Enhancement of impact toughness and damage behaviour of natural fibre composites and their hybrids through novel improvement techniques: a review

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Abstract:

The importance of natural fibres over synthetic fibres have gained significant attention in the research area, due to their higher specific strength, stiffness, lightweight and inexpensive. Natural fibre composites used in various applications are often susceptible to moisture absorption and various critical loadings scenarios during their service life such as low-velocity impact damages which is a concern for structural and non-structural applications. For enhancing the toughness of natural fibres hybridisation with synthetic fibres is essential. This paper examines the essential information critically from the published literature influencing the morphological characteristics, fracture toughness, damage tolerance and impact resistance of natural fibre reinforced and their hybrid composites. Following this, this review paper critically analyses the novel improvement techniques suitable for natural fibre composites for damage tolerance and impact resistance behaviours.

Keywords: Natural fibre; Synthetic fibre; Hybrid composites; Morphological characteristics; Damage tolerance; Fracture toughness; Impact resistance.

List of Nomenclatures

PA	-	Propionic Anhydride
MA	-	Methacrylic Anhydride
PP	-	PolyPropylene
LDPE	-	Low-Density Polyethylene
HDPE	-	High-Density Polyethylene
PS	-	Polystyrene
PLA	-	Polylactic Acid
PU	-	Polyuretane
HMPP	-	High Modulus Polypropylene
UHMWPE	-	Ultra-High-Molecular-Weight Polyethylene
rHDPE	-	recycled High-Density Polyethylene
vHDPE	-	virgin High-Density Polyethylene
EG	-	Ethylene Glycol
ASTM	-	American Society of Testing Materials
DCB	-	Double Cantilever beam method
MBT	-	Modified Beam theory
MCC	-	Modified Compliance Calibration
CC	-	Compliance Calibration
ENF	-	End Notch Flexure test
ELS	-	End- Load Split test
NFRPCs	-	Natural Fibre Reinforced Polymer Composites
NFRC	-	Natural Fibre Reinforced Composites
NFPC	-	Natural Fibre Polymer Composite
HFRE	-	Hybrid Fibre Reinforcement
DD	-	Dyneema Dyneema
CC	-	Carbon Carbon
CD	-	Carbon Dyneema
CGC	-	Carbon Glass Carbon
CG	-	Carbon Glass

CGGC - Carbon Glass Glass Carbon

SCP - Sandwich Composite Panels

FVE - Flax Vinyl Ester

FBVE$_s$ - Flax Basalt Vinyl Ester Stitched

FBVE$_u$ - Flax Basalt Vinyl Ester unstitched

WCC - Woven Commingled Composites

KCC - Knitted Commingled Composites

WC - Woven composites

UD - Unidirectional

CFRP - Carbon Fibre Reinforced Polymer Laminate

ALE - Arbitrary Lagrangian Element

CEL - Coupled Eulerian-Lagrangian

STF - Shear Thickening Fluid

CNT - Carbon Nanotubes

CS - Cold Spraying Technique

FML - Fibre Metal Laminates

MSO - Methacrylated Soybean Oil

1: Introduction

Throughout the years, the research field has been involved in strengthening the fibre-reinforced composites. Several researchers suggested various techniques to make these materials as damage resistant and less brittle [1]. Strategies were identified to increase the matrix toughness in the fibre-reinforced composites, which has a substantial influence on the matrix- dominated composite properties [2]. However, to find a new toughness mechanism for the biological composites, there has been an increase in structural properties relation that is resilient to failure [3].

Fibres are categorized as synthetic and natural. Among the synthetic fibre, Carbon and glass are gaining more attention over the last two decades [4]. Synthetic fibre-reinforced composites provide high strength, stiffness, and extensively used in aerospace and automobile applications [5]. Synthetic fibres are consumer-friendly to waterproofing, stretching, and stain-resistant [6]. Whereas natural fibres grow rapidly in research interest for their sustainability in resources, biodegradability, low cost, and less in weight [5]. The replacement of artificial fibres with natural fibres is considerably increased in engineering applications [7]. This replacement helps to preserve the environment by reducing utilisation of artificial fibres concerning weight reduction, low material cost, and renewability [8]. Although natural fibres are weak in mechanical properties, they have good absorption capability in impact tests. Therefore, for advanced applications, high strength is required with higher mechanical properties. In such cases, the hybridisation is the possible solution for increasing the mechanical properties [9]. According to Dhakal [7], natural fibres are used for high-performance applications by hybridising with other composites. Though, few limitations in non- structural applications are high moisture content, lower strength, and stiffness properties [10]. As a solution, hybridisation with chemically modified fibres is recommended [11]. Because by adding another form of hydrophobic fibre to the hydrophilic fibre composites, it provides an efficient method and also enhances the resistance to moisture and reduces the degradation of the properties of NFPC [9].

Over the last decades, researchers are focussing on the impact and damage tolerance of composite structures, which has been revealed several design problems [7,11–14]. Such problems are complicated since the results are not solely dependent on the material parameters but also test configuration and sample configuration [15]. Also, the damage occurs in various forms, such as fibre/matrix debonding, surface micro buckling, matrix cracking, and fibre breakage [16]. Therefore to produce a practical design for a structure, it is vital to know the impact response, impact energy, and impact strength absorbed by the material before its failure [17].

The Impact test is an approach to evaluate the fracture toughness and notch sensitivity in composite fabric. The critical feature that affects the impact characteristics of a composite is the fracture toughness of the resin system [15]. The fracture toughness, such as Mode I and Mode II, governs the impact response of the compact material [18]. Therefore, under impact loading, the composite material can absorb a large amount of energy in the full range of damage modes [19]. The other common failure in the composite laminates is delamination. It occurs by interfaces of the laminate layers. The most critical feature in the laminates is caused by matrix cracking, shear cracks, and bending cracks due to the dynamic loadings such as low and high-velocity impact tests [20]. Generally, there are four types of impact test with respect to velocity: low velocity (0-11 m/sec), high velocity (> 11 m/sec), ballistic impact (> 500 m/sec) and hypervelocity impact test (> 2000 m/sec). Besides, the impact events are grouped into two notable cases: low-velocity impact test with a huge mass (for instance, dropped object) and a small mass for the high-velocity impact test (for instance, runway debris). A detailed summary of types of impact tests concerning velocity shown in Table 1. Also, the stress in the composites can initiates crack between the layers and causes delamination. Besides, interlaminar performance is characterised by weakness under both shear and tensile stresses. Therefore, such stresses affect the whole performance of material discontinuities and geometry [21]. Moreover, the different aspects damaging of composite materials are illustrated in Fig. 1.

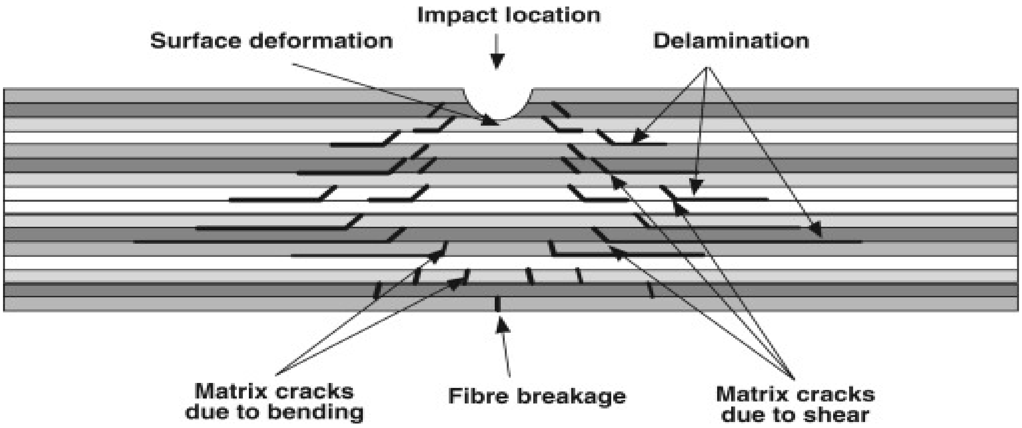


Fig. 1. Schematic description of impact location and damage mode of laminates' composite [22]

Therefore, identifying the various damage modes and their progression towards impact is vital. The primary purpose of this paper is to review the information from the various literature articles published in impact mechanics and damage tolerance of hybrid composites. Initially, the morphological characteristics, structure and surface treatment are discussed with the challenges and comparison between natural and synthetic fibres. Following this, a detailed review on natural fibres related explicitly to materials, geometry and the damage tolerance and impact resistance of natural fibre composites and their hybrids under different loading conditions, exposed to different natural environments. Finally, a novel improvement technique on improving the damage tolerance and impact resistance for usage in various engineering applications with future perspectives are presented.

Table 1

Types of Impact tests with a velocity range [15]

Velocity range	Test equipment	Applications
Low velocity 0-11 m/sec	Drop hammer Pneumatic accelerator	Dropped items Vehicle impact crash
High velocity >11 m/sec	Compresses air gun Gas dun	Free falling bombs Fragments owing to the explosion
Ballistic impact >500 m/sec	Compressed air gun Gas dun	Military
Hypervelocity impact >2000 m/sec	Powder gun	Military, space vehicles

2: Natural fibres, their structures and morphological characteristics as reinforcements in composites

The characteristics of the natural fibres depend on various factors including soil conditions, climate, fibre extradition techniques and fibre processing. However, the knowledge of fibre properties is vital, which are particularly useful for the mechanical and impact performance of composites. It is, therefore, the knowledge must be permanently integrated into a processing technique that allows high volume production and processing of natural fibre composites.

Naturals fibres are neither synthetic nor human-made; they are plants and animal sources such as oil palm, flaxseed, and jute [23]. It can be applied as a reinforcement or fillers [24]. A block diagram with a classification of natural fibres depicted in Fig. 2. Vegetable/ plant fibres such as jute, kenaf, flax, ramie, sisal, and hemp are commonly used in textile and other industrial applications [10]. Also, all vegetable fibres consist mainly of cellulose and lignin. Cellulose contains a linear chain of anhydroglucose units with semi-crystalline polysaccharide hydrophilic components that contain alcoholic hydroxyl groups [25]. Because hydroxyl is present in all-natural fibres and other polar groups in their constituents, natural fibres exhibit higher moisture absorption [26,27]. However, few disadvantages are lower impact strength, weak moisture resistance, less durability, and poor matrix fibre adhesion [28]. Irrespective of its shortcomings, natural fibres are rapidly increasing in research interest due to their sustainability in resources [29]. Also, natural fibres have a low environmental impact, low cost, low density and low machine wear.

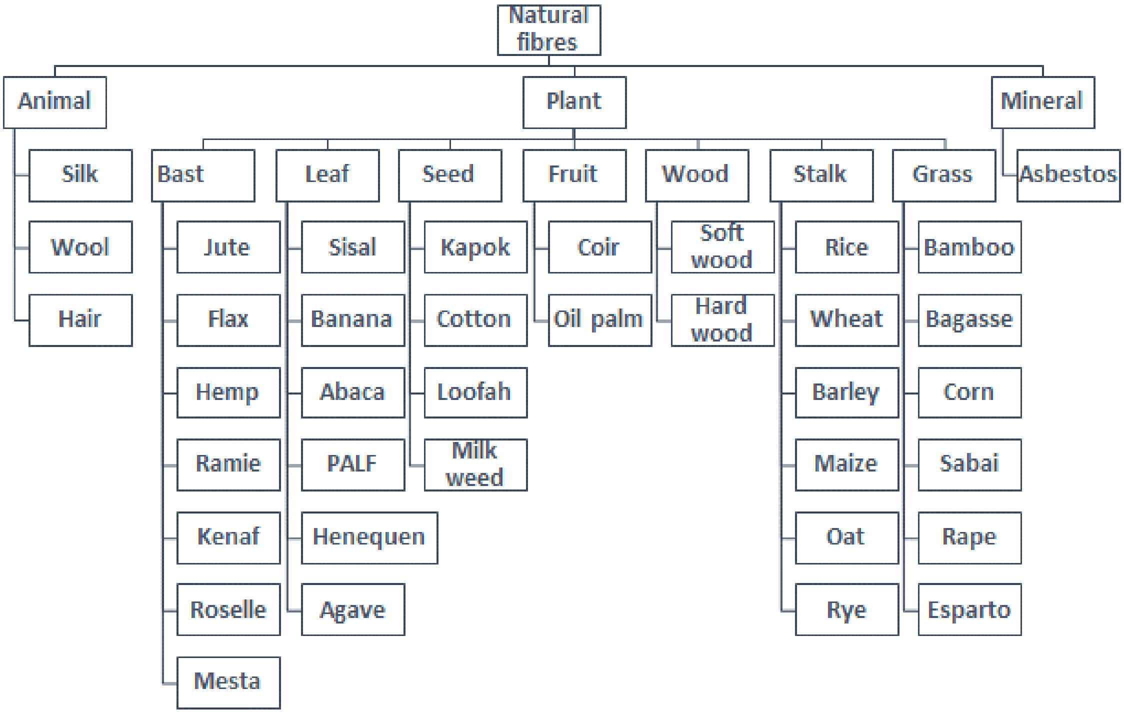


Fig. 2. Natural fibres and their classifications [30]

2.1 Structure of bast fibres

Bast fibres are obtained from the Phloem of the fibrous plant [31]. A flax plant stem, for instance, displayed in Fig. 3. consists of Bark, Fibre bundle and group of xylem, shives and woody core. The topmost layer is bark/skin, which is mainly responsible for the plant protection against moisture evaporation and unexpected changes in the temperature. The middle layer is where the fibres are located in the Phloem, appears as a bundle. The bottom layer contains xylem, shives, and woody core has a significant role in transferring water and nutrients from the centre of the fibre. As in Fig. 4. Single fibres are connected by middle lamella, a substance composed of pectin, which functions as a glue [32]. The single fibres consist of two significant walls such as primary and secondary wall; these walls surround a channel filled with protein and pectin called the lumen [32]. The primary wall possesses a rigid framework of cellulose microfibrils within a network of hemicellulose, pectin compounds and glycoprotein. Whereas the secondary wall is largest with three-layered structure (S1, S2, S3). All these layers consist of cellulose, hemicellulose, and lignin. The middle layer (S2) forms the maximum bulk of the fibre (about 70-80% of mass) [33,34]. Thus, the highest attribute of a single fibre is controlled by the S2 layer, and it determines the overall mechanical strength [35].

The chemical composition of the plant fibres are cellulose, hemicellulose, lignin, pectin, and wax, as shown in Fig. 5. All-plant fibres have distinctive properties because they are cultivated in a natural environment with water, sunlight, soil, and air. However, the peculiarity is that all they have the same constituents with different compositions [29]. Table 2 shows the compositions and percentage of plant fibres, amongst which banana, coir, flax, hemp, jute, pineapple, and sisal are the primary resources for industrial materials [30].

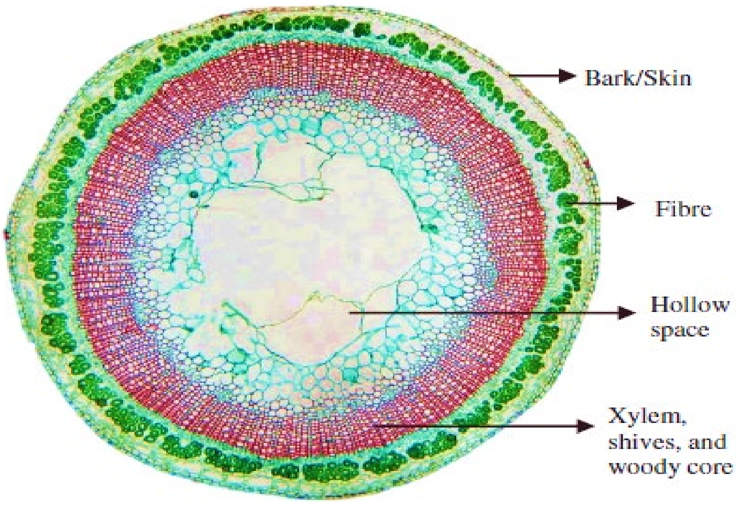


Fig. 3. Cross-section of Flax stem [31]

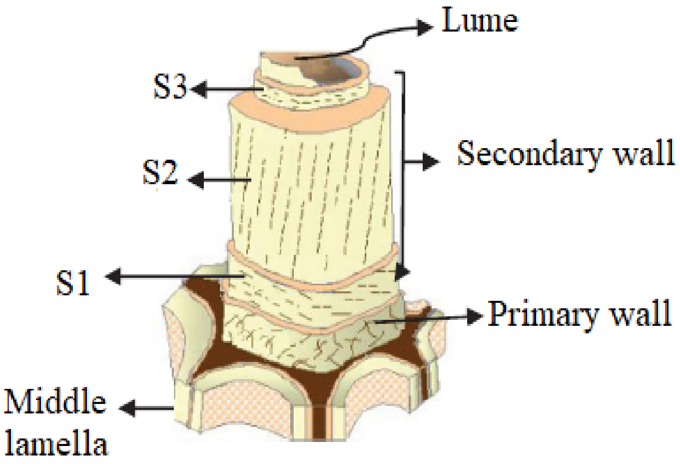


Fig. 4. Single element fibre source [31]

Table 2

Chemical composition of plant fibres [36,37]

Name of fibre	Cellulose	Hemicelluloses	Lignin	Pectin	Waxes
Abaca	62.5	21	12	0.8	3
Banana	62.5	12.5	7.5	4	-
Bamboo	34.5	20.5	26	-	-
Coir	46	0.3	45	4	-
Cotton	89	4	0.75	6	0.6
Flax	70.5	16.5	2.5	0.9	-
Hemp	81	20	4	0.9	0.8
Jute	67	16	9	0.2	0.5
Kenaf	53.5	21	17	2	-
Pineapple	80.5	17.5	8.3	4	-
Ramie	72	14	0.8	1.95	-
Sisal	60	11.5	8	1.2	-

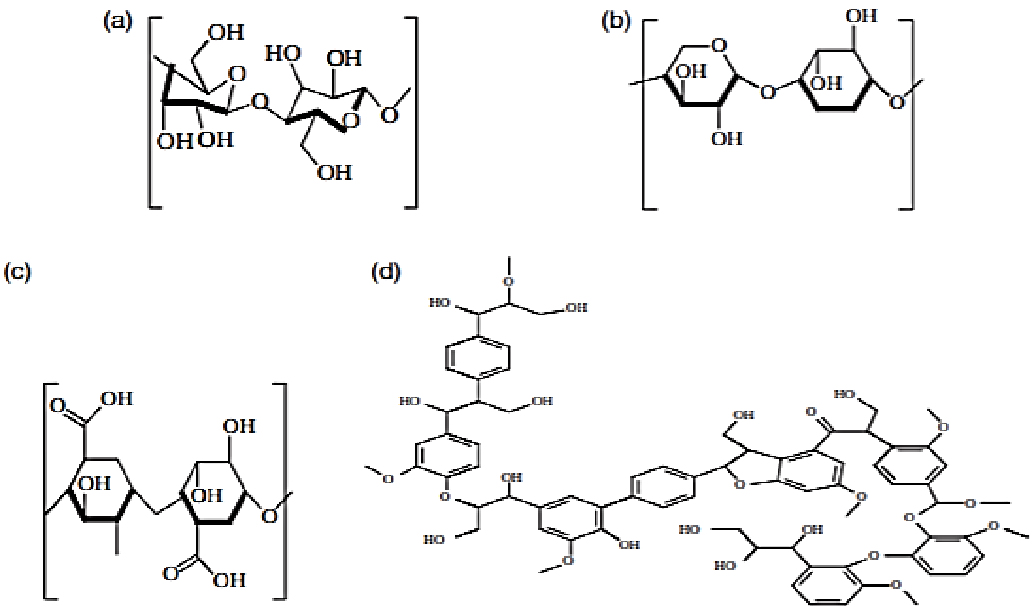


Fig. 5. Structural representations of (a) cellulose, (b) hemicellulose, (c) pectin, and (d) lignin [38].

Cellulose is the essential components of natural fibres for increasing mechanical properties. The higher the cellulose content, the greater will be the mechanical characteristics [39]. However, other compositions, such as hemicellulose and pectin, increase the moist absorption [40]. Besides, pectin has a drawback that affects natural fibres' structural and morphological properties [41].

2.2 Morphological characteristics of natural fibre

Bast fibres have a typical cell structure, which consists of a lumen and a thick wall [42]. Generally, the cross-sectional shapes of all-natural single fibres consist of a rounded polygonal, outer shape [43]. Perhaps, a non-uniform and irregular shape differs from one fibre to another. Typically, they are between 10 μm and 50 μm in diameter and 8 – 14 μm in length [44] shown in Table 3. The plant maturity determines the lumen diameter ranging from 0.5 μm to 10 μm, which can be short, cylindrical or elliptical [44,45]. According to Yamada [46], the fibre density, mechanical and dimensional characteristics will have a more substantial influence on the single fibre shape and lumen diameter. Baley [47] reported that lumen size increases with increasing diameter for flax fibre bundles. Increasing the lumen size can increase the porosity of the fibre and reduce the fibre density. Schäfer and Hornermeier [44] observed that lumen cross-section was around 70-130 μm^2, which is 13%-16% of the overall fibre cross-section area. Many researchers observed that tensile strength and rigidity are mainly dependent on their diameter [48–50]. As the diameter of flax [48,49], hemp [51] and jute [52–54] increases, (See

Fig. 6) the tensile strength decreases because of Griffith's theory [55], which is used in the fracture mechanics of brittle materials. Moreover, in the case of diameter dependent on tensile strength, the author Fan M [51] reported that there are several studies related to the Ultimate tensile stress concerning the fibre diameter, but the questions related to the tensile elastic modulus dependent on diameter remains unresolved. Indeed, several authors have experimentally observed a substantial decline in modulus associated with increasing fibre diameter in the case of elemental flax [56], hemp [57] and jute [53,54].

Three techniques were commonly used for the characterisation of fibre diameter [58] they are SEM., Laser diffraction and light microscopy. Hu W [48] stated that in actual practice SEM observations of the fracture surface do not allow the surface area of the lumen to be determined. The fractography image (Fig. 7.) gives a piece of clear evidence that the geometry changes in the fibre portion, where the lumen is almost disappeared in the rupture field [42]. However, under microscopic observation (Fig. 8.) use of polarised light can identify the dislocations of the lumen in hemp fibres [59,60].

Table 3

Morphological characteristics of natural fibres [61–63]

Plant	Hemp fibre	Flax fibre	Jute fibre	Ramie fibre	Sisal Fibre	Cotton Fibre
Length mm	5-60	2-40	2-3	40-150	2-7	20-70
Diameter µm	20-40	20-23	16	30	20	20-30
L/D	100-2000	100-2000	160	40-150	140	1250

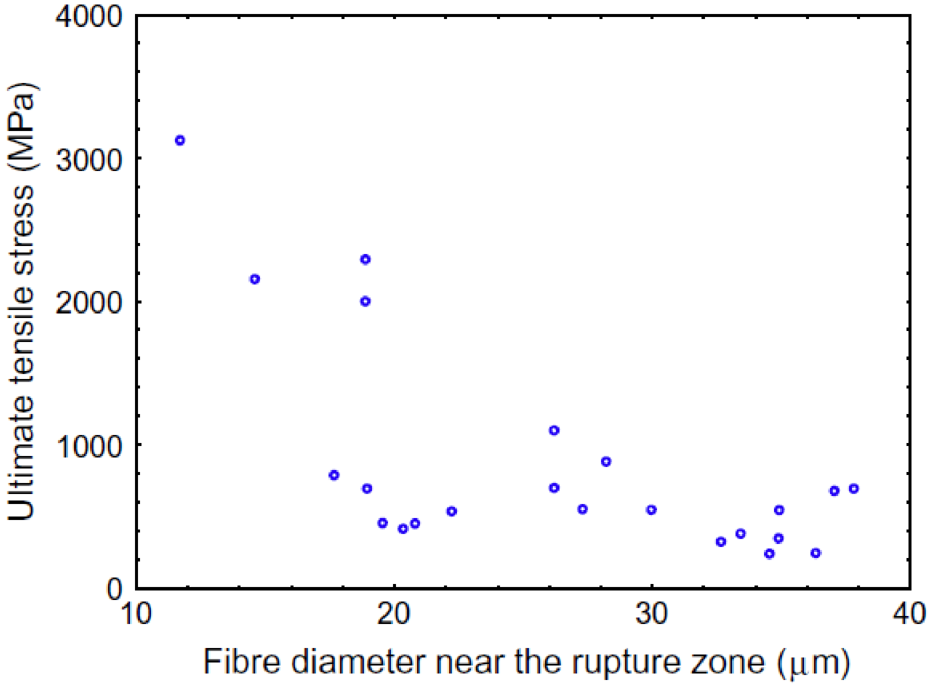


Fig. 6. Ultimate tensile strength vs Fibre diameter of hemp fibre [42]

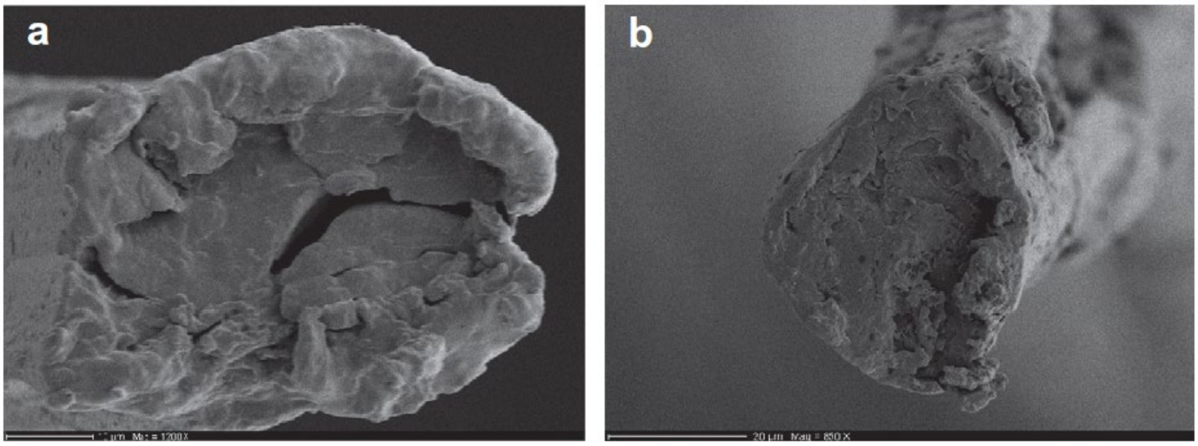


Fig. 7. SEM images of hemp fibre cross-section (a) virgin fibre and (b) failed fibre [42]

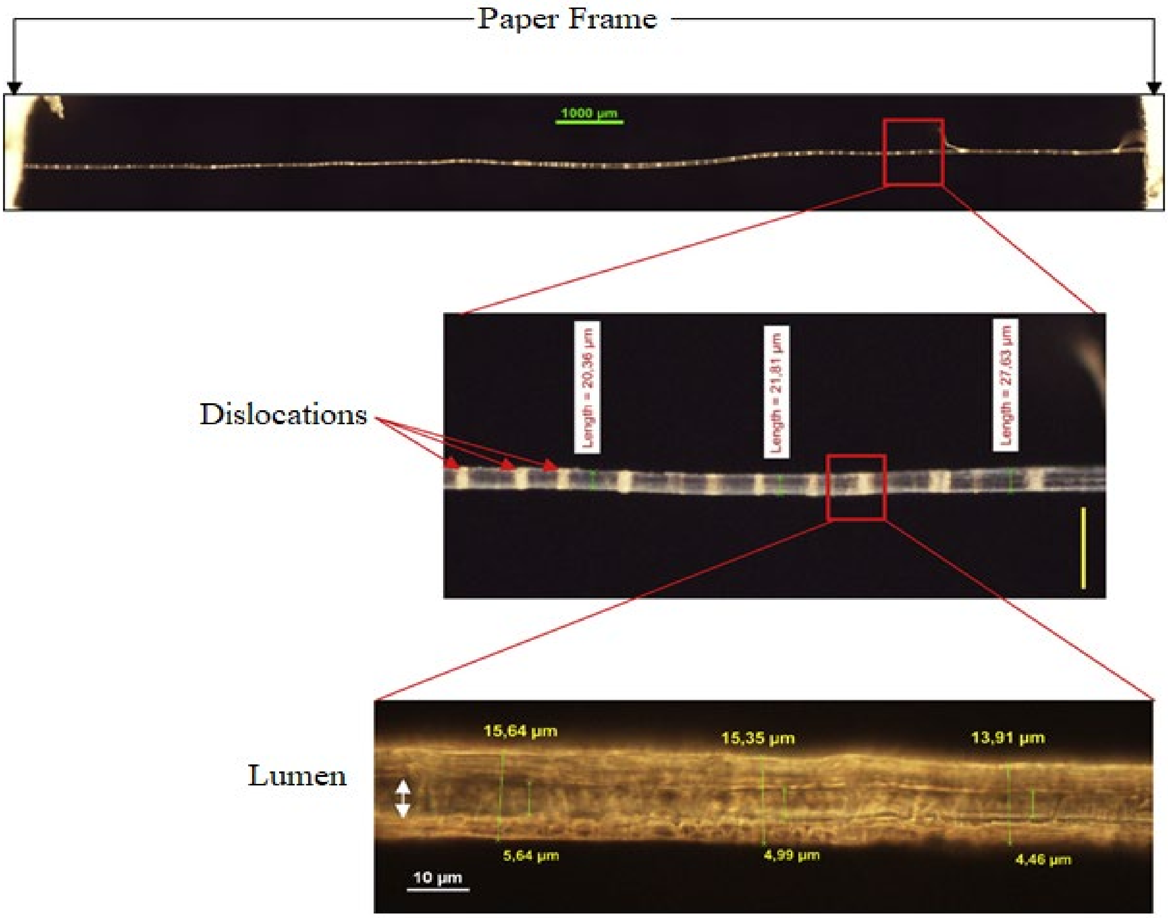


Fig. 8. Microscopic observation of polarised light images of a hemp fibre [42]

2.3 Surface treatment of Natural fibres and their effects

Natural fibres are prone to water absorption due to rich cellulose and hydrophilic in nature [64]. Many researchers [65,66] observed that moisture content would cause significant degradation of natural fibre-reinforced composites concerning a crack in brittle matrices. To overcome the moisture content and for better bond strength, it is, therefore, primarily to apply some treatment to the surface, either physically or chemically. Trujillo et al. [67] observed that treatment of fibres partially removes the lignin, hemicellulose, waxes, and other impurities. However, there is a lower chance of moisture entering into the material by reducing the empty content of composites and improving the adhesion between the fibres and matrix. Currently, there are various techniques for natural fibre surface modifications, which helps in improving their properties. These techniques include treatment with Alkali [68], Acetylation [69,70], silane [71], Ozone [72,73], plasma [74–77], mercerisation [78–80], benzoylation [78] and grafting [81,82].

Sánchez et al. [83] presented findings on mercerisation, ozone, and plasma treatment and checked the properties of bamboo fibres- reinforced composites. They observed a substantial decline in the average surface rough value (about 30% for plasma treatment fibres and 80% for Mercerization and ozone treatment). Even Barra et al. [84] also obtained similar results. Valadez-Gonzalez et al. [85], observed that alkaline treatment has two major effects on the fibre: (1) it improves the properties, and surface roughness (2) improves cellulose treatment on fibre surfaces. Jähn et al. [86], reported that alkaline treatment has a lasting influence on the flax fibres, particularly fibre strength and stiffness. A study from van de Weyenberg et al. [87], noticed that a 30 % rise in tensile properties for flax fibre – epoxy composites with the elimination of pectin. Negawo et al. [88], studied the surface morphology and structural composition of alkali (NaOH) treated 2.5%, 5%, and 7.5% Ensete stem fibres reinforced unsaturated polyester composites. The alkali increases the roughness of fibre surface and also provides higher mechanical performance in all concentrations. Fiore et al. [89], examined the pretreated solution of 6% NaOH on kenaf- epoxy composites at room temperature for two different periods, 48h and 144h, respectively. The untreated fibres (Fig. 9.a) indicates the impurities on the surface, at 48h in the alkali removes the impurities (Fig. 9.b), after 144h heavy damage of fibre was found since the soaking time has increased. (Fig. 9.c)

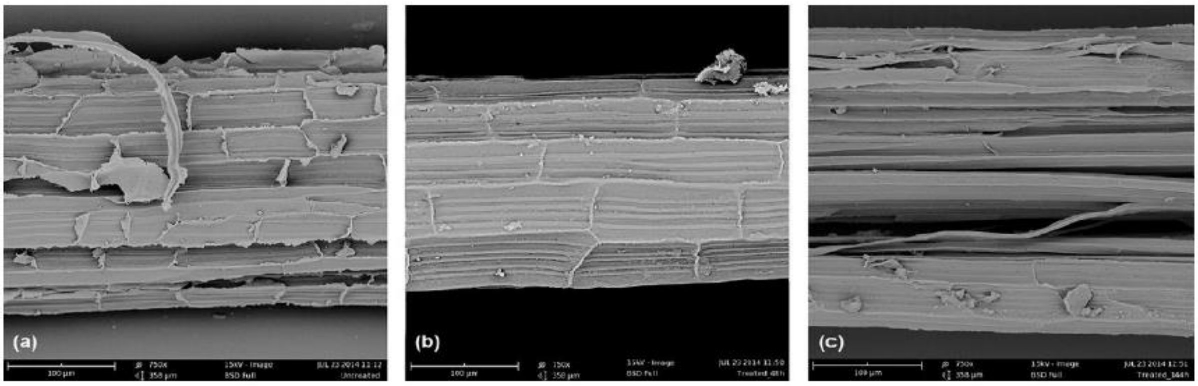


Fig. 9. S.E.M. images of Kenaf fibres (a) untreated Kenaf fibres; (b) after 48 h in NaOH solution, and (c) after 144 h [89].

Therefore, alkaline treatment or mercerisation is often utilised chemical treatments for natural fibres along with reinforced thermoplastics and thermoset composites [90]. Many researchers recommended that silane treatment is commercially available inorganic compounds and suitable for the surface fibre modifications [91–93]. Gang [94], discovered that silane treatment has excellent composite-bonding interfaces with increased tensile and impact properties. Liu et al. [95] observed the corn stalk fibre-reinforced composites treated with silane reaction could drastically reduce the water absorption and porosity, which makes poor friction performance in the silane treated corn stalk fibre composites. Therefore, for enhancing friction performance, acetylation has been documented to strengthen the adhesion of fibre-matrix [90]. Yao et al. [96] investigated the acetylation of flax/PP composites. They reported an 18% rise in acetylation had a considerable rise in the flexural and tensile strengths. Hughes et al. [97], observed that flax fibre showed better properties after coating with propionic anhydride (PA) and methacrylic. Similar results also obtained by Cantero et al. [98], by coating a vinyl group of methacrylic anhydride (MA) and hydrocarbons. A detailed summary of the chemical treatments on the function of natural fibre is illustrated in Table 4.

Table 4

Summary of the surface treatment of natural fibres and their effects

Treatment	Effect	References
Alkali	Improve fibre- matrix adhesion and helps in improving the properties	[99]
Acetylation	Better progress in tensile and flexural strengths	[38]
Benzoylation	Superior in surface modifications and better improvement in the hydrophobicity	[38]
Enzyme	Helps to decrease the lignin content from 35% to 24%	[38]
Grafting	Better enhancement in properties and ultraviolent protection properties	[78]
Mercerisation	Helps to reduce the moisture content and gives a better interface and significant result in flexural and tensile strengths.	[83]
Methacrylate	Better improvement in tensile and flexural strengths	[98]
Ozone	Increases the strength properties of polyethylene and pulp composites	[100]
Plasma	Superior in surface modifications and better improvement in the hydrophobicity	[101]
Silane	Better improvement in the hydrophobicity and mechanical properties	[102]
Sodium chloride	Significant increase in the properties such as tensile strength, youngs modulus and extension of the break	[103]

2.4 Benefits and challenges of natural fibres over synthetic fibres

In response to the growing demand for environmentally sustainable materials and to minimise the costs of conventional (e.g. steel, glass, carbon, and aramid), researchers have started to concentrate on NFRPCs. Moreover, natural fibres show many advantages in mechanical properties that facilitate the substitution of synthetic fibres in polymer composite. J. Agarwal et al. [104], stated that natural fibre polymer matrix composite is manufactured either using thermoplastic or thermosetting reinforced polymers. Synthetic thermoplastics with NFRPC such as (polyethylene, polypropylene, and polyamide) along with natural fibres forms partially biodegradable composites that are equal to the used percentage of natural fibres [105]. The fabrication of these composites can withstand temperatures of 170 ºC during long processing time and around 210 - 220 ºC during the short time. If the temperature is not under control, discolouration, weak interfacial adhesion, and cellulose damage may occur. The properties of composites depend on the synergetic effect within the matrix, and these effects are attained by adding suitable coupling agents [106]. Therefore, the coupling agents are modifiers for the matrix, which helps the poor wettability and weak the interfacial bonding. Maleic anhydride grafted polypropylene is the compatibilizer used for polypropylene matrix [107]. Polypropylene possesses low density, less expense, long durability, and recyclability due to which it is used as thermoplastic matric in automobile industries [104]. One the other hand, epoxy, polyester, and vinyl ester are thermosets used in manufacturing the natural fibre reinforced composites. Primarily epoxy was used in high-performance applications to minimise health hazards. Epoxy-reinforced composites were used in crash applications to minimise the damage of the passengers by placing the intended part between bumper and side rail to absorb maximum dynamic energy [107]

Currently, aluminium and steel are now replaced by NFPC, because of lightweight and release less carbon dioxide footprint without compromising passenger safety. Though automobile industries recommended epoxy for its high performance and durability, few disadvantages are associated, for instance, high cost and long curing time. Because of these drawbacks, the vinyl ester is used among the automobile industries. Vinyl ester is made of epoxy and unsaturated carboxylic acid. They have excellent properties, thermal degradation and can be easily treated in a short time. [21]. However, several factors could affect the properties of NFRP composites, such as incompatibility between natural hydrophilic fibres and hydrophobic thermoplastic matrix that makes it undesirable [108]. In such cases, processing techniques and parameters should be selected precisely, as it has significant influence in the composite properties and interfacial characteristics [27]. There are several types of polymers are available in the market, and the common thermoplastic and thermoset polymers are outlined in Tables 5 and 6.

Table 5

Properties of the thermoplastic polymer used in natural fibre composite [105,109,110]

Property	P.P.*	LDPE	HDPE	PS	PLA
Density (g/cm^3)	0.899–0.920	0.910–0.925	0.94–0.96	1.04–1.06	1.252
Water absorption-24 h (%)	0.01–0.02	<0.015	0.01–0.2	0.03–0.10	N/A
T$_g$ (C)	10 to 23	125	133 to 100	N/A	55
T$_m$ (C)	160–176	105–116	120–140	110–135^0	165
Heat deflection Temp (C)	50–63	32–50	43–60	Max. 220	55
Coefficient of thermal expansion (mm/mm/C10^5)	6.8–13.5	10	12–13	6–8	N/A
Tensile strength (MPa)	26–41.4	40–78	14.5–38	25–69	59
Elastic modulus (GPa)	0.95–1.77	0.055–0.38	0.4–1.5	4–5	3.5
Elongation (%)	15–700	90–800	2.0–130	1–2.5	7%
Izod impact strength (J/m)	21.4–267	>854	26.7–1068	1.1	26

Table 6

Properties of the thermoset polymer used in natural fibre composites [105,110]

Property	Polyester resin	Vinylester resin	Epoxy
Density (g/cm^3)	1.2-1.5	1.2-1.4	1.1-1.4
Water absorption-24 h @ 20C	0.1-0.3	0.1	0.1-0.4
Tensile strength (MPa)	40-90	69-83	35-100
Elastic modulus (GPa)	2-4.5	3.1-3.8	3-6
Elongation (%)	2	4-7	1-6
Izod impact strength (J/m)	0.15-3.2	2.5	0.3
Compressive strength (MPa)	90-250	100	100-200
Cure shrinkage (%)	4-8	n/a	1-2

2.5 Can natural fibre replace synthetic fibres?

Natural fibres are gaining more attention in composite industries, owing to its specific properties and environmental benefits [111]. Despite poor strength in properties, they partially replace synthetic fibres in structural applications for its enhanced characteristics such as specific modulus and elongation at break (Fig. 10.) [112]. The density of natural fibres shown in Table 7 is less than that of glass fibre, resulting in composites called lightweight fibres. Consequently, much scope has been increased for the industrial consumption of NFPC in different manufacturing sectors [113]. Flax, jute, kenaf, hemp and sisal are used for the manufacture of lightweight composites [114–116]. In general, fibres with higher length – to – diameter ratio and cellulose content have been achieved superior mechanical properties [117]. Nevertheless, the structural properties of NFRP composites depend mainly on fibre material, fibre distribution and fibre-matrix adhesion [27]. Ku et al. [118] stated that chemical modification would improve the interfacial matrix-fibre bonding, which results in the significance of tensile properties of natural composites. Moreover, Ahmad et al. [119] also observed that an increase in fibre loading would have a significant increase in tensile properties. Natural fibres provide outstanding thermal and acoustic insulation with beneficial properties over traditional fibres for their low density and cellular structure [120].

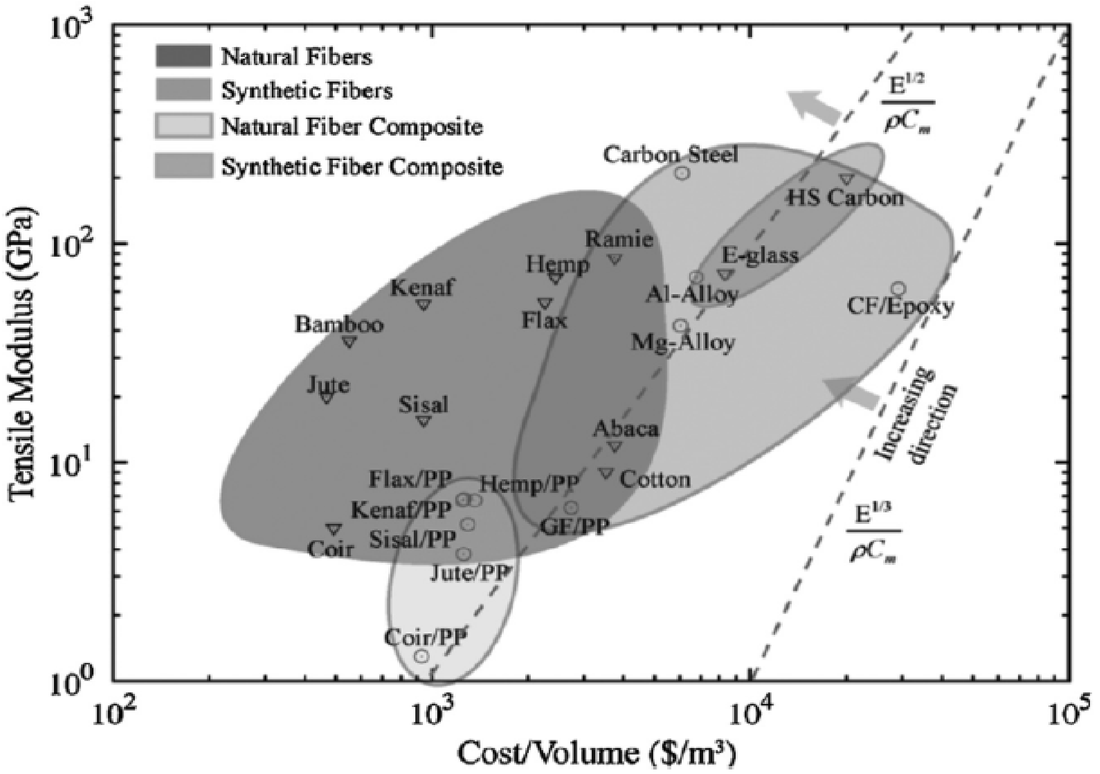


Fig. 10. Tensile modulus versus cost/volume comparison for synthetic and natural fibres [112].

Table 7

Mechanical properties of natural, synthetic and aramid fibres

Fibre	Density (g/cm^3)	Elongation (%)	Tensile strength (MPa)	Elastic modulus (GPa)	References
Cotton	1.5–1.6	7.0–8.0	400	5.5–12.6	[105,112,121,122]
Jute	1.3	1.5–1.8	393–773	26.5	[105,112,121,122]
Flax	1.5	2.7–3.2	500–1500	27.6	[105,112,121,122]
Hemp	1.47	2–4	690	70	[105,112,121,122]
Kenaf	1.45	1.6	930	53	[105,112,121,122]
Ramie	N/A	3.6–3.8	400–938	61.4–128	[105,112,121,122]
Sisal	1.5	2.0–2.5	511–635	9.4–22	[105,112,121,122]
Coir	1.2	30	593	4.0–6.0	[105,112,121,122]
E-glass	2.58	4.8	3100-3800	80-81	[123]
Carbon T-800	1.80	2.2	5516	234.5	[124]
Kevlar 149	1.47	1.5	3400	179	[125,126]
Continuous Basalt fibre	2.66	3.56	2016	61.9	[127]

3. Important parameters for measuring the impact toughness of the composite

In general, composite laminates under fracture toughness usually expressed in critical energy rates (Gc). Based on the loading type, the energy releases the rate at which delaminations actually begins and therefore extends drastically [128]. Fracture toughness Mode I and II governs the impact response of the compact material [18]. As a result, under impact loading, the composite material can absorb a significant amount of energy in the full range of damage modes. [19]. Mode I loading is amongst the most current studies for the delamination fracture type. Many authors [129–131] recommended that ASTM D5528-13 (American Society of Testing Materials), Double Cantilever Beam (DCB) is the standard specimen to measure Mode I fracture toughness G_{ic} of the fibre-reinforced composites. The three different methods to calculate the Mode I fracture toughness G_{ic} are Modified Beam theory (MBT), Compliance Calibration (CC), and Modified Compliance Calibration (MCC). Among them, the author Caprino, [132] observed that Eq. (2) was better than Eq. (1) and Eq. (3). Similarly, Prasad et al. [133] perceived that the latter method Eq. (2) had yielded 80 % of the repeated values of G_{ic} trialled during ASTM round-robin testing. In recent studies, Almansour et al. [134], stated that MBT gives the lowest possible value of the samples related to initiation and propagation. As shown in Fig. 11, the flax fibre of wet samples, G_{ic}, is comparatively lower than the dry sample. Further, the high absorption of moisture and the water content in the flax fibre makes a weak fibre matrix interface and creates surface degradation and reduces fracture strength. Even, Bensadoun et al. [135] found that flax composites showed higher interlaminar fracture strength. Whereas Zulkifli et al.[136], found that multiple fibre layers have a significant impact on the woven silk/epoxy composites toughness.

Delamination often propagates in Mode II under the low-velocity impact [21]. The fracture characterization of such materials can be easily predicted, and propagation is often dominated by a fracture in Mode II [137]. However, it has a few disadvantages, such as unstable crack and predicaments in monitoring crack propagation [137]. Currently, there has been little progress to Mode II [138]. It follows specific delamination methods to test the G_{IIC} fracture toughness. Many researchers [21,138] recommended the End- Notch Flexure test (ENF) and End- Load Split test (ELS) as the methods to determine the toughness of the reinforced composite. Due to simplicity, Ferreira De Moura [137] stated that the ENF is the standard method for analysing the Mode II fracture toughness G_{IIC}. As per Russell et al. [139], the ENF equation from the simple theory method is given below in Eq. (7).

$$G_{ic} = \frac{P^2a^2}{bc} \tag{1}$$

$$G_{ic} = \frac{3P\delta}{2ba} \tag{2}$$

$$G_{ic} = \frac{9PI\delta^4}{4ba^4} \tag{3}$$

$$G = \frac{P^2}{2b}\frac{dc}{da} \tag{4}$$

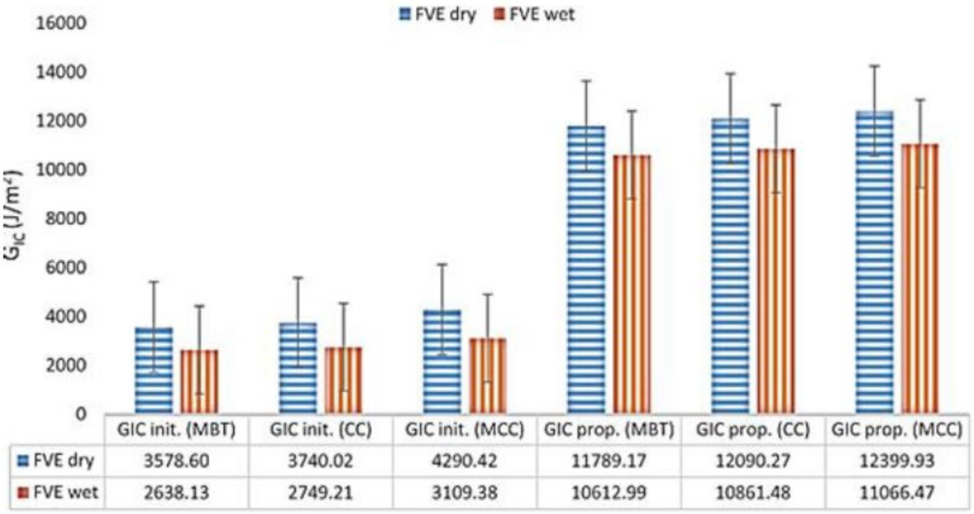
$$C = \frac{\delta}{P} \tag{5}$$

$$C = \frac{2l^3 + 3a^2}{8E_1bh^3} \tag{6}$$

The interlaminar fracture toughness can be obtained by Eqn (4) and (6)

$$G_{IIc} = \frac{9P\delta\, a^2}{2b(3a^3 + 2l^3)} \tag{7}$$

Where P is the applied load, b is the delamination length (mm), b is the sample width (mm), c is the compliance ratio of the load point displacement to the load applied, and δ is the load point displacement (mm)



	GIC init. (MBT)	GIC init. (CC)	GIC init. (MCC)	GIC prop. (MBT)	GIC prop. (CC)	GIC prop. (MCC)
FVE dry	3578.60	3740.02	4290.42	11789.17	12090.27	12399.93
FVE wet	2638.13	2749.21	3109.38	10612.99	10861.48	11066.47

Fig. 11. Initiation and propagation G.I.C. graph against wet and dry flax fibre [134]

Moreover, many researchers are focussing on hybridisation by embedding two or more fibres into one polymer matrix [140]. Composites such as synthetic- synthetic, natural – synthetic, and natural – natural fibre types achieve a possible combination of hybrid composites [141]. Over the past few years, hybrid composites have gained interest in many research areas [142], and it has a vast application in engineering science due to low cost, strength to weight ratio, and ease of manufacturing [143]. However, in contrast to single fibre composites, the hybrid composites possess better fracture toughness, increased fatigue life, and lower notch sensitivity[144]. Furthermore, it has a unique characteristic in mechanical properties and has various advantages over traditional materials, are utilised in many structural or engineering applications[145,146].

3.1 Hybrid Mode I fracture toughness

Saidane et al. [147], performed a study on the toughness of hybrid flax and glass-reinforced composites, by using a DCB. They noticed that the hybridisation of flax fibres together with glass fibres during crack propagation G_{ic} exhibits a superior interlaminar fracture toughness. However, they found that difference between glass fibres and hybrid fibre reinforcement (HFRE) was closer to the initiation value of G_{ic}. Moreover, in the propagation stage, the HFRE showed a more considerable value of G_{ic} in contrast to other composites. Zhang et al. [148] also published the same findings that flax fibre had higher interlaminar shear and fracture toughness compared to glass fibre composites. Jung et al. [149] in their study observed that increased glass fibre could reduce hybrid composite fracture toughness. Also, they found that the glass layer tends to withstand/absorb load when the fracture occurred with some carbon fibres.

Almansour et al. [134] experimented the fracture toughness of hybridised flax/basalt reinforced vinyl ester composites. They found that water immersed flax vinyl ester had better performance in the fracture toughness in contrast to dry composites. Besides water immersed flax/basalt stitched improved 15 to 17% than dry specimens. Moreover, the author Almansour et al. [134], also noticed that basalt hybridisation provides a better shield to the swelled glass fibres. Hybridising basalt as an external layer on flax composites enhances interlaminar fracture toughness. A detailed summary of the toughness of hybrid composites shown in Table 8.

Table 8

Outline of Mode I fracture toughness of hybrid composites.

Hybrid composite materials	Specimens	Matrix type & manufacturing of composites	Mode 1 – fracture toughness Critical strain energy release rate, $G_{I.C.}$ (J/m^2)		Critical stress intensity factor K_{ic} (MPa.m½)	References
			Initiation	Propagation		
Flax fibre (10 piles)	(D.C.B.)	SR 1500 Epoxy resin & Compression Molding Technique	1073.2 ±66.4	2340	---	[147]
Flax/Glass hybrid (6,6 piles)			944.8 ± 76.7	2080		
Glass fibre (20 piles)			922.9 ± 30.2	1040		
Flax – Vinyl ester	(D.C.B.)	Vinyl ester & Vacuum infusion	Dry – 3578.0 Wet - 238.13	Dry – 11789.1 Wet – 10612.1	---	[134]
Flax/ Basalt vinyl ester unstitched			Dry – 4236.6 Wet –2821.7	Dry – 9556.55 Wet – 10187.6		
Flax/ Basalt vinyl ester stiched			Dry – 2669.9 Wet –2806.1	Dry – 4429.36 Wet – 5475.13		
DD	D.C.B.	Epoxy resin &	993.0 ±162.3	1294.3	----	[150]
CC			388.1	659.2		
CD			729.1 ± 63.3	1027.3		

HH – Interwoven carbon & Dyneema		Vacuum-assisted resin transfer moulding	69.1 ± 62.4	1011.4		
CGC. (10 Piles) CGC (20 Piles) CGC (30 Piles) CGC (40 Piles)	Strain fracture toughness test	epoxy resin & Vacuum-assisted resin transfer moulding	---	---	36.457 32.588 30.013 30.246	[149]

3.2 Hybrid Mode II fracture toughness

Almansour et al.[151], investigated Mode II fracture toughness on hybridised flax/basalt reinforced vinyl ester composites. They observed basalt fibre hybridised composites performed better interlaminar toughness for initiation and propagation compared to flax vinyl ester dry specimens. Audibert et al. [152], studied the damage mechanism and mechanical characterisation of new flax – kevlar hybridisation composite. They observed that flax and Kevlar fibre shows a stable non – linear anisotropic behaviour. Besides, flax fibre provides better resistance to compressive load compared to kevlar fibre. Kevlar fibre performed better fracture toughness owing to the plastic strain in the non – linearity behaviour. Whereas in flax fibres, the damaged modulus was hugely affected due to the fibre pull-out. Similarly, Salman et al. [153] reported that irregularities in mechanical behaviour caused more damage in the kevlar and kenaf fibres. However, the indication shows that damage occurred in the epoxy resin, particularly in areas where the crack had developed and initiated (Fig. 12). Also, Zhou et al. [154], highlighted that presence of Dyneema fibre improves the fracture toughness of the hybrid composites and delamination growth. A detailed description of Mode II fracture toughness of hybrid reinforced composites shown in Table 9

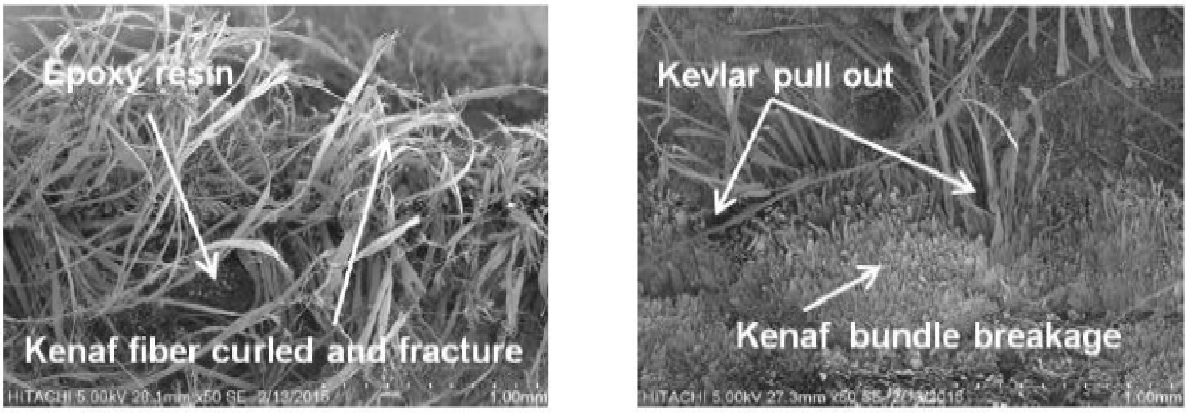


Fig. 12. S.E.M. image of the Kenaf/Kevlar caused by fibre pull-out and fibre breakage [153]

Table 9

Outline of Mode II fracture toughness of hybrid composites.

Hybrid composite materials	Specimens	Matrix type & manufacturing of composites	Mode 1I – fracture toughness Critical strain energy release rate, $G_{.I.C.}$ (J/m^2)		Critical stress intensity factor K_{ic} (MPa.m½)	References
			Initiation SBT	Propagation S.B.T.		
FVE dry FVE wet FBVEs dry FBVEs wet FBVEu dry FBVEu wet	3ENF	Vinyl ester & Vacuum-assisted transfer moulding	239.85 328.32 615.29 685.75 369.54	3648.82 5039.4 2779.02 3577.82 4498.74	--	[151]
Flax -epoxy Kevlar-epoxy Kevlar/flax -epoxy	3ENF	Epoxy resin & Vacuum infusion process	---	---	2.5 % 1.6%} Tensile Elongation fail	[152]
Cc CD HH DD	3ENF	Epoxy resin &Vacuum assisted resin transfer moulding (VARTM)	---	2128 3499 3192 4458	--	[154]

4. Factors affecting the impact response of natural fibre composites

The critical factor that affects the impact characteristics are material characteristics, geometrical factors, impactor shape/size and environmental service conditions (Fig. 13.).

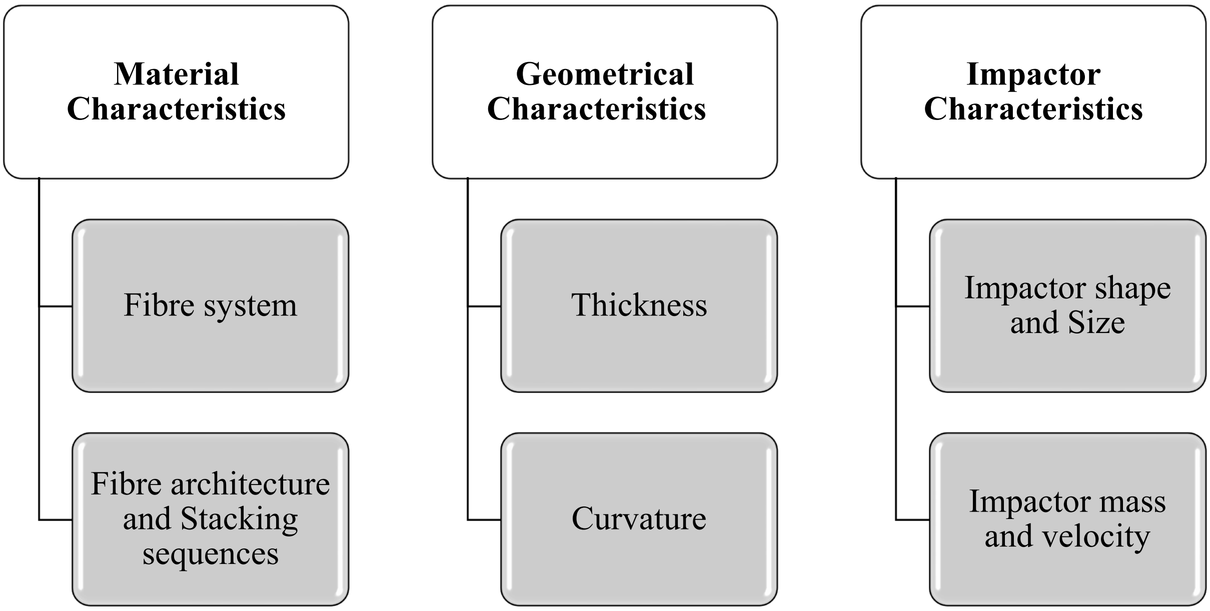


Fig. 13. Factors affecting the impact response of natural fibre composites modified from Andrew et al. [15]

4.1 Material characteristics

4.1.1 Natural fibre system

The nature of the fibre system relies on the fibre reinforcement, which provides crucial load-bearing elements offering structure with greater strength and rigidity [155]. The impact properties are influenced by the size used, their distribution, fibre volume percentage and volume portion orientation within the matrix [156]. Bar et al. [157], studied the mechanical properties of flax-polypropylene reinforced composites and observed that impact strength decreases with higher flax content. Similar results obtained by George et al.[158], for the flax fibre-reinforced epoxy composites, where the tensile strength and modulus increase with fibre loading. On top that by adding 35 Vol.-% fibre, the tensile strength was noted to increase to 20% compared to the neat epoxy system, but the strain to failure rate was decreased due to fibre content. Nevertheless, three reinforcements can be accomplished by aligning the fibre-filled composites, orientation and position of fibres within a matrix (i) longitudinally – could obtain high tensile strength and low compression strength by the buckling of fibres (ii) transversely – could achieve low tensile strength and (iii) short fibre randomly oriented – challenging to predict the mechanical properties, due to the weak distribution of load [159–161]. Therefore, Shesan et al. [156] stated that high-performance NFPCs could generally be obtained primarily by using materials with high fibre content, and hence fibre loading on NPCs is essential. Moreover, the additional fibre content of the composites inevitably leads to increased tensile properties [162,163].

4.1.2 Natural fibre architecture and stacking sequences

Restricting plant fibre composites to the large volume and structural elements is due to the restricted information about how these materials fail under bi-directional impact loading [164]. In general, the plant fibre architecture on impact behaviour is uncertain, and still, it is unstated. Moreover, alternative considerations on the existence of 3-D fibres on impact properties have always opposed the usage of plant fibre mats for application purposes [165]. In addition, inserting loose plant fibres into the composite by applying adequate pressure will allow a higher amount of reinforcement in the composite. Santulli C et al. [164] investigated the falling weight impact properties on different types of Hemp/ epoxy composites; one is unidirectional (0/90°) hemp fibres and hemp mat (M laminate). They observed the unidirectional laminate gives the better performance in the Quasi-static results. Though, they noticed that mat fibre laminates absorbed significant energy during the impact event. However, in the hysteresis cycle, hemp mat fibre may have an improved impact resistance followed by a barely noticeable drop in load and considerable rebound energy released at quasi-constant rate after penetration. Similar observation also made by Dhakal et al. [166], observed that fibre volume fraction of hemp/polyethene composite had a better energy absorption in the unidirectional laminate since the impact resistance was highly dependent on the hemp layers. According to Bensadoun et al., unidirectional fibre architecture has a significant impact on the damage resistance, whereas, in energy absorption, the results were not significant [167]. However, interwoven fibre architecture is also helpful in minimising the escalation of fibre damage and increasing laminate tolerance [168]. Awais et al. [168] fabricated hemp, jute and flax fibres with woven composites (W.C.), woven commingled composites (WCC) and knitted commingled composites (KCC) (Fig. 14.) and observed that WC laminates (refer Fig. 15.) performed excellent impact resistance than the others. Besides, woven composite laminates (W.C.) showed the lowest energy absorption. Lebaupin et al. [169] investigated the impact resistance and stacking sequence of the flax/polyamide composites. Unidirectional $(0^{o})_{8}$, cross-ply

$(0^{\circ}/90^{\circ})_{2s}$, sandwich-like $(02^{\circ}/90_2^{\circ})_s$ and quasi-isotropic $(45^{\circ}/0^{\circ} -45^{\circ}/90^{\circ})_s$ were fabricated. They observed that cross-ply and quasi-isotropic samples had less damage visible in front sides, refer Fig. 16 (b) and (d). Also, they noticed that unidirectional and sandwich layups absorbed higher impact energy than other configurations— moreover, Muralidhar et al.[170] stated that the increase of flax/epoxy composite preformed layers leads to the decline of the absorbed energy. Simultaneously, the layup angle seems to affect the impact properties of the preformed composite. Even, Kannan et al. [171] also noticed that the energy absorption of the flax/ polypropylene composite was highly dependent on the composite's fibre orientation.

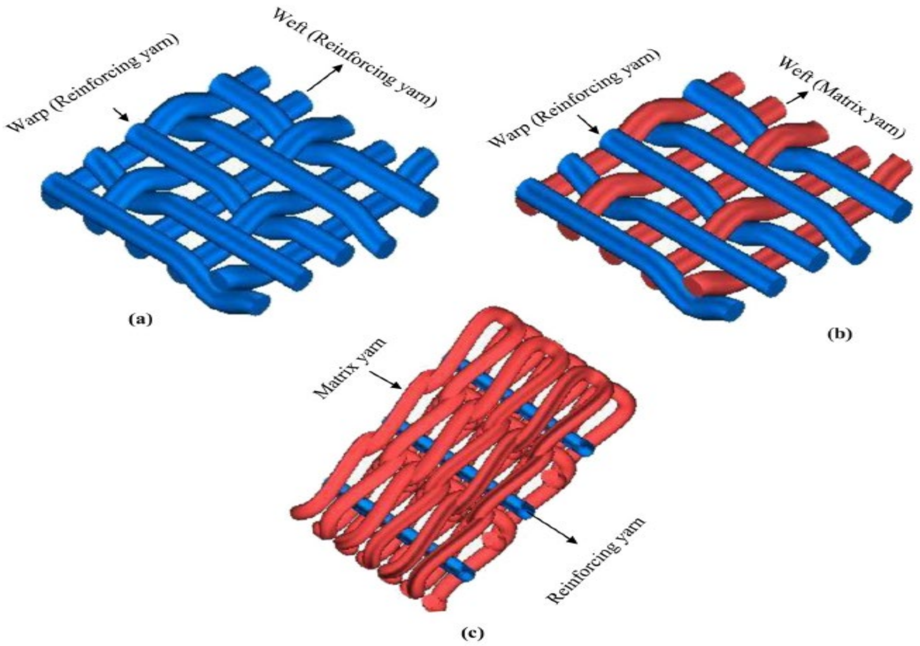


Fig. 14. Graphical representation of fabric preforms (a) woven, (b) woven commingled, and (c) knitted commingled [168].

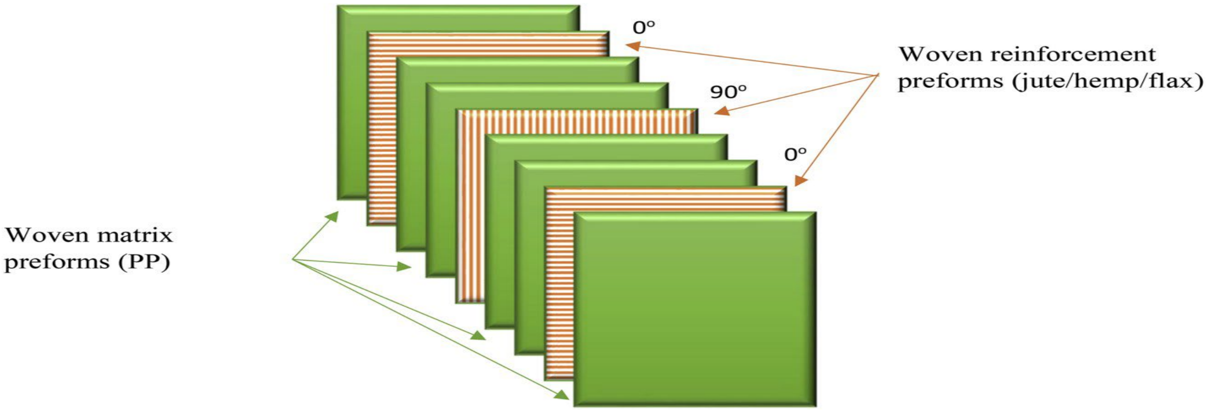


Fig. 15 Stacking sequence of woven composite (W.C.) laminates [168].

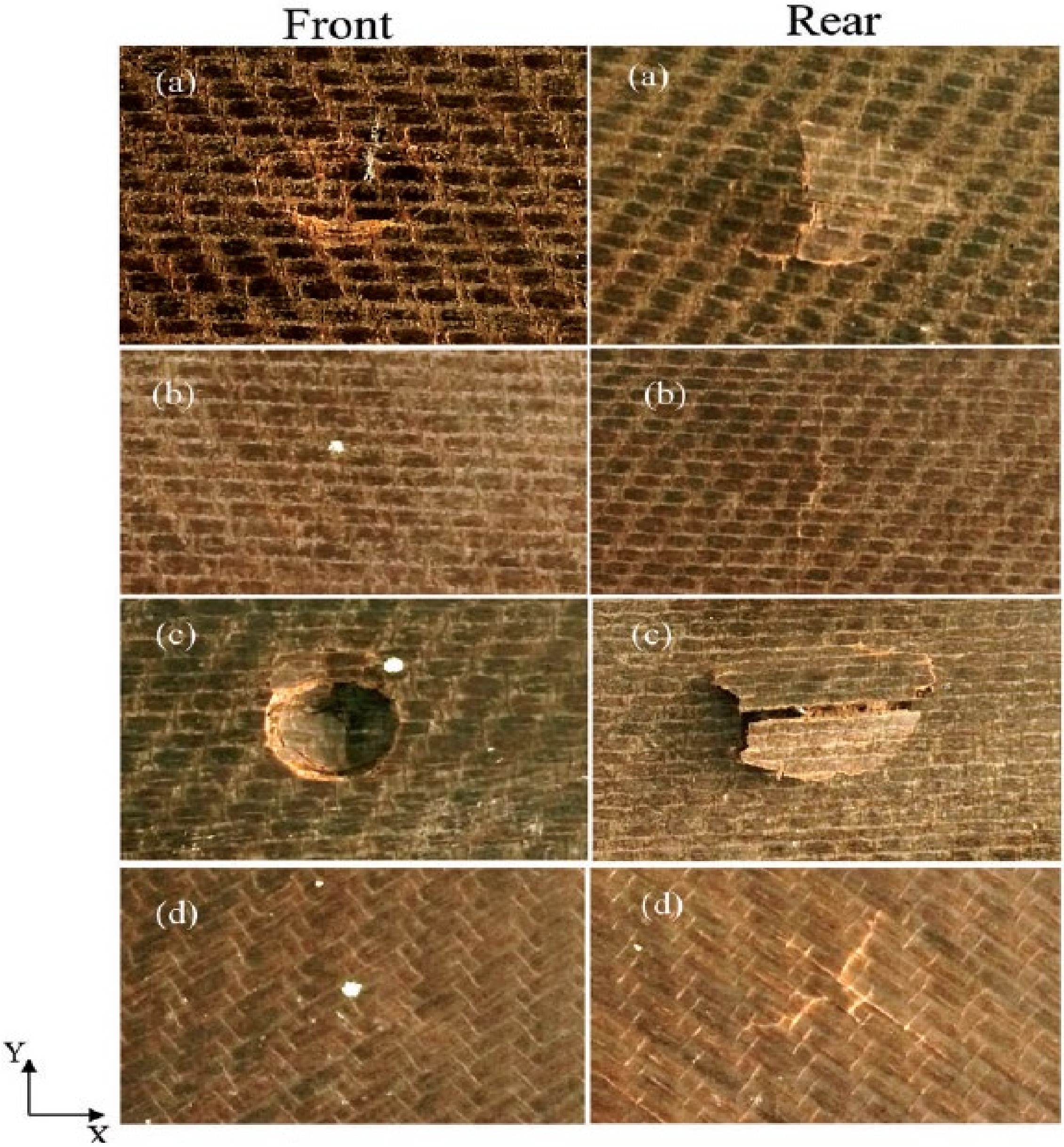


Fig. 16 Damage images on the front and rear faces of (a) unidirectional, (b) cross-ply, (c) sandwich-like and (d) quasi-isotropic composites impacted at 3.6 J. [169]

4.2 Geometrical characteristics

4.2.1 Thickness

Thickness is the essential parameter because it modifies the energy absorption mode and the area of composite failure. The predominant failures were determined during the impact event, mainly by the composite laminate thickness [172]. Therefore by considering thickness in NFPC composites, Wang et al. [173], noticed that impact force and the Hertzian force of flax – fibre reinforced polymer was found to increase with increasing thickness. Whereas the ductility index decreases with increased composite thickness. Dhakal et al.[174] stated that the thickness is essential in the behaviour of jute (methacrylated soybean oil) MSO bio-composites in low-speed impacts. However, the connection between impact response and damage profile against laminate thickness was not adequately recognized. Zaman et al. [175] studied the influence of fibre volume fraction on coconut fibre reinforced composites. They observed that increasing 15% of fibre content leads the composites more flexible and easy to deform; in fact, it could be used to reduce the high resonant effect. A similar observation made by Singleton et al. [176] on high-density polyethylene composites with flax and concluded that better impact strength is found at 10 % – 20 % of fibre content. Garkhail et al. [177] observed Charpy tests on flax fibre length ranging from 5, and 10 mm yielded the highest impact strength in flax/polypropylene composites. Bax and Mu¨ssig [178] noticed the impact resistance of flax fibre/polylactic acid increases with the fibre mass fraction.

4.2.2 Curvature

Curvature has a significant influence on the NFPC with the response to impact loads, due to high shear stress induced by the extrusion or injection moulding process, the fibre can easily bend into the composite. However, it is rational to think that micromechanical models require a factor of adjustment that considers the natural fibre curvature to estimate the better mechanical properties of the composite [179]. Though the reports in the literature consider other contributing factors, these findings do not comply with the experimental values [180–183]. Saghafi et al. [184], analysed the preload effect of the impact behaviour of the curved composite panels by placing samples under tensile and compressive stress. Their findings exhibit that the preload of the plate had a significant influence on the impact factors like final displacement and damage. Over the past two decades, several studies have studied the effect of curvature on the impact behaviour of composite, but attention is focused primarily on low-speed impact tests [15]. Laminates with higher curvatures showed lower deflection and higher contact strength constantly than a flat plate. The impact behaviour of curved composite plates yet to be explored experimentally. Invoking these experimental experiments will provide ample practical and reliable details on complicated damage mechanisms of curved composites.

4.3 Impactor characteristics

Impactor characteristics were widely examined for the study of impact mechanics of composites, including shape, size speed, mass and angle. Fig. 17. signifies several forms of the impactor and different pattern of perforation in the target lamina by different projectiles, respectively. Lee et al. [185] performed a low-velocity impact test on four different impactors:

flat, hemispherical, semi-cylinder, and conical on sheet moulding laminates. They noticed that flat and hemispherical impactor exhibited similar damage and energy dissipation levels. Zhou et al. [186] noticed that the impactor shape influences the failure mechanism of laminates under low-velocity impact tests. Mines et al. [187] performed a high-velocity test with flat and hemispherical conical impactors. They observed that flat and hemispherical impactors generated higher delamination compared to the rest. Dhakal et al. [7] studied the effect of varying impactor geometries, where they observed that different impactor shapes would produce various failure mechanisms and the damage areas in the composite structures; thus, the residual material will differ upon the impactor shape. Ambur & Kemmerly [188] performed the impact test on woven composite plates for investigating the effect of impactor mass for forty-eight piles of laminated composites at four different masses ranging from 1.1 to 9 kg. They noticed that there is no change on impactor mass for the impact range investigated from 10-33 J. Bucinell et al. [189], observed that even with increasing mass ratio from highest to lowest up to 5, there would be no changed on the mass impactor.

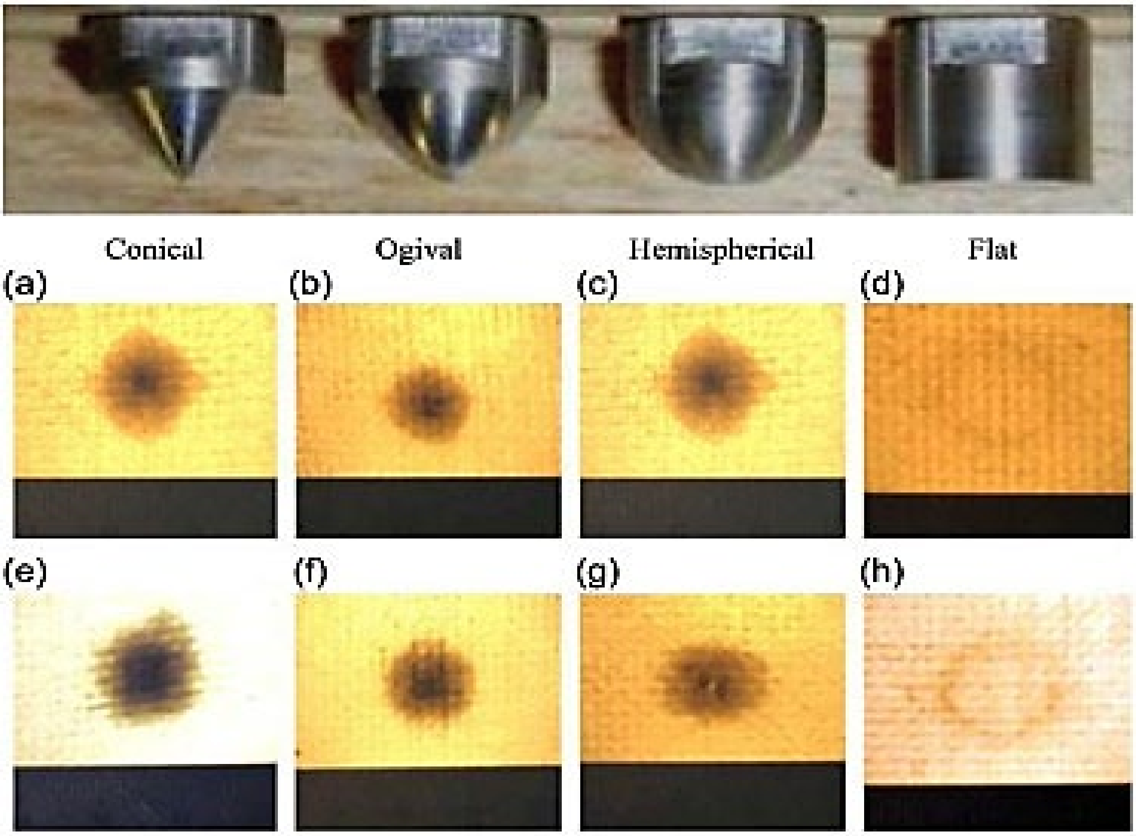


Fig. 17. Result of impactor size and type on the resulting damage to composite laminates [190].

5. Properties of natural fibre in terms of impact damage characteristics

Couture et al. [191], Investigated the mechanical properties of flax and polylactic acid. Tensile, flexural, and impact tests were observed during the process. The compression moulding fabrication technique was used to impregnate U.D. reinforcement. During the tests, the tensile strength of the UD – Flax/PLA showed a higher value than U.D. flax- paper/PLA. The maximum tensile strength for UD Flax/PLA was (339 ± 23/62 ± 1.4 MPa) by a factor of 5.5 and for U.D. flax-paper/P.L.A. (316 ± 14/62 ± 1.4 MPa) by a factor of 5.1. Sathish et al. [192], examined the properties of flax fibre-reinforced epoxy composites with the influence of SiC and Al2O3. The obtained tensile, flexural and impact strength were 44.56 MPa, 112.56 MPa and 28.57KJ/m2 at 20% weight of flax fibre and 8% weight of SiC. However, these results have a vital influence on the mechanical properties of flax composites.

Sair et al. [193], studied the rigid polyurethane and hemp fibre at different loading rates (5%, 10%, 15%, 25%, 30%) to determine the mechanical properties and thermal conductivity. The result showed that composite with 15%wt fibre loading provided a 40 % increase in tensile strength, and also thermal conductivity increased linearly with density. Haghighatnia et al.[194] studied on the fibre length, fibre content, and alkali treatment of 40% hemp fibre and thermoplastic – polyurethane composite. They noticed that the hemp fibre content in composite increases in flexural and tensile strength with an increase in modulus. The tensile properties of hemp composite are better than pure thermoplastic polyurethane. Lu & Oza [195], compared the mechanism of hemp fibre with virgin and recycled high-density polyethylene matrix. Composites were prepared using recycled high-density polyethylene (rHDPE) and virgin high-density polyethylene (vHDPE). Before the fabrication, the hemp fibres were treated with 5 %wt of NaOH. The indication of 5%wt NaOH treatment significantly improved the fibre-matrix interface resulting in improved mechanical properties. Neves et al. [196], studied the mechanical properties of epoxy and polyester composite reinforced by hemp fibre. The fibre content of 10, 20, and 30 vol % of continuous and aligned fibres were mixed with either epoxy or polyester composites. The result showed that for a higher amount of 30 vol% of fibre content along with epoxy composite gives a superior strength than polyester composite. The flexural and tensile strength of epoxy composites reinforced with hemp fibre is 76 MPa, and 50.5 MPa are higher than 49.1 and 25.4 MPa respectively for polyester composites. The author Neves et al. [196] recommended hemp fibre/epoxy composite as an essential application for multilayer armour ballistic protection system.

Akhtar et al. [197], investigated alkaline treatment and the properties of untreated and treated kenaf/polypropylene reinforced composites. By using an injection moulding technique, the result appeared that the alkaline treated kenaf/PP composites performed better in tensile, modulus, and flexural strength than the untreated kenaf/PP composites. The author Akhtar et al. [197], also observed that 40 % of fibre loading had a significant increase in tensile strength which is beneficial in automotive applications. Abu Bakar et al. [198] observed kenaf fibres weighing 5, 10,15 20, and 25% in the hot-pressed method with 4% of NaOH and they reported that the highest flexural strength and modulus improved by almost 24% and 83%, at 25%wt of treated fibre loading. While with the epoxy composites, treated fibres yield better results than untreated with the help fibre-resin bond, which is enhanced by NaOH.

6. Natural fibre composites and their hybrids: benefits and critical issues

Natural fibres are hydrophilic because their hydroxyl groups swell in water, leading to inappropriate characteristics. However, the combination with synthetic hydrophobic fibres ensures the absorption is minimised and hence improving the material properties [120,199]. Basalt fibres have also recently been considered to be an appropriate substitute for glass fibres [200]. Since they have a lower environmental impact, they can compete effectively with glass in sectors such as automotive, for instance, improved acid resistance [201]. Nisini et al. [200] observed that basalt layers reduce the bending of flax fibre laminates and improve their rigidity, which is advantageous for flexural and interlaminar strength. Besides, other configuration was not significantly increased. Fragassa et al. [202] noticed that the reduction in the brittleness of basalt fibre provides some evidence of plastic behaviour, in particular, the hybridization with flax fibre reinforced composites provides a relatively long period of quasi-constant load during the impact tests. Thus lead to delayed failure time, while extensive damage is caused. Petrucci et al. [203] studied the impact characteristics of hemp, basalt, glass and flax hybrid composites. They noticed that GHB. (Glass, Hemp and Basalt) achieved the worst impact performance compared to FHB. (Flax, Hemp and Basalt) hybrid composites. Akhil et al. [204] observed that glass fibre hybridised with the jute fibre reinforced composites reduces the absorption and improves the tensile and flexural properties. Similar findings have been reported in other literature studies [205,206]. Currently, researchers are focussing on natural-natural fibres reinforced hybrid composites as appropriate alternatives to carbon and glass Fibres [202,207–209]. Natural fibres may also be combined with other natural fibres and incorporated into a polymer matrix to create a hybrid composite. This can balance the costs, boost the material's efficiency and properties [210]. Cavalcanti et al. [211] investigated hybridisation and chemical treatments of Jute, sisal and curaua fibres. They noticed that the hybridisation process enhanced the properties of pure jute fibre composite treatments, which also influenced impact strength. Compared to flexural tests, alkaline treatment increased the impact strength of jute+sisal, while the treatment damaged jute+curaua hybrid composites. Similarly, there are numerous researches on hybrid composites enhanced with natural fibres (e. g. sisal, jute, curaua, ramie, banana leaf) or natural fibres combined with glass fibres [27,212–226].

6.1. Damage mechanisms of natural fibre composites and their hybrids under impact loadings

Understanding of composite material damage mechanisms involves accurate mapping of failure actions using reliable technology. This study of literature analysis the low and high velocity falling weight impact damage behaviour of hybrid composites. Papa et al. [227] investigated a low-velocity impact behaviour on carbon/glass hybrid composites laminates by using a cylindrical impactor with a 19.8 mm diameter hemispherical nose. They observed that stacking of 10 glass and 50 carbon layers with glass on the impacted side showed the highest load of 4713.26N with the least absorbed energy of 6.20J. However, similar results also observed by Zhang et al. [228] for a low-velocity impact behaviour of carbon- glass hybrid composite by drop-weight impact test. They noticed that 1.1 (CG) and 1.4 (CGGC) of hybrid ratio achieved higher peak force with less absorbed energy. Also, Chen et al. [229] studied the hybridisation effect and fabric structure of carbon/basalt/glass-reinforced plastic composites. Low-velocity impact behaviour was performed under the energy levels of 50J and 60J, respectively. The stacking sequence of 16 piles of unidirectional or plain weave fabric showed a considerable effect on the impact resistance. However, on the hybridisation of three different fibres, the placement of carbon layers achieved higher energy absorption compared to the same stacking configuration by hybridising with carbon, glass, or ballast fibres. Tirillò et al. [230] performed a high-velocity impact test on hybrid basalt- carbon epoxy composites. Four different stacking sequences were impacted by a spherical tempered steel projectile of mass 1.725g and diameter 7.5mm with the impact velocity ranging from 165 – 475 m/sec were used and performed using a gas gun. They reported that BCBI (Basalt Carbon Basalt Intercalated) sandwich configurations performed an excellent combination of impact properties and static properties. However, the Basalt limits are comparatively higher even though the flexural strength and stiffness are the same as C laminates.

Concerning Natural fibre hybridisation, Papa et al. [231] presented an impacted behaviour on basalt/flax hybrid composites with twill laminates. They stated that flax fibre exhibits an excellent solution as an impact absorber by reducing the peak stress and absorbing energy by an inelastic modulated response. In contrast, hybridisation with basalt and flax (refer Fig. 18) were efficient than other laminates in short recurring impacts and reducing cumulative failures. Even, Ismail et al. [232] performed a low-velocity impact behaviour of kenaf/carbon hybrid composites with various weight ratios. They reported that 75% of glass fibres and 25% of kenaf fibres could withstand impact energy up to 40J. Barouni & Dhakal [11] investigated a damage assessment in flax/glass hybrid composite plates reinforced with vinyl ester composites. Low-velocity tests performed for the specimens subjected to the energy level of 25J and 50J to examine the damage perforation and complete penetration. They observed that in Fig. 19. flax specimens exhibit higher energy absorption during the impact event. Simultaneously, they showed a more significant damage extension at lower energy levels compared to the glass- flax specimen. Bar et al. [233] compared low-velocity impact behaviour on flax-polypropylene hybrid woven composites with plain woven glass fabric reinforced P.P. composites. Resulted that glass P.P. exhibits higher energy absorption with a maximum peak load than flax-based P.P. composites. A detailed summary of the impact tests illustrated in Table 10.

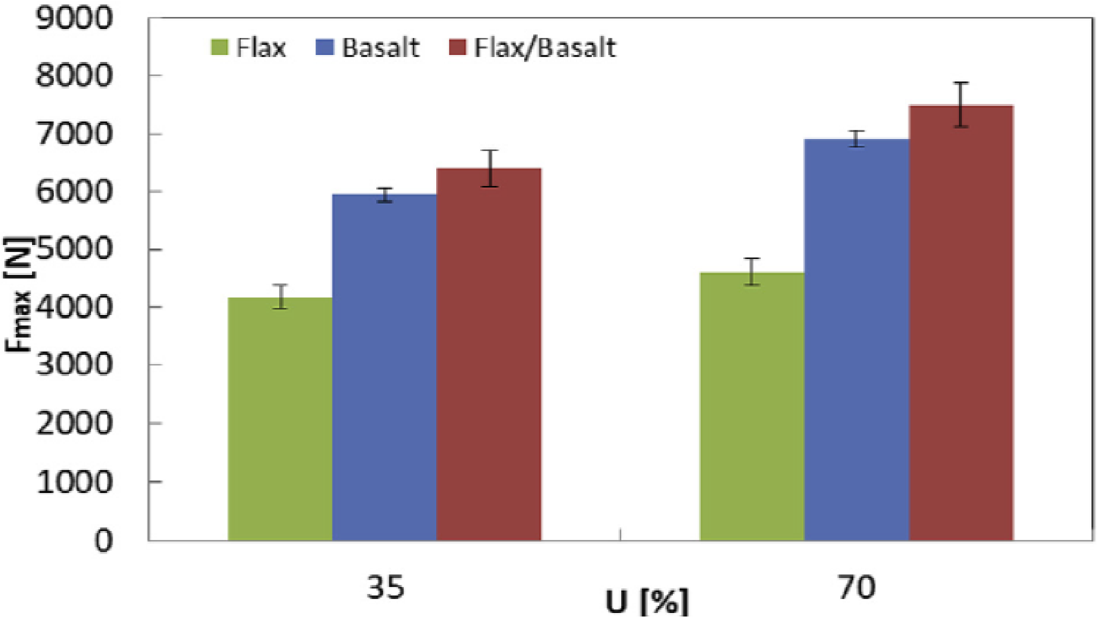


Fig. 18. Maximum load, Fmax, vs impact energy, U. [231]

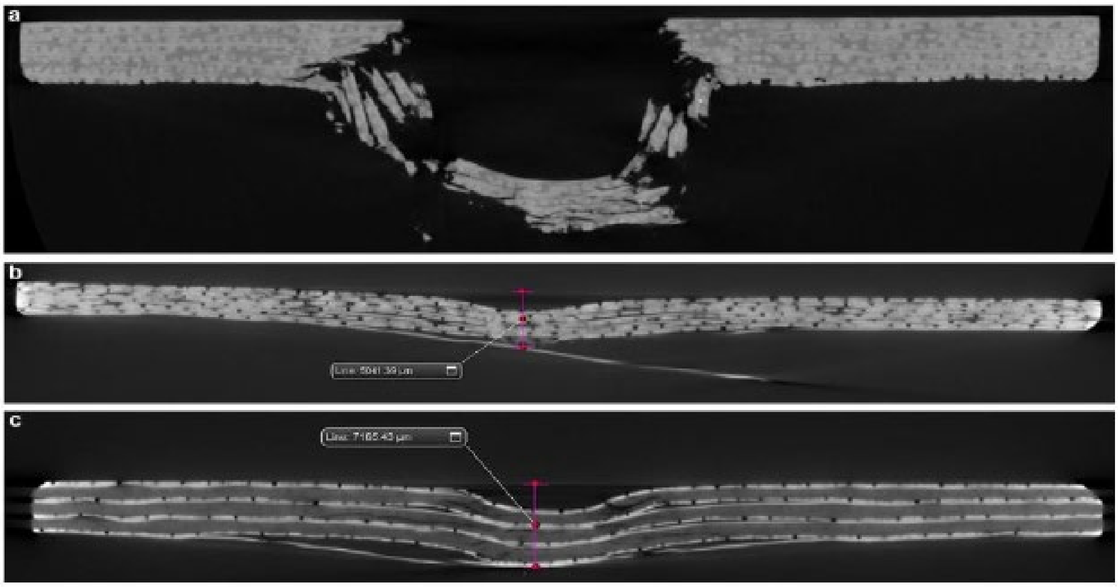


Fig. 19. Cross-sectional X-ray image of the X.Y. plane of the (a) flax fibre, (b) glass fibre, and (c) hybrid glass/flax reinforced composite specimen [11]

Table 10

Summary of the impact test of Natural and Synthetic Hybrid composites.

Hybrid composite type	Matrices	Manufacturing of composites	Dimension of specimens mm	Impactor type and size	Type of impact	Hybrid sample type/sequence	Maximum energy absorbed J	Maximum load N	References
Carbon/glass	Vinyl ester	Vacuum resin infusion	100 x 150	Cylindrical impactor D= 19.8 mm	Low-velocity impact	G10, C5	96	9944,.22	[227]
Carbon/Glass Hybrid 4 layers [0/45, -45/90]	Epoxy resin	Resin transfer molding	100 x 150	Hemisphere impactor D = 12.7 mm	Low-velocity impact	C.G. (1:1) CGGG (1:4)	28-30 30	9000 8600-8800	[228]
Carbon/ Glass/basalt	Epoxy resin	Vacuum-assisted resin infusion	100 x 100	Hemispherical impactor D =12.7mm	Low-velocity impact	B-50 B-60 UD-B-50 UD-B-60	42.71 59.43 42.70 55.00	9.17 KN 8.97 8.35 8.92	[229]
Basalt – Carbon	Epoxy resins	Prepeg lay-up process	100 x 100 Thickness = 4-4.44	Gas gun D = 7.5 mm M =1.725 kg	High-velocity Impact	BCBI	Flexural modulus 765 ± 16.77	The velocity of 247 m/sec	[230]

Basalt/Flax twill laminates	Epoxy resins	Resin infusion fabrication technology	100 x 100	Cylindrical impactor D = 19.8 mm M = 3.64 kg	Low – velocity impact	Flax/Basalt	20	8000 N	[231]
Flax/Glass hybrid	Vinyl ester	Vacuum infusion process	Square specimens 4.5 mm t 60 x 60	Hemispherical nose impact D = 19 mm M = 23.11 kg	Low – velocity impact	Flax/VE Glass/VE Flax/Glass/VE hybrid	50	16000 5500 13000	[11]
Flax/ Glass Polypropelene	Polypropylene	Vacuum-assisted compression molding	150 x 150 x 3.5	Hemispherical nose D = 12 mm	Low – velocity impact	Glass- P.W. Roving – P.W. Roving – U.D.	18.71±1.29 13.81±1.53 23.10±1.11	2.72±0.29 1.61±0.55 2.72±0.29	[233]
Kenaf/Glass	Epoxy resin	Hand-layup process	100x 150	Drop test Height – 0.8 m	Low – velocity impact	Kenaf/Glass	23.33	9.3 KN	[232]

7. Novel improvement techniques of natural fibres composites and their hybrids to enhance damage tolerance and impact resistance.

The main purpose of this paper is to identify the effective techniques or methods for enhancing the damage tolerance and impact resistance of natural fibre hybrid polymer composites and their hybrids. This section will broadly discuss the critical parameters of polymer matrix composites such as STF, 3D metallic coating, stitching, Z-pinning and hybrid techniques.

7.1. STF: working mechanisms, benefits in comparison to other techniques

7.1.1 Background of STF

STF also called as the dilatant fluid has gained attention for the impact protection, for its unique viscosity [234]. It a Non-Newtonian flow of behaviour when the viscosity of the fluid increases with the shear rate or applied stress [235]. The fluid behaviour has been observed mainly in the concentrated colloidal suspensions [236]. Fig. 20 shows the schematic illustration of the shear–thickening behaviour. At rest, the liquid present is enough to fill the void when space is minimum. Then, the liquid slowly fills into a paste and suddenly becomes a substantial under low shear stress. At high shear stress, the viscosity of fluid behaviour increases drastically and turns solid. It is entirely reversible; the suspension could return to the easy flowing state or flow like any other liquid after removing impact stress [237–240]. There are different theories for explaining the mechanism of shear thickening, and many researchers proposed order-disorder theory and hydrodynamic clustering are the two widely accepted theories [241].

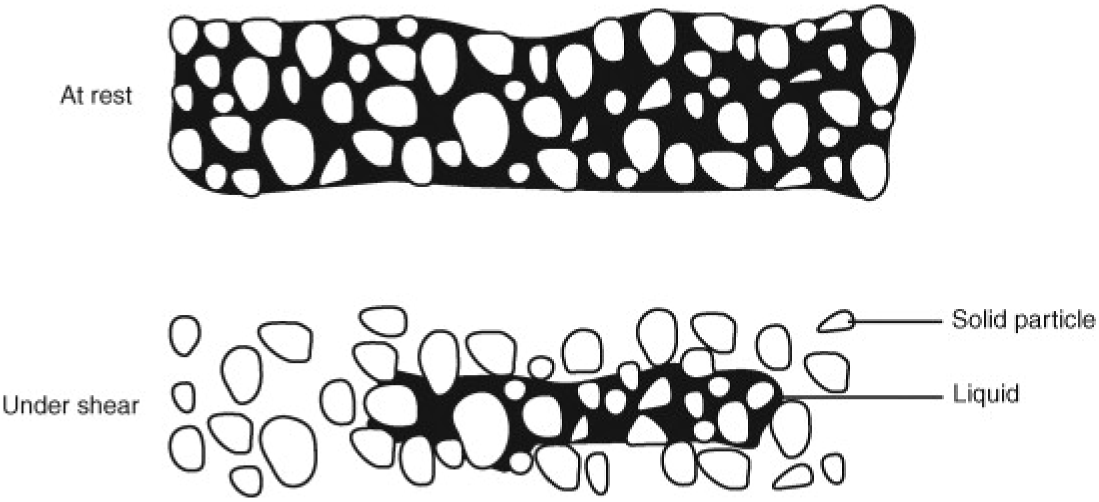


Fig. 20. Schematic representation of the shear- thickening behaviour [236]

7.1.2. Rheological behaviour of STF that improves the impact properties of the composites.

Balali et al. [242] investigated a rheological behaviour of STF for the glass fibre reinforced composites under a low-velocity impact test. Initially, they diluted ethanol with the STF for the weight ratio of ethanol and STF at 3:1. Then, they fabricated glass fabric layers with 16 cm x 16 cm soaked with the ethanol and STF for 1 min and baked at 60° C until they were dry and ethanol had evaporated. Later, they observed that different combinations of STF, STF nano-silica, and STF nano clay treated with glass fibres shown in Table 11 achieved a superior energy absorption with maximum load toleration. Fu et al. [243] studied the rheological behaviour of STF and STF filled sandwich composite panels (SCP) with carbon fibre reinforced composites under the low-velocity impact test. It was noted that 58 vol % of styrene/acrylate particles were mixed with ethylene glycol (EG) showed a higher energy absorption capacity than an aluminium foam. However, the experimental resulted that SCF with a thin core exhibits the higher absorption energy with 99.3%, and a thick core showed impact damage was effectively suppressed on the back surface of SCPs. Y. S. Lee et al. [244], stated that kevlar fabric with ethylene glycol showed a lower energy dissipation results owing to the decreasing friction. However, the presence of STF impregnated with kevlar fibre exhibited a significant increase in energy dissipation with an increased volume of STF. Selver [245] observed that during the manufacturing process (Fig.21.), the carbon fabric (d) absorbs more STF than the glass fabric in Fig. 21 (b) with better STF holding. Further, the author noticed with silica particles; the shear thickening did not show any behaviour on glass/epoxy composites. However, this changes the interfacial bonding and impact resistance of the composite laminates.

In recent studies, polyurethane (PU) performs excellent energy absorption and resilience for the functional protective materials [246–248]. Wu et al. [249], experimented STF with 30%, 50%, and 70% for flexible woven composite with polyurethane glycol (PEG). They observed that higher STF content yields a positive influence on the dynamic resistance of the composites. Moreover, the volume fraction of 70 % wt achieved a higher impact strength to that of the composites treated without STF. Even Caglayan et al. [250] produced similar results in the Shear thickening fluid-filled with Polyurethane (PU) foam core sandwich composites. They stated that STF filled with PU exhibits a higher impact and compressive strength compared to neat PU foams. Furthermore, they observed that 1wt% STF-40/PU was the active foam core in terms of impact resistance. Also, the highest percentage of energy absorption for the received impact energy was achieved with the lowest possible damage of 13%.

Table 11

Combinations of Glas fibres treated with STF [242]

Samples	Weight (gm)
Four layers of neat glass fabric	34.8
Four layers of glass + hybrid STF (20% of nano-silica)	45.7
Four layers of glass + hybrid STF (30% of nano-silica)	47.9
Four layers of glass + hybrid STF (30% of nano-silica and 1% nano-clay)	48.9
Four layers of glass + hybrid STF (30% of nano-silica and 3% nano-clay)	50.2

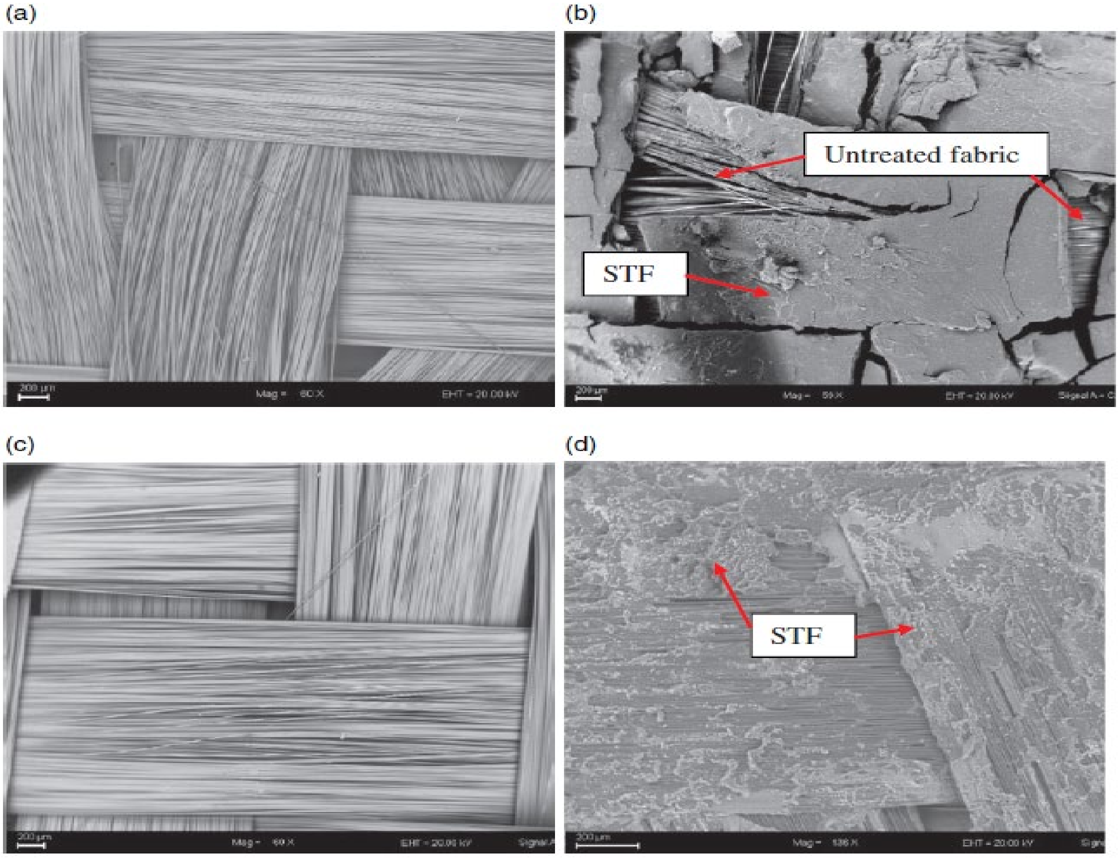


Fig. 21. SEM images of: (a) GF, (b) GF20, (c) CF and (d) CF20 fabric samples [245].

7.1.3 Experimental and Numerical analysis of the impact test of hybrid composites filled with STF - Review

Experimental analysis

Selver [245] studied the impact and damage tolerance of carbon and glass fabric composites treated with silica ratios of 10%, 15%, and 20%, respectively. A drop weight impactor with a 16 mm diameter striker was used. The experiment showed that glass and carbon treated with STF achieved more energy in the low impact test of 20 -30 J with a load of 2900 – 3500 N than pure glass and carbon composites. However, Balali et al. [242] noticed similar results in the glass fibre reinforced hybrid with STF. The maximum energy for the neat glass fabrics was absorbed in 2.57J, and the highest of 140.825 N. While the STF/glass and STF/nano-clay/glass fabrics exhibit a superior penetration compared to neat glass fabrics. For 30 wt% concentration STF and 3 wt% concentration nano-clay, the accepted load was at 1773.7 N with the absorbed energy of 43.5J.

Pinto & Meo, [251] worked on improving hybrid composite by adding silica-based liquid in a carbon fibre reinforced polymer laminate (CFRP). A drop test rig of a spherical head geometry of 20mm diameter with a mass of 12.684 kg as an impactor. They noticed the peak remained linear at low-velocity impact (6J). While at 20J, it gradually moves from 4252 N to 4742 N with STF (Fig. 22.). However, the curve showed a dramatic increase in the 40J impact test because of hybridisation. The peak moves from 5197 N for the unreinforced samples to a highest of 6929N for the STF based ones (~30%). Therefore, the results could not achieve for low-velocity impacts as the hydrocluster activation mechanism requires a relatively high solidification. Even Asija et al. [252], produced a similar result for UHMWPE variant shield and Spectra shield. They observed that STF treated panels absorb less energy at low impact tests compared to the untreated panels for the deflections at peak load. Nevertheless, the peak load absorbs the higher energy at higher impacts when the composites treated with STF. According to author Lu et al. [253] stated that the ballistic performance of STF under a high velocity of 450- 510 m/sec, does not show much improvement due to increased mass and the degradation of maximum perforation velocity of the yarns in the fabric. However, they noted that aramid fabric panels under STF provide a better shielding performance in a low- velocity impact test.

Fu et al. [243] examined the mechanical performance, namely rheological, low-velocity behaviour of STF, and STF filled with sandwich composite panels (SCP). It was noted in Fig 23(a), the STF stiffness is initially low when the striker impacts the front-facing, because of densification. Whereas when the striker reaches the backplate, see Fig 23 (d), the STF already gets compressed severely, and they are fully densified. Therefore, a higher resisting force is observed due to high supporting stiffness of STF in the second peak. However, the author also observed that the impact behaviour of SCP was significantly improved with the STF concentrations

Regarding Natural fibres treated with STF, Abhishek et al. [254], investigated Jute/E-glass with epoxy resins by adding STF as a fourth element. They observed that jute fibre loading with STF performs good results in tensile and elastic properties in contrast to other composites. A detailed summary of the impact test of composites treated with STF shown in Table 12.

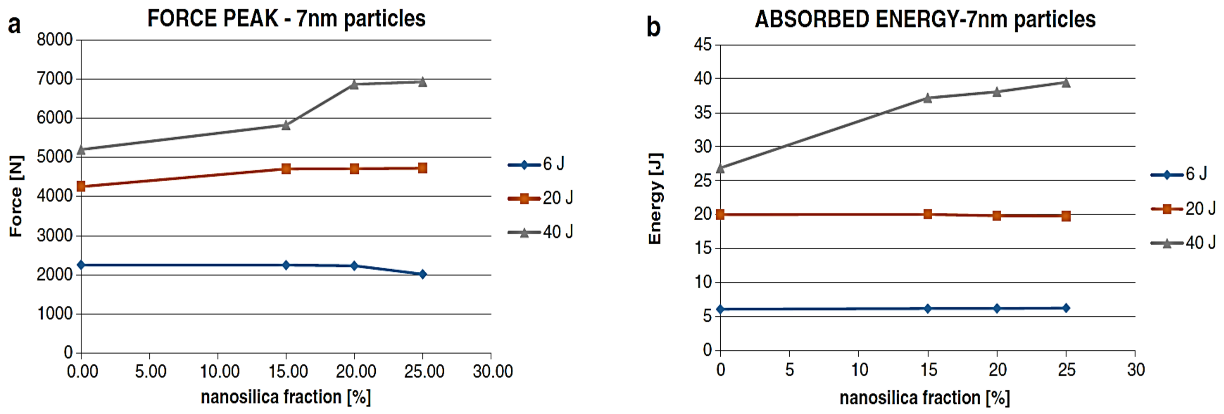


Fig. 22. (a) Force Peak variation at different energies and different silica concentrations in STF; (b) Energy absorption variation at different energies and different silica concentrations in STF [251]

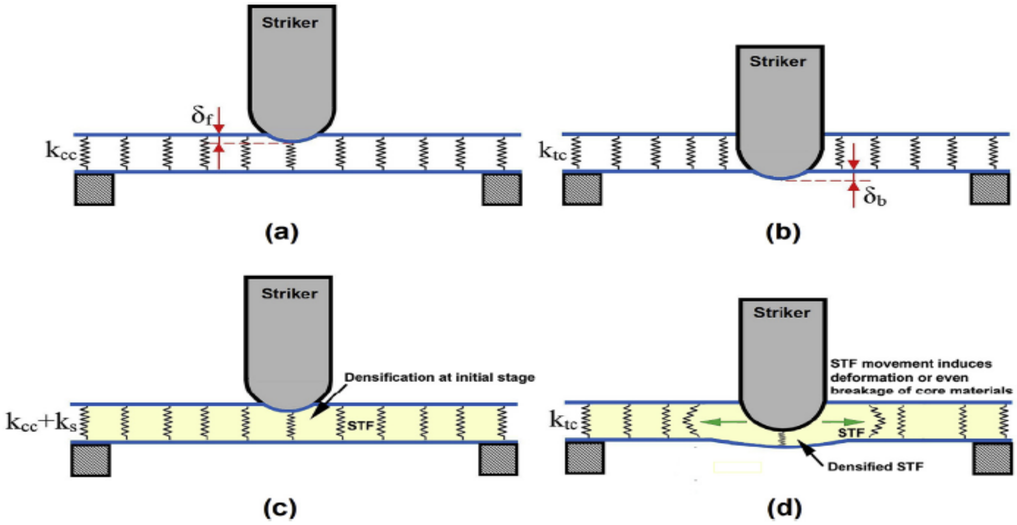


Fig 23. Schematic impact on (a) pristine front-facing (b) pristine back facing (c) STF filled front-facing (d) STF filled back facing [243]

Table 12

Summary of impact test of composites treated with STF

Composite samples	Type of Impactor	Specification of impactor	Type of STF	Impact-low/high	Peak force (N)	Maximum energy absorbed (J)	References
Carbon and glass fibre composite	Circular	16 mm in diameter striker	Silica nanoparticles and polyethylene glycol	Low – velocity impact test	Glass 3580 ± 159 Carbon 3118 ± 146	Glass 28.95 ± 1.4 Carbon 3118 ± 146	[245]

Glass fibre reinforced STF	Circular/ Round	16 mm in diameter	200 g of silica nanoparticles and 400 g of polyethylene glycol	Low – velocity impact test	1773.6	43.5	[242]
Sandwich composite with polyurethane grid	Circular flat compressive heads	80 mm in diameter	200 g of polyethylene glycol and 200 g of SiO_2	Low – velocity	3000	4827.46 MJ for 70% STF	[249]
Sandwich composite panels with STF	Cylindrical striker	12.7 mm in diameter	58wt% styrene/acrylate + ethylene glycol	Low – velocity	1069.3	26.718	[243]
UHMWPE variants gold shield and spectra shield	Hemispherical impactor	15.585 mm in diameter	400 g/mole of polypropylene glycol and ethyl alcohol	Low and high velocity	1283.1	14.45	[252]
Sandwich composites	Hemispherical metal crosshead	10 mm in diameter	Silica nanoparticles + ethylene glycol	Low velocity	1550	18.50 ± 0.53	[250]
Carbon fibre reinforced STF	Semi-spherical head geometry	20 mm in diameter	10 to 25wt% of silica particles with polyethylene glycol	Low velocity	6929N	40 J	[251]

Numerical analysis

Numerical modelling is comfortable and straightforward ways of solving highly dynamic and complex geometries cost-effectively [255]. Also, the material can be modelled from 2-Dimensional to 3-Dimensional at different resolutions. This includes many informative results, such as stress-strain behaviour, residual velocity, contact force, and failure patterns. There are different approaches to model the behaviour of fabrics under low/high impact tests, amongst which shell and solid- element method are the most preferred by researchers [255,256]. For modelling the STF, Arbitrary Lagrangian Element (ALE) is mainly considered for applications where fluid-structure interaction must be considered [257]. Rizzo et al. [257] investigated a numerical analysis of CFRP plates with silica-based STF. A rectangular plate of 150mm x 150mm x 2mm was modelled using a 3D Solid finite element method and simulated by using a quasi-isotropic stacking sequence [0/90/+45/-45]$_s$. An LS-Dyna code made the 3D model with a constant stress formulation of 0.25mm thickness, and the impactor was modelled as a hemispherical static body of mass 12.864 kg and 20 mm in diameter. STF was placed at three different locations shown in Fig. 24. ALE formulation was used to model the STF in LS-Dyna. They found the numerical model could simulate the damaged area of mechanisms of STF/CFRP with an error of 9 to 12% between the experimental and simulated results. Also, it can anticipate the impact behaviour of the STF/CFRP system allowing the optimisation of the STF to maximise the impact properties.

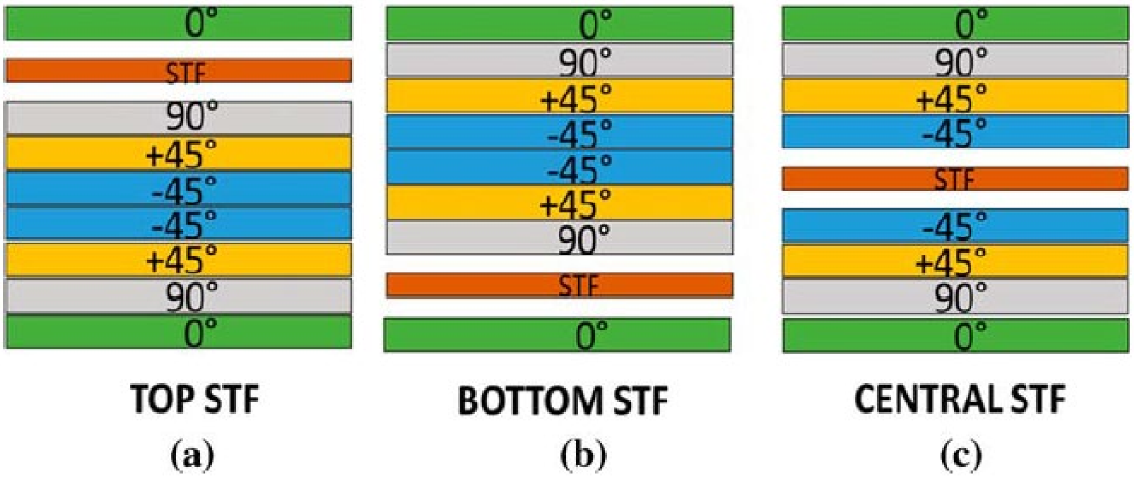


Fig .24. Location of STF layer with CFRP thickness (a) STF placed close to the contact surface (b) STF is placed far from the contact surface (c) STF placed on the midplane of the laminate [257].

Sen et al. [258] developed the kevlar-STF model by using 3D solid elements, taking into account actual weave geometry and inter yarn friction. The inter yarn friction was developed by using Abacus element M3P4R (four noded membrane element with reduced integration) was used to mesh the geometry, and the kevlar fabric was modelled with a rectangular cross-section with a thickness of 0.23mm and a wavelength of 1.548mm respectively as shown in Fig. 25. A coupled Eulerian-Lagrangian (CEL) was used to model the STF, and the properties of STF are defined by using the Mie-Grüneisen equation of state described in Eq. (8). Initially, they modelled an STF- fabric composite by using a Lagrangian approach as fabric experienced in the smaller deformation, and later, they modelled STF by using an Eulerian approach due to the fluid at a lower strain, which acts as a larger deformation. While comparing the experimental and numerical results, the author noticed that the proportion of energy absorption showed similar results in all the test cases. Even Wetzel [259] also observed similar results for experimental and numerical behaviour of STF-kevlar composites.

The equation of Mie-Grüneisen is given by:

$$p = \frac{\rho_0 {C_0}^2 x}{(1 - sx)^2}\left(1 - \frac{\Gamma_0 x}{2}\right) + \Gamma_0 E \tag{8}$$

Where,

p = pressure

C_0 = sound speed through the medium

X = 1 – (ρ_0 / ρ) where ρ_0 is the initial energy and ρ is the current density

Γ_0 = material constant

S = dU_s / dU_p is a linear Hugoniot slope coefficient

U_s = shock wave velocity

U_p = partial veloicity

E = internal energy per unit reference volume

Hasanzadeh et al. [260] investigated the numerical analysis of multilayered plain-woven HMPP fabrics filled with STF by using LS- Dyna. An eight noded solid element was used to simulate the woven fabric and projectile in contrast to the original study, which used shell elements by B. W. Lee & Kim [261]. In both studies, STF was used as a polyethylene glycol weighing 200 g/mol. However, the particle size and the fabric were different for both the studies. Hasanzadeh et al. [260] used HMPP fabric with a particle size of 12nm while B. W. Lee & Kim [261] used kevlar fabric with a particle size of 100-500nm. The only difference was the yarn pull – out speed was observed, and it showed a massive difference in both investigations. Hasanzadeh et al. [260] observed speed of 1.67 mm/sec, which was lesser than the original study by B. W. Lee & Kim [261] in which the STF impregnated kevlar fabric was claimed to be invoked at 23.3 mm/sec. These factors are likely to cause a difference in the shear thickening behaviour of the two systems. Besides, there is no information available on user-defined friction subroutines used in Hasanzadeh et al. [260] simulations, even though they say that simple Coulomb friction has been implemented and updated to account for the shear thickening effect. Moreover, Hasanzadeh et al. [260] compared the numerical and experimental results of HMPP fabric impregnated with STF for the depth of indentation. They observed that the numerical findings were in close agreement with the corresponding experimental results.

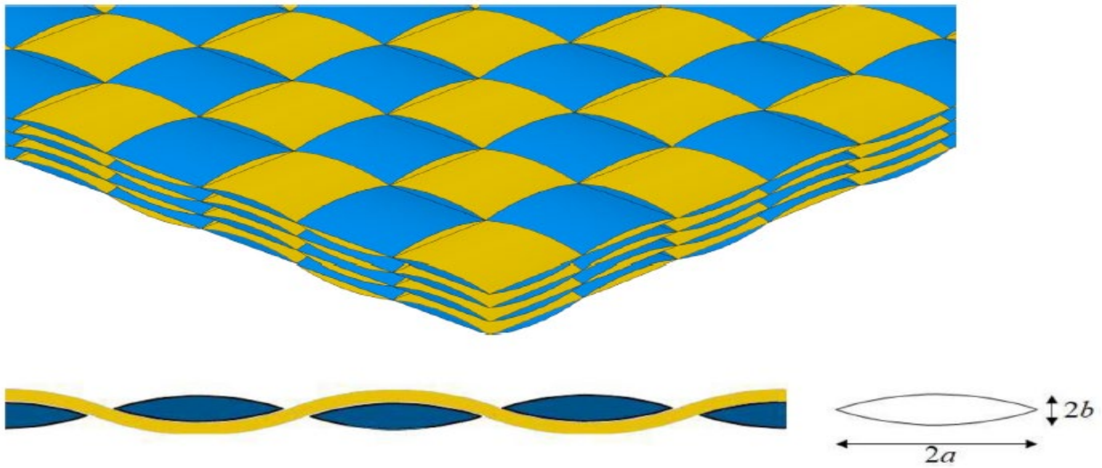


Fig .25. Schematic diagram of cross-sections of the fabric [258].

Chauhan et al. [262] studied the Lagrangian method for examining the characteristics of STF with an eulerian carrier fluid. Water was chosen as the carrier fluid with a viscosity of 0.0091 poise, and a silica density of 20196 g/m^3 was modelled by using ANSYS CFX. As shown in Fig. 26, the diameter of each spherical SiO_2 was 2 mm with an applied flow velocity 2m/s. They observed that by applying higher velocity and acceleration to the STF domain, there would be a possibility of achieving a higher shear thinning effect. On the other hand, the use of sand particles could improve the viscosity of the STF, resulting in an improved shear thickness of the dilatant fluid.

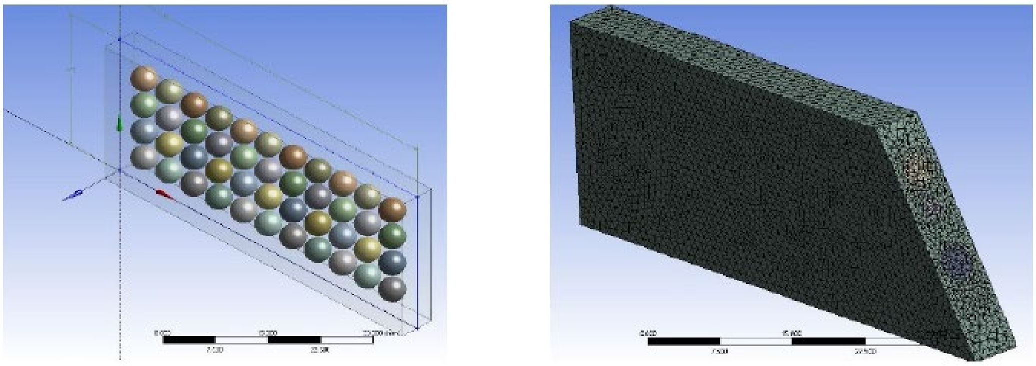


Fig.26. Geometry and meshing of Shear thickening fluid model [262].

7.1.4. Natural fibres with STF benefits in contrast to other techniques.

A detailed study on the shear thickening fluid with natural fibres are inadequate. It is, therefore, essential to develop and optimise the use of STF in the hybrid combinations with natural fibres along with the traditional fibres. However, there are numerous types of application of composite structures built with STF, such as Kevlar, ceramic, and UHMWPE. Previous experiments were conducted to develop the performance of materials used in defence applications, such as bulletproof jackets and armoured helmets [263,264]. Further, advanced materials like aerogel and carbon nanotubes (CNT) coupled with conventional composite materials, would enhance the capability of resisting bullet impact to a certain extent [265]. Currently, there is a significant interest in developing impact-resistant fabrics such as Kevlar built on the shear thickening fluid (STF) [244,258,259,266,267]. Besides, STF has shown promising results in protection and flexibility. Also, the use of STF with composites/fabrics still suffers several drawbacks like evaporation, leakage of carrier fluids, and air-moisture permeability for the clothing purpose. However, some fibres impregnated to STF faces many issues due to evaporation and leakage [268] As a result, evaporation of the STF fluid will be a vital issue in existing STF research. Remarkable efforts are being made to solve this problem in recent years. One of the efforts was to fill a hollow fibre with an STF [269]. However, by its nature, the STF is not easily pumped into the hollow fibres. In order to pump the STF into fibres, the viscosity of STF must be reduced, but in the case of shear response, STF will be acted accordingly.

7.2. Metallic coating technique

Additive manufacturing techniques are currently booming in the research interest due to the unique potentialities, among these the cold spray deposition technique is the most commonly used in the production of metallic layers on polymeric substrates [270]. In this technique, a uniform metallic coating is produced by accelerating solid particles in supersonic gas flow at a velocity of 300-1200 m / sec and depositing them onto substrates [271,272]. When the powder particles have an impact on the target surface, the kinetic energy is converted into plastic deformation; the solid particles are deformed and bonded together, making it possible to form and produce coatings [273]. Extensive researches are done in the past on 3D coatings on metal and alloy substrates [274–278], but much less attention has been paid to polymeric substrates and in particular, composite materials [270,279,280]. The significance of this technique on polymer structures is to prevent damage to sensitive polymer substrates requiring metallization. In addition, electrically conducting coatings is produced because of minimal particle oxidation during the deposition cycle, which is an additional advantage over the thermal spray process [281]. Astarita et al. [282] studied the Metallic Coating on hemp-PLA laminates through the Cold Spraying Technique (CS). The coating was found to be very thick, and there were no visible voids or cavities present. Also, the properties of the hemp- PLA laminates were not affected by the deposition process, if the parameters are correctly set. Lupoi and O'Neill [283] investigated metallic coatings of glass fibre composites using cold spray deposition of copper, aluminium and tin on various thermoplastics (polyamide-6, polypropylene and polystyrene). They detected that CS of tin resulted in the successful deposition due to its low critical velocity resulting in low theoretical impact energy (10.7 times lower than copper). Whereas, erosion in copper was the most common effect. Ganesan et al. [270] compared the deposition mechanisms on thermoplastics and thermosetting substrates. They found that deposition of thermoplastics was possible through mechanical interlocking (particle embedment), while localised fracture was noticed for thermosetting substrates. Che et al. [284] investigated the carbon fibre reinforced polymer composites with both high-pressure and low-pressure CS systems. They concluded that erosion was considered to be a critical barrier to the development of coatings on CFRP substrates, as exposed and fractured carbon fibres were observed after particle impacts. Resulting, only individual particles embedded in the epoxy matrix. However, Fallah et al. [285] also noticed that delamination of the carbon fibre-reinforced polymers (CFRPs) by cold spray deposition occurred at higher gas pressures. The Copper coatings protect the CFRP from any possible damage but also improves the overall properties of the composites. Recently, Papa et al. [286] investigated novel composite structures of the basalt-PA6 substrate made of a polymer matrix composite with a superficial cold sprayed aluminium layer. Micron-sized aluminium particles have been used to metallise laminate surfaces. Different energy levels of impact tests have been carried out. They noticed that only a few damages were recorded on the treated laminates. Also, in Fig. 27, the energy absorbed by the neat system is significantly higher, indicating higher susceptibility to damage to the composite coating. Besides, the aluminium deposition damages the mechanisms that cause a lower indentation depth and a considerable plastic deformation. The author further concluded that the development of a 3D printed metal coating on a composite surface designed to minimise the damage caused by low-speed impacts, leading to a novel composite structure, would lead to a significant improvement in impact strength.

Therefore, the coating techniques could satisfy the mechanical and impact properties of the composites. However, this novel technique could be challenging for natural fibre composites, especially for its poor wettability, excessive level of moisture absorption and inadequate adhesion. If this could be successful, then this method would be the rapid replacement for synthetic fibres and makes the composites stronger, resulting in competitive advantages.

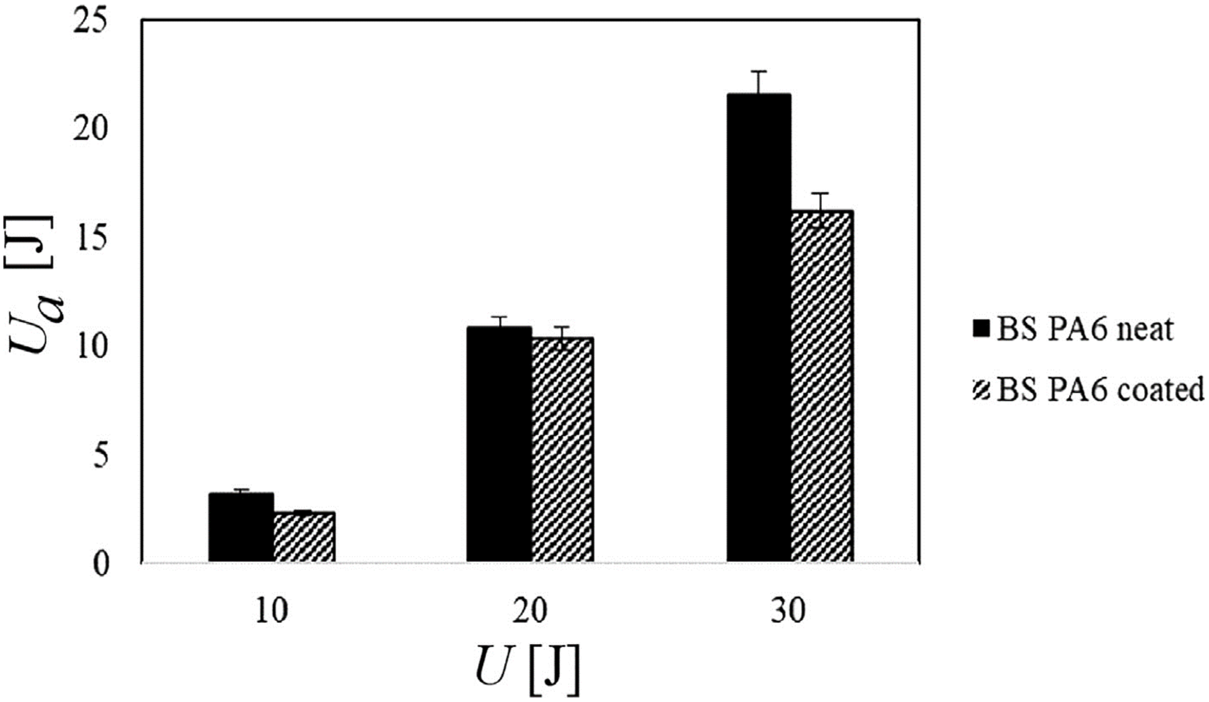


Fig. 27. Comparison of absorbed energy, U_a, versus impact energy U for BS/PA6 neat and coated samples [286]

7.3 Stitching

An efficient method for increasing the delamination resistance of fibre-reinforced polymer composites is through-thickness stitching. In the past, several studies were proposed on the stitching of synthetic reinforced fibre composites [287,288]. However, reports on NFRC were rare [289,290]. Amongst the studies, Rong et al. [291]investigated the fracture toughness of unidirectional sisal/epoxy laminates stitched with nylon and Kevlar threads. They noticed the expansion of the fibre bridging zone improved the delamination resistance, while the tensile and flexural properties were not affected by the stitching. Fig. 28 represents the schematic view on the stitched preform of woven flax fibre-reinforced composites in which Ravandi et al. [292] observed that the laminates stitched with flax yarn and cotton thread showed right consistency and well impregnated stitched fibres. Also, they noted that flax yarn stitches showed a superior fracture toughness compared to cotton thread stitches. However, in the other studies, Ravandi and co-workers, found stitching causes a decrease of 16% in the woven flax intralaminar fracture toughness. Alternatively, a decrease of 5% is recorded in cotton yarn stitching [293]. Even, Almansour noticed a similar result in the flax/basalt hybrid vinyl ester composites. Due to crack propagation, they pointed out that the stitching of FBVE composites significantly decreases the fracture resilience in the stitched zone than un-stitched ones.

More recently, Li et al. [294] fabricated a novel hole arrangement- threading glue filling method to examine the mechanical characteristics of NFRP composites. The idea of this novel technique is to adjust the volume fraction, and the cross-sectional shape of the NFRP epoxy composites (refer Fig. 29). Also, adding natural fibres to epoxy caused the neutral axis to shift downwards, allowing a fair stress distribution and significantly improving the flexural properties and energy absorption capacity of NFRP epoxy composites. In other studies, Habibi et al. [295] presented a novel natural fibre reinforcement manufacturing technique by using wet lay paper making process as a replacement of stitching or weaving UD yarns. Short fibres have been used as binders to maintain cohesion between UD yarns. Habibi and co-workers note the short fibres mat layers offers a considerable increase in the transverse modulus and strength for the majority of the loading configurations. Besides, the surface of the tested specimens slowly transformed from a serrated fracture surface to a flat fracture surface with fewer matrix cracks produced on the surface of the specimen parallel to the load direction. However, a noticeable decline in longitudinal splitting and delamination with UD yarns.

Moreover, the application of stitching in the NFPC could improve the intralaminar fracture resilience and the damage tolerance. Therefore, it is vital to develop the stitching techniques in the NFPC by carefully considering the nature of fibre length, thickness and the volume of the fibre. Certain factors in the stitching may affect the impact properties of the composites due to the areal density or stitching scale. Nevertheless, the influence of stitch thickness and the stitch size could enhance the performance of the NFPC.

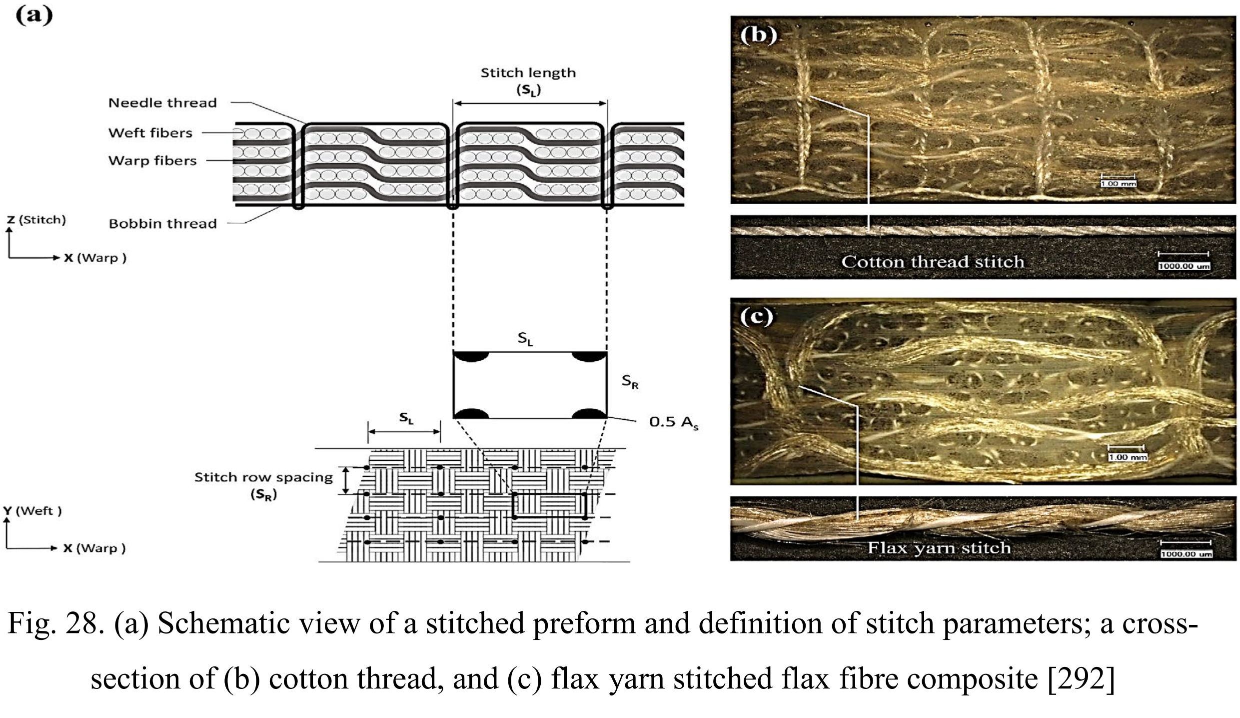


Fig. 28. (a) Schematic view of a stitched preform and definition of stitch parameters; a cross-section of (b) cotton thread, and (c) flax yarn stitched flax fibre composite [292]

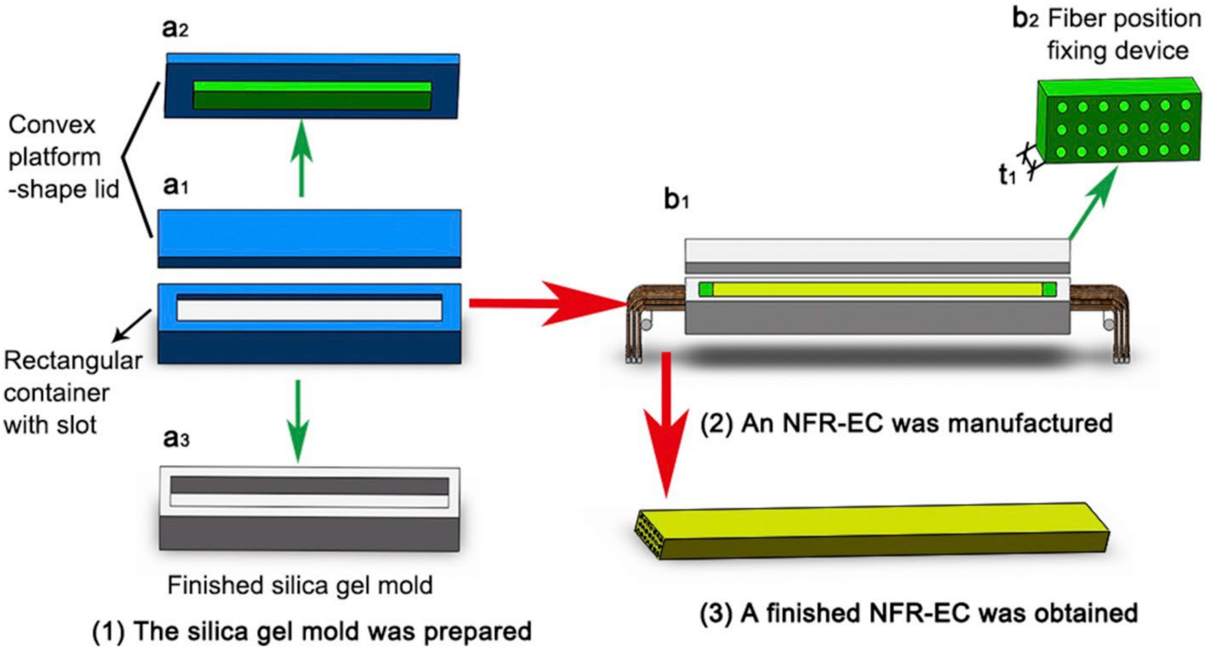


Fig. 29. Method for manufacturing an NFR-EC [295]

7.4. Z - pinning

Z- pinning is one of the most effective methods for enhancing composite impact resistance. This method is entirely new to the research field in the composite structures, especially for the impact test. Also, in recent studies [296], this novel technique was used in synthetic fibres to investigate Z-pinning on the composite laminates. Amongst the studies, Francesconi and Aymerich [296] found Z-pinning of carbon/epoxy prepreg laminates cannot delay the initiation and propagation of delamination damage. Nevertheless, it can improve resistance and reduces delamination size for high-speed impacts. Even Zhang and co-workers [297] noticed that this method is more useful for the thicker specimens and enhances the delamination resistance for the low-speed impacts. More recently, Hoffmann et al. [298] presented a novel technique for the integration of Z-pins in the thermoplastic reinforced composites. Ultrasonic vibrations used to insert metallic z-pins into the glass fibre reinforced polypropylene composites. While examining the mechanical properties of z-pinned thermoplastics composites, they found that there is a low pull out force, by the weak bonding and lack of friction between the steel pin and the thermoplastic polymer.

However, this method is quite challenging for natural fibres, and mostly, natural fibres are hydrophilic that affects the adhesion to a hydrophobic polymer matrix, and perhaps reduces the impact on composite material.

7.5 Hybrid technique

Hybridisation techniques are superior in strengthening the composite structures and are classified into three types. (i) interply, (ii) Intraply and (iii) Fibre metal laminates shown in Fig. 30. Naik and coworkers [299] claimed that the impact strength of glass-epoxy is sensitive to the interply sequence and the modulus of the hybrid composites. In comparison with Inter -ply and Intra-ply hybrid laminates, Pegoretti et al. [300] noticed that intra-ply hybrid laminates had superior impact performances in the E-glass-poly vinyl alcohol/polyester laminates. However, Wang and coworkers [301] noticed a different observation in the basalt-aramid epoxy composites. They stated that low-velocity impact response of inter-ply composite had a more significant ductile index, with least contact force and superior specific absorption than the intra-ply composites. Even, Pegoretti et al. [302] noticed the same result in the E-glass poly (vinyl alcohol) hybrid composites. They depicted that crack growth was higher for intra-ply composites compared to inter-ply hybrids because the ductility index reached maximum values. Moreover, many researchers [303,304] are trying to adopt a modern hybrid technique to improve delamination resistance and crack propagation. In that, fibre metal laminates showed a promising technique to improve the fatigue crack resistance of the composite material and besides, this hybrid technique possess excellent damage tolerance characteristics compared to metallic alloys. Dhar Malingam et al. [305] studied the properties of kenaf/glass hybrid reinforced metal laminates with different fibre orientations and stacking configurations. They observed that fibre orientation ±45° with metal laminates showed an outstanding impact resistance in contrast to fibre orientation of 0°/90°. Also, FML with hybrid kenaf/glass composites exhibited superior characteristics in impact and tensile performance. Kaun et al. [306] stated that fibre metal laminates obey the rule of mixture approach related to tensile strength and modulus properties. They further stated that adding aluminium layers to the outer side of the basalt/flax/hemp hybrid reinforced polypropylene composites yield more considerable improvements.

Although these techniques are obtainable in the existing literature studies, it is essential to recognise the best properties for natural fibres, which can perform equally with synthetic fibres in terms of strength and rigidity. Therefore, by selecting the proper matrix reinforcement and also using alternative techniques like metal laminates combined with the natural and synthetic fibres could enhance the mechanical and impact characteristics of the ductile fibres.

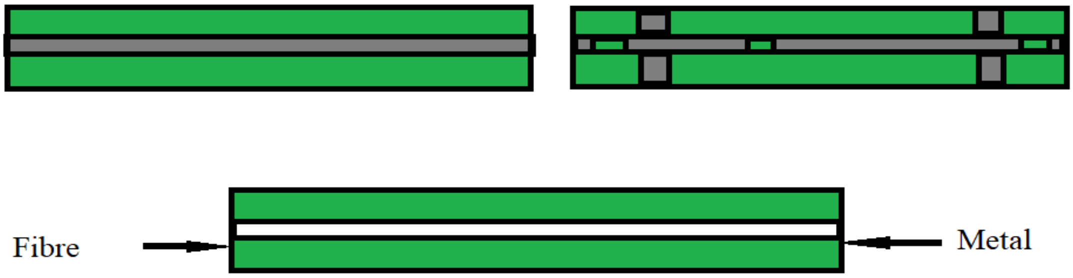


Fig. 30. Hybridisation technique (i) Inter-ply (ii) Intra-ply (iii) Fibre metal laminates, adapted by [15]

8. Critical aspects of natural fibres exposed to different natural environments and their influence on the impact of damage characteristics.

The various environmental factors that affect the impact damage of natural fibre composites are explained below in the following sections.

8.1. Effect of temperature

Temperature is a critical factor in environmental service conditions and plays a vital role in the impact of composite materials [307]. Only a few reported works on the effect of temperature and residual strength assessment for natural fibre hybrid composites subjected to low-velocity impacts. According to Suresh Kumar et al. [308], the impact damage of hemp/basalt fibres has adverse effects with an increase in temperature. Also, they observed that at 50 °C, hemp and hybrid/epoxy composites performed better than the basalt/epoxy composites. At the Same Time, Mueller [309] also noticed that all composites regardless of any type of fibre showed comparable performance with a maximum impact strength in the medium temperature range. Also, there is a more or less distinctive decline in the lower and higher processing temperatures. An increased processing temperature leads to a lower viscosity and improved flowability of the binding portion. This leads to the better embedding of fibre during consolidation and thus to more excellent composite stability. However, Mueller [309] also stated that a comparatively large diameter and rough surface morphology, natural fibres often counteract free flow during the consolidation of the binder portion. Therefore, for higher shares of natural fibre, a lower binder viscosity is essential for reasonable fibre embedment. Thus, the peak value of impact strength is attained at higher temperatures (Fig. 31), and the reinforcing influence of improved fibre embedding is more prevalent over the weakening thermal decomposition.

Shen et al. [310] noticed that moderate high temperatures could improve the impact damage of flax fibre composites. Dhakal et al. [311] investigated the temperature, and the impact response of jute fibre reinforced unsaturated polyester (UP) composites. They observed that jute/UP composites tolerate higher loads at 30°C. David-West et al. [312] identified that there is a certain amount of plasticity seen in the after impact state of the samples at higher temperatures in natural fibre – styrene polyester matrix composites. In flax fibre composites, they noticed the sudden drop in load at temperatures tested at 40 °C and 60 °C respectively. Perhaps this could be associated with some damage by loss of stiffness and energy stored in the composites, which later may be subsequently dissipated. Singh et al. [312] investigated the effect of curing temperatures ranging from 80 °C to 130 °C on different samples of natural fibre reinforced composites. Based on the experimental results, the author found that changes in the curing temperature reduces the impact strength, but changes tensile strength and flexural strength and subsequently decreases, reaching a maximum value at 100°C.

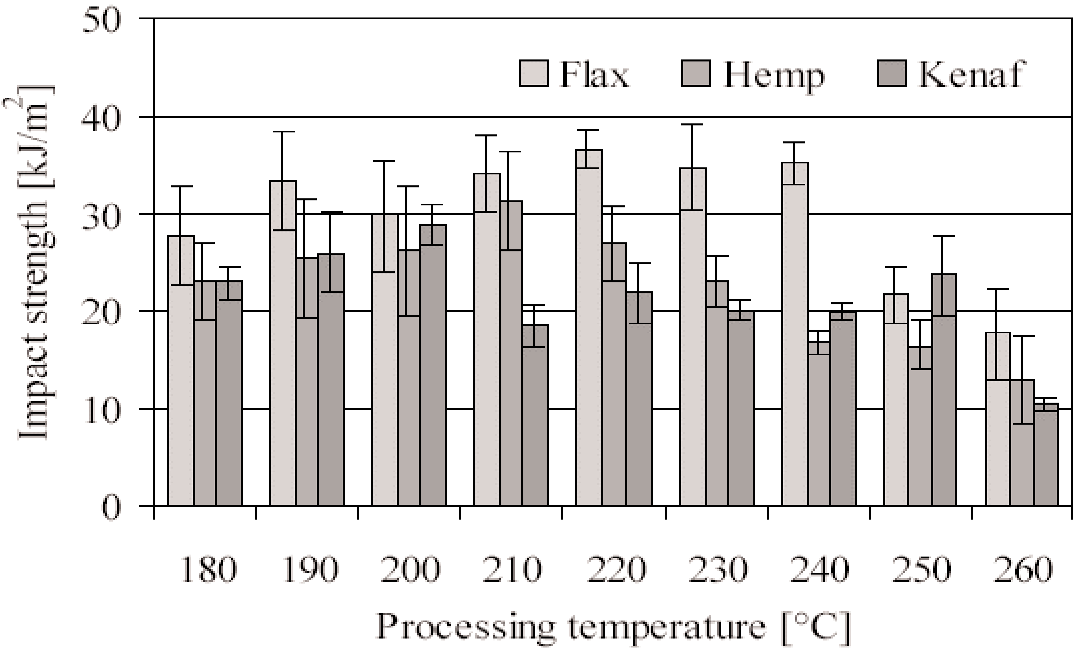


Fig. 31. Impact strength versus Temperature of composite manufacturing improved by various natural fibres [309]

8.2. Effect of moisture absorption

The effect of water absorption mainly depends on the particular factors such as fibre volume fraction, types of matrixes used, absorption time and temperature. Numerous works have been reported on the effects of moisture penetration related to NFPC [313–317]. Živković et al. [318] investigated the influence of basalt and flax fibre reinforced hybridised with vinyl ester composites. They noticed that a higher percentage of water absorbed by the flax fibre reinforced (5.92%) compared to basalt fibre reinforced composites (0.70%). In the case of impact tests, due to increased ductility, flax fibre showed better performance in impact resistance after accelerated ageing. On the other hand, basalt protection showed the lowest moisture intake and better residual in fibre/matrix adhesion. However, similar results were also obtained by Fiore et al.[319]. They observed that basalt layers as protection could improve the ageing resistance of flax fibre reinforced composites. More recently, Wang et al. [320] studied the hydrothermal and ageing properties of flax/carbon hybrid composites. It was observed that the water content and diffusion rate of the water molecules in the reinforced flax/carbon composites showed better performance than the flax fibre composites. Besides, the addition of carbon fibre decreases flax fibre content and provides a barrier to water molecules, enhancing FFRP's hydrothermal ageing behaviour. Dhakal et al. highlighted that the addition of carbon fibre with flax substantially reduces the amount of water absorbed [321]. Islam et al. [322] stated that the impact strength of both treated and untreated hemp-PLA composites increases due to the hygrothermal ageing is attributed to the swelling of fibre surface due to the moisture absorption, thus improving the frictional work of fibre pull-out from the PLA matrix.

9.0. Application of natural fibres with hybrids in structural and non-structural components

9.1 Automotive applications

Hybrid composites have boomed around the global market in recent years as an alternative material for conventional composites [323]. For example, the automobile industry has implemented the hybrid composites in many of the interior and exterior applications. Also, The growth of nature fibre polymer composite is humongous in engineering fields. Many well-known automobile industries such as german auto companies (BMW, Audi groups, ford, and Mercedes), proton company, and the Cambridge industry have been using these NFPC for their production to date. Apart from automobile industries, the NFPC used in construction industries, aerospace, and windows frames [29]. Hung et al. [324] discovered the impact response of hybrid carbon/glass fibre-enhanced composites for automotive applications. They observed that a hybrid composite with carbon fibre layers on the surface would help to minimize the risk of damage, in terms of damage size and deflection subject to impact load. Likewise, in the case of natural fibres, hybridized with synthetic fibres, the authors, Loganathan et al. [325], experimented with coconut coir and glass fibre to check their unique mechanical properties under various circumstances. They reported that mechanical properties such as impact strength, hardness, and tensile strength showed an excellent performance in natural fibre, which will be useful for automotive and aerospace engineering.

9.2. Military helmets

Over the past two decades, there has been a significant increase in armour materials and designs [264]. Later, composites played a massive role in manufacturing helmets and armour materials due to lightweight in design reinforced with aramid, ultra-high molecular weight polyethylene, and other type fibres, especially in hybrid combinations [259,326,327]. Campbell & Cramer [328] investigated a hybrid thermoplastic composite for fabricating an anti-ballistic infantry helmet. The helmet was constructed by using an inner aramid composite anti-ballistic liner and an outer-carbon fibre thermoplastic shell shown in Fig. 32. The findings of the study show potential for cycle time changes using thermoplastics, but more research will be required to increase heat transfer during pre-heating of the material before formation and to automate multiple process steps.

Recently, many researchers proposed that having natural fibres would be an added advantage in the reduction of the component's weight [329,330]. Besides, NFRP has excellent properties in impact and compression strength. Bajpai et al. [329], experimented a hybrid glass/jute/epoxy composites fabricated by using a hand-lay up method. They observed that 3-layers of glass and one layer of jute resulted in achieving a maximum impact strength pf 72.24 J/m, which can be used to replace the existing industrial safety helmets. However, Murali & Nagarani, [330] also observed similar results in hybridizing the different types of natural fibres, stating that sisal and jute showed better performance in impact and compressive tests. Also, it possesses adequate strength in the hybrid materials and reduces half of the original weight.

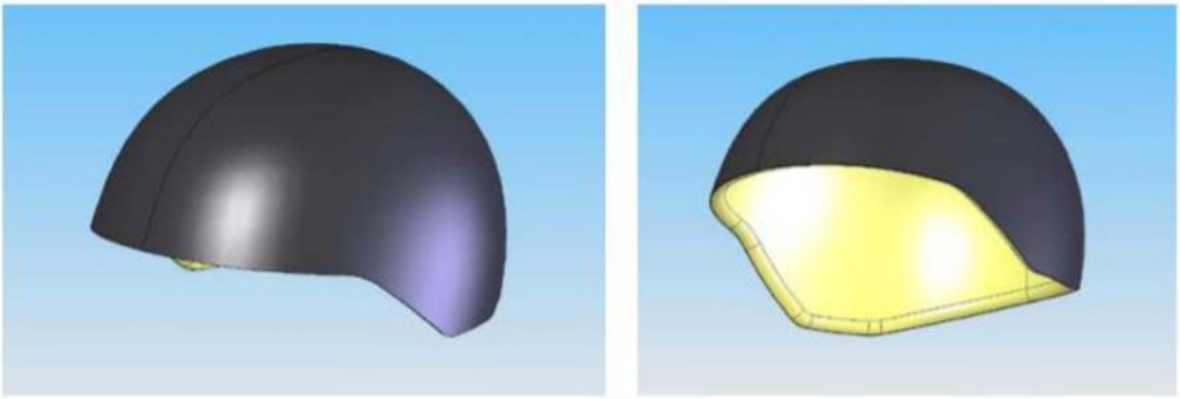


Fig. 32. Hybrid Helmet design [328]

9.3. Bulletproof vest

There are many types of bulletproof vest armour plates made from different types of materials, namely steel, Kevlar, and ceramic and Ultra High Molecular Weight Poly Ethylene (UHMWPE). The development of efficient body armour for better protection against the dangerous ammunition threat has been the main goal for many researchers in this field. The continuous change in military tactics calls for the military body to undergo constant evaluation and improvement. Many experiments have been carried out to examine the efficiency of ballistic-impacted armour-plates using various types of advanced materials ranging from polymeric composites to materials such as metals, ceramics, and aerogel [331]. The para-aramid fabrics have few disadvantages in most body armours, including the large ceramic plates, which are inflexible and restricting the comfort and agility of the wearer. Therefore, to overcome these problems, liquid armours were developed, such as STF. An STF is a non-Newtonian fluid material such as "oobleck" has proven to resist impact loading significantly. There has recently been a growing interest in using this form of material as part of body armour. R, G, & Alexander, [263] conducted a study on different types of shear thickening fluid materials made of oobleck, Polyethylene Glycol & Silica mixture for testing the impact performance. The STF materials were sandwiched between many layers of Kevlar fabrics and manufactured and tested different types of configurations. It was discovered that the samples that contained the STF material were best in resisting penetration due to bullet impact. Besides, many researchers have started to focus on natural fibre. For instance, Da Luz et al.[326] found that even kevlar could cope with the ballistic output of an epoxy composite alongside the jute fibres. By comparing the energy absorption ratio, however, Wambua et al. [327] found that the hybrid structures have a substantial advantage over the composites of mild steel, plain flax, hemp, and jute composites.

10. Conclusions

This paper highlights the various literature articles published in impact mechanics, damage tolerance of natural fibre reinforced composites. Besides this critical review, the paper has identified and discussed numerous factors for damage tolerance and impact resistance of natural fibre reinforced composites such as fibre architecture, geometry, surface treatment and novel improvement techniques. The main purpose is to highlight the numerous factors that could influence the composite's impact resilience and the challenges where the manufacturing application of composites in future could replace artificial fibres with natural fibres. Many researchers portrayed that natural fibres possess weak mechanical properties. With the existing studies, few gaps are observed that can benefit NFPC properties. Moreover, some techniques remain unresolved and challenging for further research. They are explained in the below section as follows.

1. The hybrid technique perhaps is one among the challenges for enhancing the impact characteristics of the NFPC. Though these techniques are already available, it is essential to recognize the strength and rigidity of natural fibres, which is equivalent to synthetic fibres. Therefore, the mechanical and impact strength of ductile fibres is improved by choosing the appropriate matrix reinforcement and by using alternative techniques by combining fibres with metal laminates.

2. There are numerous findings related to impact tests hybridised with synthetic fibres. Many studies suggest that STF plays a significant role in hybridising with synthetic and aramid fibres. However, studies related to STF with natural fibres is yet to be found. Moreover, STF has shown promising results in protection and flexibility. Besides, the STF with composites/fabrics still suffers several drawbacks like evaporation, leakage of carrier fluids, and air-moisture permeability. Therefore, STF evaporation will be a vital issue in the current STF research.

3. Extensive studies are done earlier on metallic coatings on metal and alloy substrates, but much less attention has been paid to polymeric substrates and composite materials. The main advantage of using 3D metal coating is to minimise the damage caused by low-speed impacts, leading to novel findings in the composite structure, for improving impact strength. However, more findings on metal coating technique with NFRPC could improve the fibre impact resistance and toughness characteristics

4. Z- Pinning is a reliable technique to improve the impact toughness of the composites. However, it is quite challenging to predict in natural fibres as its hydrophilic nature may affect the adhesion to a hydrophobic polymer matrix, and perhaps reduces the impact on composite material.

5. Stitching in the composite could improve the intralaminar fracture toughness and the damage tolerance. Therefore, stitching technique enhancement is essential by carefully considering the fibre length, thickness, and volume. Besides, other factors in the stitching may affect the impact properties due to areal density or stitching size. However, the thickness and size of stitching could influence the improvement of the impact toughness of NFRC.

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