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EFFECT OF FIBRILLATION OF FLAX MAT BINDER ON THE IMPACT RESPONSE OF UNIDIRECTIONAL FLAX/EPOXY COMPOSITES AND COMPARISON WITH A GLASS/EPOXY COMPOSITE

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Abstract: *In this study, the behavior of flax fiber (FFC) and glass fiber composite (GFC) laminates under a series of low velocity impact loads from a drop weight impact tower is analyzed and compared. All laminates are made by stacking unidirectional (UD) layers of reinforcement, where the UD flax fibers are combined to short flax fibers used as a mat binder to form the base architecture of this reinforcement denoted by UD-mat. Two types of short flax fibers are taken into consideration for the mat binder: one is unmodified fibers while the second one undergoes a refining modification. When comparing the two types of flax/epoxy laminates, the FFC with refined short flax fibers shows the highest permanent and maximal displacements. Results show that, for all impact energies the maximum load for FFC with unmodified short fibers is higher than that with refined fibers. The flax/epoxy composites absorb more energy than the glass/epoxy ones.*

Keywords: Natural composites, flax, impact behavior, fibrillation, damage.

1. Introduction

Composite materials are widely used in structures and as substitutes for metals in various sectors like aerospace and automotive. However, their response to localized impact is one of their limitations [1]. The impact properties of a given composite material depend on several factors. Namely, the type of matrix and its ductility, the type of fibers and their architecture as well as their orientation, the quality of the interface between the fibers and the matrix and the thickness of the laminate [2]. The most used method to characterize the response of a composite to a low-velocity impact is that of the drop tower [3]. This method simulates tools drops and other scenarios during materials manufacture and maintenance [4]. The use of natural fibers in high performance applications has been limited due to a lack of information on their durability facing fatigue loads and impact scenarios. Because of the natural origin of bast fibers, the mechanical behavior of their composites varies remarkably, unlike glass fiber reinforced composites. This is the result of variations in the fiber properties due to many factors such as the seasonal variations, fiber location along the plant stem and fiber defects, to name a few [5]. Ramakrishnan et al. [6] found a similarity in the performance of flax fiber reinforced composites made from a PP matrix and an epoxy matrix. For the epoxy composite, a fiber dominant damage

is detected. Fiber breakage in the back face appeared with limited delamination. Panciroli and Gianni [5] compared the impact response of epoxy composites (using quasi-isotropic configurations) reinforced with non-crimp and twill flax fabrics and UD glass fabrics. They showed that the damage region expands with the increase of the impact energy while fiber breakage, for the flax/epoxy composites, took place at each impact energy. Habibi et al. [7] studied flax/epoxy laminate with UD-mat reinforcements (the same reinforcements used in this work). They found that for low energy levels the cracks on the back face of the specimen are parallel to the UD fibers. As the impact energy increases, cracks in the direction transverse to the UDs (resulting in fiber breakage) begin to appear. Finally, the damage area widens with the increase in the impact energy. Liang et al. [8] also investigate the effect of low-velocity impacts on quasi-isotropic flax/epoxy composites. A microcrack initiation energy is found at 4 J for their materials. The increase in the impact time is linked to an increase in the damage accentuation. This phenomenon is explained by the development of the damage in the material, which is perceptible by the growth of the macrocracks length.

Surface fibrillation consists in the refinement of short fibers with a partial peeling of the microfibrils located at the surface of fibers. Fiber refining is well known in the paper industry, and Afra et al. [9] have found an enhancement of the bonding between fibers resulting in increased tensile strength of the produced papers. For this purpose, the present work is conducted on reinforced epoxy composites using flax UD-mat reinforcements made of non-fibrillated and fibrillated short fibers for the mat phase. The results are then compared to laminates made of UD E-glass reinforcements. The impact behavior is analyzed using force-time and force-displacement curves and photos of the impacted faces of the specimens are shown in support to the analysis.

2. Materials and methods

2.1 Reinforcements preparation and laminates fabrication

The same fabrication procedure for the UD-mat flax reinforcement, described in previous works [10, 11], is applied for this study. The UD phase consists in Tex 400 flax strands supplied by Safilin Inc. (France). Tex 5000 flax strands, also supplied by Safilin, are chopped into short fibers of 6 ± 1 mm, which are used for the mat phase of the reinforcement. It is important to note that this length is chosen to stay over the minimum length/diameter fiber aspect ratio below which fiber debonding and loosening could occur during testing. There is also a length limit of about 10 mm, after which the projection of short fibers/water mixture is difficult considering the risk to obstruct the projection nozzle in the fabrication process. For the preparation of the reinforcements with refined short flax fibers, the fibrillation process is performed according to the following procedure. First, the desired quantity of short fibers is placed in a disintegrator (Figure 1a), water is added, and the mixture homogenized. Next, a quasi-homogeneous thick sheet is obtained using a static former (Figure 1b). The sheet is then tear into small pieces and placed inside a PFI mill (Figure 1c) consisting in a rotor and stator assembly which, by rotation of the rotor, allow the refinement of fibers (surface fibrillation). The refinement level is controlled by the number of rotor revolutions and for this study, this number is set to 400 based on ongoing works. Finally, after fibrillation, the fibers are mixed with water in a proportion 0.06% by weight. The mixture is used next to fabricate the reinforcement, procedure described in previous works [10, 11]. After fabrication, the short fiber mat acts as a binder layer ensuring a natural adhesion between the UD yarns while keeping them well aligned. The average surface

density of the reinforcements is $299 \pm 11 \text{ g/m}^2$, including the 50 g/m^2 mat binder. Before molding, the reinforcements are dried at 105°C for 15 minutes to remove humidity. The glass UD reinforcements, supplied by Texonic Inc. (Québec, Canada), have been chosen to have a surface density of $295 \pm 2 \text{ g/m}^2$, close to that of UD-mat flax reinforcement. The same laminate sequence of $[0]_8$ was chosen in both types of composites to allow comparisons in the analysis.

The laminates are produced using the resin transfer molding (RTM) process. The resin SikaBiresin CR72 and the hardener SikaBiresin CH72-3, both supplied by Sika Industry (USA) are mixed at a resin to curing agent weight ratio of 100:18 according to the technical data sheet. Degassing vacuum was applied for 10 minutes before the start of injection. After injection, part was cured at a temperature of 80°C for 12h. The measured thickness of the flax/epoxy specimens with unmodified (standard) and refined short flax fibers was respectively of $4.28 \pm 0.09 \text{ mm}$ and $4.07 \pm 0.06 \text{ mm}$ corresponding to fiber volume fractions of $38.84 \pm 2.10 \%$ and $40.84 \pm 1.47 \%$ respectively. The measured thickness of the glass/epoxy specimens is $2.34 \pm 0.04 \text{ mm}$ with a fiber volume fraction equal to $37.98 \pm 2.17 \%$ measured by calcination. Note that the flax/epoxy with standard and fibrillated short fibers are termed FUMOS, FUMOF, respectively. While the glass/epoxy composites are called GUD0.

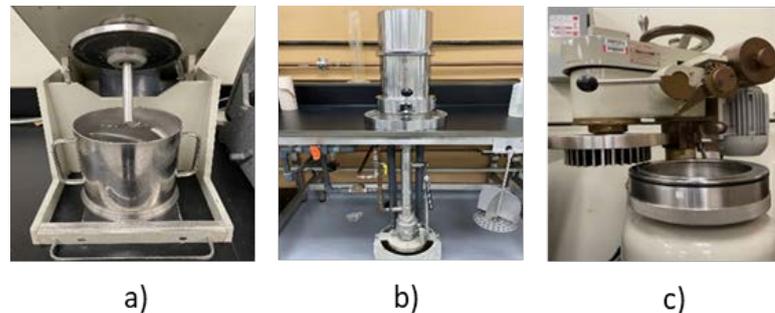


Figure 1. The different equipments used for the short flax fibers refining: a) Desintegrator, b) Static former, c) Refiner (PFI mill).

2.2 Impact tests

The sample dimensions follow the recommendations of ASTM D7136/D7136M standard. An Instron CEAST 9350 drop tower (Figure 2a) is used to perform the impact tests with a load cell of 22 kN. Three impact curves are recorded: load-time, load-displacement, and energy-time. Using the information of these curves, the maximum load and displacement at impact are determined along with the permanent displacement and absorbed energy. The samples had the following dimensions: 150 mm x 90 mm x measured thickness for the FFCs and 150 mm x 100 mm x measured thickness for the GFCs. For reasons related to the reinforcement fabrication, the width of the flax samples was 90 mm instead of the 100 mm recommended by the standard. Considering the relatively smaller size of damage after impact, this had no influence on the results. The length of the sample corresponds to the longitudinal (UD fiber) direction of the composite. The impact energies applied in this study are 3 J, 5 J, 8 J and 11 J. For each impact energy, three specimens are tested for the FFCs and two specimens for the GFCs, which are more repeatable. The impacts are performed with a hemispherical impactor (Figure 2b) of a diameter of 20 mm. The samples are fixed between a lower fixed support and an upper movable support, at a clamping pressure of 7 bar, and the internal and external diameter of supports are 40 mm and 60 mm respectively.

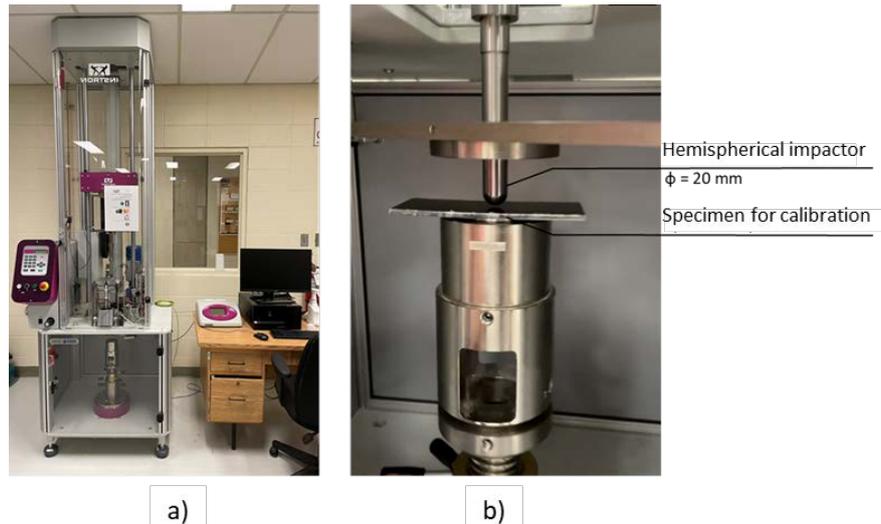


Figure 2. The impact machine used in this study: a) Instron drop-tower and the data acquisition system, b) the impactor with a specimen between the supports

3. Results and discussion

3.1 Force-time curves

The force-time curves of the materials impacted at 8J are shown in Figure 3 for the three types of composites. The peak load of the FUM0S is around 2900 N, with an impact time equal to 8 ms, while the FUM0F have a slightly lower peak load around 2700 N with the same impact time. The GUD0 have the highest peak load at approximately 4000 N (40% higher than the flax/epoxy composites) with a slightly lower impact time of 7.3 ms.

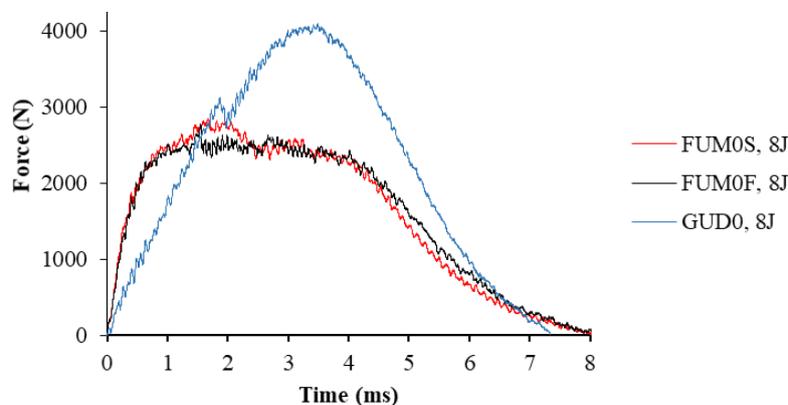


Figure 3. Force-time curves of the FUM0S, FUM0F, and GUD0 laminates.

When comparing the force-time curves, those of FUM0S and FUM0F reach a plateau with a quasi-constant force maintained for the same duration, followed by a decrease. This plateau corresponds to progressive damages taking place in the material. On the other hand, the glass/epoxy curve has a sinusoidal shape, which means a more brittle damage occurs leading to a lower energy dissipation [5]. The small, abrupt load drop occurring at 3000 N can signify that a sudden damage occurred such as delamination which can be observed by visual inspections. In this case, the damage threshold is exceeded without the occurrence of a plateau in the force-time curve. Figure 4 shows the peak loads reached for each type of laminate at each impact

energy. Comparing the response of the FFCs (FUM) and the GFC (GUD), the peak load of the glass/epoxy composite is always higher, the difference being larger at higher impact energies (see Figure 4). Finally, and compared to the standard flax UD-mat reinforcement, it appears that surface fibrillation of short mat fibers did not have an evident influence on the peak load between 3 J and 8 J. Otherwise, fibrillation seems to reduce the peak load at the higher 11 J impact energy.

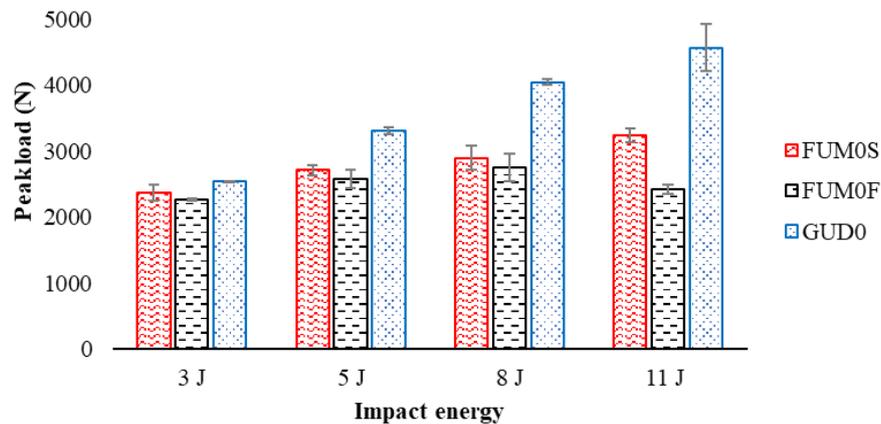


Figure 4. Peak loads registered at the four impact energies (3 J, 5 J, 8 J and 11 J) for the FUMOS, FUMOF and GUDO

3.2 Force-displacement curves

Typical force-displacement curves at 8 J are shown in Figure 5. It is observed that the fibrillated flax/epoxy specimens show a slightly larger permanent and maximum displacement than the specimens with standard mat. The average permanent displacement of FUMOS is 2.045 ± 0.290 mm, 15% lower than that of the FUMOF which is 2.417 ± 0.197 mm. However, this result must be considered with care considering the overlap of standard deviations. The force-displacement history of FUMOS and FUMOF are very similar showing a plateau with a larger surface area under these curves compared to GFC. Instead, the curve shows lower dissipated energy, and the damage threshold is exceeded with a continuous load increase. Finally, the GUDO showed a maximum displacement comparable to that of the FUMOF, but the permanent displacement of the GUDO is much less pronounced than that of the FUMOS and FUMOF, so more energy was restituted in the glass-epoxy laminate compared to the flax-epoxy ones.

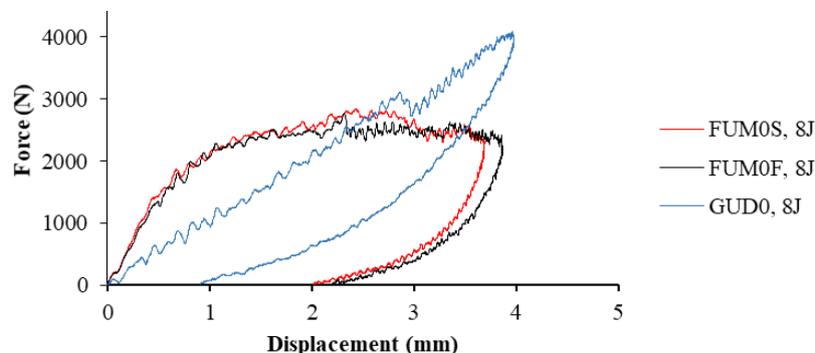


Figure 5. Force-displacement curves of the FUMOS, FUMOF, and GUDO laminates.

Figure 6 shows the mean permanent displacements for all tests. In line with Figure 5, the permanent displacement is always lower for the glass-epoxy laminate. The permanent displacements of the FFCs are similar at low energies, but that of fibrillated composites (FUM0F) becomes larger at 8 and 11 J. The standard flax UD-mat reinforcement seems to perform better than the fibrillated one as concern the impact behavior of UD flax laminates.

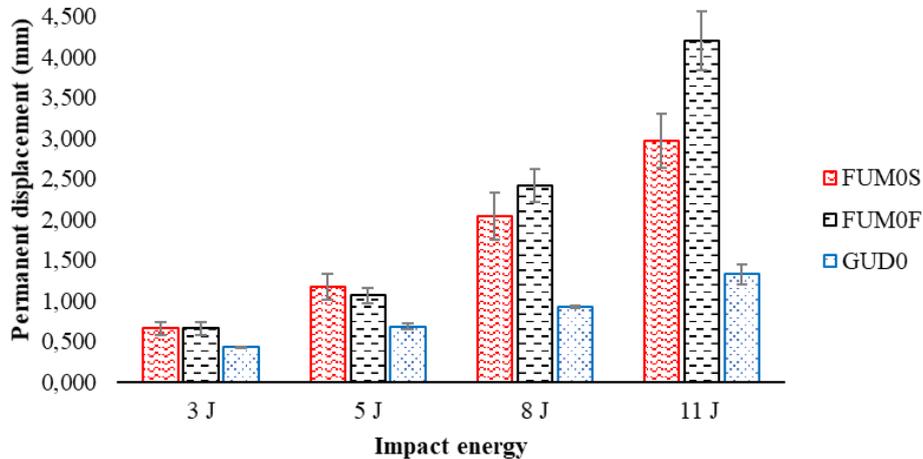


Figure 6. Permanent displacements registered at the four impact energies (3 J, 5 J, 8 J and 11 J) for the FUM0S, FUM0F and GUD0

Globally, the energy absorbed by the biocomposites is higher than that absorbed by the GFC, as shown in Figure 7. The absorbed energy and the energy absorption ratio increase progressively with the applied impact energy for the three types of laminates. It should be noted that no full penetration occurred.

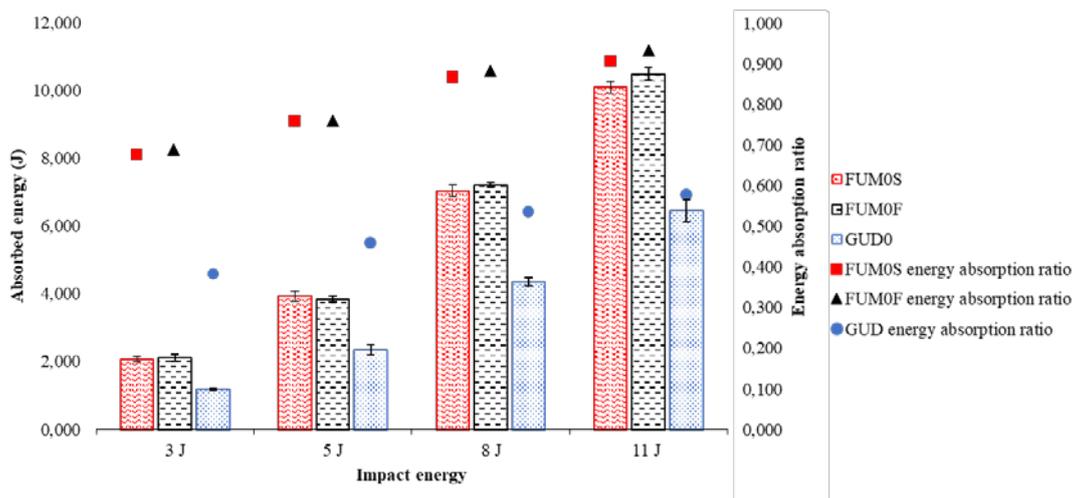


Figure 7. Absorbed energy and the energy absorption ratios for the four impact energies (3 J, 5 J, 8 J, 11 J) for the FUM0S, FUM0F and GUD0

3.3 Damage analysis

Figure 8 shows example images of the front and rear faces of the different specimens impacted at 8 J. It can be seen that the impacted flax/epoxy specimens present on the front face two large longitudinal cracks that spread on a length of about 36 mm. On the rear face, the damage is

highlighted by a central longitudinal crack of about 41 mm with other smaller longitudinal cracks, and a large transverse fracture. Moreover, delamination has been visually detected on the rear face of the FUM0S and FUM0F specimens impacted at 11 J. The damaged surface takes an elliptical shape with the longitudinal and transverse cracks being its primary and secondary axes, respectively. In a study conducted by Habibi et al. [7] on the same type of flax UD-mat/epoxy composites oriented at 0° with the same V_f of 40%, the form of damage that were identified are matrix tension cracks, fibers breakage, delamination, and shear matrix cracks. The difference in the size of damages in their material compared to the ones of this study can be explained by the use of a conical impactor in their work. They also used thinner 2.95 mm plates, compared to the 4.07 and 4.28 mm thick fibrillated and standard flax laminates in the actual work. The main damages occurring in the glass/epoxy impacted specimens are matrix cracks and delamination (see GUD0 in Figure 8). Delamination was detected due to the transparency of this composite. In line with previous observations in Figure 6, it is important to note that fiber breakage occurred only in the FFCs, which may explain the difference in permanent displacement between FFCs and GFC. Visual inspections of the damages at the front and rear faces of impacted flax/epoxy specimens (at 8 J) show the presence of fiber breakage, which can explain the appearance of the plateau in Figures 3 and 5.

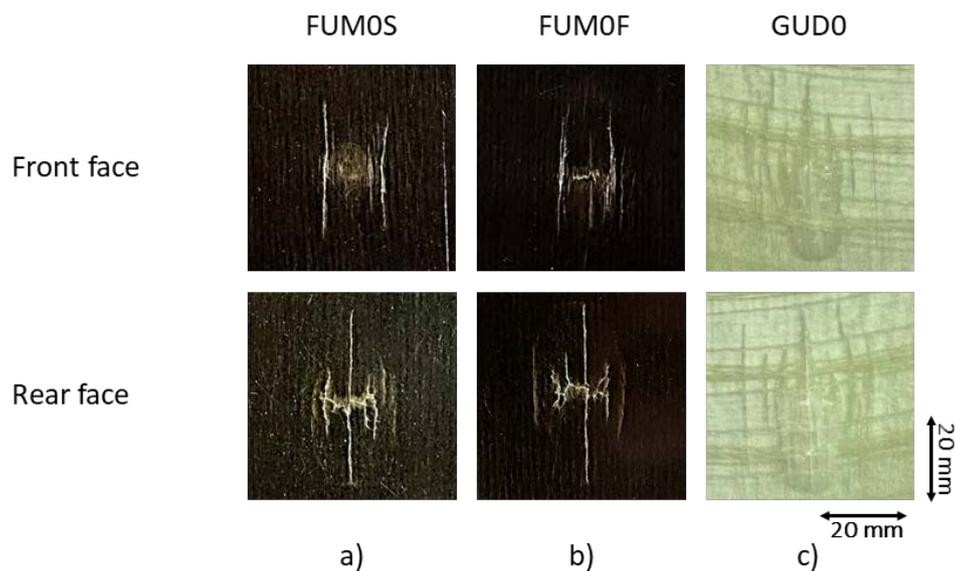


Figure 8. Front and rear faces of the impacted specimens at 8 J : a) FUM0S, b) FUM0F, and c) GUD0

4. Conclusion

In conclusion the flax/epoxy laminates with refined short flax fibers exhibit the highest permanent displacement for the impact energies of 8 J and 11 J when comparing to standard FFCs. In addition, the maximum load for FFC with unmodified short fibers is slightly higher than that with refined fibers, and the difference becomes remarkable for the impact energy of 11 J. For this reason and for the energy levels tested, it can be concluded that the fiber refining does not provide a higher performance of the FFC under impact loading. On the other hand, the GFC exhibits the highest maximal load and absorbs less energy than the FFCs. In terms of damage mechanisms, delamination and matrix cracks are observed in GFC while matrix cracks, fiber breakage and delamination occurred in FFCs. It may be concluded, at least for the studied

parameters and UD laminate types, that the fibrillation did not offer an improvement for the flax/epoxy UD composites and the glass fiber is more resistant than flax fiber facing a low velocity impact. For the upcoming studies, the effect of fibrillation will be explored while considering cross-ply flax/epoxy composites.

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