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Scrutinizing The Effects Of Fibre Ratio And Laminate Lay-Up Configuration

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DESIGNING STIFF AND TOUGH BIOCOMPOSITES BY HYBRIDIZATION OF FLAX AND SILK FIBRES: SCRUTINIZING THE EFFECTS OF FIBRE RATIO AND LAMINATE LAY-UP CONFIGURATION

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Keywords: Biocomposite, Hybrid composites, Ductility, X-ray Computed Tomography, Flax fibre, Silk fibre

Abstract

In this study, the hybridization of flax and silk fibre reinforced composites, at ply level, were studied and compared to their monolithic counterparts. Hybrid FRP laminates were produced via the film-stacking compression moulding method using the highly ductile thermoplastic high-density polyethylene grafted with maleic-anhydride as the matrix. The fibre volume fraction ratio between the two fibres and the lay-up configuration were studied, resulting in hybrid composites with different degrees of distribution of the flax and silk plies within the laminate. The results showed that more balanced properties in terms of stiffness, strength, strain to failure, and impact energy absorption can be achieved by varying those two parameters, thus increasing the designing freedom tailored to specific engineering applications. Additionally, the findings revealed that by optimizing those parameters, multiple fractures of the flax plies or "fragmentation" can be achieved as a toughening mechanism, leading to a pseudo-ductile hybrid composite with a gradual or delayed failure development. The fibre fragmentation mechanism, apart from the increased ductility, can be potentially used as a failure detection criterion for structural health monitoring.

1. Introduction

Flax fibre composites offer a sustainable alternative to traditional synthetic fibre-reinforced composites. Known for their relatively high stiffness and strength along with a low density, these composites find applications in industries such as automotive and construction, combining performance with eco-friendliness [1]. Additionally, flax fibres with negative CO2 emissions during their growth phase, are (potentially) low-cost and have additional outstanding physical properties such as remarkably good acoustic and mechanical vibrational damping and low coefficient of thermal expansion [2, 3]. As the demand for sustainable bio-based solutions grows, flax fibre composites present a compelling option for diverse applications.

However, flax fibre composites have limited impact resistance and toughness due to the intrinsic brittleness of the reinforcing fibres reaching a tensile strain at failure in the range of 1%-2% [4–6]. Therefore, this results in brittle catastrophic failure with little to no signs of failure before breakage and poor impact performance typically reaching energy values of around 4-6 J/mm (normalized to plate thickness, with a 16-20 mm diameter striker on a circular plate of diameter 80mm) [7]. On the other hand, silk, a protein-based (animal) natural fibre is attracting growing attention in the composites research community because it exhibits high ductility resulting in outstanding toughness and impact damage resistance.
In general, silk fibres can be divided based on their origin to spider or silkworm produced fibres with spider silk outperforming silkworm produced silk essentially in any mechanical property, often being described as a “super” fibre. This outstanding combination of properties is a result of continuous evolution by nature for 300-400 million years which allowed the development of fibres capable of stopping larger flying insects with high kinetic energy [8–10]. However, mass production of spider silk is not possible, particularly in the scale required for high performance composites due to the cannibalistic nature of the spiders to control their population dynamic in an area [11]. Therefore, hereafter any mention to silk will refer to silkworm produced silk, specifically produced by the *Bombyx mori* species, which can reach exceptionally high strain to failure of above 16% [12], remarkable toughness (70 MJ/m^3, surpassing traditional highly tough synthetic fibres like Kevlar 40 (50 MJ/m^3) [13, 14]), and high impact resistance [15].

Therefore, it seems promising to combine the high stiffness flax fibres together with the highly tough silk fibres to create a hybrid composite without compromising the bio-based nature of the composite, thus obtaining synergistic properties as well as introducing various modes of damage propagation which can affect the toughness and impact resistance. The two types of fibres can be combined in many different configurations, however the hybridization at the ply level, here indicated as interlayer configuration, where the different layers/plies of each fibre are stacked onto each other, has the advantage of the manufacturing simplicity and low cost for producing hybrid composites, compared to intralayer hybrids where the two fibres are combined within the same fabric and the yarn-to-yarn where the fibres are mixed at the fibre level [16]. The combinations of different lay-up stacking sequence reportedly have a strong effect on the resulting properties of the hybrid composites due to the degree of ply dispersion [17–19] between the two types of fabric layers as well as the fibre volume fraction (V_f) ratio between the fibres, thus resulting in different damage propagation mechanisms. Controlling these parameters in an intelligent combination can result in development of synergistic effects and in some cases appearance of pseudo-ductility [20].

In this study, silk fibres are combined with flax fibres in an interlayer configuration at different fibre volume fraction (V_f) ratios between the two fibres and different lay-up stacking sequences, thus resulting in a range of mechanical behaviour due to the resulting damage initiation and propagation. Both non-hybrid (monolithic) and hybrid laminates were subjected to tensile testing and low-velocity impact and were thereafter inspected using X-ray computed tomography (XCT) to observe the resulting characteristics of the failure modes.

2. Materials and Methods

2.1. Materials

Unidirectional flax fabrics with an areal density of 110 g/m^2 under the commercial tradename FlaxTape were sourced from Ecotechnilin, France and silk weave fabrics with an areal density of 200 g/m^2 were supplied by Sport Soie, France. The silk fabric was degummed by the supplier before used in the manufacturing of the composites. High density polyethylene grafted with maleic anhydride (HDPE-MA), grade Bynel 40E529 was select as the polymer matrix system which was provided by DuPont, Belgium and extruded by Amcor flexibles, Belgium in a thin film with thickness of 65 µm. The reasoning of the matrix selection system is the high strain to failure of above 200% which based on the results of [12, 21] is a crucial parameter to allow the silk fibres to reach their full potential in terms of tensile strain and impact resistance. Furthermore, the melting point of the matrix is around 135 °C, which is positive since it is highly recommended that the processing temperature is kept below 160 °C due to the thermal degradation temperature of the silk, which is accompanied by a transition in colour of the fabric from white to yellow.

2.2. Composite manufacturing

Because of the hydrophilic nature of both types of fibres, all fabrics together with the matrix films were pre-dried at 60°C for 24h prior to the composite manufacturing step. If this is not done, this may result in high porosity due to the formation of steam because of the applied processing temperature. The composites were manufactured using the film stacking compression moulding method where polymer films of the matrix are stacked alternately between each fabric layer. The stacking of the layers occurs in a stainless steel frame, which is used to control the final thickness of the laminate, thus ensuring high control of the produced thickness of the laminate and hence precise control of the final fibre volume fraction (V_f), which was aimed to be close to 40% for the pure composites and for the hybrids (flax and silk). For the compression moulding process the stack was pressed in a pre-heated hydraulic hot press (Fontijne, Belgium) where pressure of 15 bar and temperature of 150 °C were applied for 15 min, followed by a cooling step to room temperature using the internal cooling system of the press with a cooling speed of approximately 30 °C/min. Both non-hybrid and hybrid composites were manufactured following the same process. The produced laminates were cut into testing coupons using a hydraulic guillotine knife.

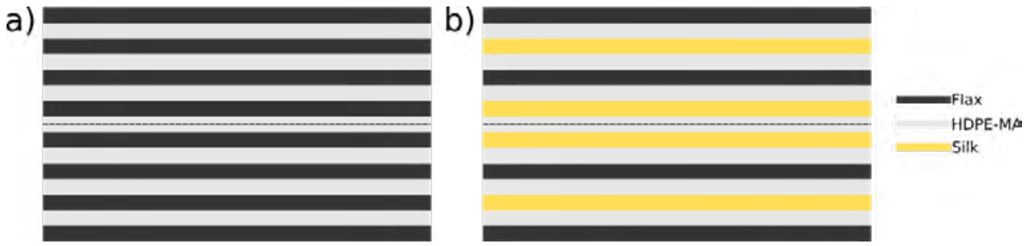

Figure 1. Representative manufacturing layup configurations of a) non-hybrid and b) hybrid composites.

2.3. Tensile testing

Tensile testing was performed using an INSTRON 5567 frame equipped with a 30 kN loadcell and an extensometer to measure the strain. The testing rate and dimensions of the produced specimens were in accordance with ASTM D3039. No end tabs were used apart of sandpaper, however the statistical majority of the specimens failed within the gauge length and samples failing within the gripping area were excluded from the analysis.

2.4. Low-velocity-impact testing

Impact properties were determined using an INSTRON CEAST 9350 following the falling weight impact method, where in this test specimens of 100×100 mm plates were punctured using a rail guided striker with a hemispherical head of 20 mm diameter. The combination of the striker mass and drop height resulted in a kinetic energy level before impact of 100 J. The composite plates were pneumatically clamped with a ring-shaped fixture with an inner diameter of 80 mm. The force was recorded using a 22 kN loadcell installed in the striker and the displacement was recorded using a laser device monitoring the position of the striker. Together with the monitoring of the time, force-displacement and displacement-time diagrams were used to determine the actual kinetic energy before impact and after impact, thus leading to a precise calculation of the absorbed impact energy. The specimens used for the impact testing had a crossply configuration to properly characterize the composite behaviour in impact.

2.5. X-ray computed tomography (XCT)

The post-impact inspection of the composites was performed using the XCT scanner TESCAN UniTom HR equipped with a 160 kV/25 W X-ray tube with a tungsten reflection target and a detector of 2916 x 2280 pixels, with a 50 µm pixel pitch. The scans were performed at 60 kV and 4 W with a voxel size of 4 µm for the tensile and 30 µm for the impact specimens, respectively. The reasoning behind the larger voxel size for the impact specimens is the need for a larger field of view due to the large area affected by the impact load. In total, 3000 radiographic projections were acquired over a 360° angle rotation of the sample each with a 210 ms exposure time and with a frame averaging set to 5. After acquisition, the radiographic projections were reconstructed to tomographic slices using the TESCAN reconstruction software Panthera, which is based on a filter back-projection algorithm. The results analysis and further 2D or 3D visualization were performed in the Avizo software (Thermo Fisher Scientific).

3. Results and discussion

The mechanical behaviour of the hybrid composites compared to the non-hybrid flax and silk composites is shown in Figure 1, using the representative stress-strain diagrams. The reference non-hybrid composite shows the reasoning behind the combination of these two fibres with flax being significantly stiffer and stronger but with a drastically lower tensile failure strain, whereas the silk is compliant but has a higher failure strain. It is observed that depending on the stacking sequence (Figure 1a) the hybrid composites achieve different behaviour and more noticeably strain to failure, despite having the same flax and silk ratio. When the silk fibres are positioned on the outside in a “sandwich” configuration with the flax fabrics on the inside, the composite exhibits sudden catastrophic failure when the hybrid composite reaches the failure strain of the flax composites. On the other hand, when the layers are positioned inversely, the hybrid shows an initial drop at the strain to failure level of flax composites, however then pseudo-ductility is noticed where the stress remains on a fairly stable level while the composite can reach nearly 5 times higher strain to failure than non-hybrid flax composites. This pseudo-ductility is a result of the damage mechanisms within the composite where after the initial fragmentation, delaminations develop and spread around the fragmented brittle fibre fabric [16, 20]. Additionally, the more dispersive configurations often showed a more classical hybrid behaviour where an initial drop is observed and is followed by a transition of the load to the remaining silk fabrics which to a little extent delays the failure of these hybrid composites.

Similarly, in Figure 1b, the effect of the V_f ratio between the two fibres on the mechanical response is presented. It is observed that at flax V_f level of above 21%, the failure of flax layers essentially determines the fracture of the hybrid composite, because the silk layers are getting damaged due to the energy release during the breakage of flax. However, this strain energy release due to flax fibre breakage can be avoided at lower flax V_f content, thus allowing the silk layers to survive since there are less flax layers failing, and thus keep carrying load. Moreover, at flax V_f content of 7%, the hybrids can retain almost all the strain to failure compared to the pure silk composites coupled with a noticeably higher stiffness due to the presence of the much stiffer flax layers leading to significant synergistic effect.

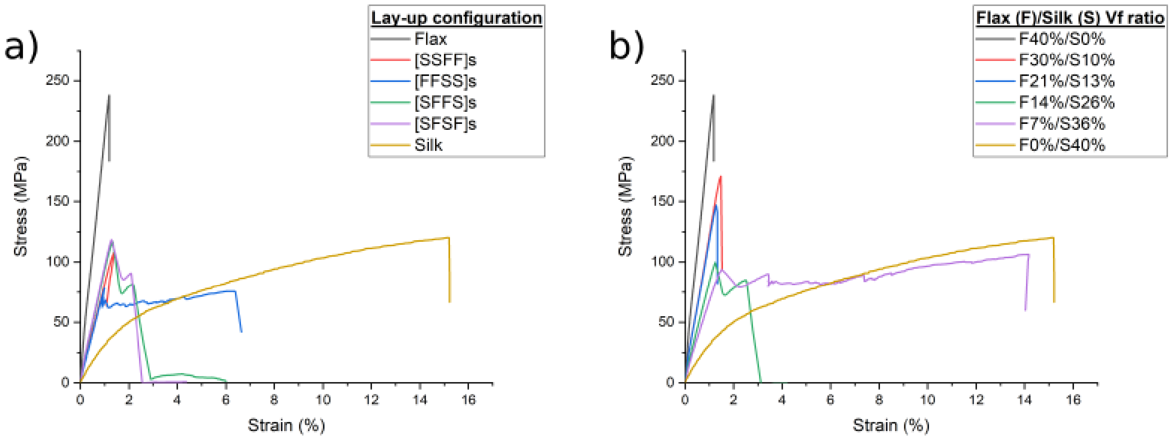

Figure 2. Representative stress-strain diagrams of hybrid composites and their non-hybrid counterparts: a) effect of layup configuration for the same V_f ratio of flax 14%/silk 26% and b) effect of the V_f ratio.

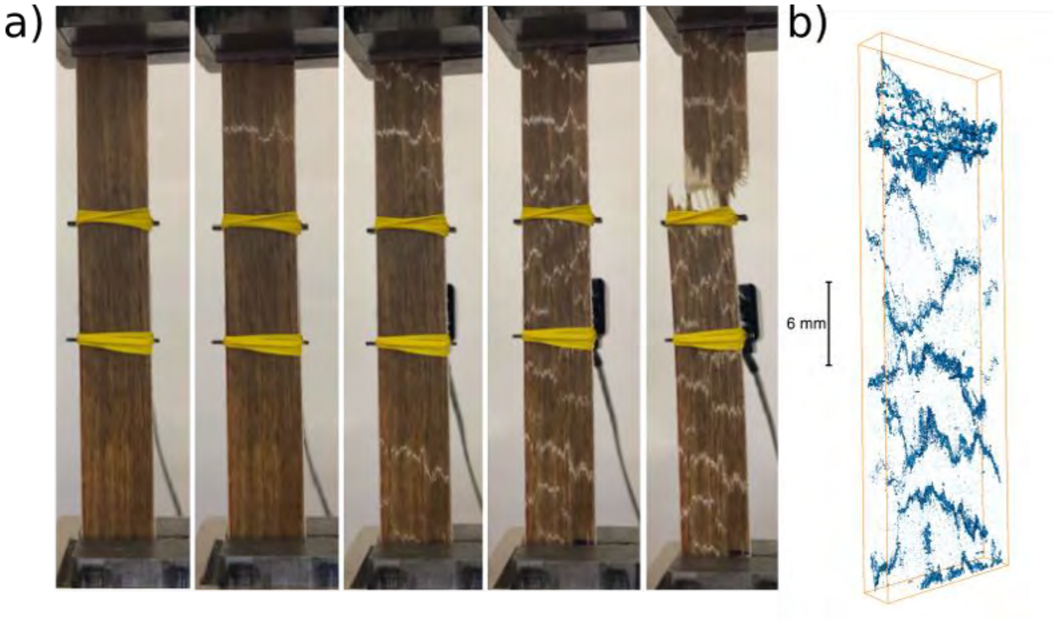

Figure 3. a) Visual development of the fragmentation process and b) internal image using XCT observation of the damage mechanism which resulted in the appearance of pseudo-ductile behaviour,

Multiple fractures of the flax layers were achieved in specimens by optimizing the combination of the two parameters of lay-up stacking sequence and V_f ratio between the two fabrics. The fragmentation behaviour (Figure 3a) starts with one fragment when the strain to failure of flax is reached, however with further increase of the strain the stress starts building up again resulting in the creation of follow up failures on the same flax layers, which are often accompanied by delamination of flax near the fragmented area (Figure 3b). The samples which exhibited this fragmentation process had a stress-strain curve where a pseudo-ductile behaviour was visible.

The impact resistance is one of the key reasons for performing hybridization in this study. Figure 4 shows the impact force-displacement recorded together with the low-velocity kinetic energy absorption at penetration. It can be seen that increasing the V_f content of silk higher than the flax content increases the kinetic energy required for penetration. However, from the tensile stress-strain curve, it can be observed that this comes at a cost of primarily stiffness. Overall, the sandwich structure with flax at the outer layers showed consistently, for all V_f ratios, the best performance amongst the different stacking sequences. One potential explanation is that due to the stiff and strong fibres being on the outside, the bending stiffness of the laminate increases and since a bending moment is induced when the striker impacts the specimen, higher bending stiffness increases the resistance to the impact load.

Another observation is that when the flax/silk fibre content ratio increases from 14%/26% to 21%/13% and above, the impact energy absorption is practically the same as for the pure flax composites, which is rather limiting towards application of these materials where high stiffness is essential for the structure. Overall, the flax/silk system with a ratio of 14%/26% shows an in between behaviour with ~130% improvement in impact energy compared to the flax composites while retaining a stiffness higher than the silk composites, thus allowing more design freedom in the use of bio-based composites due to its more balanced properties.

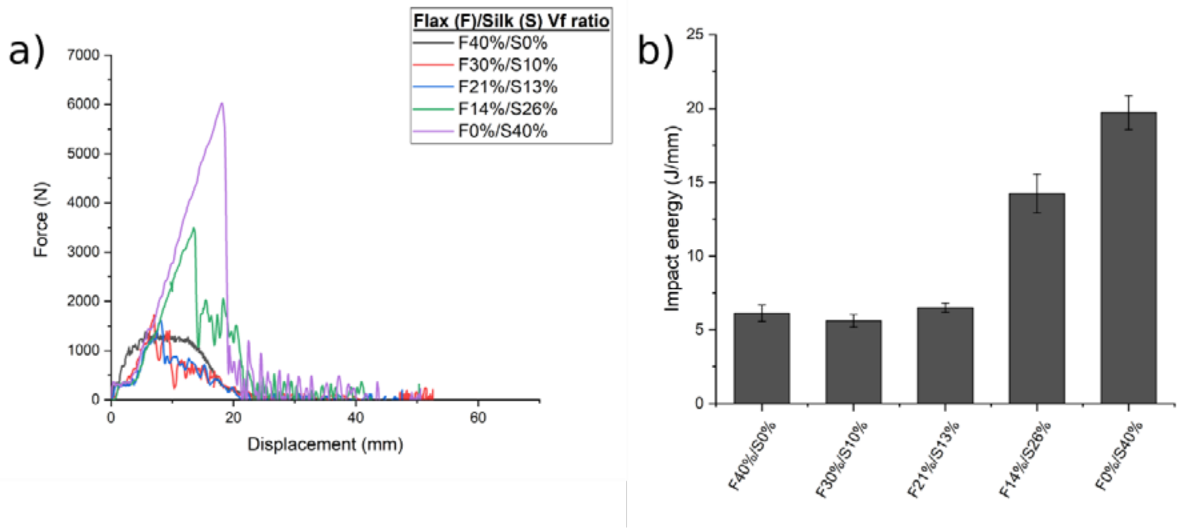

Figure 4. Impact properties of produced hybrids compared to non-hybrids: a) force-displacement representative curves and b) calculated impact energy absorbed normalized to laminate thickness.

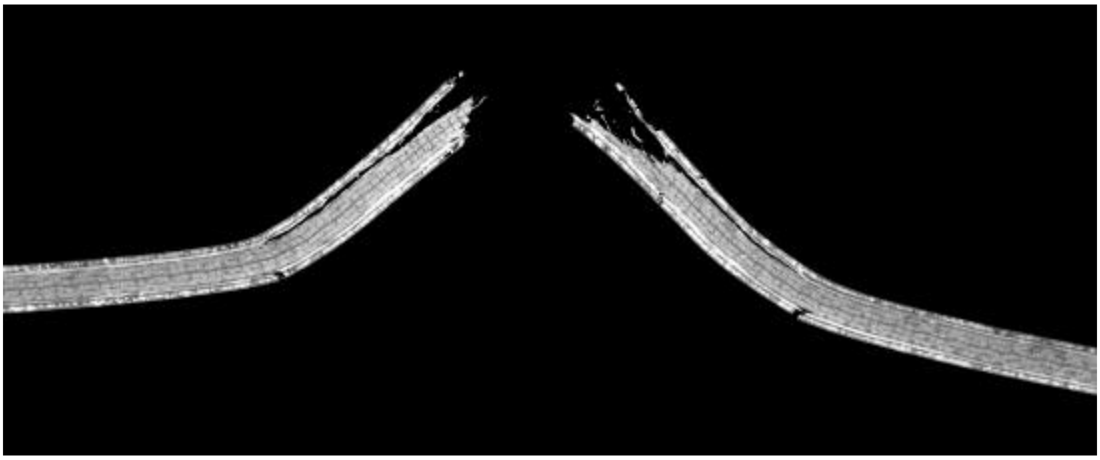

Figure 5. Internal damage observation of best performing hybrid (F14%/S26%) in impact using XCT.

Furthermore, XCT was used to study and explain the high, compared to non-hybrid flax, energy absorption in low-velocity impact of the F14%/S26% sandwich-type hybrids where the flax is located at the outer plies. Using this method the internal damage of the specimens is visualized in Figure 5 where the appearance of large delamination areas is observed indicating that this combination of design parameters for the hybrid material results in damage spreading over a larger area.

4. Conclusions

In this work, the interlayer hybridization of stiff and strong flax fibres with highly tough silk fibres was explored using different lay-up stacking sequences with a range of V_f ratios between the two fibres. By controlling these parameters, different stress-strain behaviours and balance in properties such as stiffness, strength, tensile strain to failure and impact energy absorption was achieved. It was noticed that when the fibre content of flax is higher than 21%, compared to the total 40%, the failure of flax fibres determines the failure of the hybrid composites due to the damage to the surrounding silk layers, explained by the violent nature and rapid energy release from the flax fibre breakage. Furthermore, in specific cases, namely F14%/S26%, only for the [FFSS]s configuration, and for the composites with volume fraction ratio F7%/S36%, the fragmentation of flax layers in multiple locations across the specimen's length was observed, which resulted in around 5 times higher strain to failure compared to flax fibre composites. This introduction of pseudo-ductility was further studied using XCT where flax delamination around the fragmented areas was observed, thus introducing a damage mechanism which can be used to control the materials' behaviour and additionally these local delaminations add to energy absorption mechanisms. Furthermore, laminates with fibre content of 14% flax/26% silk showed more than 130% increase in impact energy absorption compared to monolithic flax fibre composites while retaining a higher modulus than silk composites. This observation was explained using XCT suggesting that large delamination areas lead to spread of damage over a larger volume in this hybrid configuration.

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