propagating transverse to fibers with fiber failure at high impact energies.

In this study and for the same UD flax-mat reinforcement, the impact behavior at low impact energies was performed to investigate the effect of mat binder on the impact performance of $[0]_4$ and $[SFFM/0]_4$ composite laminates denoted as UD and UD-mat composites, both at V_f around 41%. The tests were realized using a drop test tower, mounted with a 20 mm diameter hemispherical impactor, for three energy levels of impact energy, i.e, 3.4

J, 8.6 J and 12.6 J. By comparing the different load-displacement curves and absorbed energies, the objective is to clarify how the mat binder contributes to ameliorate the impact performance of UD composites.

2 EXPERIMENTAL

2.1 Materials

The flax fibres used in this study were supplied by Safilin (France) as a Tex 1000 (1000 g/km) untreated low twist fibre yarn (30 turns per meter) and a Tex 5000 uncompressed flax ribbon. The UD flax/mat reinforcements (Figure 1) are made of Tex 1000 yarns, placed side-by-side to form the UD layer, held together by a flax fibre mat made of short fibres (6 ± 1 mm in length) cut from the Tex 5000 ribbon. The reinforcement fabrication is well described in previous works [2, 3] and will not be described in details here. The matrix was a low viscosity Marine 820 epoxy/824 hardener system supplied by ADTECH Plastic Inc. The hardener ratio (18% by weight) is in conformity with the recommendation of the supplier.

Figure 1. Typical UD flax-mat reinforcement used. The mat binder (left) and UD layer (right) are shown.

2.2 Composites manufacturing

The resin transfer molding (RTM) process was used to manufacture the composite plates with epoxy resin. Prior to composite fabrication, the stack of reinforcements was oven dried for 2 h at 70°C. The composite plaques are manufactured by stacking four layers of reinforcement with the longitudinal direction of UD fibers in the longitudinal direction of the mold. To evaluate the influence of mat binder on the impact properties of laminates, a few plaques were made of layers containing only UD fibers (no mat binder), which were used for comparison with plaques made of UD flax-mat layers. A $[0_4]$ stacking sequence was used for all plaques, where a thin mat binder was present for each layer in the UD flax-mat plaques. Steel spacers are used to ensure constant thickness plaques. The RTM mould used and the procedure details are described in previous works [2-4]. The produced plaques are 270 mm x 150 mm with a fiber volume fraction of 40.9 ± 1.2 % and 41.2 ± 1 % for the UD flax-mat and UD flax plates, respectively. The mold containing the reinforcements was preheated at 100 °C for one hour prior resin injection. After degassing for 10 minutes, the resin is injected at a pressure slowly increased from 1.5 to 3 bars, up to full filling of the mold. The injection and vent ports are finally clamped and the mold placed inside an oven for 3 hours at 110 °C for final cure of resin.

2.3 Testing

Low-energy impact tests have been carried out using a IM10 ITS drop tower (from Imatek Systems, U.K.) mounted with a 9 kg head and a hemispherical impact nose of 20 mm diameter. 75 mm x 75 mm square impact specimens were fixed at their periphery using an annular clamp with inner diameter of 40 mm. The impact tower is equipped with a 30 kN load cell. The displacement of the sample is measured using a laser displacement sensor. Figure 2 shows details of the testing setup. The measured parameters are the impact force (F), the nose displacement (d), the impact energy (E_P), the absorbed energy (E_A) and the restituted energy (E_R).

Figure 2. Impact testing setup showing a composite coupon clamped over the base supporting plate.

At least five coupons per testing condition were evaluated following ASTM D7136 standard. Table 1 shows the coupon type and testing details. In the rest of this work and for the sake of simplicity, the tests will be referred to as tests at 3, 8 and 12J even though they were not carried out at these exact values as specified in Table 1.

Table 1. Impact testing details.

Material	Impact energy (J)
UD-3J	3.4
UD-8J	8.6
UD-12J	12.6
UDMat-3J	3.4
UDMat-8J	8.6
UDMat-12J	12.6

3 RESULTS

3.1 Influence of mat binder on the absorbed energy

Figure 3 compares the maximum displacements, absorbed energies and ratio absorbed/impact energies between the UD and UD-mat composites for all impact energies. The absorbed energy and the ratio absorbed energy/impact energy are found to increase with the impact energy. Up to 8 J, the displacements and absorbed energies are almost the same for the UD and UD-mat composites. The higher stiffness of UD-mat in the transverse direction (due to the presence of the mat) shows a slightly lower displacement (statistically insignificant however) but this doesn't have any evident effect on the absorbed energies and the absorbed/impact energy ratios. The difference becomes evident only at an impact of 12 J where, as will be shown in the next section, full penetration was observed in some of the UD tests while this never happened for the UD-mat tests.

In Table 2 shows the precise values of impact energy, peak load reached, maximum displacement, absorbed energy and absorbed/impact energy ratio. At a higher energy of 12 J, the combination of mat binder to the UD fibers reduces the impact-induced damage; with the absorbed energy and ratio absorbed/impact energies reduced by about 11.1% and 15.2 % respectively. The plate displacement is lower, no full penetration was observed and so the absorbed energy is also lower, producing less interior damage with consequently more recovered energy in this case (see Figure 7). The addition of a thin mat binder over the UD yarns has improved the impact performance of UD flax laminate and can be used advantageously to reinforce composite structures for dynamic loading. Future works should verify if this positive influence of the mat binder is maintained at higher impact energies. It is known that the fiber-matrix adhesion is relatively weak in natural fiber composites. So increasing this adhesion should increase the influence of the mat by reducing premature fiber debonding.

3.2 Typical load-deflection curves

A typical test curve related to impact testing of composite laminates is shown in Figure 4. It shows a loading phase, a damage phase (often represented by a load plateau with more or less instabilities representative of the damage in progress) and an unloading return phase. The area under the unloading phase corresponds to the recovered elastic energy Er and the difference between the impact (Ep) and restituted (E_R) energies, represented by the area inside the force-displacement curve, corresponds to the absorbed energy E_A. The energy balance is such that Ep = $E_R + E_A$. Such a curve has normally a closed shape except when the displacement continues to increase when the impact load decreases, typical of a full perforation of the laminate by the impactor.

Figures 5 and 6 show typical force versus time and force versus displacement curves for the UD and UD-mat composites tested at 8 J. The load-time history starts with a perfect linear load increasing with time without oscillation. It is noted that after the initial linear part, which corresponds to elastic deformation of the composite plate, the successive drops of load in the damage phase are clearly visible. The most significant of them can be attributed to important damage mechanisms. However, some irregularities in the curve could be attributed to the vibration of the material or test bench but in the present case such vibrations have been removed by locally smoothing the curves taking care not to affect their interpretation. The ratio of absorbed to impact energy is 76.3% for the UD composite and 78.8% for the UD-mat composite, so it is very similar. On the other hand and as observed in Table 2, the UD composite shows more variability in the absorbed and absorbed/impact energy ratio results. Figure 7 shows, for all energy levels, the front and back faces of UD and UD-mat coupons after impact testing. As shown, some longitudinal cracks are observed on the back face of 8 J impacted UD and UD-mat specimens. At the 12 J impact energy, damages are visible on both front and back sides, with matrix cracks propagating in both longitudinal and transversal direction with a more pronounced propagation along the longitudinal direction. The

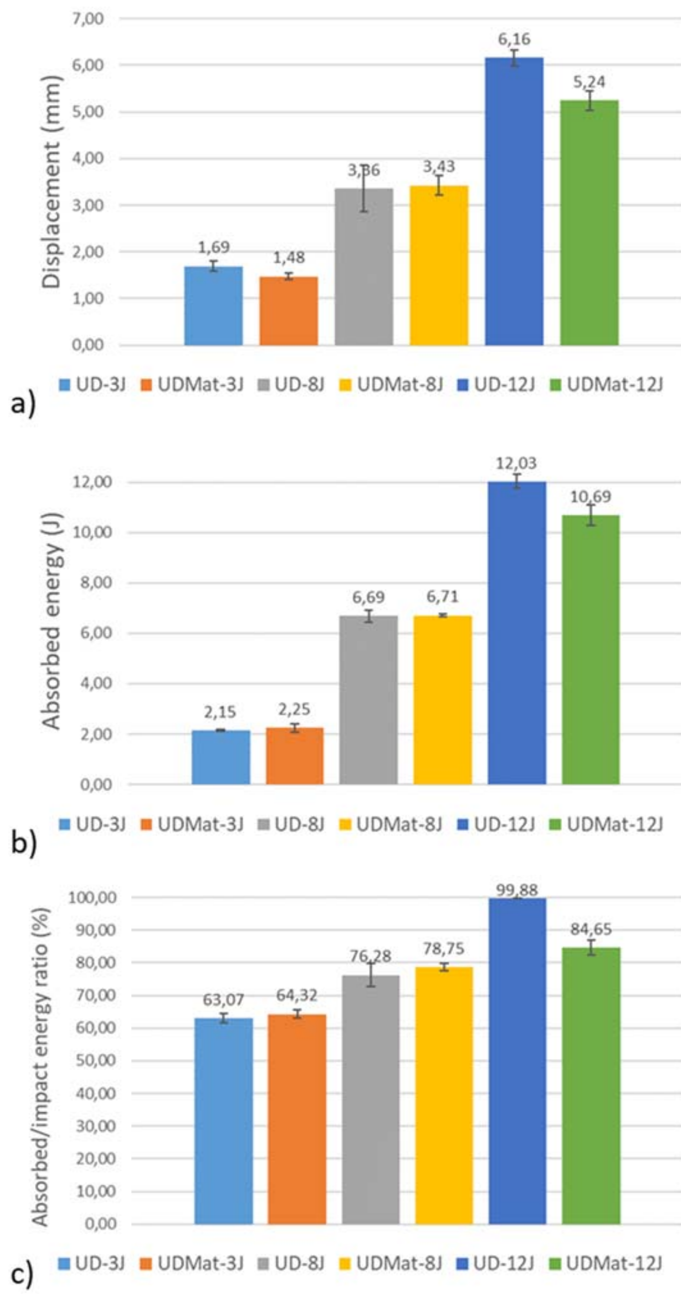


Figure 3. Influence of the mat binder on the maximum displacement (a), the absorbed energy (b) and the absorbed/impact energy ratio (c).

cracks propagating through the fiber yarns lead to the deterioration of the interface between the fibres and the matrix and fiber breakage. Among the 12J impact UD-specimens, some of them were completely destroyed into two parts (as shown in Figure 7). Separation and full penetration of the impactor were never observed for the UD-mat coupons, due to the presence of the mat phase contributing to increase the stiffness and impact strength in the transverse direction of coupons. This is supported by the lower maximum displacement at 12 J for the UD-mat composite (Figure 3), where the mat phase stiffens the composite and reduces the displacement during impact.

Table 2. Results of impact tests for UD and UD-mat composites.

	UD-3J	UDM-3J	UD-8J	UDM-8J	UD-12J	UDM-12J
Impact energy (J)	3.43±0.06	3.49±0.18	8.64±0.19	8.49±0.29	12.15±0.12	12.62±0.19
Peak load (KN)	2.30±0.15	2,53±0,03	2.78±0.21	2.58±0.06	2.56±0.19	2.48±0.04
Displacement (mm)	1.69±0.11	1.48±0.06	3.36±0.50	3.43±0.21	6.16±0.16	5.24±0.21
Absorbed energy (J)	2.15±0.04	2.25±0.15	6.69±0.24	6.71±0.06	12.03±0.28	10.68±0.41
Energy ratio (%)	63.07±1.32	64.32±1.35	76.28±3.47	78.75±1.13	99.88±0.20	84.65±2.21

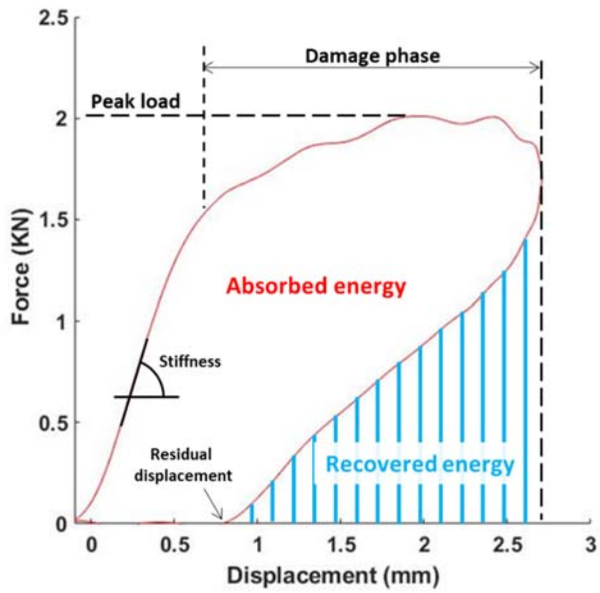


Figure 4: Typical impact force versus displacement.

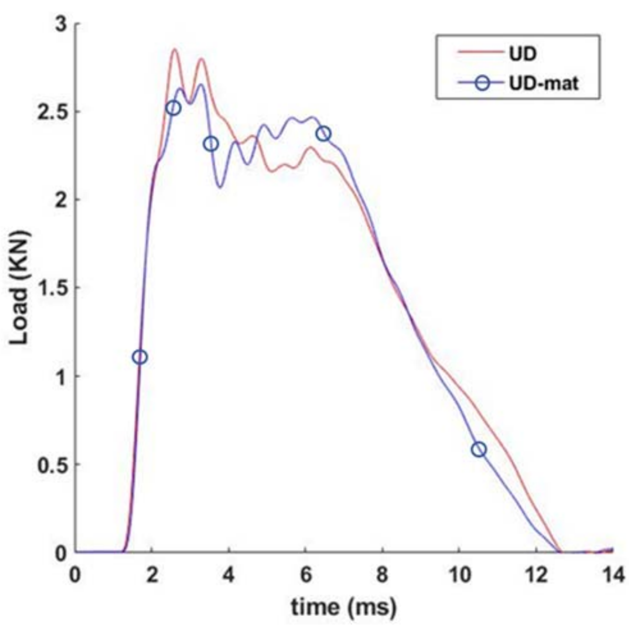


Figure 5: Typical force versus time curves for the UD and UD-mat composites for the 8.6 J test.

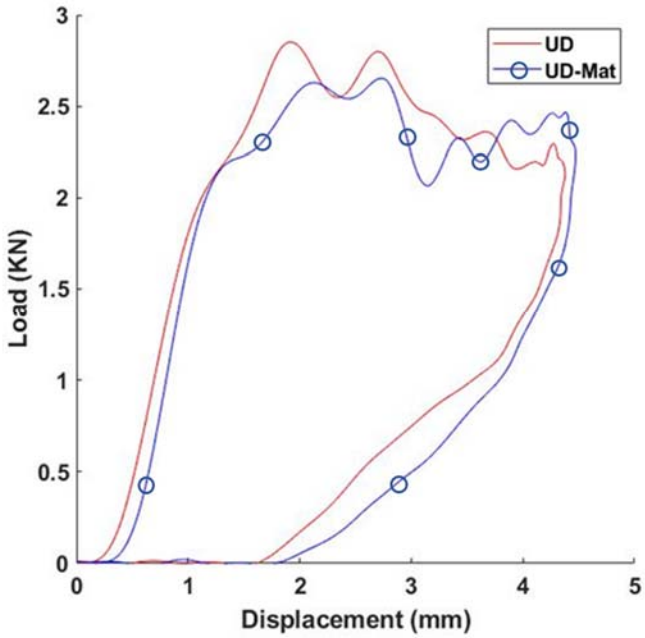


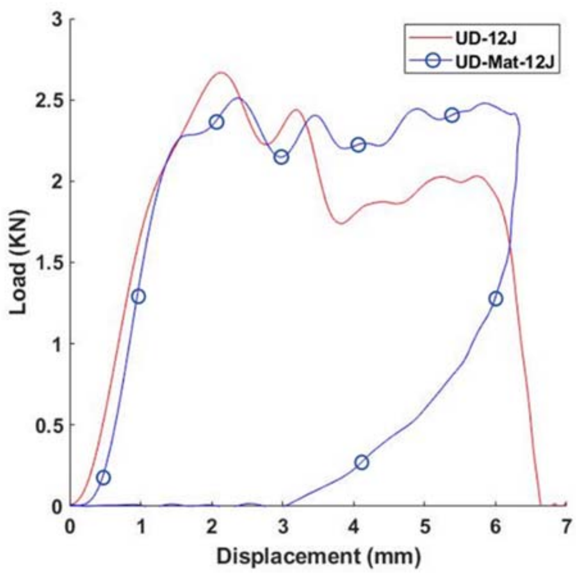
Figure 6: Typical force versus displacement curves for the UD and UD-mat composites for the 8.6 J test.

Front face	**Back face**	**Front face**	**Back face**	
				**3 J**
				**8 J**
				**12 J**
UD		**UD-Mat**		

Figure 7. Typical front and back faces photos of the UD and UD-mat coupons after impact testing for the three energy levels.

Damaging characteristics are also observed in the force-time and force-displacement curves of Figures 5 and 6 where the larger oscillations in the damaging phase suggest the creation of important damages like cracking in the fibre direction, interlaminar cracking and fiber-matrix debonding.

Figures 8 shows typical force versus displacement curves (Figure 8a) for the UD and UD-mat composites tested at 12 J and an enlarged view of the back face of UD-mat coupon tested at this energy level (Figure 8b). The UD composite shows two significant drops in the force-displacement curve indicating important damage mechanism such as delamination and longitudinal cracking. Second, it is clearly observed that the UD composites are much more damaged, sometimes going as far as a full perforation as the displacement keep increasing while the impact force drops. Finally, the UD-mat composites show smaller maximum displacement with a relatively smoother curve in the damage phase. The UD specimens are therefore more damaged than the UD-mats for which the mat phase contribute considerably in avoiding perforation as suggests the enlarged view of the back face in Figure 8b.



a)

b)

Figure 8. a) Typical force versus displacement curves for the UD and UD-mat composites for the 12 J tests and b) enlarged view of the back face of UD-mat coupon after impact at 12 J.

4 CONCLUSION

In this work, the influence of a mat layer (used as binder for the UD phase of a UD-mat reinforcement) has been evaluated for its influence on the low energy impact behavior of unidirectional flax composites. At low energy levels (3 and 8 J), the influence of mat was negligible, with no clear effect on the maximum displacement, absorbed energy and ratio absorbed/impact energies. At the higher energy of 12 J however, it was shown that the mat phase contributes to avoid the full penetration of impactor. This was clearly observed when comparing the load-displacement curves along with the absorbed energy and ratio absorbed/impact energies, which were reduced by about 11.1% and 15.2 % respectively when the mat phase is present in the laminate. These reductions are attributed to the higher stiffness and strength in the transverse direction of the UD-mat composite, which avoid the full penetration of impactor and allow a partial elastic recovery of the impact energy.

5 REFERENCES

[1] Y. Lebaupin, M. Chauvin, T-QT. Hoang, F. Touchard and A. Beigbeder. “Influence of constituents and process parameters on mechanical properties of flax fibre-reinforced polyamide 11 composite”. *J Thermoplast Compos Mater*, 30(11), 1503-1521, 2017.

[2] M. Habibi, L. Laperrière, G. Lebrun and L. Toubal. “Combining short flax fiber mats and unidirectional flax yarns for composite applications: Effect of short flax fibers on biaxial mechanical properties and damage behavior”. *Composite Part B*, 123, pp 165-178, 2017.

[3] R.S. Mbakop, G. Lebrun and F. Brouillette. “Experimental analysis of the planar compaction and preforming of unidirectional flax reinforcements using a thin paper or flax mat as binder for the UD fibers”. Composites Part A, 109, pp 604-614, 2018.

[4] D. S. De Vasconcellos. ‘’Comportement en fatigue avant et après impact de composites tissés chanvre/époxy’’. Ecole Nationale Supérieure de Mécanique et d’Aérotechique (ISAE-ENSMA) - Poitiers, 2013.

[5] F. Bensadoun, D. Depuydt, J. Baets, I. Verpoest and A.W. van Vuure. “Low Velocity Impact Properties of Flax Composites,” Compos. Struct., 176, pp. 933–944, 2017.

[6] R. Panciroli and O. Giannini. “Comparing the Impact Resistance of Flax/Epoxy and Glass/Epoxy Composites through Experiments and Numerical Simulations,” Compos. Struct., 264, pp 1-12, 2021.

[7] K.R. Ramakrishnan, S. Corn, N. Le Moigne, P. Lenny and P. Slangen. “Experimental Assessment of Low Velocity Impact Damage in Flax Fabrics Reinforced Biocomposites by Coupled High-Speed Imaging and DIC Analysis,” Compos. Part Appl. Sci. Manuf., 140, 14 p., 2021.

[8] C. Scarponi, F. Sarasini, J. Tirillò, L. Lampani, T. Valente and P. Gaudenzi. “Low-Velocity Impact Behaviour of Hemp Fibre Reinforced Bio-Based Epoxy Laminates,” Compos. Part B Eng., 91, pp. 162–168, 2016.

[9] B.L. Sy, Z. Fawaz and H. Bougherara. “Damage Evolution in Unidirectional and Cross-Ply Flax/Epoxy Laminates Subjected to Low Velocity Impact Loading,” Compos. Part Appl. Sci. Manuf., 112, pp. 452–467, 2018.

[10] M. Habibi, S. Selmi, L. Laperrière, H. Mahi and S. Kelouwani. “Experimental Investigation on the Response of Unidirectional Flax Fiber Composites to Low-Velocity Impact with after-Impact Tensile and Compressive Strength Measurement,” Compos. Part B Eng., 171, pp. 246–253, 2019.