The Potential of Biocomposites in Low Velocity Impact Resistance Applications

Fabrizio Sarasini, Jacopo Tirillò, Claudia Sergi, and Francesca Sbardella

Abstract The urgent need to make natural fibre composites suitable for semi-structural applications demands a thorough assessment of their behaviour under different loading conditions and strain rates. In this regard, low velocity impact represents a severe hazard to the composite industry due to the resulting complex damage scenario able to markedly impair the mechanical properties of composite structures. The aim of this chapter is to provide a comprehensive review of the resistance to low velocity impacts of natural fibre composites, with a view to highlighting the effects of the various factors that influence the impact resistance of traditional fibre reinforced composites. The potential of natural fibre composites and differences with the behaviour of the synthetic counterparts are addressed, along with the areas that need improvement for a better exploitation of natural fibre composites in semi- or structural applications. Literature survey highlighted that also for natural fibre composites the toughness of the matrix dictates the energy absorbed at perforation, the damage resistance and tolerance, which are largely independent of fibre architecture. Another important feature, for energies far from perforation, is the less detrimental role played by delamination compared to synthetic laminates.

Keywords Natural fibres · Natural fibre composites · Low velocity impact · Impact resistance · Damage tolerance

1 Introduction

Over the last twenty years the use of biocomposites, intended as conventional polymer matrices sourced from fossil resources reinforced with natural fibres, has recorded an enduring increase in several industries due to global awareness and promotion of sustainable development (Pickering et al. 2016; Sanjay et al. 2018; Gholampour and Ozbakkaloglu 2020). In this regard, the global natural fibre composites market size was esteemed at USD 4.46 billion in 2016, while it is envisaged to grow with a

F. Sarasini (✉) · J. Tirillò · C. Sergi · F. Sbardella
Department of Chemical Engineering Materials Environment, Sapienza-Università Di Roma, Via Eudossiana 18, 00184 Roma, Italy
e-mail: fabrizio.sarasini@uniroma1.it

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CAGR (Compound Annual Growth Rate) of 11.8% from 2016 to 2024, in accordance with a recent market report (Grand View Research Inc. 2018). The most important raw materials are wood, flax, kenaf, cotton, and hemp, but wood is still governing the market with a 59.3% share of the total revenue in 2015 (Grand View Research Inc. 2018). Another important contribution is offered by flax, which in 2015 had a market share of 13.0% thanks to its CO_2 neutrality, vibration damping ability and high specific mechanical properties (Yan et al. 2014; Bourmaud et al. 2018).

The need for lightweight and sustainable composite materials is governing the rise of applications of natural fibre composites in two specific industrial sectors, namely the automotive and construction, where biocomposites are usually applied in cosmetic applications (door panels, decking, frames, etc.). Natural fibre composites find applications in the automotive field because the parts, in addition to adequate mechanical performance, offer a weight saving (by 30%) that allows to reduce the fuel consumption and diminish CO_2 emissions. It is not surprising that this segment accounted for a revenue share of over 30% in 2015 (Grand View Research Inc. 2018), the main objective being the replacement of glass fibres with wood or non-wood fibres such as flax and hemp (Koronis et al. 2013). Natural fibres represent also inexpensive and sustainable alternatives to synthetic fibres used as building materials, supported by a share of 56% of the overall market volume in 2015 (Dittenber and GangaRao 2012; Grand View Research Inc. 2018).

This development, especially in the transportation field, has exacerbated the need for a thorough understanding of the damage and fracture mechanisms of natural fibre composites when subjected not only to quasi-static but also to dynamic loads. The shift from static to dynamic loading in heterogeneous and anisotropic materials, like composites, is much more complicated compared to traditional metallic materials. Two or more constituents with varying mechanical properties and potentially different fibre/matrix adhesion quality influence the propagation of stress waves that in turn results in a complex and unpredictable damage initiation and subsequent propagation. In this regard, a major threat to composite structures is represented by their proneness to low velocity impacts, as they produce significant internal damage as delaminations, matrix cracks and fibre breakages, which can go easily undetected but that considerably influence their residual mechanical properties. This occurs also if barely visible impact damage (BVID) is produced. In fact, BVID can involve delaminations and back-face splitting, which result in residual strength reductions by as much as 50–60% (Shah et al. 2019). It is therefore of utmost importance to prove that composite structures can bear loads even when already damaged, which is included in the well-known issue of damage tolerance. The problem with composite materials lies in the meaning of the word damage, as it involves an accumulation of matrix cracks of different size-scales, shapes and orientations, voids, broken fibres and delaminations. Therefore it is expected that this “state of damage” can originate the loss of diverse design-induced functionalities and the loss of strength is just one of them (Talreja and Phan 2019). This scenario is even much more complex for natural fibre composites, in which the inherent variability in natural fibre properties is often coupled with a non-reliable and in-depth understanding of their mechanical behaviour, in particular in terms of their impact damage resistance and damage tolerance. This complexity can partly explain why information on low velocity impact behaviour of composites including natural fibres, such as jute, flax, hemp, etc., is still quite restricted. Nevertheless, this is a valuable and required property for assessing their suitability to semi- or structural applications. The aim of this chapter is to outline a comprehensive overview of the low velocity impact behaviour of natural fibre composites with reference to the several factors that influence the impact resistance of traditional fibre reinforced composites (Fig. 1), to identify the missing points

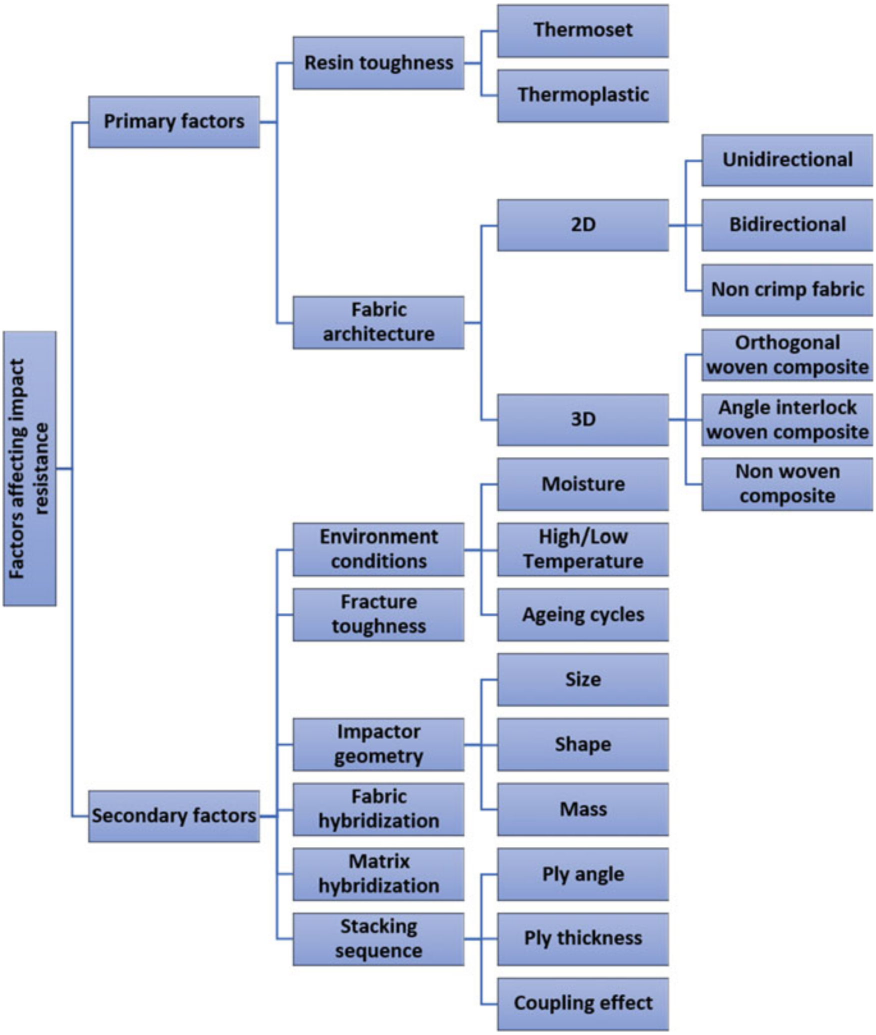

Fig. 1 Factors affecting impact resistance of fibre reinforced composites (adapted from Shah et al. (2019))

in the available literature and recommend potential directions for future researches in this field.

2 Low Velocity Impact of Natural Fibre Composites

2.1 Effect of Matrix Toughness on Impact Behaviour

Among the different factors summarized in Fig. 1, matrix toughness represents an important one, and its effect has been investigated in conventional laminates based on synthetic fibres. From the available results, composites based on thermoplastic matrices usually show a better impact resistance compared to thermoset-based ones in terms of smaller delaminated areas (Jang et al. 1991; Schrauwen and Peijs 2002; Vieille et al. 2013; Arikan and Sayman 2015). Matrix plasticization is particularly active in matrix-rich zones because it stimulates local deformation, while fibre-bridging hinders the Mode I opening of plies and retards the growth of interlaminar and intralaminar cracks according to Mode II and Mode III. The overall result is a reduction of their global growth that is accompanied with the development of limited delaminations in size (Vieille et al. 2013). This is the typical damage scenario created by an impact: at first the high out-of-plane shear stresses under the impactor-surface contact point generate matrix cracks that once they reach the lower interface, promote the opening of the interface and trigger the damage following the Mode I. The delamination grows mainly under Mode II.

This trend seems to be confirmed also in natural fibre composites. Bensadoun et al. (2017a) performed a detailed investigation on flax fibre reinforced composites with a view to highlighting the effect of matrix type (epoxy and polypropylene (PP)) on impact properties. The authors used different levels of impact energy up to perforation, and found that for both matrices a traditional power law can be used to foretell the energy needed to achieve perforation, the same that has been validated with glass and carbon fibre composites (Eq. 1) (Caprino and Lopresto 2001):

$$U_{perforation} = K\left(t V_f D_t\right)^{\alpha} \quad (1)$$

where K and α are two material constants to be experimentally determined, t is the thickness in mm, V_f the fibre volume fraction and D_t the diameter of the striker in mm. The calculated value of α was equal to 1.3, exactly the one reported for synthetic composites (Caprino and Lopresto 2001).

The type of matrix played a significant role, as confirmed by the value of parameter K, which was equal to 12.5 and 9 for flax-thermoplastic (PP) and for flax-thermoset (epoxy) composites, respectively, thus suggesting a better behaviour of thermoplastic composites and the possibility to accurately predict the perforation energy once the thickness and the fibre volume fraction are known. During an impact that causes perforation, there is the matrix and fibres breakage, and therefore also their role has to be considered. In the specific case of natural fibre composites, natural fibres are not as strong as glass or carbon ones, therefore their contribution to the energy needed to perforate a specimen is lower than the matrix toughness. The high ductility of PP increased the perforation energy, an increase dependent on the fibre architecture, in the range 20–50% compared to thermoset based composites. In thermoplastic composites, most of the energy is dissipated after the peak force, suggesting a higher resistance experienced by the impactor while penetrating the specimens.

In another study, where an epoxy matrix reinforced with hemp fibres was compared with a polylactic acid (PLA) matrix (Caprino et al. 2015), the power law (Eq. 1) was not found to hold. In this case, a linear trend was detected (Eq. 2):

$$U_{perforation} = a \cdot \left(t V_f D_t \right) + b \tag{2}$$

where the a and b constants were reported to be equal to 0.5 and 13.4, respectively.

Interestingly, when correlating the absorbed energy (U_a) with the impact energy (U), the authors found a very simple linear relationship for both epoxy and PLA matrices (Eq. 3):

$$U_a = a \cdot U - b \tag{3}$$

The values of the material parameters, a and b, were reported as 0.745 and 3.66 for epoxy-based composites, and 0.653 and 2.71 for PLA-based composites, respectively. A similar relationship was obtained in (Sutherland and Guedes Soares 2005) for glass/polyester composites. When $U_a = 0$, the threshold impact energy (U_0) below which no energy is absorbed to produce damage can be estimated. This parameter was equal to 4.9 and 4.1 J for epoxy and PLA-based composites, respectively, higher than corresponding values for glass fibre reinforced composites (Caprino et al. 2011), thus implying that higher energies are potentially required in natural fibre composites for the onset of damage compared to synthetic laminates.

In both studies (Caprino et al. 2015; Bensadoun et al. 2017a), in non-perforating impacts, the thermoset systems exhibited greater damage area compared to thermoplastic composites. A severe through-the-thickness crack with fibre failure and limited delaminations were detected in thermoset and thermoplastic composites (Fig. 2), mainly related to the inherent low strength of the flax fibres and to the high interlaminar fracture toughness (G_{Ic}) of the flax composites fostered by specific energy absorbing mechanisms, such as crack branching and fibre bridging (Bensadoun et al. 2017b). The brittle behaviour of epoxy matrix exacerbated the presence of matrix cracks compared to delaminations, while in thermoplastic composites a more concentrated impact damage zone was detected, and the higher ductility allowed a higher energy absorption and restrained the development of cross-shaped cracks in the composites.

This behaviour represents a significant difference with respect to synthetic fibre composites (Liu and Hughes 2008). In the tensile side there is the origin of cracks oriented perpendicularly to the direction of fibres that later grow through the thickness and result in small delaminations. In conventional laminates, the glass or carbon

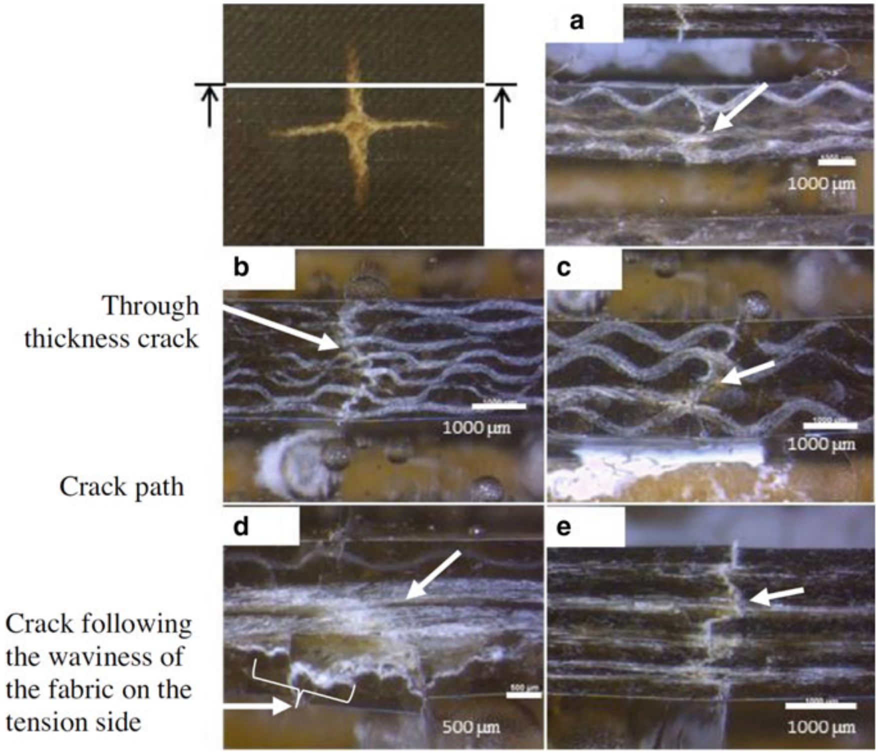

Fig. 2 Through-the-thickness damage in flax/epoxy and flax/PP composites after a 3.1 J impact: **a** plain weave with PP; **b** medium–high twist twill with epoxy; **c** plain weave with epoxy; **d** quasi-UD [0, 90] with epoxy; **e** UD [0, 90] with epoxy (reprinted with permission from Bensadoun et al. (2017a))

fibres are less prone to be fractured and only matrix cracks are generated, which can subsequently start delaminations (Shyr and Pan 2003).

To assess the impact damage tolerance, the authors (Bensadoun et al. 2017a) did not perform the standard compression after impact (CAI) tests, because of extensive buckling of the specimens. In particular, they used a flexure after impact approach, well used in literature for synthetic laminates (Sarasini et al. 2014). The reduced damage in thermoplastic-based composites resulted in a marginal decrease in flexural properties after impact and in any case lower compared to that experienced by thermoset-based composites.

2.2 Effect of Fabric Architecture and Stacking Sequence on Impact Behaviour

Fabric architecture is another key parameter influencing the impact resistance and damage tolerance of composite laminates. Unidirectional laminates (UD) offer superior quasi-static mechanical properties but suffer from poor impact resistance (Shah et al. 2019), and 2D laminates represent a better alternative due to the yarn waviness in the fabric architecture. Unfortunately, their in-plane mechanical properties are significantly lower than UDs because the crimps in the yarns act as stress concentration points. This explains the development of non-crimp fabrics, which brought improvements in delamination resistance (Greve and Pickett 2006). Delamination that can be further hindered by using techniques that introduce reinforcement through-the-thickness, such as stitching, z-pinning, etc. (Mouritz 2001, 2007; Francesconi and Aymerich 2017; Yasaee et al. 2017), at the expense of in-plane mechanical properties.

3D woven composites represent a relatively recent development in structures subjected to impact, because the yarns that link together the different plies hinder the development of large delaminations. Few experimental studies have addressed influence of such architecture on impact and post-impact behaviour of composites (Bibo and Hogg 1996; Chiu et al. 2004; Chen and Hodgkinson 2009; Seltzer et al. 2013; Elias et al. 2017). Seltzer et al. (2013) showed that 3D composites dissipated over twice the energy compared to 2D laminates and that this energy absorption capability was essentially affected by the z-yarns, which introduced energy dissipation mechanisms such as tow splitting, fibre breakage under the tup and creation of a plug by out-of-plane shear.

Contrary to synthetic fibre reinforced composites, the effects of stitching on the mechanical properties of natural fibre composites have not been widely investigated. Rong et al. (2002) analysed the factors affecting in-plane mechanical response and Mode I interlaminar fracture toughness of laminates based on an epoxy matrix reinforced with unidirectional sisal fibres but stitched with Nylon 6,6, Kevlar and sisal threads. The in-plane mechanical properties of sisal laminates were not degraded by the presence of threads because the sisal fibres showed a high damage tolerance, but at the same time the presence of stitches expanded the fibre bridging zone and improved the resistance to delamination.

Ravandi et al. (2016) addressed the effect of stitch areal fraction on the Mode I interlaminar resistance and tensile properties of flax fibre/epoxy laminates stitched with flax yarns and cotton threads. Interestingly, both stitch materials caused a similar decrease in tensile properties of the composites, but the presence of flax yarns improved the interlaminar fracture toughness by at least 10%. The results highlighted the need to optimize the areal fraction of stitch to offset the increases in interlaminar fracture toughness and the decrease in tensile properties. The same authors in (Ravandi et al. 2017) investigated the effect of through-the-thickness natural fibre stitches (twistless flax yarn and twisted cotton thread) on the response to low velocity impacts of epoxy laminates reinforced with woven flax fabrics (Fig. 3). In all woven flax fibre composites, cross-shaped cracks were displayed in both the front and the

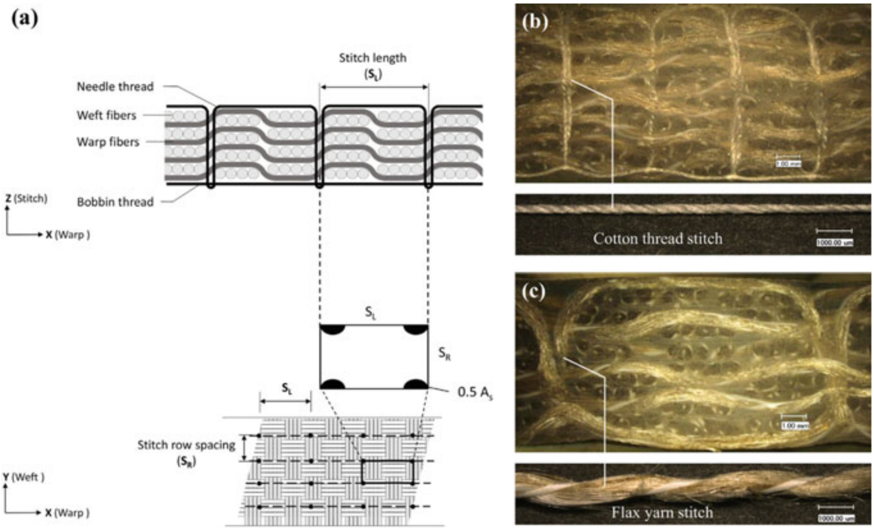

Fig. 3 **a** Schematic view of a stitched preform and definition of stitch parameters; a cross-section of **b** cotton thread, and **c** flax yarn stitched flax fibre composite (reprinted with permission from Ravandi et al. (2017))

rear surfaces of the specimens, and no visible differences were detected between unstitched and stitched specimens, with the exception that crack lengths were longer for the stitched specimens. It is supposed that these cracks were caused by defects originated from the stitches, such as fibre fracture, crimping of in-plane fibres, as well as resin-rich spots. Delamination was only detected in unstitched cross-ply laminates $[0/90]_{4S}$.

Prior to the delamination, in-plane fibre dominated breakages occurred, because of the high interlaminar toughness and relatively poor in-plane strength of the woven flax fibre lamina. For non-perforating impacts, the ratio of the absorbed energy and the kinetic impact energy for the stitched woven composites was about 12–18% higher than that of the unstitched woven laminates, ascribed to the defects generated by stitching. Stitching promoted the development of in-plane cracks because delamination was not the dominant damage mode in woven laminates. The matrix-rich areas located between the stitch loops are characterized by a lower resistance to cracking that requires a lower force during impact to initiate damage. The authors concluded that flax yarn stitching of woven flax laminates did not improve the structural behaviour of the composites in response to low-velocity impacts. In natural fibre composites, fibre failure and matrix cracking have been recognized as the governing mechanisms during a transverse impact, contrary to what happens in standard glass or carbon fibre reinforced composites.

In addition to the fabric architecture, the stacking sequence is another key design parameter for conventional fibre reinforced composites in order to face impact loading, and many investigations are available in this field (Hitchen and Kemp

1995; Hosur et al. 1998; Aktaş et al. 2013; Riccio et al. 2014; Hazzard et al. 2017; Caminero et al. 2017). As a general comment, the damage resistance can be improved by reducing the mismatch in stiffness between neighbouring plies (Caminero et al. 2017), therefore quasi-isotropic laminates are usually characterized by better performance in terms of damage resistance. The use of dispersed configurations with small mismatch angles can result in a superior response in terms of indentation, dissipated energy and residual compressive strength (Sebaey et al. 2013a, b). It is also worth mentioning that the damage modes are influenced by the ply thickness. In thin ply laminates the failure originates in the bottom plies due to bending stresses, which then propagate through the thickness up to the impacted face. Thick laminates are characterized by matrix cracks that emanate from the front side due to the high contact stresses and subsequently travel toward the bottom plies, leading to the typical pine tree damage pattern.

The influence of the stacking sequence on the low-velocity impact and damage tolerance of natural fibre-reinforced composites was investigated by Li et al. (2020). The authors considered three different stacking sequences, i.e. cross-ply $[0/90]_{6s}$, quasi-isotropic $[0/45/90/-45]_{3s}$ and multi-directional ply $[0/30/60/90/-30/-60]_{2s}$. For comparison purposes, a similar glass fibre reinforced composite with the quasi-isotropic configuration was manufactured and tested. The authors highlighted differences with the results usually found for synthetic laminates (Caminero et al. 2017). In particular, the cross-ply composite showed the highest peak load, followed by the quasi-isotropic (by 6%) and multi-directional ply composite (by 7.1%). When compared with the glass fibre composite, synthetic composite displayed the highest peak load indicating the highest impact resistance. This different behaviour resulted in a higher penetration resistance of the glass fibre composite compared to the flax fibre reinforced laminates, among which the best performing laminates were the cross-ply ones. The authors also discussed the damage development and failure mechanisms of the composites. Cross-ply laminates displayed a delamination with the shape of a cross on the impacted surface coupled with two crossed cracks on the rear face, while the other two configurations exhibited a circular indentation on the front surface and an extended delaminated area on the rear surface. Cross-ply laminates absorbed lower energy due to the limited extension of the delaminated area. In cross-ply laminates, once the cracks originate on the back surface, they propagate through the thickness, but they are hampered by the flax fibres oriented at 90° that lie in the neighbouring layer. In this way, a new intra-laminar crack in the fibre direction is triggered and propagate in the adjoining layer. It can be summarized that the interaction between two perpendicular intra-laminar cracks stemming from neighbouring plies is negligible, hence resulting in lower inter-laminar stress and limited delamination. This is not the case in multi-directional or quasi-isotropic laminates, where intra-laminar cracks stemming from adjacent layers display only slightly different angles, and this results in a stronger interaction. This suggests that the through-thickness intra-laminar cracks can readily change their direction and spread through the interface, inducing extensive delamination between the two neighbouring intra-laminar cracks. Synthetic composites with a quasi-isotropic configuration exhibited a different damage pattern, characterized by an overall lower damaged area mainly in the form of a small circular delaminated area. During low velocity impact events, the main drawback of natural fibre composites is related to their inherent lower mechanical properties compared to synthetic fibres, which promote the initiation of transverse cracks at the back surface due to the high bending stresses. These cracks then propagate transversely and through the thickness direction. On the contrary, the glass fibre composites show higher stiffness and strength that result in a lower deflection not able to fracture glass fibres, and delamination is the preferred damage mode. As regards the damage tolerance, the compression after impact behaviour of the cross-ply flax fibre composites outperformed those belonging to the other stacking sequences, namely $[0/30/60/90/-30/-60]_{2s}$ and $[0/45/90/-45]_{3s}$.

In another study, Sy et al. (2018) compared the impact performance of flax fibre reinforced laminates with two different stacking sequences, namely a symmetric unidirectional flax/epoxy laminate $[0]_{8S}$ and a symmetric cross-ply flax/epoxy laminate $[0/90]_{4S}$. In this case, the instrumented drop-weight impact testing machine was replaced with a modified Charpy impact pendulum apparatus. In line with experimental results on synthetic fibre reinforced composites (Ahmad et al. 2015), cross-ply flax/epoxy laminates absorbed higher energy compared to unidirectional flax/epoxy laminates. The latter showed a penetration energy of 10 J, while the former was not completely penetrated even after a 30 J-impact. The two different stacking sequences were characterized by different visible damages on the front and back faces (Figs. 4 and 5). Unidirectional laminates were characterized by a similar damage on the front and rear faces, though it was more pronounced on the rear side. This damage consisted in a critical longitudinal crack starting from the centre of the specimen, a smaller transverse crack running across the longitudinal one decorated with two additional small cracks at its edges (Fig. 4).

On the contrary, impacted and rear faces of cross-ply laminates displayed different damage patterns (Fig. 5). The impacted face damage consisted of a central longitudinal crack combined with a delaminated region with the shape of a butterfly coming from the outermost layer close to the impact location, whereas the back-face damage was made of two cross-shaped matrix cracks. The cross-ply configuration featured the absence of delaminations in the rear face, contrary to what reported in synthetic fibre composites (Namala et al. 2014), due to lower fibre strength compared to synthetic fibres.

In conventional laminates, is the matrix resistance in the rear face that governs the damage development, which is basically in the form of delamination and matrix cracking. In flax/epoxy laminates, the damage produced on the back-face is governed by the fibre properties that are poor, this leading to fibre breakage without significant delaminations. Figures 6 and 7 show an illustrative view of the damage development in through-the-thickness direction for unidirectional and cross-ply flax/epoxy laminates, respectively. In cross-ply laminates the delamination on the impacted side is generated by the interlaminar shear stresses at 0°/90° interfaces, but no delamination is possible in the rear face because the lower strength of flax fibres governs the failure.

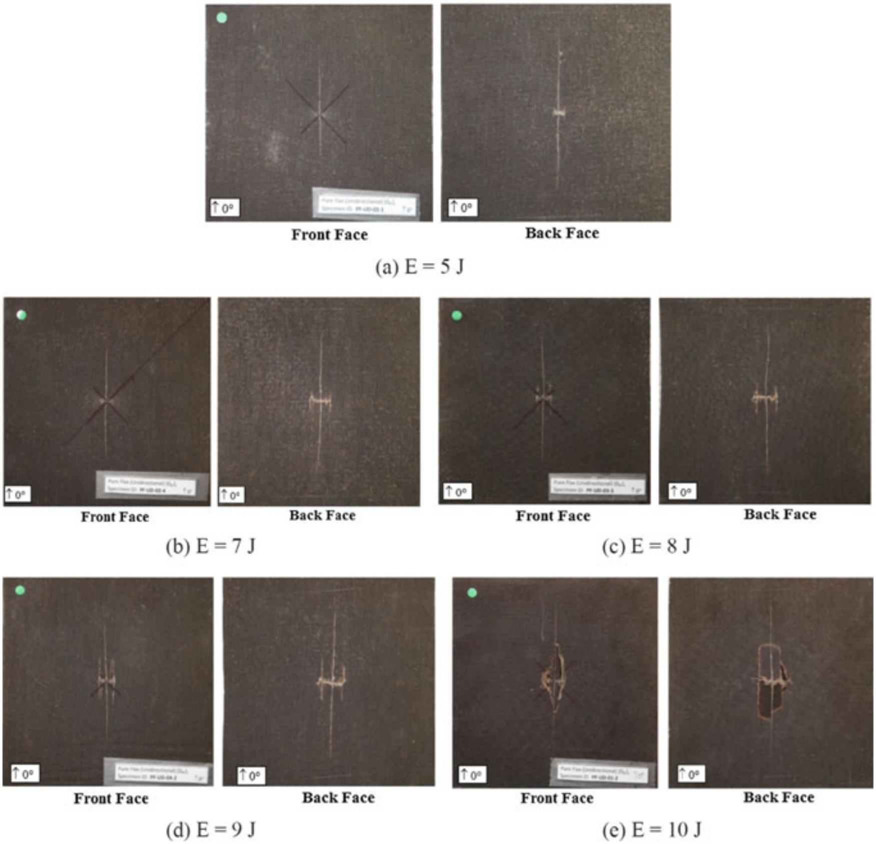

Fig. 4 Front and back face damage on the unidirectional flax/epoxy composite (reprinted with permission from Sy et al. (2018))

Also in quasi-isotropic flax/epoxy composites ($[0/90/45/-45]_{2S}$), Liang et al. (2015) reported fracture mechanisms controlled by the development, in the specimen's rear face, of intra-laminar transverse cracks and a macrocrack due to the failure of the flax fibres (Fig. 8), once again pointing out the prominent role played by the inherent low mechanical properties of natural fibres. After a CAI test, the authors reported a decrease in compression strength of around 30%, similar to that of epoxy composites reinforced with glass fibres (17–34%) (Icten et al. 2013).

Despite the poor mechanical properties of natural fibre composites compared to synthetic counterparts, they still can be used as inexpensive and sustainable energy absorbing structures, even in the ballistic regime (Wambua et al. 2007). Meredith et al. (2012) compared the specific energy absorption (SEA) of jute (plain weave), flax (satin weave) and hemp (chopped strand hemp mat)-based/epoxy composites with that of carbon (plain weave) fibre/epoxy composites. The authors manufactured by vacuum assisted resin transfer moulding cones with an angle of 15°. This geometry

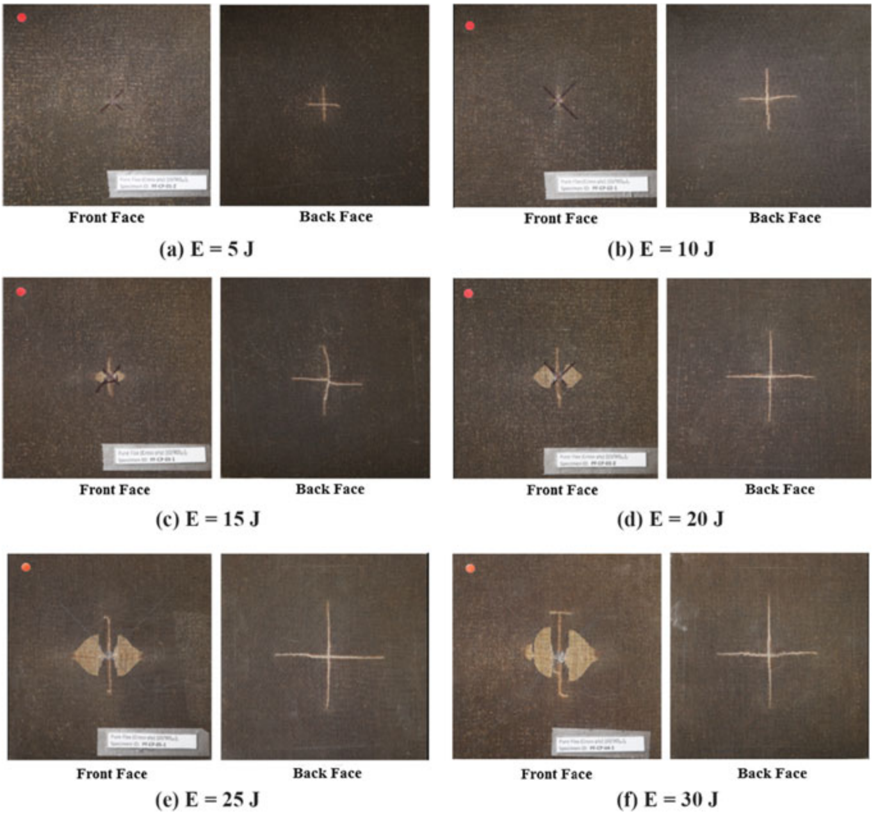

Fig. 5 Front and back face damage on the cross-ply flax/epoxy laminate (reprinted with permission from Sy et al. (2018))

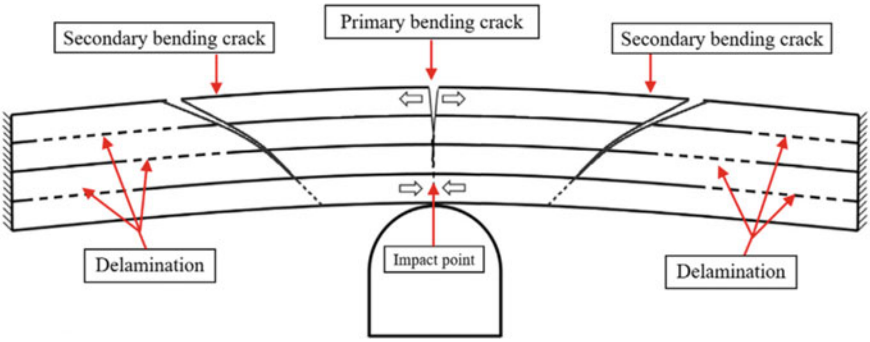

Fig. 6 Schematic of a typical cross section damage on unidirectional flax/epoxy laminate (reprinted with permission from Sy et al. (2018))

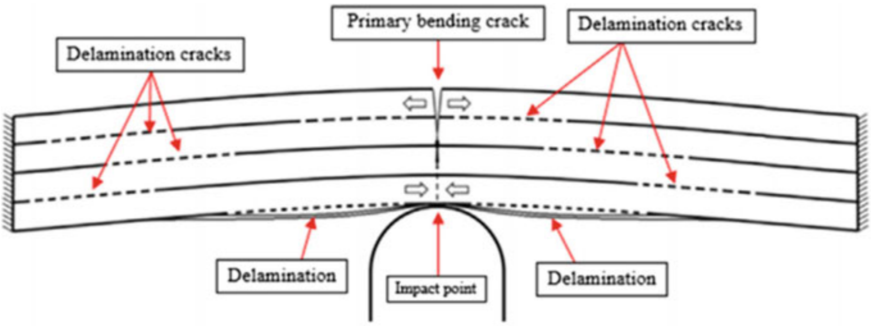

Fig. 7 Schematic of a typical cross section damage on a cross-ply flax/epoxy laminate (reprinted with permission from Sy et al. (2018))

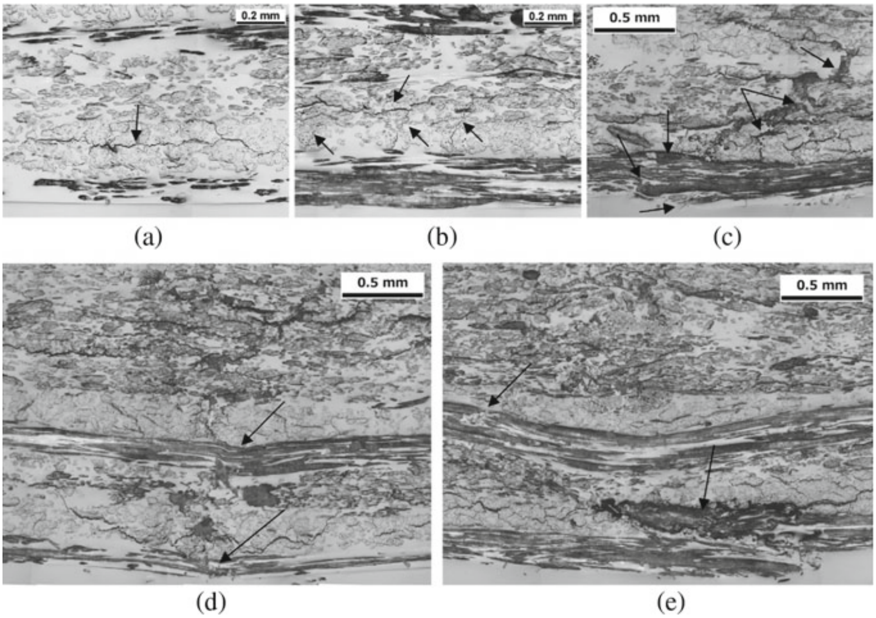

Fig. 8 Damage evolution in quasi-isotropic specimens impacted at different energy levels: **a** 2 J, **b** 4 J, **c** 6 J, **d** 8 J, **e** 10 J. Arrows point to the cracks (reprinted with permission from Liang et al. (2015))

was selected to ensure vertical crush resistance while a wall thickness of 3 mm helped avoiding instability issues during the progressive crushing. Samples were impacted with impact velocities ranging from 3.78 up to 6.70 m/s (Fig. 9). Both woven flax and jute specimens displayed a brittle fracture. In contrast, the non-woven hemp exhibited a progressive collapse with no evidence of terminal longitudinal crack formation upon crush initiation, which resulted in a higher specific energy absorption value (54.3 J/g)

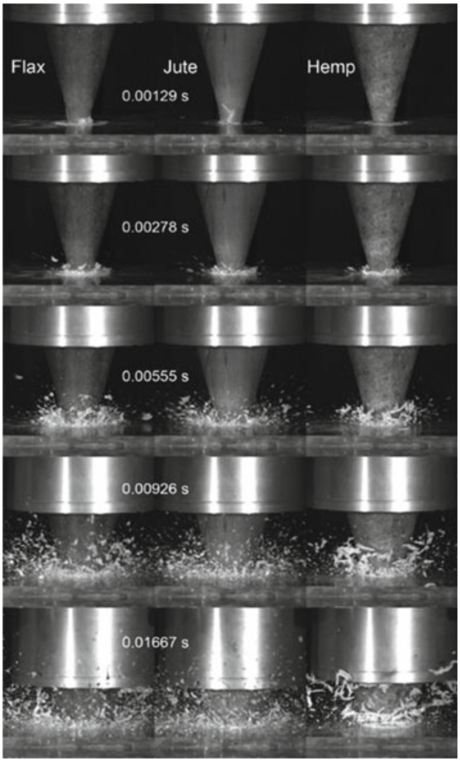

Fig. 9 Comparison of all samples at different time points during impact tests (reprinted with permission from Meredith et al. (2012))

similar to that of carbon fibre (55.7 J/g) composites, whereas woven flax displayed a specific energy absorption of 48.5 J/g and woven jute 32.6 J/g.

2.3 Effect of Fibre Hybridization on Impact Behaviour

Hybridization is a common technique used to improve the properties of composite materials, also when impact performance is taken into consideration (Sevkat et al. 2009; Swolfs et al. 2014, 2018; Bandaru et al. 2016). For natural fibres, hybridization is a significant opportunity to achieve a sufficient mechanical performance for semi-structural applications whereas reducing the carbon footprint of synthetic composite materials (Santulli 2007; Jawaid and Abdul Khalil 2011; Dong 2018; Ravishankar et al. 2019). Glass fibre is the most common synthetic fibre used in combination with natural fibres, and these hybrid composites are usually characterized by improved mechanical properties, reduced property variability and moisture sensitivity (Almeida Júnior et al. 2012; Atiqah et al. 2014; Fiore et al. 2016).

An important parameter, when considering impact behaviour of hybrid composites, is the positioning of the layers (Safri et al. 2018). In particular, two main types of hybridization are possible: (i) the intraply hybridization, when a ply is formed by mixing yarns of two different fibres, (ii) and the interply one, where plies belonging to two different reinforcements are stacked. In the typical configuration, i.e. the interply, the dispersion is completely determined by the lay-up. The positioning of the layers in an interply hybrid composite is of utmost importance, as this affects the flexural stiffness, strength, and the resulting damage mechanisms. Santulli et al. (2005) showed that for E-glass/flax hybrid epoxy composites, the sandwich configuration with glass fibre facesheets and flax fibre core represents the best configuration for improving the impact resistance. Also Ahmed et al. (2007) used a sandwich-like configuration in jute/glass hybrid composites. Jute composites showed higher absorbed energies than jute–glass hybrid laminate, but poorer damage resistance and tolerance that were improved by glass fibre hybridization. Shahzad (2011) investigated the effect of hybridization of hemp fibres with glass fibres on the impact properties of hybrid composites. Two sandwich-like configurations were manufactured: hemp skin and glass core, and glass skin and hemp core. From the results, the replacement of only 11% by volume of hemp fibres with glass fibres resulted in a remarkable increase in residual strength and stiffness of hybrid laminates compared to hemp fibre composites, while the impact damage tolerance of the configuration with glass skins and a hemp core was better than that of composites with hemp facesheets and a glass core, ascribed to the greater mechanical properties of glass fibres. Similar conclusions were achieved by Fragassa et al. (2018), who reported a better impact performance for hybrid composites featuring a flax core sandwiched between basalt fibre facesheets without a non-significant increase in weight.

Glass and basalt fibres (Petrucci et al. 2015; Dhakal et al. 2015; Papa et al. 2018; Ricciardi et al. 2019) have been widely used to increase the impact performance of natural fibre composites, while the combination with carbon fibres has received comparably less attention. This is due to the marked difference in cost and stiffness between natural and carbon fibres (Noorunnisa Khanam et al. 2010; Fiore et al. 2012; Dhakal et al. 2013; Flynn et al. 2016). In this regard, carbon/natural fibre combination has some potential because natural fibres might introduce different modes of damage propagation and energy dissipation, with a view to alleviating the inherent limited toughness of carbon fibre composites. Al-Hajaj et al. (2019) assessed the impact response of an hybrid composite displaying a sandwich structure with woven carbon fibres and flax fibres (i.e., unidirectional and cross-ply) in an epoxy matrix. This particular configuration, as previously mentioned, was chosen because it was supposed that the presence of two carbon/epoxy plies as facesheets was enough to enhance the impact properties compared to neat flax/epoxy laminates. As a general conclusion, both hybrids displayed better resistance to penetration compared to non-hybrid composites, while the composites with the cross-ply flax core performed somewhat better featuring lower absorbed energy, higher penetration energy, smaller crack lengths, smaller indentation depths and smaller damage areas. The penetration energy for the hybrid laminate with the cross-ply flax core was equal to 40 J, higher than neat unidirectional flax fibre (10 J) and neat cross-ply flax fibre (25 J) composites investigated in (Sy et al. 2018). The cross-ply architecture allowed more energy absorption by restraining matrix crack initiation and propagation through the thickness. Fibre bridging at cross-over points between warp and weft yarns might distribute the stress, thus increasing the energy absorption while retaining the structural integrity (Naik et al. 2000).

Hybridization affects the development of damage inside an impacted laminate as well as its damage tolerance and this demands an in-depth analysis of the stacking sequence. Sarasini et al. (2016) addressed this issue by impacting carbon/flax hybrids with two different stacking sequences, i.e., FCF ($[(0_2/90_2)^F/(0_2/90_2)^C/0^C]_S$) and CFC ($[(0_2/90_2)^C/(0_2/90_2)^F/0^F]_S$). The results suggested that the requirements of flexural and impact loading are different in terms of the respective positioning of flax and carbon plies. In particular, the presence of flax fibres as facesheets is useful for impact performance because they restrain crack propagation but not for flexural properties. In terms of residual properties, FCF configuration displayed a better flexural strength and a similar stiffness when compared with laminates made of carbon fibres. The role of outer flax layers in reducing impact damage can be appreciated in Fig. 10, which shows micro-CT scans for the different laminates after a 10 J-impact. While pure carbon (C) fibre reinforced composites displayed the well-known pine tree damage pattern that includes shear cracks, bending cracks and extensive delaminations, pure flax (F) specimens exhibited a damage pattern mostly based on severe transverse matrix cracks, both inter- and intra-yarn in nature. These damage modes were markedly affected by hybridization. In CFC laminates, both carbon facesheets and flax core appear to be damaged in the form of delaminations and bending cracks mainly found in the carbon facesheets. The flax core showed delaminations not only

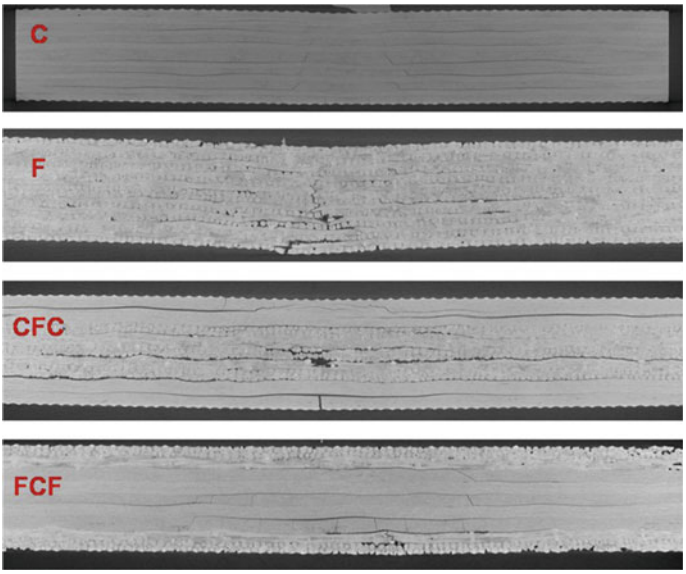

Fig. 10 Micro-CT scans for the different carbon/flax configurations after a 10 J-impact (reprinted with permission from Sarasini et al. (2016))

at the 0°/90° interfaces but also at the flax/carbon interfaces. In FCF composites, only the carbon core showed the typical pine tree damage pattern, while the flax skins were not extensively damaged apart from bending cracks located in the 0° lower plies. The compliant flax skins restrained the propagation of cracks originated in the carbon core, which resulted in a higher damage tolerance after impact.

Compared to the most common interply hybrid composites, intraply hybrid composites have received limited attention. Despite contrasting results in literature (Pegoretti et al. 2004; Wang et al. 2008), the intraply hybrid composites should offer improved resistance to crack propagation during an impact event. Zhang et al. (2018) investigated how different hybrid configurations made of interlayer and intralayer warp-knitted fabrics with carbon and glass fibres can affect the low-velocity impact performance. The intralayer hybrid showed smaller peak load and higher damage area at the same hybrid ratio and level of impact energy compared to the intralayer configuration, thus pointing out that a better impact resistance can be obtained by using an intralayer hybridization. This strategy can be exploited for supporting the introduction of natural fibres in at least semi-structural applications (Audibert et al. 2018). Recently Sarasini et al. (2019) proposed a new hybrid intraply woven fabric based on flax and basalt fibres to reinforce epoxy and polypropylene matrices. Laminates with the thermoset matrix compared positively with results available in literature for pure flax laminates. Bensadoun et al. (2017a) addressed the impact behaviour of laminates as a function of flax fibre architectures and matrix types. By taking into account the differences in impact test parameters and similarities in terms of laminate thickness (2 mm) and a total fibre volume fraction (0.40), flax/basalt intraply hybrid (15 J) displayed a perforation energy (15 J) about two times higher than that of pure flax laminates, whose values are in the range 5.7–7 J (Bensadoun et al. 2017a). The basalt hybridization was able to counteract the poor transverse strength of flax fibres, hindering the growth of diamond-shaped cracks. Also in this study the superior performance of thermoplastic matrix was confirmed, because the plastic deformation is able to restrain further propagation of the cross-shaped cracks.

3 Conclusions and Future Perspectives

The objective of this chapter was to provide a detailed review on the potential use of natural fibre composites in energy-absorbing applications. The impact response of composites is a topic of significant complexity, as it is governed by multiple factors that are often interrelated. The significant amount of literature available on the subject for synthetic composites do not reflect the same interest in natural fibre composites. In addition, natural fibres display peculiarities in their microstructure and mechanical properties compared to synthetic fibres that often deserve specific tests and the results found for synthetic laminates cannot be easily transferred to natural fibre composites. The similarities include the major role played by the matrix toughness on the energy needed to achieve perforation, on the damage resistance and tolerance, which seem largely not affected by the particular fibre architecture in natural fibre composites. For impacts performed at energy levels far from perforation, delamination is not recognized as frequent and detrimental as for synthetic composites. This different behaviour can be ascribed to the high interlaminar fracture toughness of natural fibre composites in combination with some extra energy absorbing mechanisms and the low transverse strength of natural fibres, which tend to localize the damage and trigger the initiation and development of significant cross-like macro-cracks. While the effects of fabric architecture and stacking sequence have been adequately addressed, there are other parameters that have received limited attention. In real cases, impact events occur in a random way and can involve different levels of impact energy, impactor masses, geometries, and velocities. Such studies are definitely limited in number for natural fibre composites (Wang et al. 2016; Habibi et al. 2018), and to address this issue extensive experimental campaigns are needed, and possibly the development of methods to simulate them (Sy et al. 2019) in order to limit the number of tests.

Another important area is the assessment of the residual strength of impacted specimens and its correlation with the visible dent depth, which might make easier not only the inspection but also the decision on whether to repair the composite part or to replace it, especially important if the applications of natural fibre composites will abandon the cosmetic field. Damage can be detected by referring to different thresholds that depend on the type of inspection, which are usually classified as follows: BVID (barely visible impact damage), Minor VID (visible impact damage) and Large VID (visible impact damage). These thresholds are connected with the dent depth left by the impactor on the specimen: 0.3–0.5 mm for BVID, 2 mm for Minor VID and 50 mm diameter preformation for Large VID (Talreja and Phan 2019).

A major concern in the field of natural fibre composites is their resistance to hygrothermal ageing, and limited studies are available trying to correlate the effects of temperature and/or moisture on their impact resistance. In this regard the role of fibre hybridization is of paramount importance. Živković et al. (2017) reported how basalt fibre hybridization with flax fibres in a vinylester matrix enhanced the impact behaviour compared to single composites, especially for conditioned samples (35 ppt salt water at 80 °C for 912 h without protection). Fiore et al. (2017) aged under salt fog conditions basalt/jute epoxy composites with two lay-ups (i.e., intercalated and sandwich-like). For the quasi-static properties, the sandwich-like structure enhanced the durability of specimens subjected to salt fog aging conditions, while alternating layers of jute and basalt allowed to reduce the strength loss after accelerated aging. The damage tolerance assessment of aged composites is lacking in the available literature and this gap needs to be bridged.

Another unexploited area but full of potential with a view to increasing the impact resistance of natural fibre composites, deals with the combination of 3D composites with thermoplastic matrices, in order to merge the through-the-thickness reinforcement offered by 3D woven fabric and the toughness of thermoplastic matrices.

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