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Natural Fibers to Composites

Process, Properties, Structures

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Engineering Materials

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This work is dedicated to the team members of National Center for Composite Materials

Preface

The recent hike in petroleum prices and depletion of non-renewable resources have fostered the research on natural fiber reinforced composites. These composites are a preferred alternative to conventional composites due to their sustainability and environment-friendly nature. However, their market share is limited due to poor mechanical properties of fibers, availability in limited quantities of natural fibers, diversity in fiber structure, moisture absorption, susceptibility to microbial attacks, and Cellulose degradation at higher temperatures around 200 °C.

This book covers the conversion of Natural Fibers to Composites, including the Process, Properties, and Structure relationship. It also addresses the problems discussed, to explore the potential of natural fibers to replace synthetic fibers as sustainable reinforcement material for Green Composites. We believe that this book will be essential reading for professionals and students working in the domain of natural fiber reinforced composites.

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Contents

Alternative Natural Fibers for Biocomposites

Bushra Mushtaq, Sheraz Ahmad, Faheem Ahmad, and Yasir Nawab

Abstract The increasing cognizance of the ecological harm produced by synthetic materials has given rise to the rapid increase in the consumption of environmental-friendly materials. There has been a great upturn in demand for marketable consumption of ecological natural fiber-based composites in several manufacturing industrialized sectors, such as automotive, building construction, furniture, and aerospace in recent years. Natural environment-friendly fibers are renewable resources which are abundant in nature with benefits of low cost, lightweight, renewability, biodegradability, and higher specific characteristic as compared to synthetic materials. The enduring feasibility of natural fiber-based composite materials has increased their usage in several manufacturing sectors. In this chapter, different sources of natural fibers, their characteristics, and morphological structure are discussed in detail. Moreover, the primary use of ecological natural fibers, as well as their effective utilization as reinforcement in polymer composite materials were also summarized.

Keywords Seed fibers · Bast fibers · Leaf fibers · Grass fibers

1 Introduction

Natural fibers are the valuable, ecological, and renewable raw material source in the textile sector characterized as environment and human-friendly materials [1]. Currently, several world researchers, scientists, and practitioners are working on the development of high-quality, sustainable, biodegradable natural fibers-based products for the well-being of the people and society [2, 3]. Natural fibers have numerous advantages like less cost, lower density, lightweight, renewability, and biodegradable characteristics as compared to man-made fibers [4–6]. Moreover, natural fibers are abundantly available which as some promising properties such as high stiffness, high

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modulus, lower health risk during processing, and energy recovery with less carbon dioxide (CO_2) emission [7–10]. Owing to their specific characteristics, natural fibers are used as an excellent raw material in automotive, aerospace, building construction, furniture, packaging, military, railway coaches, and cosmetic industries [11].

Composites are specially designed multi-functional materials exhibiting special functional characteristics which are not found in other materials. Composite materials have interconnected structures formed by substantially combining a minimum of two or more compatible constituents that exhibit different compositions, properties, and, in some cases, forms [12, 13]. Among the different composites, fiber reinforced composites (FRC) have been used in multiple end products for many years, and their market share is constantly expanding. It is well recognized that adding fibers to polymers has numerous advantages, particularly in terms of composite mechanical properties. Synthetic Carbon and glass fibers are reinforced in polymers for use in high-performance end applications such as automobile and aircraft manufacturing industries [14, 15]. Different researchers worked continuously to enhance the performance-based characteristics of the FRC by using various combinations of matrix and reinforcement. However, all these high-performance materials are non-biodegradable and very difficult to recycle. The composites made from these high-performance materials are disposed of by landfills and incineration process which causes environmental impact [16–18]. In recent decades owing to concerns related to ecological issues, the research on the new alternative materials was increased to interchange the traditional FRCs with lesser environmental impact [19, 20]. This will draw attention to natural resources which can be used as reinforcements or fillers in composite materials. These natural fibers-based composites are termed Bio-composites/Green composites or Eco-composites [21]. Netravali et al. were the first to use the phrase "green composite." They developed and tested the mechanical characteristics of the coir fiber-based reinforced Polyhydroxybutyrate-co-volerate (PHBV) resin composites [22, 23]. European researchers referred these materials as biodegradable composites rather than green composites.

The furthermost significant characteristic of green composites is the complete 100% biodegradability. The NFR composites can be entirely resolved into water and carbon dioxide via microorganism degradation in the soil or burn up without the emission of toxic/hazardous gases. As a result, green composites are renewable, environmentally friendly, and biodegradable materials [24].

A view of some natural fibers is shown in Fig. 1. Cotton, jute, flax, sisal, ramie, and hemp are examples of vegetable fibers which are primarily composed of cellulose. These natural fibers can be further categorized as given below:

Seed fibers: These fibers are derived from seeds or seed cases for example Cotton and Kapok.

Bast/stem fibers: These fibers are extracted from the bast that surroundings the stem of the plant. Their tensile strength is higher than other fibers. As a result, these fibers are used to make strong yarn, fabric, packaging, and paper products. Flax, jute, kenaf, hemp, ramie, and banana fibers are an example of bast fibers.

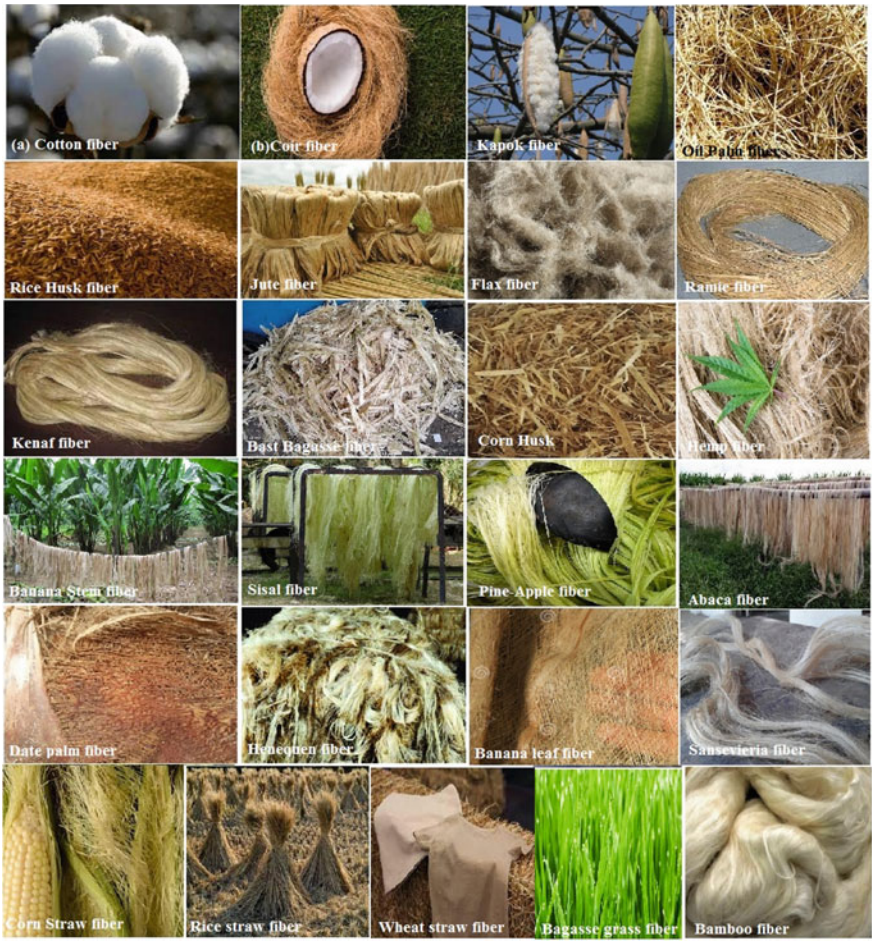

Fig. 1 View of some natural fibers

Leaf fibers: These fibers are obtained from the plant leaf, for example, sisal, pineapple, abaca, etc.

Straw fibers: These fibers are the straw or stalk of the plant, for example, corn straw, rice straw, wheat straw, etc.

Grass fibers: The bamboo and bagasse are examples of the grass fiber.

The comprehensive categorization of the plant/vegetable-based natural fibers is [25] shown in Fig. 2.

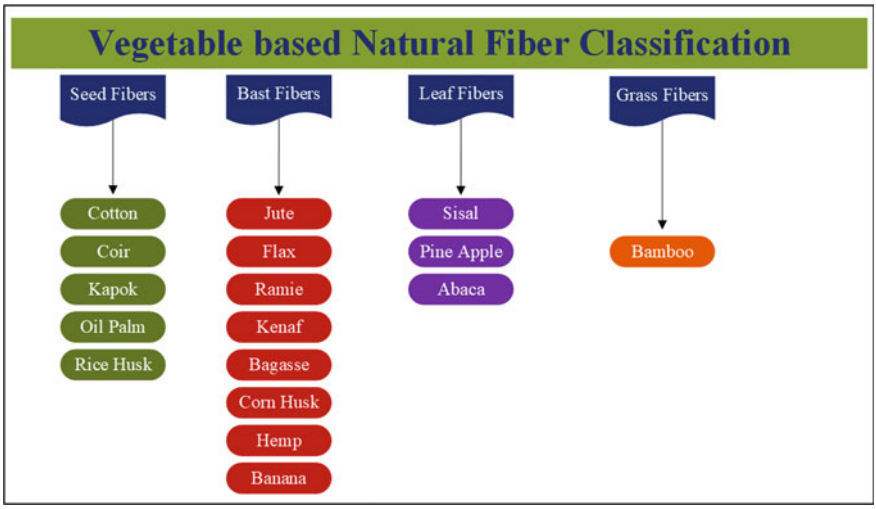

Fig. 2 Classification of vegetable-based natural fibers

2 Seed Based Fibers

*2.1 Cotton (*Gossypium *Genus) Fiber*

Cotton is a soft staple fiber which covers the seeds of the cotton plant (*Gossypium* genus), a tropical and subtropical plant native to the Americas, India, and Africa [26]. China, India, the United States, Pakistan, Brazil, Australia, Uzbekistan, Turkey, Greece, Turkmenistan, and Syria are the top ten cotton producers in the world in 2007 [27]. These countries account for roughly 85% of global cotton production. The diameter of the fiber is approximately 15–25 μm, while the length is approximately 10–50 mm [28]. The waxes and fats on the fiber surface makes it smooth, even and flexible.

Cotton fibers have a low elongation, which help the fabric in shape retention during use. A cotton fiber looks like twisted ribbon-like structure under a microscope. These twists in the cotton are termed as convolutions, and approximately 60 convolutions/cm are present in the fiber structure. The cross-section of cotton fiber is often described as kidney-shaped [29].

Cotton fiber also has several applications in field of interior design. A highly absorbent bath towels and robes, roller blinds, small pillows, and so on are the excellent examples of cotton products [30]. Cotton is normally blended with other natural and synthetic fibers to make high quality comfortable fabrics. Organic cotton is cultivated without the use of insecticides or any chemical fertilizers [31]; cotton from organic resource is used to make more expensive products ranging from tissues to kimonos [32].

2.2 *Coir (*Cocos nucifera*) Fiber*

Coir is the coconut fiber derived from the coconut shell/husk [33]. Coconut fiber is the coarser fiber among the various natural fibers. Coconut plants are mainly cultivated in tropical macroclimates [34]. In the world the coir fiber production is estimated to be 250,000 tones [28]. Coir fiber, in particular characteristics, is a light weight and strong/durable fiber that has gained scientific and commercial importance due to its exceptional characteristics [35]. Its high micro-fibrillar angle, imparts valuable characteristics such as resilience, resistance to weathering, strength, damping, and high breaking elongation [36]. The single fiber internal structure is slender and hollow, with thick walls of cellulose, and every cell is approximately 1 mm long and 10–20 μm in diameter [37].

Coir's properties are less affected by wet conditions than those of other hard fibers. The thickness of coir fiber limits the coarseness and weight of coir products [38]. Coir fiber is used to make Brushes, mattresses, bags, ropes, and upholstery in the automotive industry when combined with other materials [39, 40]. In recent developments, the coir fibers are used in roof greening, road embankments, bio-engineering, woven geotextiles, soil erosion control, capping landfills, mining, landscaping, ski slopes/ski lift tracks, re-vegetation, shoreline stabilization, stitched erosion control blankets, etc. [41].

2.3 *Kapok (*Ceiba pentandra*) Fiber*

Kapok is a member of the Bombacaceae family. It grows in tropical climates [42]. Kapok fiber is silk cotton that is yellowish or light brown. Kapok seeds are encased in the fibers. The fibers are composed of cellulose, light in weight, and exhibiting hydrophobic characteristics [43]. Kapok fiber is commonly used as oil-absorbing material, a buoyancy material, adsorption material, reinforcement material, biofuel, and so on [42, 44].

According to various sources, the kapok fiber varies in chemical composition. According to a study, kapok fiber is chemically composed of 64% cellulose, 13% lignin, and 23% pentosan by weight, while another study found that kapok fiber is composed of 35% cellulose, 21.5% lignin, and 22% xylan, with a high ratio of syringyl/guaiacyl units (4–6) and a high level of acetyl groups (14.0%) when compared to normal plant [45, 46]. These variations could be due to the different in kapok sources and different processing techniques.

Optical microscopy revealed a cylindrical shape, and smooth surface fiber of kapok [47]. The oval to round cross-section with a large lumen and thin cell-wall [48]. This hollow fiber structure distinguishes kapok fiber from other natural fibers and exhibiting the porosity of more than 80% [49].

2.4 *Oil Palm (*Elaeis guineensis*) Fiber*

Oil palm fiber (OPF) is considered as secondary material that is extracted from empty bunches which can be used as reinforcement in bio-composites. The fibers can also be gathered from some other parts of the plant, but the other part's fiber yield is lower than that of the fruit bunch [50]. Empty fruit bunches cause waste dumping issues [51]. Abdul Khalil et al. investigated the anatomy, chemical composition, cell wall structure, and lignin distribution, in depth [52]. These fibers are tough, with characteristics similar to coir fibers. The OPF is composed of approximately 29% hemicellulose and 65% cellulose [53].

The longitudinal structure shows the slender shape with the tapering and sealing end. The fiber cross-section is filled with the lumen [53].

2.5 *Rice Husk Fiber*

Rice husk is the rice grain's rigid shielding cover. After the rice harvesting, the grain and husk are further processed to obtain the rice husk. Until late 1800s, husk was a manually separated. Burning of rice husk (ash) produces amorphous reactive silica and it has potential applications in material science [54]. Panthapulakkal et al. investigated the improvement of processing of rice husk/PE composites, as well as swelling, density, water absorption, and extrusion rate [55]. Because of its mechanical properties, rice husk is unsuitable as a filler in a variety of applications [56, 57].

The longitudinal view shows the tip-like structure on the top with micro-bumps. The cross-sectional view indicates the inclined *epidermis* fibers, which resulted in a bulge on the external surface. The region between the bulges and contiguous inclined areas containing the amorphous silica concentrations [58].

3 Bast Based Fibers

3.1 *Jute (*Corchorus capsularis*) Fiber*

Jute fibers (*Corchorus capsularis* and *C. olitorius*) varies in color from off-white to brown. This fiber is obtained from the plant's bast and skin. Jute bushes can reach heights of 1.5–4.8 m and have stem diameters ranging from 1.25 to 2.0 cm [59]. Furthermore, the retting technique can be used to draw those fibers [60]. Jute fibers with an excessive tensile elasticity and occasional extensibility might be grown in 4–6 months.

Jute fibers are attractive owing to their biodegradability, recyclability, and environmental friendliness [61]. These fibers are brittle and have very low creep value. The coarse nature of the jute fiber limits the fineness of the yarn. Jute fiber applications

include packaging, sack material, tapestry coatings, and electrical insulation. This fiber is not appropriate for food packaging applications due to broken hair problem, which may add impurities to the food [62].

On the longitudinal view, the crosswise marks are clear on the fiber surface which are called nodes or joints. A cross-sectional view indicates that the fibers have a small central canal similar to the lumen present in cotton. The sides have a polygonal shape with round edges [63].

3.2 *Flax (*Linum usitatissimum*) Fiber*

Flax fiber is extracted from the stems of the *Linum usitatissimum* plant. Fibers are approximately 1 m tall and 2–3 mm in diameter [64]. Flax fiber, like cotton fiber, is a cellulose based fiber, with more crystalline structure. This results in stronger fiber which are stiffer to handle, and more easily wrinkled. The molecular fine structure of flax fiber determines its properties, which are influenced by cultivation conditions and the type of retting procedure [65].

Their diameter is about 20 μm. Flax fibers are much less twisted than cotton fibers with a lumen in the centre [66]. On longitudinal images of fibers, several dislocations are seen, that are regions of the cellular wall in fibers where the path of the microfibrils differs from the microfibril angle of the encompassing secondary wall. These distortions appear during the extraction process. Fibers may be polygonal, oval, or irregular in form. The shape of fibers is decided via fiber length, plant growth situations, and adulthood [67].

3.3 *Ramie (*Boehmeria nivea*) Fiber*

Ramie (*Boehmeria nivea*) is an Asian bast fiber that is ordinarily produced in China and Brazil. It has been grown for hundreds of years in China and is commonly called "China Grass" [68]. It's far a perennial herbaceous plant in the Nettle family that can be harvested three to six times consistent with year. The plant lives for about 7–20 years and grows to a top of 1–2.5 m [69]. Due to the presence of gum, pectin, and different substances in the bark, chemical remedy is required before using the fibers [70]. Nam et al. investigated the physical characteristics of ramie fibers, and the composites were created through the identical authors [71]. To create bio-composites, ramie fibers are used as a reinforcement in variety of thermoset and thermoplastic resins [72–74]. Ramie fibers are less explored than the alternative noted bast fibers due to the aforementioned problems (availability and impure). Summerscales et al. and other researchers reviewed bast fibers and their use as composite reinforcements [66, 75].

Fibers are oval to cylindrical in form and white with a high luster. The surface of the fibers is rough, with small ridges, striations, and deep fissures. Ramie fiber is outstanding by way of its coarse, thick cell wall, loss of twist, and surface properties.

3.4 *Kenaf (*Hibiscus cannabinus*) Fiber*

Kenaf fiber is derived from the *Hibiscus cannabinus* plant stem, which has been grown as part of the ISKARA (intensification of community sack community) program since 1979/1980 [76]. Those plant life are adaptable and may develop on a huge variety of surfaces, such as peat and flooded soil [77, 78] Kenaf fiber production can range from 2.0 to 4.0 tons of dry fiber/ha, dependent on the plant variety and cultivation climate conditions [79]. They thrive in unfastened, nicely-draining soil and are planted inside the same manner as jute. They can be harvested 4–5 months after blooming [80].

There are various capacity unique packages for kenaf complete stalk and outer bast fibers, including paper products, textiles, composites, constructing substances, absorbents, and so on [81]. The lumens are predominantly huge and oval to spherical in shape, and the cross-sections are polygonal with rounded edges.

3.5 *Sugarcane Bagasse Fiber*

Bagasse is the fibrous residue that remains after crushing sugarcane stalks to extract juice. It changed into primarily a waste product that caused disposal charges for sugar turbines. Due to the low calorific value and sucrose content material of bagasse, strength and bioethanol production is a low-performance system. Bagasse manufacturing in the international is excessive (75×10^6 tones) and is frequently produced in Brazil, India, South Africa, and China.

When as compared to bast and leaf fibers, bagasse fiber has lower tensile characteristics. Extrusion, compression molding, and different composite approaches were used to create bagasse reinforced thermoset and thermoplastic composites. Luz et al. compared compression and injection molding in terms of mechanical behavior and microstructural analysis of bagasse composites. Bilba et al. prepared and analyzed bagasse reinforced cement composites. The potential of bagasse fiber and its composites was thoroughly investigated [82–91].

3.6 *Corn Husk Fiber*

Corn is a broadly planted crop in many Asian international locations. Corn plant consists of the stems, leaves, and skins which has excessive ability to be considered a

natural fiber. Amendment of corn husk fiber with 0.5–8% sodium hydroxide (NaOH) solution is understood to reduce hydrophilic properties whilst growing crystallinity, tensile energy, and thermal resistance.

The addition of corn husk fiber to polymer composites can improve their tensile strength, bending strength, and toughness properties. Despite being immersed in water and exposed to ultraviolet (UV) light, the mechanical properties of the corn husk fiber composite were significantly higher than those of the "pandan wangi" fiber-reinforced composite. This corn husk fiber composite has previously been found to be suitable for use as a substitute for wood, soundproofing panels, and building materials in several studies [92–94].

*3.7 Hemp (*Cannabis sativa*) Fiber*

Hemp (*Cannabis sativa*), a species of annual green plant native to Europe and Asia. Hemp fibers are known for their high tenacity and low homogeneity. Hemp fiber has an average length of 17–24 mm and an elementary fiber diameter of 10–17 μm. Hemp is mostly used to make rope. Nonetheless, its fibers are increasingly being used in clothing and technical products such as composites. Hemp is also used in the food (hemp seed oil) and cosmetic industries.

The fibers have thick walls and a polygonal cross-section with rounded edges. The fiber is roughly cylindrical in longitudinal view, with surface irregularities and lengthwise deformations caused by dislocations. The fiber ends are slightly tapered and blunt. Hemp fibers are coarser than flax fibers and more difficult to bleach. The fibers are extremely moisture resistant and rot very slowly in water [28, 95, 96].

*3.8 Banana (*Musa acuminate*) Fiber*

Musa plants (*Musa acuminata*) are native to South East Asia and are members of the Musaceae family. This plant produces biomasses such as bunches, pseudo-stems, leaves, and stalks, which are classified as useful materials with high fiber content. In tropical countries such as Malaysia and South India, banana is widely available. Banana trees are cut down and dried before being processed to extract the fiber. Some of the benefits of banana fiber include its high low elongation at break, lightweight, good fire resistance, strong moisture absorption, low density, high tensile strength, and modulus [97–103].

Longitudinal sections reveal parallel-oriented unit cells with tube-like structures made of cellulose, hemicellulose, and lignin. This structure aids in the transportation of water and nutrients throughout the stem. The fracture section shows the fiber's broken parts and entangled microfibrils. It could be due to the fiber extraction process. Furthermore, the fiber has a rough surface due to the presence of hemicellulose, lignin, and waxy components [104].

4 Leaf Based Fibers

*4.1 Sisal (*Agave sisalana*) Fiber*

Sisal fiber is derived from the leaves of Sisalana, a Mexican perennial plant. Sisal plantations can be found in South and Central America, as well as Eastern Africa. In cross-section, the technical fiber appears as a crescent or a horseshoe. Polygonal elementary fibers with a round canal can be seen inside.

It is typically obtained through the decortication process, in which the leaf is crushed between rollers and mechanically scraped. The sisal fiber's length ranges from 0.6 to 1.5 m, and its diameters range from 100 to 300 μm. Because of its high cellulose and hemicellulose content, lower-grade fiber is processed by the paper industry. In the cordage industry, medium-grade fiber is used to make ropes, balers, and binders twine. After treatment, the higher-grade fiber is converted into yarns and used in the carpet industry [63, 105–107].

*4.2 Pineapple (*Ananas bracteatus*) Fiber*

Pineapple fiber (*Ananas bracteatus*) is a South American product. It has an intense luster and fineness. PALF extraction is limited due to low production, making it difficult to use in industrial applications.

The addition of fiber reduced the thermal conductivity and thermal diffusivity of the composite, which is actual for all plant fiber composites [108–111].

*4.3 Abaca (*Musa textilis *Nee) Fiber*

Abaca fibre is derived from the fibrous banana, an evergreen, perennial tropical plant. The fiber is commonly called Manila. The plant is broadly grown inside the Philippines, in addition to on Java, Sumatra, Borneo, and in relevant and South American nations. The plant trunks are reduce as low as possible to obtain the fiber. Plaiting, thick fabrics, fishnets, sails, ship ropes, paper, and eventually creation boards are made from the fiber. The diameter of fundamental fiber is approximately 10–30 μm [112–115].

5 Grass-Based Fibers

5.1 Bamboo Fibers

Bamboo is a considerable resource that has been used in agriculture, handicraft, paper production, fixtures, and structure. Tries have lately been made to provide textile fiber from bamboo. Due to the fact a single bamboo fiber is most effective 2 mm long, it is utilized in textile manufacturing as a fiber package deal [116]. Bamboo is a grass that grows in no time. Environmentally friendly fibers derived from bamboo, which is renewable, speedy growing, degradable, and does no longer require cultivated land, are fee powerful and mainly beneficial for developing in hilly areas.

The fiber cell walls contained almost axially oriented cellulose fibrils. This fibrillar arrangement maximizes the fibers' longitudinal elastic modulus, while lignification increases their transverse rigidity [67, 117–120].

The surface of the single fiber is irregular and stripy with tree bark. The longitudinal direction of single fiber contains a small amount of lumen. Because the lumen is squeaky, degumming caused an intermittent disappearance of lumen length in the fiber structure. The cross-section of single bamboo contains a small round lumen. Moreover, the cell walls of most bamboo fibers were multi-lamellate with multiple layers [121].

Fiber has micro-cavities that make it softer than cotton and indulge the hygroscopic characteristics. The fibers are flexible, ecological, and decomposable. The fiber has the bacteriostatic, antifungal, antibacterial, hypoallergenic, hygroscopic, natural deodorizer, and UV light resistant characteristics. Moreover, the fiber is also exceptionally strong, durable, stable, and tough, with high tensile strength properties. The fibers are mainly used in the textile industry for clothing, towels, and bathrobes due to their versatile characteristics. It is also having applications in bandages, masks, nurse wear, and sanitary napkins due to its antibacterial characteristics. Fibers are also used UV-proof/antibiotic/ bacteriostatic curtains, television covers, and wallpapers, among other things, to decrease the special effects of microorganisms and the harm of ultraviolet radiation on human's skin. Bamboo fibers are furthermore used for decorative purpose [122].

6 Conclusion

Natural fibers are the effective reinforcement material in polymer matrix composites as a result of augmented environmental/ecological awareness. Natural fibers have excellent properties exhibiting materials that can be used in place of manmade fibers in numerous applications. The fibers that are naturally derived from plants and animals often exhibiting the excellent moisture absorption. The incompatibility of these fibers with polymers turns out to be the main disadvantage. To overcome this

problem, natural material characteristics had been modified through chemical treatments of natural fibers. The chemical modification can improve the adhesion among the fibers and the resin with enhanced mechanical characteristics. Natural fibers can become the major reinforcement fibers for the composite and the capability of replacing synthetic fibers in various applications.

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Treatment of Natural Fibers

Khubab Shaker, Azam Ali, and Jiri Militky

Abstract Natural fiber-reinforced composites (NFRC) are being widely researched to provide a sustainable and eco-friendly solution. Natural fibers are an annually renewable resource and provide additional advantages of low density and biodegradation. However, there are certain limitations associated with these fibers, e.g., moisture absorption, flammability, low strength, etc. To overcome these limitations, natural fibers are subjected to various physical and chemical treatments. The current chapter reviews different treatments used for natural fiber and their effect on the performance of NFRC. The chemical treatments for hydrophobicity include silane treatment, acetylation, etherification, benzoylation, dicumyl peroxide, and enzyme and peroxide treatment. Fiber treatments for other functional properties including flame retardancy, electrical conductivity, and antipathogen properties are also discussed in the chapter.

Keywords Treatments · Natural fibers · Flame retardancy · Hydrophobic · Electrical conductivity · Antibacterial

1 Introduction

The twentieth century is attributed to profound political changes, tremendous industrialization, and technological innovations. A key area of technological innovation was the introduction of new materials, as a replacement for conventional metals. The research on coal tar derivatives and theoretical understanding of complex molecular structures resulted in the manipulation of molecules to produce materials with

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desired properties. The linear polymers were used to produce stronger fibers and as film formers (paint), while bulky polymers were used to form solid plastics. These materials replaced conventional materials in construction, automotive, household products, textiles, agriculture, packaging, and other areas.

The greater dependence on petroleum-based materials and their products has not only resulted in the rapid depletion of these resources but has also created environmental issues like GHG emissions and waste management. The Earth is under threat from a massive amount of waste produced as a result of irresponsible production and consumption [1]. Many attempts are being made to address these issues, including the use of sustainable and biodegradable materials. A potential replacement is the natural fiber-reinforced composites (NFRC). These composites are fabricated using a natural fiber as reinforcement material. Natural fibers are an annually renewable resource and provide additional advantages of low density and biodegradation [2].

Natural fibers are environmentally beneficial reinforcements for composites that can be used in a variety of applications. These fibers (such as jute, sisal, kenaf, flax, jute, coir, and others) provide numerous advantages over synthetic fibers since they are sustainable [3]. Cost-effectiveness, renewability, reusability, low density, excellent thermal characteristics, lower tool wear, non-irritation of the skin, and better energy recovery are some of the distinctive properties of natural fibers. The satisfactory mechanical performance of NFRC, including flexibility, maximum breaking force, flexural modulus, impact resistance, acoustical absorption, manufacturing adaptability, and crash behavior, boost their demand for vehicle components [4]. Thermoset or thermoplastic polymers are reinforced with biocomposites. Numerous researchers have looked into a thermoplastic matrix-based NFRC for transportation applications.

2 Natural Fibers

Natural fibers are attracting more study attention as a result of their possible usage in advanced applications. These fibers are obtained from a variety of sources, including plants, animals, and mineral [5]. Figure 1 depicts their brief classification of natural fiber according to their source. Jute, sisal, flax, bamboo, hemp, and coir are the most regularly cited natural fibers in the literature for composite materials [6]. However, researchers are also exploring the potential of other natural fibers to serve as reinforcement for the composite materials.

The key properties of these natural fibers include their density, diameter, fiber length, tensile strength, modulus, breaking elongation and moisture absorption. These properties determine the performance of resulting NFRC, and are compared for some selected natural fibers in Table 1.

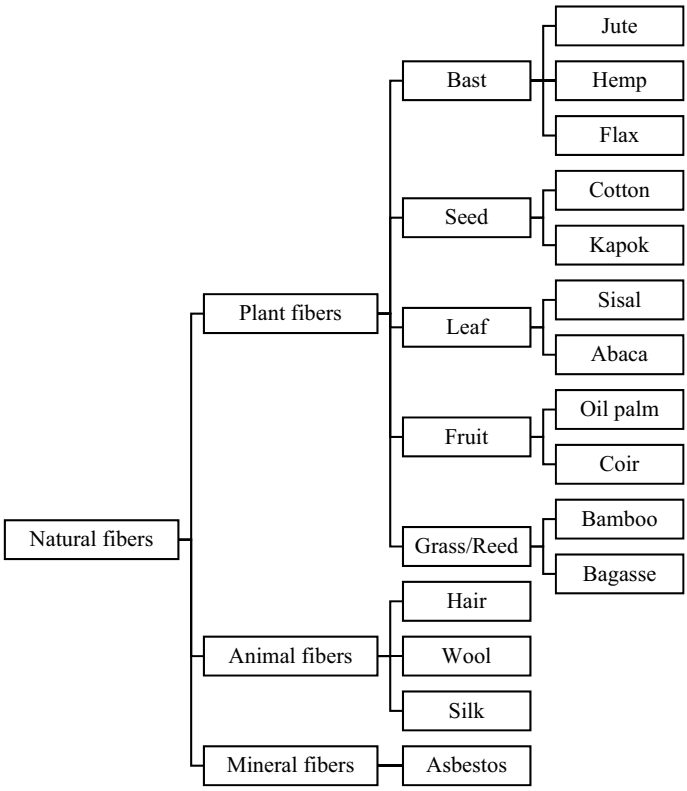

Fig. 1 Classification of conventional natural fibers [7]

2.1 Cotton

Cotton is a seed fiber maninly composed of cellulose and hemicellulose. Except for cotton, almost all fibers are primarily composed of lignin, cellulose, hemicelluloses, waxes, and several water-solvent substances. Because plant fibers have a higher cellulose content, they have better mechanical characteristics and yield strength [10]. Due to the presence of hydrogen bonds and various interactions in cellulose, the cellulose content provides the strength and stiffness to the fiber [11]. The mechanical properties of fibers are largely determined by the manufacturing method, final composite production process, and chemical processing [12].

Table 1 Properties of selected natural fibers [8, 9]

Fiber	Density (g/cm^3)	Diameter (μm)	Length (mm)	Tensile strength (MPa)	Young's modulus (GPa)	Elongation at break (%)	Moisture content (%)
Coir	1.2	7–30 (21)	0.3–3 (1.65)	175	6	15–25 (20)	10
Abaca	1.5	10–30 (20)	4.6–5.2 (4.9)	430–813 (621.5)	31.1–33.6 (32.35)	2.9	14
Sisal	1.2	7–47 (27)	0.8–8 (4.4)	507–855 (681)	9–22 (15.5)	1.9–3 (2.45)	11
Pineapple	1.5	8–41 (24.5)	3–8 (5.5)	170–1627 (898.5)	60–82 (71)	1–3 (2)	14
Bamboo	0.6–1.1 (0.85)	25–88 (56.5)	1.5–4 (2.75)	270–862 (566)	17–89 (53)	1.3–8 (4.65)	11–17 (14)
Hemp	1.47	10–51 (30.5)	5–55 (30)	580–1110 (845)	30–60 (45)	1.6–4.5 (3.05)	8
Cotton	1.21	12–35 (23.5)	15–56 (35.5)	287–597 (422)	6–10 (8)	2–10 (6)	33–34 (33.5)
Flax	1.38	5–38 (21.5)	10–65 (37.5)	343–1035 (689)	50–70 (60)	1.2–3 (2.1)	7
Banana	1.35	12–30 (21)	0.4–0.9 (0.65)	529–914 (721.5)	27–32 (29.5)	5–6 (5.5)	10–11 (10.5)
Kenaf	1.2	12–36 (24)	1.4–11 (6.2)	295–930 (612.5)	22–60 (41)	2.7–6.9 (4.8)	6.2–12 (9.1)
Ramie	1.44	18–80 (49)	4–0-250 (145)	400–938 (669)	61.4–1228 (94.7)	2–4 (3)	12–17 (14.5)

Note The values in parenthesis are the average values of that property

2.2 Coir

Coir a natural fiber that has attractive mechanical properties [13]. Coir fiber is inexpensive and widely used in a variety of applications. Coconut trees, which have been prevalent in tropical areas, provide coir fibers. Furthermore, the coir comprises flexible lignocellulosic fibers with 40–45% lignin, up to 42% cellulose, and 0.2% hemicellulose [14].

2.3 Jute

Jute fiber has a positive perspective because of its high strength-to-weight ratio and better impact strength. Bangladesh, India, and China are the top jute producing countries. Many applications for jute fiber—reinforced polymer composite materials have

been documented, including I-shaped beams, windows, water pipes, furniture, skateboards, subterranean channel drains, and floor tiles [15].

2.4 Bamboo

Bamboo fibers are abundant in Asia and The Middle east, but their full potential has yet to be explored, despite their status as a standard engineering material [16]. Bamboo has traditionally been used in offices, living spaces, and musical instruments because of its excellent strength to weight ratio attributable to the longitudinal orientation of fibers. Because of the extra lignin content coating the bamboo strands, bamboo fibers offer greater mechanical features but are weaker than other natural fibers. Bamboo fiber is also more elastic than most natural fibers, including sisal and jute [17].

2.5 Hemp

Hemp has been grown in East Asia on a yearly basis for over 12,000 years and possesses exceptional modulus and tensile strength, making it suitable for use as reinforcement in polymer composites materials. Hemp fibers are used in furniture, geotextiles, and the production of pipes [18].

2.6 Kenaf

Kenaf, like other natural fibers, can be used to substitute wood, which is harvested once every 20–25 years, whereas the kenaf plant is harvested twice or three times per year. In four to five months, kenaf plants can reach a height of 3–4 m and have three layers: bast, pith, and core [19]. The kenaf plant accounts for 33% of the bast layer, while the rest is made up of pith and core. The mechanical properties of kenaf bast fiber have been reported to be superior to those of other fibers [20].

2.7 Banana

The banana plant is an abundant source of fruit; however, it fruits only once during its life span. The plant is usually cut and disposed after getting the fruit, though cellulosic fibers can be extracted from its stem by mechanical decortication [21]. Researchers have used the extracted fibers as reinforcement with different matrices, and investigated their mechanical performance [22]. The specific modulus of banana

fiber reinforced composites was 2.39 GPa, which is comparable to that of glass fiber reinforced composites. It was reported that the fiber length has a direct effect on the properties of composite material [23].

3 Different Treatments for Natural Fibers

The treatments used for natural fibers can be categorized into physical and chemical treatments.

3.1 Physical Treatments

To explore the potential of using cellulosic fibers for high-end applications, it is necessary to understand the structure and properties of the natural fiber. The cellulosic fibers have a high hydrophilicity, which results in poor processability, mechanical qualities, and a porous structure, limiting their engineering potential. Hydrophilicity also limits the use of textiles, particularly in transportation and packing [24]. The varying treatments are another important component in NFRC manufacture that virtually impacts their characteristics and interfacial behavior. Before chemical processes, natural fibers undergo a variety of physical treatments. Fiber Beating, Corona Discharge, Plasma Treatment, Heat Treatment, and Ultraviolet (UV) processes are some of the treatments [25].

3.2 Chemical Treatment

Due to various hydroxyl group from lignin and cellulose, chemical treatments have a significant impact on NFRC's mechanical behavior. Chemical treatment techniques frequently focus on reagent functional or active groups that can respond more effectively to natural-fiber structures while also eliminating non-cellulosic material from the fiber [26]. Furthermore, hydroxyl groups produced by chemical treatments may interact with hydrogen bonds inside cellulose, confining to the matrix. As a result, chemical modifications activate these groups or introduces new groups that are capable of interlocking with the matrix and provide good adhesion. A number of surface treatments have been used to improve fiber characteristics, including acetylation, silane, alkali, and peroxide treatments. The effect of these treatments on the properties of natural fibers are given in Table 2.

Table 2 Effect of chemical treatments on properties of natural fibers [27]

Treatment	Effect
Acetylation	Enhanced flexural and tensile properties
Alkali	Improved fiber-matrix adhesion and high thermal stability, due to removal of lignin
Enzyme	Removal of lignin
Benzoylation	Enhanced hydrophobic behavior
Grafting	Improve hydrophobicity and mechanical properties and UV protective properties
Mercerization	Reduction in moisture absorption and improved mechanical performance
Isocyanate	Surface modification
Ozone	Affects contact angle and surface energy
Peroxide	Reduce moisture regain
Methacrylate	Enhanced tensile and flexural strength
Plasma	Improved hydrophobicity
Silane	Improved hydrophobicity, and mechanical performance
Sodium chloride	Enhanced tensile strength, modulus and breaking elongation

4 Treatments to Impart Electrical Conductivity

There is a growing interest in the utilization and fabrication of electrically conducting polymer composites, in many commercial applications. The fact that NFRC can demonstrate combined features like efficient insulation and high rates of suitable mechanical strength, allowing them to be good mechanical reinforcement for field carrying conductors, has sparked interest. Such composites can be employed in a wide range of applications including terminals, switches, connections, circuit boards, insulators, household and industrial plugs, and panel [28].

The dielectric characteristics of materials, on either side, have a significant impact on the conversion of electromagnetic radiations into heat. Cotton, nylon lycra, viscose rayon, polyester, and wool are among the textile fibers and textiles that are being used with conducting polymers for super-capacitor, electromagnetic interference, conductive textiles, heating devices, shielding, and antimicrobial textiles. Since a range of tissues are responsive to electrical stimulation, conducting polymers are intriguing for biological applications such as tissue scaffolds for the regeneration or repair of defective or dysfunctional tissues [29].

4.1 Conductive Polymers

With an electrical perspective, conducting polymers outperform other electro-active biomaterials (such as photovoltaic materials, electrets, and piezoelectric). They have

Table 3 commonly used conductive polymers with abbreviations [13, 32]

Sr. #	Name of polymers	Abbreviations
1	Poly (2,5-thienylenevinylene)	PTV
2	Polypyrrole	PPy
3	Polythiophene	PTh
4	Poly (3-alkylthiophene)	PAT
5	Polyacetylene	PAc
6	Polyisoprene	PIP
7	Poly (isothianaphthene)	PITN
8	Polyfuran	Pfu
9	Polybutadiene	PBD
10	Poly (a-naphthylamine)	PNA
11	Poly (p-phenylene-sulfide)	PPS
12	Polythiophene-vinylene	PTh-V
13	Poly (p-phenylene)	PPP
14	Poly (p-phenylene-terephthalamide)	PPTA
15	Poly (3-octylthiophnene-3-methylthiophene)	POTMT
16	Polyazulene	PAZ
17	Polyaniline	PANI
18	Poly (3,4-ethylenedioxythiophene)	PEDT, PEDOT
19	Poly (p-phenylenevinylene)	PPV

superior electrical stimulus control, a good conductivity to weight proportion, can have great electronic and optical features, and can also lead to the improvement of recyclable, porous, and biodegradable materials [30]. One of their unique features is that their electrical, physical, and chemical, properties may be tuned to meet the precise needs of their applications. Antibodies and enzymes can be used to accomplish this as well as other biological constituents. Furthermore, even after synthesis, such useful features of conducting polymers can be regulated and adjusted through stimulation (e.g., using different ways like pH, light, electricity, etc.) [31]. Table 3 lists conductive polymer materials that may be used to develop conductive NFRC.

5 Treatments for Antipathogen Composites

Natural fibers being a bio-based product are biologically degradable by the action of fungi, bacteria, and other microorganisms. The hydrophilic nature of cellulosic fibers makes them highly susceptible for the biotic degradation and microbial decay. Various approaches are used to overcome the decay caused by biotic factors, e.g. imparting the antibacterial activity to the fibers, or incorporation of antibiotic agents

in the NFRC. Jamili et al. [33] used a dye solution extracted from natural resource to impart antibiotic activity in the natural fibers. They bleached the natural fibers using sodium chlorite and then dyed with natural dye extracted from green walnut shells. The effect of this natural dye on the mechanical and antibacterial properties of NFRC were studied. The developed composites showed excellent resistance against E. coli and S. aureus, by inhibiting their growth on NFRC.

Thakur et al. [34] developed NFRC with antibacterial functionality and hydrophobicity. They modified the coir fiber surface using laccase biografting of eugenol, for antibacterial functionality. The antibacterial activity was evaluated by colony forming unit (CFU) method, while hydrophobicity was investigated by moisture absorption test. The treated fibers were found to have improved antibacterial activity and hydrophobicity as compared to untreated fibers. These grafted fibers were used as reinforcement in poly(butylene succinate) matrix, and it was found that mechanical properties of NFRC were also enhanced due to the finer treatment.

Shaker et al. [35] developed bioactive woven flax-based composites, with antibacterial activity. The bio-functionality was imparted in flax fiber composites using ZnO nanoparticles in different fractions. They dispersed these ZnO nanoparticles in the matrix (unsaturated polyester) to get a suspension and impregnated the reinforcement with this suspension. The developed NFRC were tested for bioactivity in terms of antibacterial activity using zone of inhibition test. It was reported that the lowest concentration (0.02 wt%) of ZnO nanoparticles developed antibacterial activity in these composites, showing a clear zone of inhibition.

Tavares et al. [36] reviewed the biofunctionalization of NFRC for biomedical applications. the biofunctionalization is achieved by immobilization of bioactive ligands, nanoparticles, peptides, enzymes, essential oils, etc. on the surface of fibers. However, there are number of challenges in this approach including understanding material properties, selection of proper immobilization technique and selection of suitable bioactive agent. Owing to the bio-based origin, the different extraction technique used for fibers, it is difficult to predict their properties accurately and variations between batches may be observed.

Smiechowicz et al. [37] developed composite cellulosic fibers with antibacterial activity. They modified the cellulosic fibers using nano-silica and silver nanoparticles for potential medical applications. The cytotoxicity and antibacterial activity of the silver nanoparticles loaded lyocell fibers was studied in the human and mouse cells. The outcome of this study showed that the silver nanoparticle modified fibers have excellent antibacterial properties and are safe for human tissues. Such composite cellulosic fibers may be used for the biomedical applications.

6 Treatments for Flame Retardant Composites

NFRC are an environmentally sustainable and cost-effective alternative to petroleum-based materials, and they are progressively being used in a wide range of industrial implementations due to its tremendous benefits, including excellent mechanical properties, cost effectiveness, recyclability, and good biocompatibility. These engineered composites, on the other hand, have intrinsic drawbacks, such as enhanced combustibility when exposed to flame initiators or heat flux, which can restrict the range of applications [38]. Consequently, certain initiatives to lessen the combustibility of biomaterials are currently being developed. Combustion of fiber—reinforced composite materials in buildings has the potential to produce life-threatening situations, resulting in significant physical and human damages. Flame-retardants (FRs) are often employed to improve the flammability of NFRC.

Limiting oxygen index (LOI), time to combustion, thermal stability index, random ignition, and ignition point temperature, extinction combustibility index, rates of heat release, smoke toxic effects, mass loss, flame spreading on the surfaces, and flame resistance are key parameters. Polymers burn rapidly due to their high calorific potential. It is possible to modify their fire behavior (e.g., by neutralizing or reducing smoke and heat) by adding FRs [39]. To address fire safety regulations, FRs have now been incorporated in the production of numerous goods for these applications.

In refractory materials for wood, boric acid, halogens (i.e., chlorine or bromine), borax, nitrogen, phosphorus, or inorganic metallic compounds are frequently used. Most FRs compounds now on the market contain mono-di-ammonium phosphate, ammonium chloride, ammonium sulphate, and calcium, boric acid, zinc, borax, aluminum chloride, and magnesium as major chemical contents [40]. FR additive properties give fire resistance in coatings, thermosets, rubbers, thermoplastics, and textiles. The flame-retardant additive disrupts the combustion cycle, lowering the combustion efficiency of the fiber and, in certain situations, extinguish the fire. FR additives could inhibit, decrease, and prevent burning. FRs are chemicals that are added to materials to stop the prevalence of fires, promote thermal stability, or suppress ignition [41].

6.1 Properties of FR in Fabricated Composites

The incorporation of FR into adhesive and coating the fibers with FR before impregnating with an adhesive are two primary methods for developing flame retardant NFRC. A novel design was investigated in this study to enhance the performance of fire-resistant adhesives used as ornamental panels for domestic needs [42]. The orthogonal analysis calculates the quantity of fire-resistant additives in the adhesive, as well as the number of layers, so that the flame-retardant adhesives can boost the effectiveness of flame-retardant film-coated wood sheets during the manufacturing

process [43]. The dyeing, surface bond strengthening, peeling, and formaldehyde release processes accord to local and industrial requirements.

The flame retardancy and thermal properties of NFRC have been extensively studied. Examples of some fire-retardant NFRC are given in Table 4. Dorez et al. [47] investigated the thermal and fire characteristics of natural fibers (bamboo, sugarcane, flax, and hemp) reinforced polybutylene succinate (PBS) biomaterials. When ammonium polyphosphate (APP), was added to flax fibers, it caused a hot hydrolysis of phosphorylation of fibers and PBS which resulted in the formation of a protective film on the biocomposites due to the char development of the matrix and retention of the fiber skeleton, which helped to extinguish the fire.

Kandare et al. [48] investigated the parameters of fire response on flax/epoxy laminates with balsa as a sandwich composite core. Ammonium phosphate was used as a fire-resistant additive, which enhanced the fire resistance of their composites. Lazko et al. [49] investigated the influence of various fire-retardant chemicals on semi-rigid panels (zinc borate (ZB), melamine borate (MMB), melamine phosphate (MMP), and aluminium trihydroxide (ATH)). Flax short fibers and pea protein adhesive were used to make the semi-rigid panels. They discovered that the modifications boosted flame resistance, with melamine borate providing the best results (heat release reduced up to 50% and ignition time enhanced six times from the reference specimen). The addition of flame retardants, on the other hand, tends to diminish the mechanical characteristics of the semi-rigid panels.

The limiting oxygen index (LOI) analysis is another quantitative method for evaluating a fire resistance of materials. It depicts the lowest quantity of oxygen required in an oxygen–nitrogen mixture to sustain combustion reaction of a vertically held specimen that ignites downwards from the top. The more efficient the flame-retardant procedure is, the greater the LOI value. Xue et al. [50] investigated several amounts of ammonium polyphosphate in their kenaf/PLA combination and found that the LOI value improved as the ammonium polyphosphate concentration was increased. Xu et al. [51] used different solutions (boron, nitrogen, and phosphorus) at different percentages to treat hemp fiber. They observed that the LOI of treated specimens increased as compared to untreated specimens, and that the values fluctuated. All of the composite materials modified with fire retardant outperformed, showing the LOI value of greater than 28.

Misnon et al. [52] used alkaline solution (NaOH), flame retardant chemicals, and a mixture of both NaOH and flame-retardant chemicals to affect the physical properties of woven hemp fiber and fabricated composites, resulting in lower mechanical performance. However, as evidenced by limiting oxygen index tests, burning tests, and thermogravimetry analyses, the treatments improved the fire-retardant characteristics of manufactured composites. According to a study based on wood and wood products, the manufactured composites are appropriate for use as building infrastructure resources alternative to wood.

Table 4 Examples of fire-retardant NFRC

Polymer/Reinforcement materials	Flame retardants	Property improvement	References
Epoxy/Cotton	Montmorillonite	Flammability of cotton fiber reinforced composites is enhanced after treatment. No residue after combustion for the control sample, while there was some residue for MMT-treated fabric	[44]
PP/Wood fibers	APP and Silica	APP and silica are good flame retardants for wood fibers/PP composites. Mechanical properties of the treated fabric degraded apart from tensile strength	[44]
Pea protein/Flax short fibers	Zinc borate, (ZB), melamine borate (MMB), melamine phosphate (MMP)	A protein binder was used to incorporate flame-retardant in the insulating material developed using flax fibers. MMB exhibits an increase in flame retardancy behaviors	[45]
PP/Sisal	Zinc borate, $Mg(OH)_2$	The addition of flame retardants in sisal/Polypropylene composites slowed down the combustion process with an increase in temperature. The incorporation of Mg $(OH)_2$ and zinc borate to them enhances its flame retardancy without affecting its mechanical properties	[46]

7 Treatments to Impart Hydrophobicity

Being hydrophilic in nature, all cellulosic fibers absorb moisture from the atmosphere until equilibrium is reached. Natural fibers regain moisture at a rate ranging from 5 to 12%, as demonstrated in Table 1. Moisture absorption affects the interface and mechanical features of the composites by causing dimensional deviations in the fiber and composites. Moisture can cause a poor fiber–matrix interaction during composite production [53]. Due to the obvious inadequate contact, load transfer will be ineffective, and the composite will degrade.

The hydrophilic nature of cellulosic fibers makes them inappropriate with the hydrophobic matrix, affecting the fiber–matrix adhesion. Furthermore, waxy and pectin compounds block the reactive groups of the fiber, making interlocking with the matrix difficult. To improve the efficacy of interfacial bonding, several chemical treatments must be applied to the fiber surface [54]. Acetylation, g rafting monomers, bleaching, and other treatments are used for this function. The thermal stability of natural fibers can also be improved by grafting them with monomers. The coupling agent or compatibilizer can also be employed to pass stress throughout the interface effectively. Compatibilizer is a polymer that has functional groups bonded onto its chain. Chemical treatment exposes more reactive groups on the fiber surface, working as sites for better interfacial adhesion with matrix, resulting in improved mechanical performance of NFRC.

Various surface modifications methods such as sodium chlorite treatment [55], etherification, silane treatment [56], benzoylation, metha acrylate treatment [57], isocyanate treatment [58], acetylation [59], peroxide treatments [60], mercerization [61], plasma treatment [62], enzymatic treatment [63], dicumyl peroxide treatment [63], ozone treatments [64], and Polyolefin oxidation [65] has been shown to enhance incompatibility among natural fiber and polymer matrix surfaces. Natural fibers are pre-treated to help chemically change or clean the fiber surface. Since cellulose and lignin contain hydroxyl groups, natural fibers can be modified. This hydroxyl group participates in hydrogen bonding inside the cellulose, lowering the activity toward the substrate. As a result, a variety of treatments are used to increase the ageing, strength, and fiber matrix adherence of biocomposites. A well-designed interface not only improves the load bearing capacity of composite, but also provides structural stability [66]. The chemistry and nature of the fiber–matrix interactions can influence the characteristics and performance of composites. Good interfacial qualities are required to ensure effective load passage from matrix to reinforcement, reducing stress absorption and improving overall mechanical behavior.

7.1 Methacrylate Treatment

Flax fibers were esterified to render them hydrophobic by Canter et al. [67]. They developed a flax/PP composite with 10% methacrylate (MA) for 25 h at 50 °C. The flax fiber composites had high flexural and tensile strength, according to the research. Kaith and Kalia [68] used 20% by weight methyl methacrylate (MMA) to treat flax fibers for 20 min before employing phenolic resin to create a flax composite. The treated fiber composites absorbed less moisture as compared to the untreated counterpart.

7.2 Silane Treatment

Silanes function as coupling agents and stabilize composite materials by allowing glass fibers to bind to a polymeric matrix. Fiber hydroxyl groups are reduced by silane coupling agents, resulting in a better interface. A hydrolysable alkoxy unit results in the production of silanols, in the presence of moisture. The hydroxyl groups at fiber surface react with silanol to generate stable covalent connections with the cell wall, which are chemisorbed on the surface of the fiber. As a result of covalent bonding, silane prevents fiber swelling by forming a cross-linked network. The reaction has been given as follows. Kalia et al. [69] examined the influence of aminopropyl triethoxy-silane (APS) on cellulosic fiber hydrophobicity. Nevell et al. [70] showed an increase in the mechanical performance and antifungal action for epoxy and UP composites, after silane treatment.

7.3 Acetylation

Acetylation is an esterification technique, causing plasticization in natural fibers, which is commonly used to protect the cell wall of wood cellulose fibers from moisture, and environmental deterioration. According to Yao et al. [71], when acetic anhydride comes into contact with a cellulose substrate, it interacts with the hydroxyl groups while also blocking reagent diffusion. They found an 18% gain in degree of acetylation, as well as significant increases in flexural and tensile strength. When treating flax fiber with 2.5, 5, 10, 15, 18, 20, 25, and 30% sodium hydroxide, Sreekala et al. [60] found that a 10–30% solution provided the highest performance. The optimal concentrations for mercerization were found to be 5, 18, and 10%.

7.4 Etherification

Natural fiber composites can be etherified to make them more functional and improve certain qualities. The nucleophilic addition of benzyl chloride, epoxides, formaldehyde, alkyl halides, and acrylonitrile is facilitated by sodium hydroxide (NaOH) generating a charged intermediates species with the fibers [72].

7.5 Enzymatic Treatment

When enzymes are utilised with mechanical and chemical procedures for material modification, enzymatic treatment is a very important and intriguing phase. Enzymes are effective catalysts that act in a highly specific manner under low-energy

settings. Peroxidases and other oxidative enzymes can be utilised to further functionalize lignocelluloses. In the presence of oxygen, laccase converts phenolic hydroxyl groups to phenolic radicals. Laccase treatment also reduced the lignin concentration of single cellulosic fibers from 35 to 24% [73].

7.6 Peroxide Treatments

Most researchers have explored peroxide treatments for the treatment of cellulosic fibers because they are simple to apply and have good mechanical qualities [74, 75]. According to Sreekala et al., organic peroxides are rapidly degraded to free radical, which then react with cellulose in the fiber and the hydrogen groups in the matrix [60].

7.7 Dicumyl Peroxide Treatment

Fibers (30 g) that had been pre-treated with alkaline solution were soaked for 30 min in a 6% solution of dicumyl peroxide (DCP) in acetone, then decanted and dried. The mechanical characteristics of composite improved, and moisture uptake was reduced significantly [76]. Dicumyl peroxide treatment, benzoylation, and silane treatment, were performed on the treated surface of the fiber.

8 Physical Treatment

Plasma and corona treatments are the most regularly utilized physical treatments for natural fibers.

8.1 Plasma Treatment

Plasma treatment is an efficient approach for altering the surface properties of natural fibers, while leaving the bulk properties unchanged. Both cold plasma and corona therapies can cause plasma discharge. Both are plasma treatment procedures that use an ionized gas with an equal number of positively and negatively charged particles that interact with the substrate surface. The fundamental distinction between the two types of plasmas is the frequency of the electric discharge.

Microwave radiation can produce high-frequency cold plasma, but an alternating current discharge at atmospheric pressure can produce corona plasma. The type of ionized gas influenced the surface changes of synthetic polymer surfaces and wood. Furthermore, the number of the polar constituents of surface energy of pine wood for

plasma modifications has recently been explored, which comprises power, sample distance from plasma source, treatment time, plasma treatment stability, and gas type [77]. Corona discharge was largely employed to treat pulp layers with moisture content of up to 85%.

8.2 Corona Treatment

Corona treatment is by far the most intriguing method for surface oxidation actuation. This treatment alters the surface energy of the cellulose strands, resulting in better suitability between fibers and the hydrophilic matrix [78]. Plasma treatment has been successful in removing dust particles from fiber surfaces, resulting in a better fiber surface. For efficient processing, pressing factor, the gas type, and concentration must all be accurately controlled [25]. UV is a relatively recent technique for removing dust particles from plant fiber surfaces. Some parameters (gas type, steam etc.) are uncontrollable in UV treatment. The strands are placed in a container for surface oxidation of fibers during the processing. Furthermore, the UV treatment increases polarization on the fiber surface, resulting in improved fiber wettability and increased NFRPC strength [78].

9 Applications of Natural Fiber Composites

Several industries, including aerospace, automotive, energy, and construction, are being driven by governments and society to produce more environmentally sustainable products and reduce their reliance on fossil fuels. In this regard, the European Commission issued "European Guideline 2000/53/EG," which set a goal of increasing automotive recycling process to 85% by weight by 2005. By 2015, this ratio had risen to 95%. This type of legislation is a major motivator for the use of NFRC. Natural fibers are an appealing choice for enterprises to tackle environmental and socioeconomic concerns in this context. Moreover, the use of natural fibers would stimulate the economy in rural and underdeveloped areas, thereby contributing to the United Nations' sustainable development goals of eradicating poverty, sustainable industrialization and building inclusive and constructing sustainable communities and cities, and responsible consumption and production. As a result, natural fibers can play a critical part in the economic growth of our society. This section presents and discusses areas in which natural fibers are now in use, where they can be utilized, and what the future brings in terms of their applicability across a wide range of industries.

The growing emphasis on renewable raw materials and product reusability or good biocompatibility is forcing a shift away from petroleum-based materials towards natural fiber materials in automotive [79]. NFRC have been used in the automotive industry since the 1940s, when Henry Ford constructed the first composite materials in a car employing hemp fiber. The next related implementation was in the 1950s, with

the development of the East German Trabant's body, and other manufacturers, such as Daimler–Benz (1994) and Mercedes (1996), followed suit. NFRC have a lot of potential in the automotive industry since people are looking for lighter, more ecologically friendly components. NFRC have been shown to reduce the weight and cost an automotive part by 30% and 20%, respectively. According to those researchers, light weight components result in lower fuel consumption, better recycling options, less waste disposal, and reduced greenhouse emissions, which are all important factors in the usage of natural fibers. Interior sections like parcel shelves, as dashboard, backrest, seat cushion, door panel, and cabin lining are generally made of NFRC, but external uses are rare.

10 Conclusions and Future Trends

The natural fibers are a sustainable source of reinforcements for polymeric composites. Researchers are exploring new sources having potential for fiber extraction from Banana, Vakka, Date palm, Okra, Palm leaf, Pineapple, Piassava, Agave, etc. Also, they are exploring the potential fibers from husk and straws, bark of plant and even agrowaste. All these fibers are obtained as a secondary product and no significant impact on the eco-system is expected. However, the moisture absorption, low strength and attack by micro-organisms are the main limiting factors in the application of these fibers. The different physical and chemical treatments have helped to overcome these problems. Researchers are exploring newer treatments as well, to get the maximum benefit of NFRC. The flame retardant and electrically conductive composites are relatively newer trends in NFRC. The explorations for developing a multifunctional NFRC will ultimately help to offer more opportunities for application in diverse areas.

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3D Natural Fiber Reinforced Composites

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Abstract Three-dimensional (3D) natural fiber reinforced polymer composite materials are gaining popularity in the modern world due to their environment-friendly nature, lightweight, low cost, life-cycle superiority, and biodegradability. Natural fiber reinforced composites are widely used in various engineering applications, and this research area is constantly evolving. However, due to the inherent properties of natural fibers, researchers face numerous challenges in the development and application of natural fiber reinforced composites. These difficulties include fiber quality, thermal stability, water absorption capacity, and compatibility issues with polymer matrices. Ecological and financial concerns are stimulating new research areas in the field of natural fiber reinforced composites. Also, significant research has been conducted recently to improve the performance of natural fiber reinforced composites. This chapter explains the types of reinforcement and composite fabrication techniques used for 3D natural fiber reinforced composites. Also, discussed the literature survey of 3D natural fiber reinforced composites to examine the effect of binder yarn, stuffer yarn, and weave design on the mechanical performance of composites. Furthermore, 3D woven-shaped preforms and their associated composites are also discussed.

Keywords 3D woven structures · 3D natural fiber reinforced composites · Sustainable composites · Mechanical performance · Natural fibers

1 Introduction

Composite can be defined as the combination of two or more distinct materials in terms of both physical and chemical nature, these constituents cannot lose their identity, but enhance the overall performance of the composite material. Normally composite is designed for specific jobs such as high strength, lightweight, and resistance to corrosion or electricity. Composite materials showed better properties

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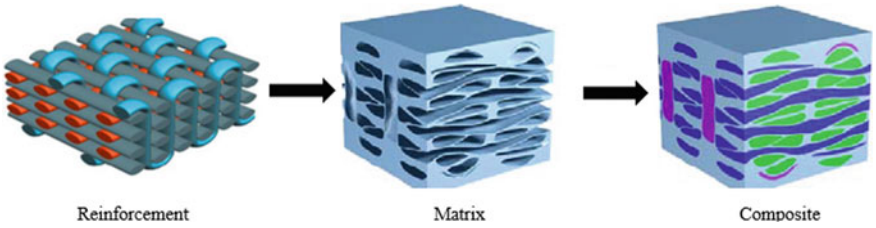

Fig. 1 Composite and its constituents, reinforcement, and matrix [4]

than their constituents in all perspectives, therefore, their applications are increased day by day with respect to conventional materials [1–3]. The main constituents of the composites are matrix and reinforcement. The functionality of the matrix is to protect the composite material from the external environment, dissipate the externally applied load throughout the structure, and keep the reinforcement together. While the contribution of reinforcement in the composite is to provide mechanical strength to the composite. Composite and its constituents are shown in Fig. 1.

The history of composite materials is very ancient, thousands of years ago people use composite materials, the first man-made composite was developed in Iraq by Mesopotamians in 3400 B.C. Ancient societies developed plywood from the combination of glue and wood strips, place the wood strips at one another and joined each layer with another through glue. The same concept was followed by Egyptians to develop death masks in 2181 B.C. from the combination of fabric (papyrus or linen) and plaster. With the time, to increase the strength of boats, mud pottery and bricks they use straw as reinforcement with mud [5]. Around 1200 A.D., the Mongols started to develop composite bows, which were extremely efficient during that period. Pine resin-bonded cattle tendons, bamboo, horn, wood, and silk were used to make these composites. Polymerization enabled synthetic resins to solidify after the industrial revolution because of the industrial revolution, synthetic resin was converted into solid form with the help of polymerization, this knowledge leads to the development of many plastics such as vinyl, polyester and phenolic. Leo Baekeland was the chemist who invented Bakelite [6], it is thermally and electrically insulator, in many industries it is used in the form of composite with other materials where thermal or electrical insulation is required. In 1930, with the invention of glass fiber, the application of composites is rapidly increased, first fiber reinforced industry was also developed in this era. Unsaturated polyester was patent in 1936, but still it is used in the composite industry. In 1960, the composite industry further boosts with the invention of high-performance fibers such as para-aramids. Successful application of composites in the Boeing 787 opened the doors for composites to use in high tech applications [7–9].

2 Reinforcement

Natural fibers, used as reinforcement in composites, are either in the form of 2D or 3D woven structures. Woven structures are dimensionally stable and having better mechanical properties as compared to knitted and non-woven structures, therefore, it is preferred to be used as reinforcement in the composites. The difference between 2D woven structures and 3D woven structures is the presence of yarns in the 3rd axis. 3D woven structures have a significant thickness due to the presence of yarns in the Z-direction. These yarns strongly joined the multilayers together and give more dimensional stability to the structure. To achieve a certain level of thickness in 2D woven structures, multiple layers are joined together, but these layers are independent of each other, therefore delamination can easily occur in 2D woven reinforced composites. 2D woven structures have poor through thickness properties due to absence of yarns in the Z-directions, as the layers are independent from each other so cracks can easily propagate in 2D woven reinforced composites. To address these issues researchers, prefer to use 3D woven structures as reinforcement in composites. 3D woven structures have better through thickness properties due to the presence of binding yarns [10].

Woven structures consist of two sets of yarns which interlace with each other at right angle. Properties of woven structures depend on the fiber's properties, yarn spinning, and weave design. Woven structures can be classified on the basis of weave design, fabric dimensions, weaving technique and end product. A weave design explains the interlacement of warp and weft yarn. Woven structures are broadly divided into two types i.e., 2D woven structures and 3D woven structures.

2.1 Two Dimensional (2D) Woven Structures

2D woven structures or reinforcements have only two dimensions, length, and width while negligible thickness. In 2D woven structures only two sets of yarns interlace with each other to develop a structure as shown in Fig. 2. The warp yarns are normally denoted by X-axis while weft yarns are denoted by Y-axis. Interlocking of yarns give dimensional stability to the structure and improve the mechanical performance of the structure. 2D laminates showed lower resistance to delamination, cracking under low and high-speed impact loading due to their inferior interlaminar fracture toughness.

2.2 Three Dimensional (3D) Woven Structures

3D woven structures or reinforcements can be defined as the interlacement of three set of yarns i.e., warp, weft, and binding yarns. 3D woven structures have a significant thickness due to the presence of yarns in third dimension. 3D woven structures are

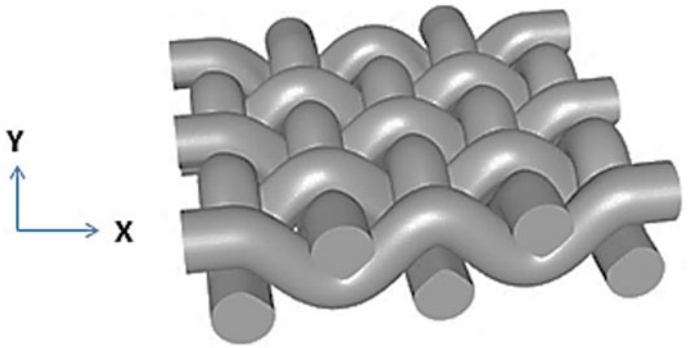

Fig. 2 Two dimensional (2D) woven structure

also called multilayered structures, because it contains more than 2 layers which are joined together through binding yarn, the distance between two binding points is larger than the ground weave of multilayered structure. Figure 3 showed the 3D woven structure, the orange color yarn is the binding yarn while black and blue yarn is the warp and weft yarn in the structure respectively.

A 3D woven structure is formed of warp (0° direction) and weft (90° direction) stuffers that are bound together by a series of warp binders. By varying the binding pattern, different 3D woven structures are produced. The performance of woven preforms depends on the orientation of the binding patterns.

Multilayer 3D woven structures

Multilayer 3D woven structures are classified as:

- Orthogonal interlock structures
- Angle interlock structures

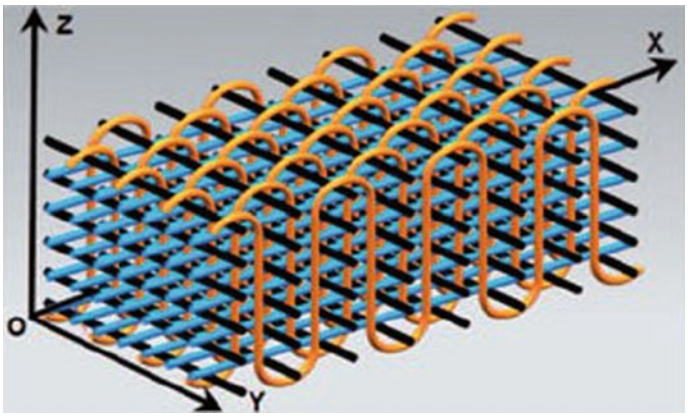

Fig. 3 Three dimensional (3D) or multilayer woven structure [11]

- Hybrid (warp-weft) interlock structures
- Bidirectional interlock structures.

Orthogonal interlock structures

The orthogonal interlock structure is produced from the two sets of yarns, no special binding yarns were used in the orthogonal interlock structure. The warp yarns are used as binding yarn, to join multi layers together. In orthogonal interlock, z-direction yarns are drawn through the warp and weft yarns, intersecting the layers at a 90° angle. The yarns are interlaced homogeneously in each of the three planes to provide quasi-isotropic properties or an unbalanced amount in each direction when anisotropic properties are required [12]. Orthogonal 3D woven structures are further divided into two types, i.e., orthogonal through the thickness and orthogonal layer to layer. In orthogonal through the thickness structure, some of the warp yarns from the top layer move down towards bottom layer and joined all the layers, similarly some of the warp yarns from bottom layer move up towards top layer and joined all the layers that come in the middle. In this way only top and bottom layer is responsible for joining all the layers in through thickness structure. In layer-to-layer structure some of the warp yarns from the first layer interlock with 2nd layer, similarly some of the warp yarns from 2nd layer interlock with 3rd layer and so on. Figure 4 showed the cross-sectional view of through thickness structure while Fig. 5 showed the cross-sectional view of layer-to-layer structure.

Angle interlock structures

Angle interlock structure can be defined as, when the binding yarn move through layers in certain angle then the multilayered structure is called angle interlock multi-layer structure. It is further divided into two types based on the depth of binding yarn. When the binding yarn moves from top layer and pass-through bottom layer then it is called angle interlock through thickness structure as shown in Fig. 6. On

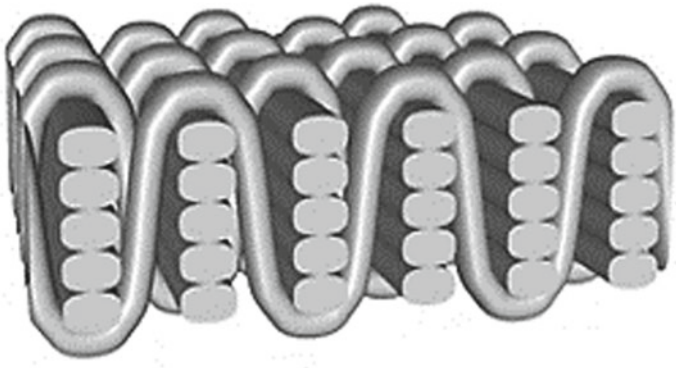

Fig. 4 Orthogonal through the thickness multilayer interlock structure

Fig. 5 Orthogonal layer to layer multilayer interlock structure

the other hand, if the binding yarns move within layers, then it is called an angle interlock layer to a layer structure as shown in Fig. 7.

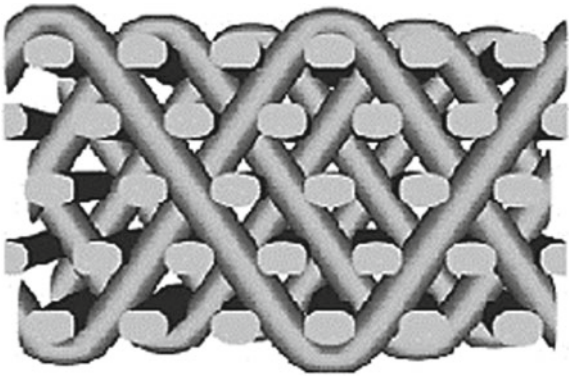

Fig. 6 Multilayer angle interlock through thickness structure

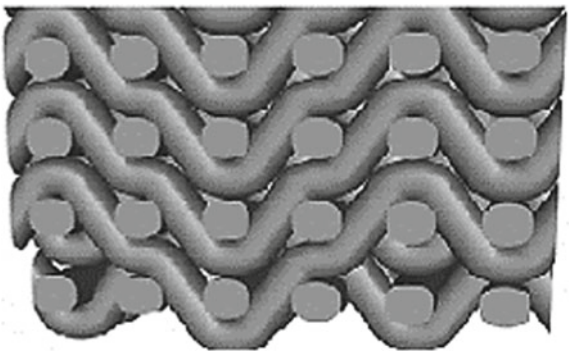

Fig. 7 Multilayer angle interlock layer to the layer structure

An angle interlock structure usually has larger geometrical repeat unit as compared to the orthogonal interlock structure. The mechanical performance of a 3D woven structure is majorly depends on the pattern of binding yarn. Angle interlock structures have higher pliability and forming capability as compared to the orthogonal interlock structure, while in case of fiber volume friction in composite orthogonal interlock structure showed better results as compared to the angle interlock structure. In multilayered structure, the thickness, strength, and stiffness of 3D woven structures significantly depend on the properties of binding yarn. 3D woven structures showed better through thickness, mechanical properties as compared to 2D woven structures.

Hybrid interlock structures

In general, 3D woven structures (reinforcements) are made up of only one type of interlocking pattern. But in hybrid interlock structures, a combination of two, three or four basic types of 3D multilayer interlocking patterns i.e., orthogonal layer to layer, orthogonal through the thickness, layer to layer angle interlock and through thickness angle interlock, can be used. Similarly, hybridization can also be done by combining warp and weft interlocks in one structure, keeping in view the required properties. The specific sequence of yarns placement can also be achieved in both directions. 3D multilayer hybrid structure can be produced by combining both; different types of interlocking patterns and yarn materials keeping in view the cost and target properties. Since interlocking and straight warp (stuffer) yarns help to improve the in plane as well as out of plane mechanical properties of the 3D woven structures. Therefore, these (interlocking and stuffer) yarns could be added in different places of the woven structures to improve their properties. These 3D hybrid woven structures with different types of interlocking patterns can also be produced on conventional dobby/jacquard looms.

Bidirectional interlock structures

In warp interlock structure, as the name suggested that warp yarn is responsible for binding the layers together, while weft yarn remains straight. Therefore, in warp interlock structure, warp yarn has higher crimps %, while weft yarn has negligible crimps %. In case of weft interlock structure, weft yarn is responsible for interlocking of layers. Therefore, in weft interlock structure the crimp % in warp yarn is reduced as compared to the warp interlock structure, while weft yarn has higher undulation or crimp % due to interlocking of layers. Bidirectional structure is the combination of warp and weft interlock structure; therefore, it has maximum crimps % because both warp and weft yarn interlock the structure at alternative sequence. Figure 8 showed the cross-sectional view of bidirectional interlock structure. In bidirectional structure the crimp % in both warp and weft wise is almost same, therefore the mechanical performance of the structure in both direction is almost same. While in case of warp or weft interlock, the structure behaves entirely different in warp and in weft direction due to differences in the undulation of the yarn in both directions. Bidirectional interlock structure-based composites are used in high performance applications due to balance performance of composite in warp and in weft direction.

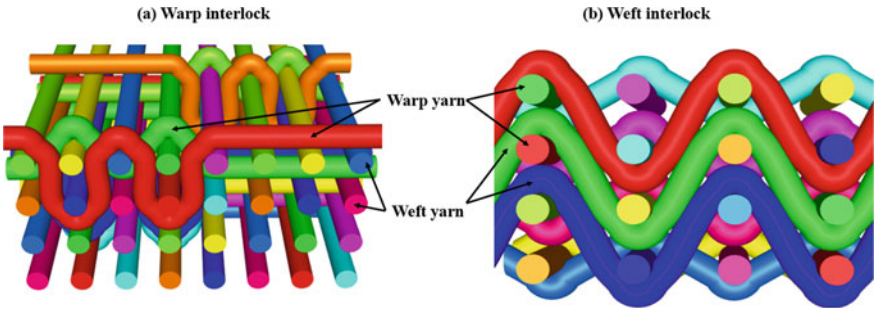

Fig. 8 Bidirectional interlock structure of **a** warp and **b** weft interlock structure [13]

3D shaped woven structures

Shaped weaving is a special type of weaving, in which certain shapes can be achieved as a final product such as V shape, T shape and H shape. In shape weaving warp and weft yarns were used just like in conventional weaving to develop shaped fabric. Shaped fabric can be single or multilayered depend on the required shape, V shape can be achieved through single layer, but T and H shape are only achieved through multilayered structures. 3D weaving loom can be used for shaped weaving, but due to some constraints such as cost, availability, productivity, and lack of training, industry and researchers prefer to use conventional jacquard or Dobby loom for the development of shaped fabrics. Figure 9 showed the cross-sectional view of T and H shaped structures.

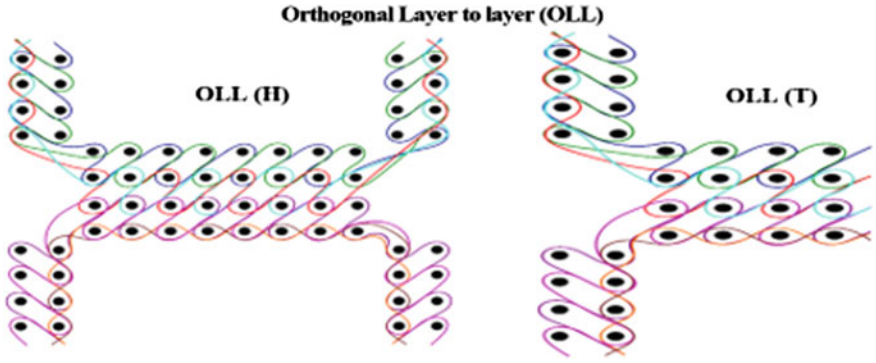

Fig. 9 Shaped woven structures (H shape and T-shape) [14]

3 Composite Fabrication Techniques

Different types of fabrication techniques are used for the manufacturing of composites. Each fabrication technique has its own set of advantages. Some allow for enormous volumes of production in a short amount of time, while others have low capital cost and materials costs. When deciding which composite manufacturing technique would produce the greatest outcomes, an engineer must make an informed decision. Molding is used in most composite manufacturing procedures to form the resin and reinforcement into the desired finished shape. When selecting a composite fabrication process, keep in mind that not all methods are equally suited for all purposes [15]. 3D woven natural fiber reinforced composite fabrication techniques are discussed below.

3.1 Hand Layup

Hand layup technique is the most basic and convenient composite fabrication technique. It is mostly used for thermoset resin because it has lower viscosity so can easily penetrate through reinforcement without need of extra pressure or temperature. In this process reinforcement layers are placed in laminate stack, then thermoset resin is poured on it and distribute throughout reinforcement so that no end remains dry. Wet layup is a variation of this procedure in which the plies are covered with resin before being laid, and the stack is subsequently "debulked" or compressed. The hand lay-up technique was used in 3D woven natural fiber reinforced composites, because in this process the applied pressure is low so the reinforcement can maintain its 3D structure. Disadvantages of hand layup technique are, difficult to achieve finished surface, higher V_f cannot be achieved in hand layup technique [16]. Yasir et al. [17] developed green composites from four layered orthogonal layered to layered reinforcement with green epoxy as a matrix. The hand lay-up technique was used for the manufacturing of composite sample. 20 bar pressure was applied to the sample to remove the air gaps and achieve the constant fiber volume friction.

Kashif et al. [18] used the same hand layup technique to develop composites from jute-based 3D woven reinforcements. Layer to layer structure, through the thickness structure and combination of both structures (Hybrid) were used as 3D reinforcement, to study the effect of reinforcement on the mechanical performance of composite. Umair et al. [19] developed T and H shaped composites from jute reinforcement by using the Hand layup technique. Eight types of 3D structures were developed in the study, orthogonal layer to layer, orthogonal through thickness, angle interlock layer to layer and angle interlock through thickness. From the above literature it can be concluded that hand layup technique is very versatile technique, different types of 3D reinforcement can be used with thermoset resins to develop composites. Figure 10 showed the hand layup process of composite fabrication.

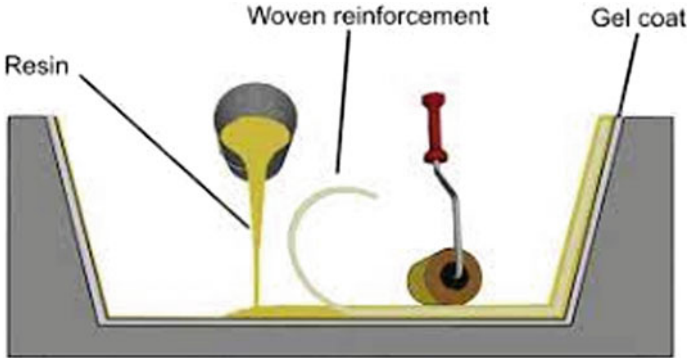

Fig. 10 Hand layup process of composite fabrication [20]

3.2 Compression Molding

Compression molding is used for both thermoset and thermoplastic polymers for composite fabrication. It is used for high scale production and the initial cost of stencil (die) is high but it can be used for long term so it can compensate the capital cost. In compression molding technique the reinforcement is sandwiched between resin and placed in the compression molding plates. Pressure and temperature are increased, resulting melting of thermoplastic resin and evenly distribution of the resin throughout the reinforcement. In case of thermoset resin, the compression molding decreases the curing time with the temperature and pressure, so the overall time of sample development is reduced. In case of thermoplastic resin, the main issue with thermoplastic polymer is high viscosity. In compression molding the applied temperature and pressure reduced the viscosity of the polymer and the pressure pushes the resin to penetrate throughout the reinforcement [21]. For 3D reinforcement the pressure is applied step wise to avoid the deformation of Z-direction yarns in the reinforcement. Imran et al. [22] developed composite sample for impact applications using para-aramid as reinforcement and PVB as matrix, while glass microspheres and silica micro particles were used as micro filler to further enhance the mechanical performance of composites. Composite fabrication technique used in this study was hand layup and then for curing and even distribution compression molding was used. In compression molding the temperature was set at 180 °C and the pressure of 2 tons was applied for 25 min. Umair et al. [23] developed green composite from green epoxy and 3D woven jute reinforcement with novel weaving designs. Novel weave design consists of orthogonal interlock, angle interlock, and hybrid structures. Green epoxy was applied through hand layup technique, and then for further curing and even distribution plates were placed in compression molding machine. The pressure in compression molding was 15 bar for 3 hours. Figure 11 showed the compression molding machine.

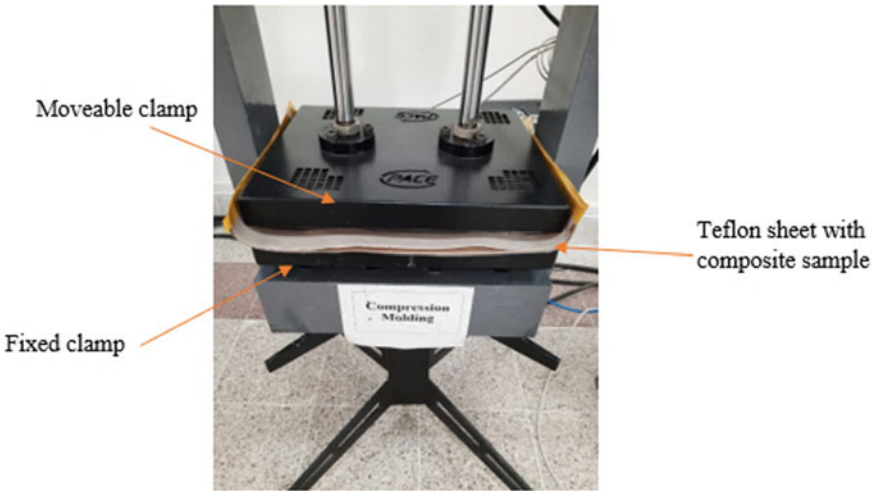

Fig. 11 Compression molding machine

3.3 Commingling Technique

Commingling technique is generally used for thermoplastic composites. Since, thermoplastic matrix based composites have higher impact strength, unlimited shelf life, can easily be reshaped, no need of extra time for curing process. But the main issue with thermoplastic resins is their higher viscosity and which hinders its penetration throughout reinforcement. To resolve this issue commingling technique is used. In this process both matrix and reinforcement are in the form of filament and weaving of these reinforcement and matrix filaments is done simultaneously. After applying temperature, the matrix yarn will melt and easily penetrate in the whole structure at low pressure [24]. Habib et al. [25] studied the effect of fabric structural design on impact and short beam shear (SBS) properties of Jute, hemp and flax fibers based thermoplastic composites. Three types of structures i.e., woven, woven commingled and knitted commingled were studied. SBS and impact tests were performed to evaluate the performance of developed samples. Polypropylene was used as matrix, the density of polypropylene is high so it is difficult to flow through structure, therefore commingled technique was used to resolve this issue and the results showed that commingled based sample showed improved SBS and impact strengths.

Feng et al. [26] used the same concept of commingling technique to form composite samples from pineapple leaf fibers (PALF) and kenaf fibers with polypropylene matrix. PALF and Kenaf both are cellulosic fibers so initially moisture was removed from the fibers to avoids the air gaps or voids issue in the composite. In the next step fibers are mixed with polypropylene fibers at different ratio ranging from 10 to 50%. After mixing the fibers pallets are placed in compression molding machine under 5 MPa pressure and temperature was set at175 °C for 8 min. High temperature melt the polypropylene fibers which joined the Kenaf and PALF fibers

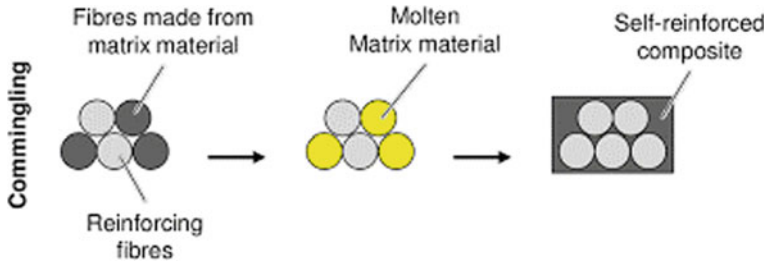

Fig. 12 Commingling process of composite fabrication at fiber stage in self reinforced composite [26]

in the sheet. Commingling can be done at fibers, yarn and at fabric levels by mixing the reinforcement with thermoplastic matrix. Figure 12 showed the commingling technique for composite fabrication.

4 Effect of 3D Woven Fabric Parameters on the Performance of Their Corresponding Composites

4.1 Effect of Z Yarn Stitching Density on 3D Woven Composites

Flax yarn of 38.5 tex was used to develop 3D woven reinforcement and EnviPOXY® 530 (epoxy) was used as matrix. In this work four types of 3D woven structures were developed to investigate the effect of stitching point density on the mechanical performance of the composites. Each structure is divided into three sections, top section (TS), core section (CS) and bottom section (BS). Top and bottom section is orthogonal through thickness while core section is layer to layer as shown in Fig. 13. Repeat cross section of design consists of 12 warp and 60 weft yarns. Each section has two layers, while the core section or middle layer is responsible for joining top middle and bottom layers together. The binding warp yarn (binding point 1) which is present in 3rd layer (L3) of the repeat makes an interlocking point with 2nd layer (L2). This interlocking is referred as stitching or binding points.

Some of the warp yarns in core section were used as binding yarn to stitch the CS layer with TS and BS. These binding points improve the delamination resistance in the structure. Sample SP1 has 3 stitching point in repeat cross section, while SP2 has 5 points, SP3 has 7 points, and SP4 has 9 stitching points in repeat cross section as shown in Fig. 14 [27].

There was 36 $\pm$ 1 and 131 $\pm$ 2 threads per centimeter warp and weft yarns, respectively. The produced fabrics had an areal density of 611 $\pm$ 5 g/m^2 for all woven reinforcement samples i.e., SP1, SP2, SP3, SP4. While plain-woven (P) reinforcement had 12 ends/cm and 42 picks/cm giving an areal density of 200 $\pm$ 2 g/m^2.

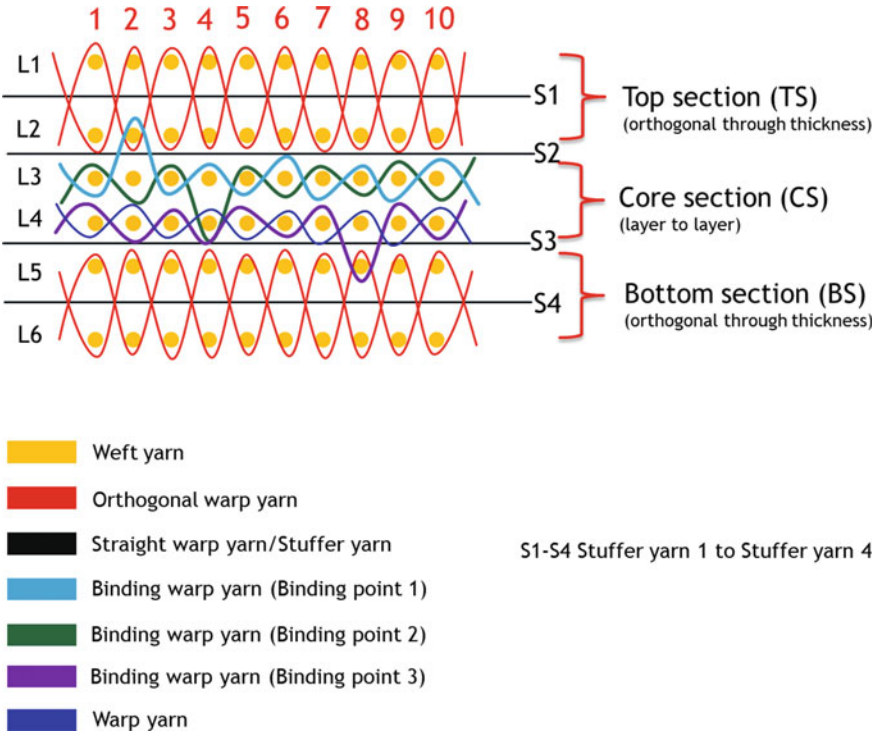

Fig. 13 Schematic of weft wise cross section of 3D woven interlock sample (SP1) [27]

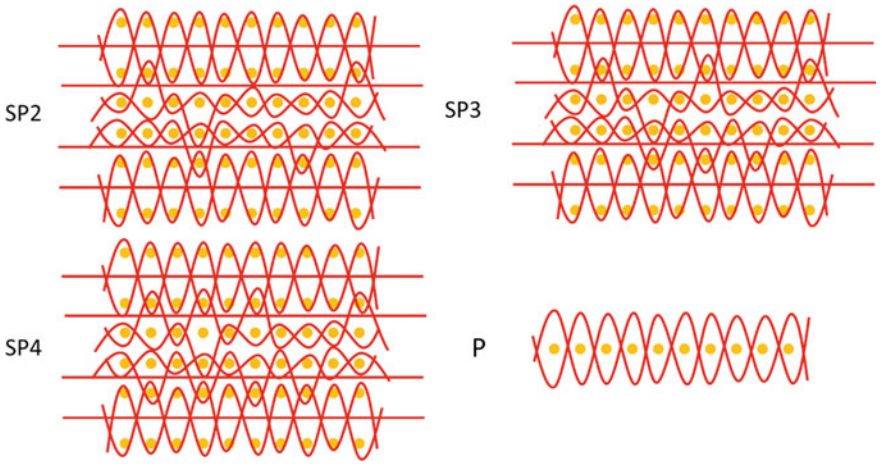

Fig. 14 Schematic of cross sections SP2, SP3, SP4 and P (1/1 Plain) [27]

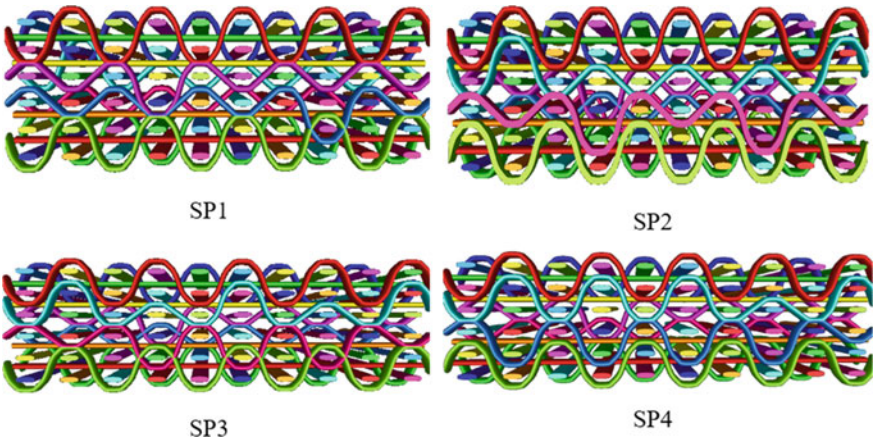

Fig. 15 Yarn architecture of 3D woven samples modelled on TexGen [28]

The warp and weft densities were selected with respect to limit of the loom and ease of manufacturing. It is important to note that in one cm^2 of the SP1, SP2, SP3 and SP4, total stitching points were 20, 33, 46 and 59, respectively. Four composites were produced from 3D interlock woven fabric. Three layers of plain fabric were used to fabricate composite sample for comparison purpose. Yarn architecture of 3D woven samples modelled on TexGen software are shown in Fig. 15.

Tensile strength results showed that the structure having minimum number of interlocking points, or a smaller number of crimps in the yarn has higher tensile strength, because the force dissipation capability of straight yarn is much higher as compared to crimped yarn. Crimp or interlocking points act as weak point because the force accumulate at that point so there is chance that the structure may collapse at that point. Out of four structure SP1 has minimum number of cross-linking points therefore it showed the highest tensile strength results as shown in Fig. 16. From SP2 to SP4 the cross-linking points gradually were increased. Therefore, the tensile strength is decreased from SP1 to SP4 samples. Sample P has the lowest tensile strength results which showed that 2D laminates has poor tensile strength as compared to 3D structures. In weft direction all the samples has higher tensile strength as compared to warp direction due to higher thread density in the weft direction [27].

Similarly, tensile modulus of composites with more stitching points decreases as compared to the sample with low stitching point. SP1 has higher tensile testing results than plain woven sample (P), which showed that 3D woven reinforced composites has better tensile properties as compared to the 2D laminated composites. Furthermore, flexural strength was higher in weft direction due to higher thread density of weft yarns while increased from SP1 to SP4 composite samples as shown in Fig. 17 because SP1 has least number of stitches per cm^2. By increasing the number of joining points per unit area, the stitching yarns tightly hold the weft yarns together and make it more compact. Structure with higher compactness required higher force to bend it, therefore SP4 showed higher value of bending stiffness as compared to the

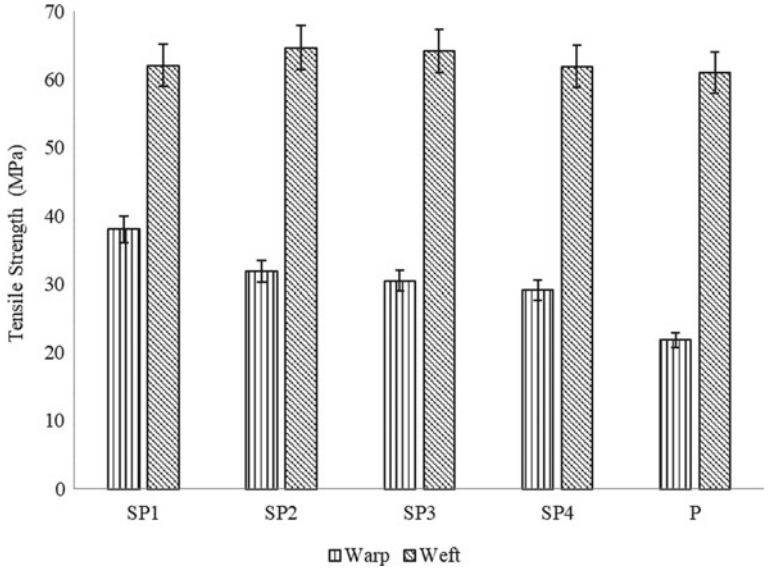

Fig. 16 Tensile strength of 3D woven and laminated composite samples in warp and weft directions [28]

structure that has lower stitching points per unit area. For example, when a structure with 4 stitching points is subjected to bending load, the yarns tend to open at 4 different points. While the structure having one stitching point resists at a single location only. Therefore, higher number of stitching points allow the structure to withstand higher out of plane bending load.

Figure 18 showed the force versus time curves of 3D woven and laminated composite samples during drop weight impact test. The initial region of the peak shows the elastic behavior, i.e., force increasing with time. In this region, maximum force was showed by SP4 and minimum by laminated composite sample. The subsequent region in the curve represents plastic behavior where sample has undergone permanent deformation. SP4 showed the highest peak force while sample P showed the lowest peak force. From the samples it was observed that initially breakage was started in matrix followed by the yarn failure. The SP1 and SP2 structure failed soon after the first peak. Due to the absence of any stitching yarns in the laminated composite P, the curve decreased gradually in the plastic region showing the sample delamination. In case of composite SP4, maximum number of stitching points offered more resistance to delamination and ultimately failure [27].

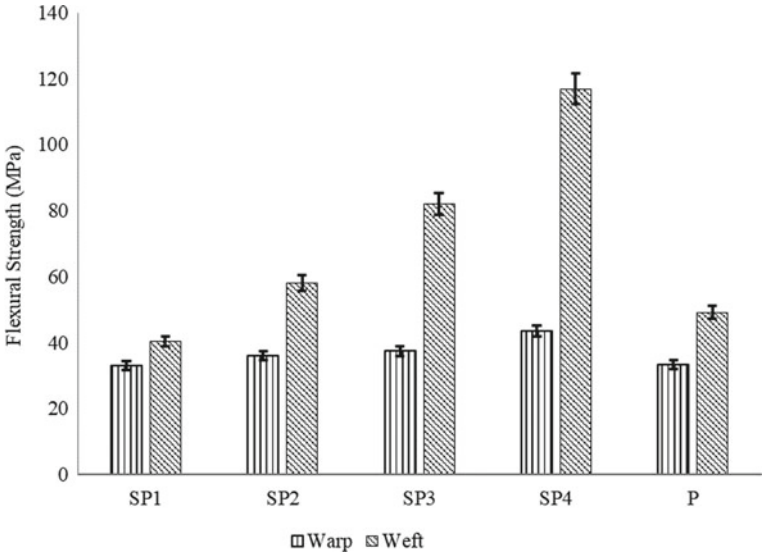

Fig. 17 Flexural strength of 3D woven and laminated composite samples in warp and weft directions [28]

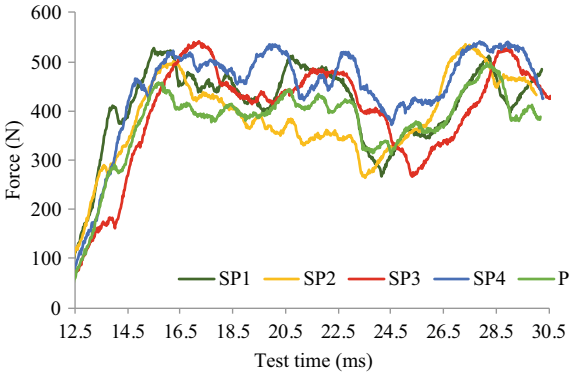

Fig. 18 Force versus time curves of 3D woven and laminated composite samples [28]

4.2 Effect of Binder and Stuffer Yarns on 3D Woven Composites

Jute yarn having linear density of 278 tex was used to developed seven types of different 3D woven structures on dobby loom. Specification and cross section of each structure is shown in Table 1 and Fig. 19, respectively.

These seven types of 3D woven structures namely F1 to F7 were used as reinforcement and bio-degradable epoxy was used as matrix to fabricate composites. For composite fabrication hand layup technique was used and for further curing and

Table 1 Specifications of 3D woven reinforcement

Sample I. D	Types of reinforcement	Ends/cm	Picks/cm	Arial density (g/cm^2)	3D structure nomenclature
F1	Orthogonal layer to layer	12.6	31.9	788	OLL
F2	Orthogonal through the thickness	12.6	32.2	794	OTT
F3	Angle interlock layer to layer	12.6	31.5	781	ALL
F4	Angle interlock through the thickness	12.6	32.2	795	ATT
F5	Hybrid of orthogonal through thickness and angle interlock through thickness structure	12.6	31.3	783	H1 (Hybrid 1)
F6	Hybrid of orthogonal through thickness and angle interlock layer to layer structure	12.6	31.5	781	H2 (Hybrid 2)
F7	Bi-directional interlock (hybrid of warp and weft interlock)	12.6	32.2	795	H3 (Hybrid 3)

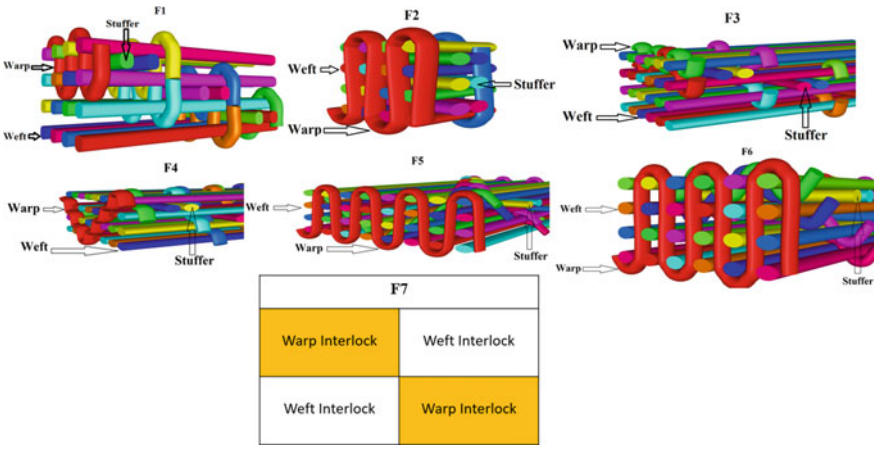

Fig. 19 Cross sectional views of F1 to F6 woven reinforcement and schematic view of F7 woven reinforcement [23]

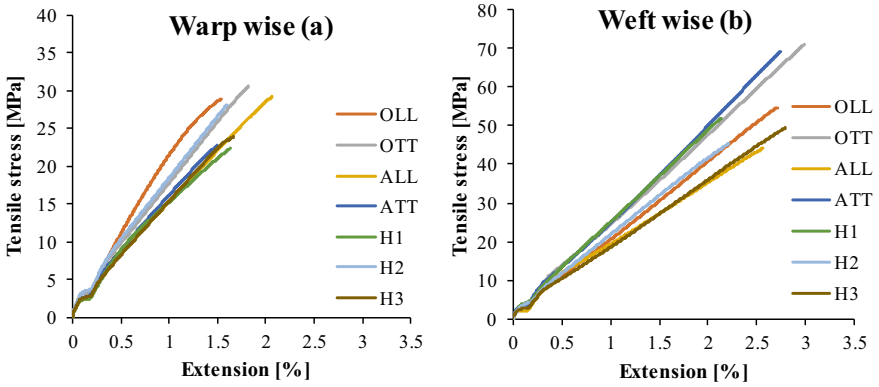

Fig. 20 Tensile stress versus extension (%) curves of 3D woven composites **a** warp wise **b** weft wise [23]

consolidation the composite plates were placed in compression molding machine under 15 bar pressure for 3 h. Fiber volume fraction (V_f) was maintained at 32% ± 0.5. Figure 20a and b showed the curves of tensile stress against extension % of developed 3D woven reinforced composites in warp and weft directions, respectively. Weft wise results of all sample were higher as compared to warp wise due to higher number of yarns per unit length in weft direction as compared to warp direction. Overall, a quasi-linear fracture behavior was observed in both directions warp and weft directions in all composite samples. The only difference is the highest value of tensile strength and elongation % because each reinforcement structure has different interlocking pattern and weave design. The curves showed that in warp direction the samples reached to fracture point after the extension of 1.2–2%, similarly in weft direction the fracture point starts after 2–3% extension. As the force applied on the composite sample, initially the sample resists against it with slightly elongation then reached to brittle fracture as shown in Fig. 20.

In 3D natural fiber reinforced composites, the failure mechanism initiates with matrix cracks followed by start of fiber failure. Delamination cannot be observed due to interlocking in the 3D structure. The breaking point of composite is sharp like brittle material. Expect H3 sample, in all other structures the stitching yarns hold the structure by moving above or below the weft yarns. The undulation in the stitching yarn produced some crimps in the structure. While in case of H3 sample, the binding yarns are present in both warp and weft directions, which generates nearly equal crimps in both directions i.e., warp and weft. The results concluded that out of seven structures, OTT structures has the lowest crimps in the yarn therefore it has the highest value of tensile strength in both directions i.e., warp and weft. Figure 21 showed the fracture behavior of OTT and H3 samples.

Figure 22a and b showed the flexural stress versus deformation % curves of composite samples in warp and weft directions, respectively. The results revealed that within elastic region, increase in stress is faster while deflection in the samples

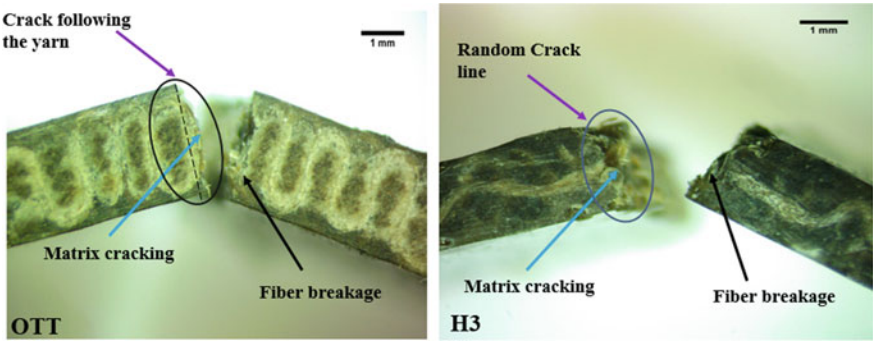

Fig. 21 Fracture analysis of composite samples [23]

is lower. After attaining the peak value of flexural stress, sudden drop in flexural stress was observed in all samples in both directions. Except OLL, all other samples showed brittle behavior at the point of fracture in warp and weft directions. In case of OLL sample, strain hardening region was observed. Weft-wise flexural stress is higher as compared to warp-wise due to the higher number of yarns per unit length in the weft direction. ATT sample showed the highest flexural strength in weft direction while in warp direction ATT and ALL showed the highest and comparable flexural stress. Within hybrid structures, H1 showed the highest value of flexural stress and H3 sample showed the intermediate value while H2 sample showed the lowest value of flexural stress [29].

Similar increasing trend of flexural modulus was observed in all composite samples. Weft wise flexural modulus was higher as compared to warp wise of all samples, it is due to a smaller number of undulations in the yarn in weft direction.

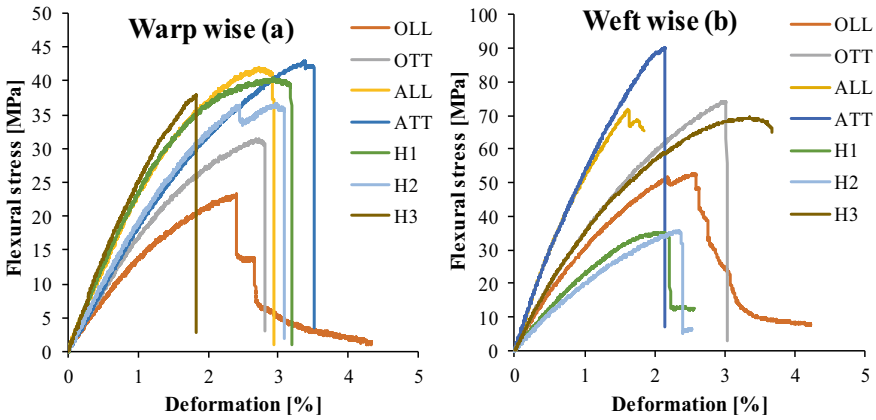

Fig. 22 Flexural stress versus deformation (%) curves of 3D woven composites **a** warp wise **b** weft wise [23]

Within the warp direction results it could be observed that ATT sample showed the highest value of flexural modulus. While in hybrid structures H1 showed the highest value of flexural modulus comparable to ATT sample. In weft direction the results revealed that ATT reinforced composite sample showed the highest value of flexural modulus. Within hybrid structures, H3 showed the highest value of flexural modulus in weft direction.

The impact performance of all seven developed composites were tested against 3 and 6 J drop weight impact energy, Fig. 23a showed the impact results of composites sample under 3 J, while Fig. 23b showed the impact performance of composite samples under 6 J impact. Figure 23a depicts that within basic four 3D woven structures, ALL sample showed the highest value of force against displacement as compared to ATT, OLL and OTT. It is due to the angular movement of binder yarns in the structure, which gave more resistance to the structure against the free fall of impact mass. Within angle interlock the float length of binder yarn also significantly affect the impact performance of the structure, binder yarn with shorter float length in angle interlock resist more to the impact force as compared to longer float binder yarn in angle interlock or truly orthogonal structure. Therefore, ALL composite sample showed higher impact force as compared to ATT based composite at similar displacement. Whereas in case of orthogonal structures, OTT sample showed better results as compared to OLL. Minor cracks were observed in all composite samples [29].

In comparison of hybrid structures-based composites, H3 showed the highest value of force at maximum displacement, while H2 showed the lowest force at maximum displacement. The float length of binder yarn in H3 is minimum, therefore it has higher impact force as compared to H1 and H2 samples. Impact force against maximum displacement of H3 is comparable to ALL sample.

Similar behavior to bear impact force was observed in case of 6 J impact force test as in case of 3 J impact test. But overall maximum force value against displacement is higher in case of 6 J as compared to 3 J impact test. Within four basic structures, ALL showed the highest value of impact force against displacement as compared

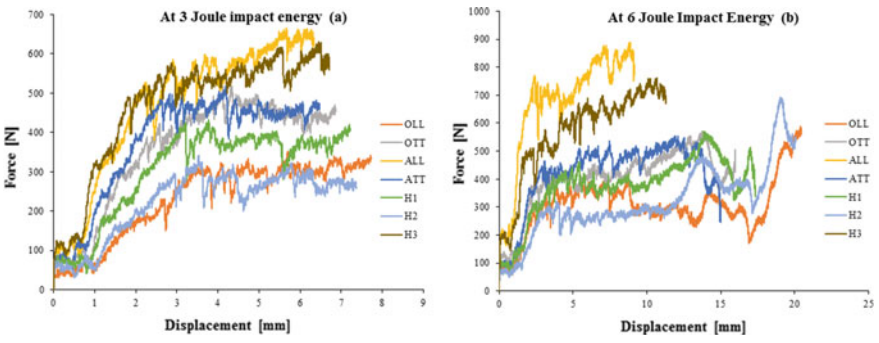

Fig. 23 Force versus displacement curves of 3D woven composites **a** at 3 J **b** at 6 J [23]

to rest of three basic structures. Within hybrid structures-based composites samples, H3 showed highest value of impact force against displacement.

Optimization of multilayered woven structures for enhanced mechanical performance

Nine (09) different types of orthogonal interlock 3D woven structures were developed on dobby loom. Jute yarn having linear density of 8 lb/spindle was used to develop all four layered orthogonal interlock structures. 3D woven structures with sample notation and description are mentioned in Table 2 and cross-sectional view of each design is shown in Fig. 24. Nine samples are divided into three groups A, B, and C. group A contains the sample of orthogonal layer to layer with warp, weft, and hybrid interlocks, similarly group B consists of orthogonal through thickness samples with warp, weft, and hybrid interlocks, and group C consists of Hybrid weave design with hybrid interlocking patterns.

Tensile strength results of all three groups of 3D orthogonal interlock structures are highlighted in Fig. 25. From the results it was concluded that by increasing the crimp % in the structure, the tensile strength decreases. There is inverse relation between crimp % and tensile strength. It is due to poor force dissipation throughout the length in crimp or undulated yarn as compared to straight yarn. From group A results, it was observed that warp interlock structure showed lower tensile strength in warp direction as compared to weft interlock layer to layer structure. It is due to a smaller number of undulations in weft yarn as compared to warp yarn in the structure. Hybrid structure of group A showed the average value of tensile strength

Table 2 3D woven samples description and notation

S. No.	Weave design	Sample ID	Interlocking pattern	Notation	Group
1	Orthogonal layer to layer (OLL)	S1	Warp interlock	LL-wp	A
2	Orthogonal layer to layer (OLL)	S2	Weft interlock	LL-wf	
3	Hybrid by combining S1 and S2	S3	Warp-weft interlock	LL-wp/wf	
4	Orthogonal through the thickness (OTT)	S4	Warp interlock	TT-wp	B
5	Orthogonal through the thickness (OTT)	S5	Weft interlock	TT-wf	
6	Hybrid by combining S4 and S5	S6	Warp-weft interlock	TT-wp/wf	
7	Hybrid by combining S2 and S5	S7	Weft-weft interlock	LL/TT-wf/wf	C
8	Hybrid by combining S1 and S4	S8	Warp-warp interlock	LL/TT-wp/wp	
9	Hybrid by combining S1 & S2 and S4 & S5	S9	Warp-weft interlock	LL/TT-wp/wf	

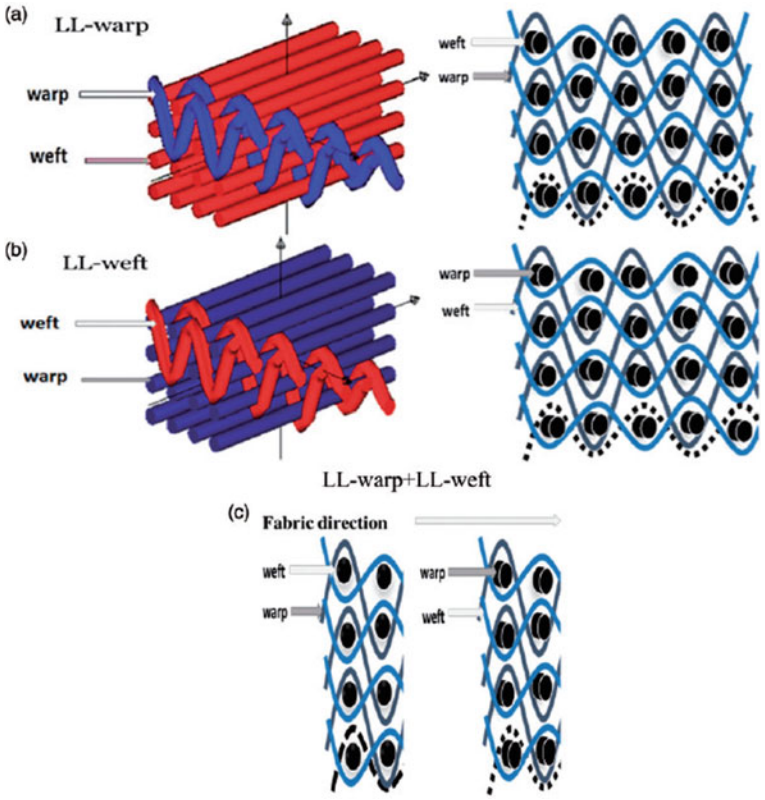

Fig. 24 Cross-sections of 3D woven preforms (i) S1 (ii) S2 (iii) S3 (iv) S4 (v) S5 (vi) S6 (vii) S7 (viii) S8 (ix) S9 [18]

as compared to warp and weft interlock structure. Similarly in group B, same trend of tensile strength was observed with through the thickness interlock samples. The structures that have higher undulations showed lower tensile strength. Warp interlock structure showed lower tensile strength as compared to weft interlock structure in warp direction. In case of weft interlock structure, the tensile strength is lower in warp direction as compared to weft direction, while undulation is higher in weft direction, it is due to higher tension on warp yarn during weaving process, which effect its mechanical properties therefore it has lower tensile strength. In group C, the lowest tensile strength was shown by LL/TT (wf/wf), it is due to highest number of undulations in the structure [18].

Figure 26 showed the elongation % of developed 3D orthogonal interlock woven structures. Elongation in the structure depends on the undulation of the yarns in the structure, higher the undulation in the structure higher will the elongation in the structure. Because after applying force on it, the crimp part of yarn tries to become straight hence the resultant length of the yarn increases, ultimately the elongation %

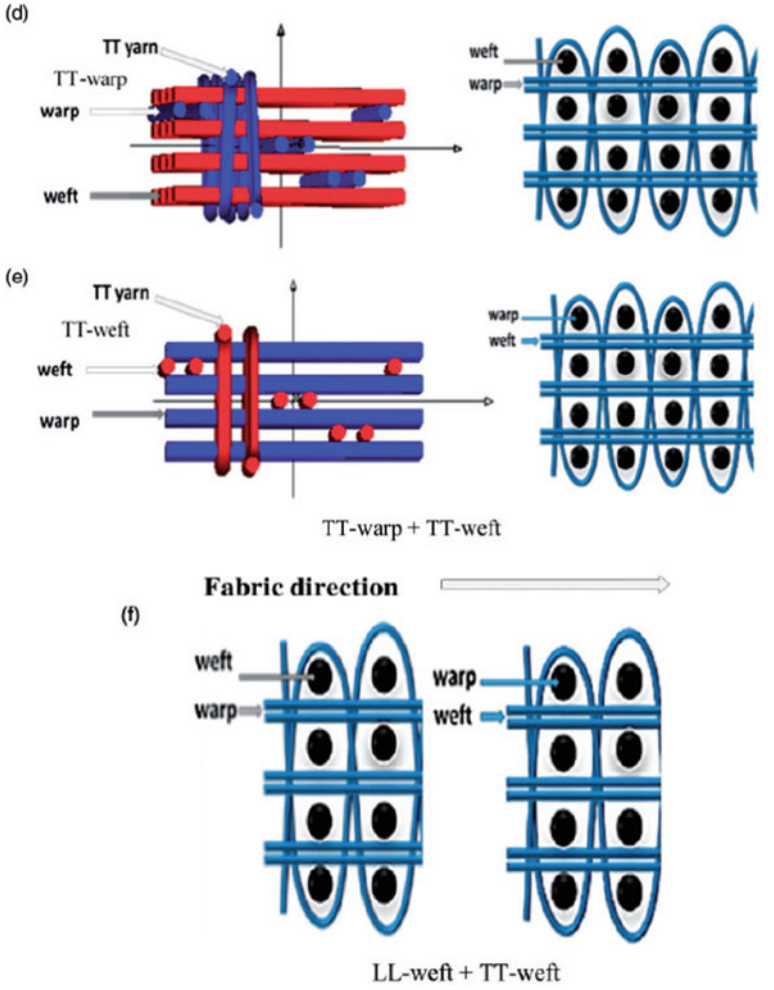

Fig. 24 (continued)

also increases. In group A the highest value of elongation was shown by LL-hybrid structure, because in this structure both warp and weft yarns interlock the structure, resulting higher crimp % in the structure. Group B consists of the structures that have through the thickness interlocking, as the binding yarn moves from top layer to bottom layer of the structure and the binding yarn faces higher tension as compared to ground weave yarns. After releasing the fabric from loom, the interlocking yarns become more relax than the ground weave yarns, due to the release of tension applied by the loom parts during weaving process. In group B TT-wf structure showed higher elongation % as compared to other structures due to lobular effect in the structure. In group C, hybrid structure of LL/TT-wp/wf showed the highest elongation due to

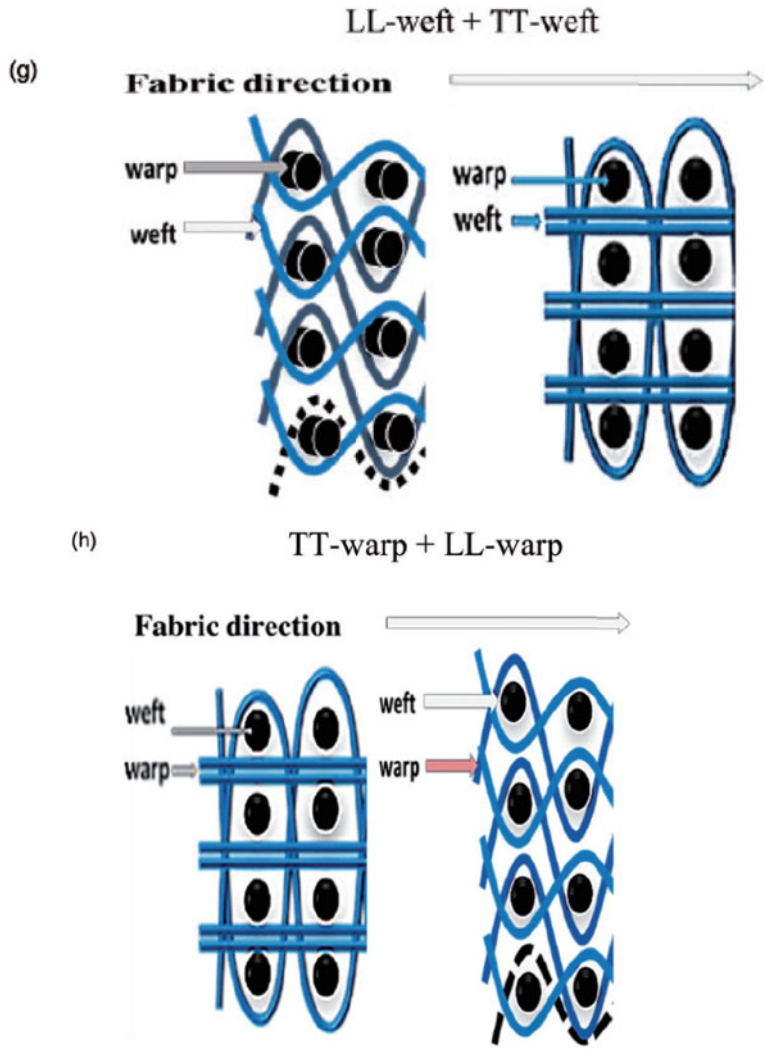

Fig. 24 (continued)

higher number of crimps in the structure, because both warp and weft yarns interlock the structure, and secondly the changing of weave from LL to TT creates more float, therefore it has higher elongation % as compared to rest of the structures [18].

Figure 27 showed the tensile modulus of developed samples in warp and in weft directions. Tensile modulus has inverse relation with the elongation % in the structure. It showed that the structure having higher crimp % will have lower modulus, because after applying force on yarn, then crimp in the yarn tends to become straight, as a result the length of the yarn increases. Therefore, it will show higher strain % and lower modulus.

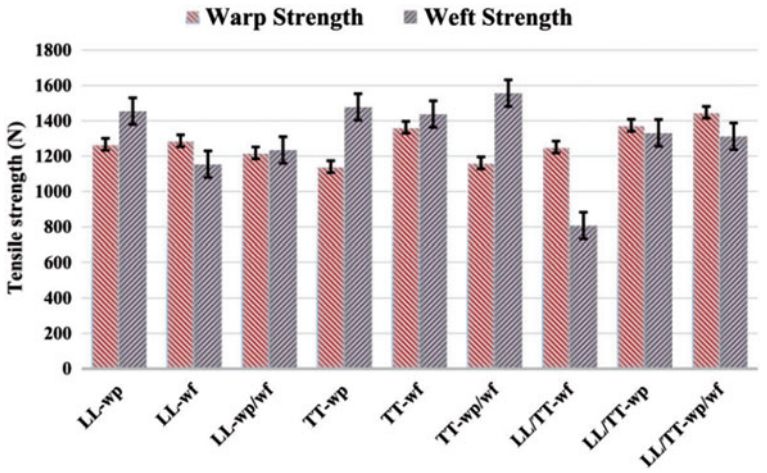

Fig. 25 Tensile strength of 3D woven structures in warp and weft directions [18]

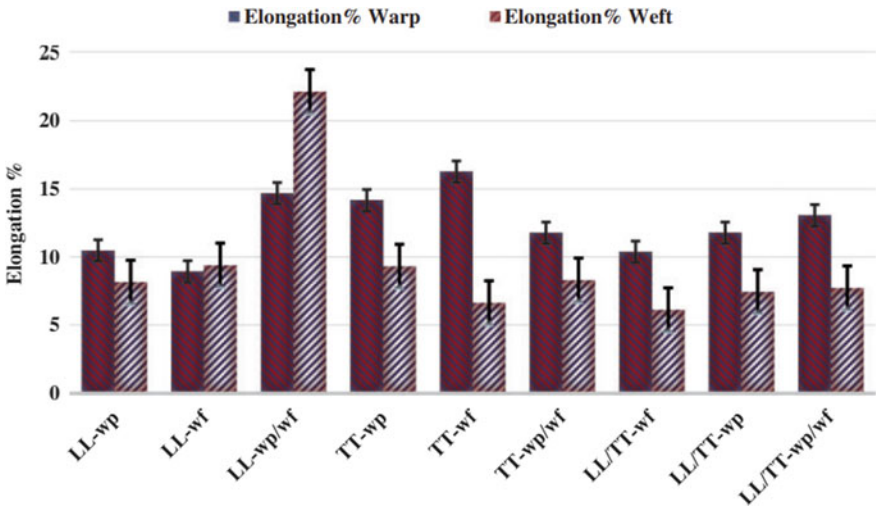

Fig. 26 Elongation % of 3D woven structures in warp and weft directions [18]

Figure 28 showed the stiffness results of developed orthogonal interlock structures. Stiffness in the structure indicates that the structure cannot easily be molded or shaped. In group A, the results showed that warp interlock layer to layer structure showed the highest stiffness value while weft interlock layer to layer structure showed the lowest stiffness. In group B, warp interlock through the thickness structure showed the highest stiffness value not only within group but it also showed the highest stiffness value among all nine samples. In group C both hybrid structures i.e., LL-TT-wp and LL-TT-wp-wf/wp-wf have comparable values of stiffness [18].

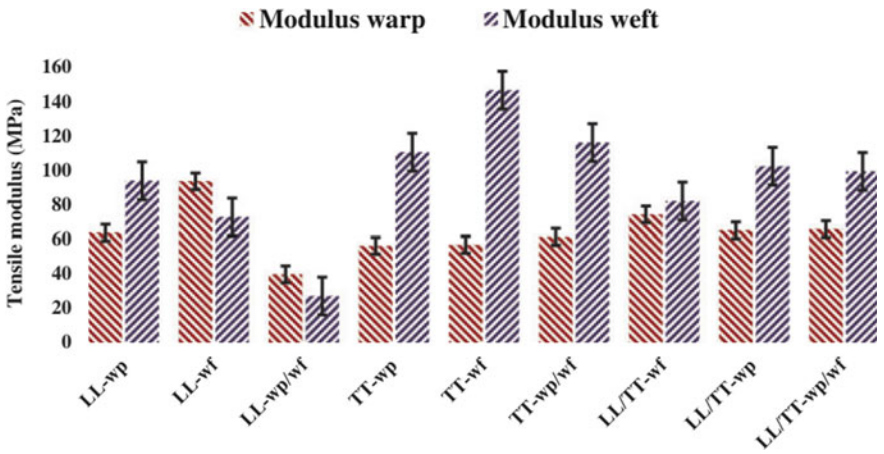

Fig. 27 Tensile modulus of 3D woven structures in warp and weft directions [18]

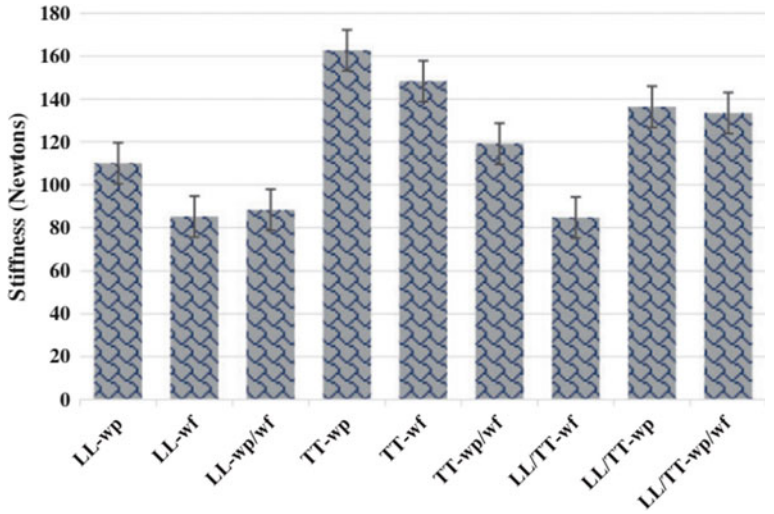

Fig. 28 Stiffness of 3D woven structures [18]

4.3 Effect of Weaving Patterns on Damage Resistance of 3D Woven T and H Shaped Reinforcements

Multi-layered T and H shaped structures were developed on conventional dobby loom, using Jute yarn in both warp and weft directions. Linear density and tenacity of jute yarn was 490 tex and 5550 cN/tex, respectively. Each shaped fabric consists of four-layer structure. Both T and H shapes were developed from four basic multilayer weave designs i.e., orthogonal layer to layer interlock (OLL), angle interlock layer to

layer (ALL), orthogonal through the thickness interlock (OTT) and angle interlock through the thickness (ATT). Total 8 samples were developed, all the samples have same warp and weft density i.e., 10 yarns/cm in both directions. Figure 29 showed the cross-sectional views of shaped fabrics while Fig. 30 showed the top view of developed shaped fabrics [19].

Figure 31 showed the sample dimensions for peel test, schematic presentation of test and the real time testing instrument used for peel strength test of developed shaped fabrics [19].

Figure 32 showed the force versus deformation curves of OLL shaped structures during peel test. This curve is divided into three zones: A zone denotes elastic zone, B zone denotes plastic zone and C zone denotes fracture zone. Real time images of sample during testing are also represent the respective zones as shown in Fig. 32. In the elastic region of the curve, it was observed that the interlocking yarn resist to the applied force, because with the increase of force the deformation is minimum in this region. At initial stage, the applied force is utilized to straighten the interlocking yarn in T-shape at the point of joint instead of breakage. Due to straightening of yarn the deformation in the structure is increased. Within elastic region, some peaks were observed it may be due to fiber failure at T-joints in the structure [19]. In zone B of the curve, plastic deformation starts. Further increase in the force initiates the failure in the interlocking yarns of the structure. In zone B of the curve maximum resistance of interlocking yarn was observed against applied force then a cluster of yarns is formed in the middle of the T-shape. In the OLL structure, interlocking yarns stitched the consecutive layers at different positions. The repeat size of interlocking yarn is different from the ground weave of the structure. During peel test only interlocking yarns start to break while the warp yarns of ground weave start to form a cluster in the middle and offer further resistance to the applied force. In the necking region of the structure, only weft yarns remain unbroken while warp yarns accumulate in the form of the cluster in the middle. While in the fracture zone complete failure of the structure is observed. In this zone the applied force was rapidly reduce because the structure shows no significant resistance to the applied force.

Figure 33 showed the force versus deformation curve of orthogonal through the thickness structures of T and H shapes. OTT curve also showed three zones like OLL shapes. The elastic region of OTT structure is steeper than OLL structure, it showed that the interlocking yarns in OTT resist more to deformation as compared to the interlocking yarns in OLL structure. Small deformation at high force is observed in elastic region in case of OTT structure, due to the interlocking pattern of stitching yarns. In OTT structure the stitching yarns bind the complete structure in the thickness direction, so there is limited space for the structure to deform under applied force. In elastic region a small peak was noticed, which indicates the initiation of breakage in the interlocking yarns. In plastic region the maximum resistance to applied force was showed by the interlocking yarns after the breakage of stitching yarns. Almost all interlocking yarns are broken in the plastic region and cluster is formed in the middle of the joint T as shown in the Fig. 33. In the last zone of the curve, complete fracture of the structure is observed [19].

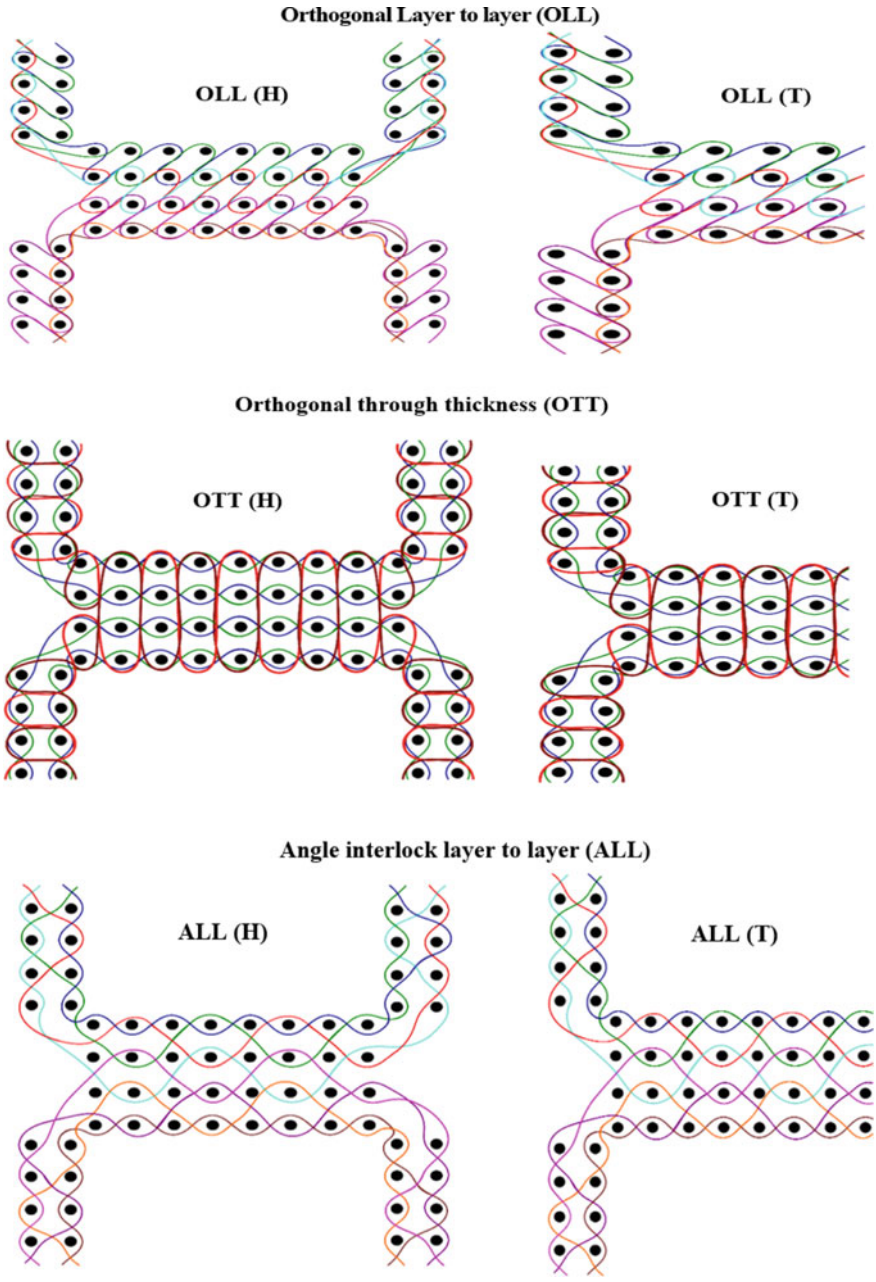

Fig. 29 Cross-sectional view of T and H shaped fabrics [19]

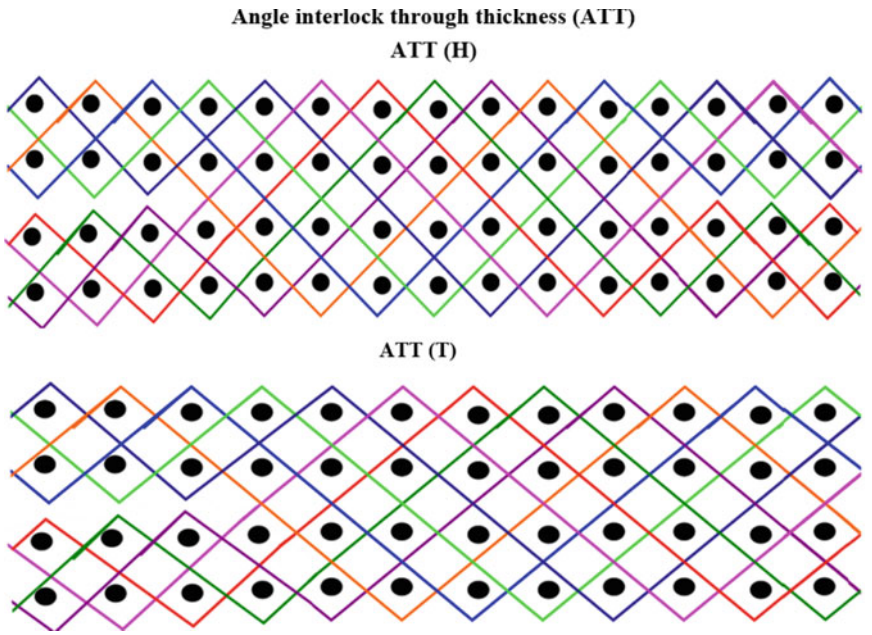

Fig. 29 (continued)

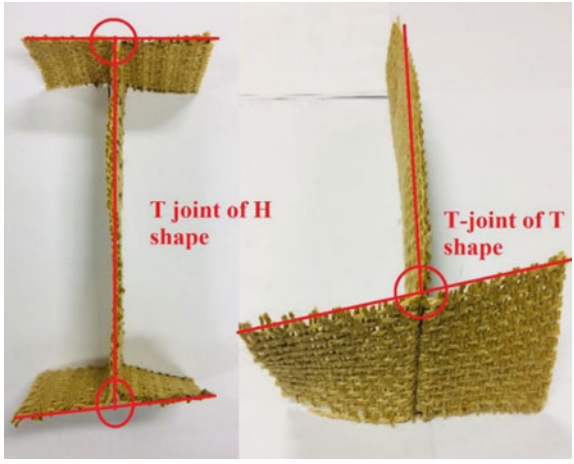

Fig. 30 Top view of developed T and H shaped fabric [19]

Figure 34 showed the force versus deformation curve of shaped structures developed from angle interlock layer to layer structure. In the elastic region the curve is sharp due to the high resistance of interlocking yarns, while deformation is due to straightening of interlocking yarns. In this region the interlocking yarns become straight in the response to applied force, while breakage of the yarn starts in plastic

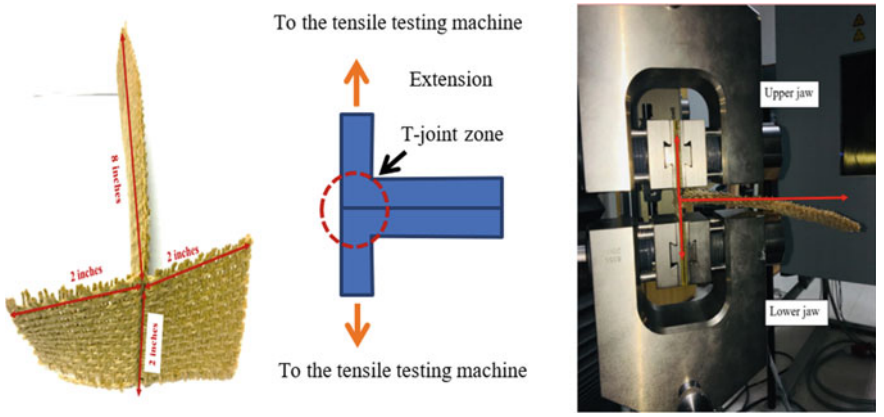

Fig. 31 T-shape sample dimensions for peel test, schematic presentation of test and real-time testing instrument for peel test [19]

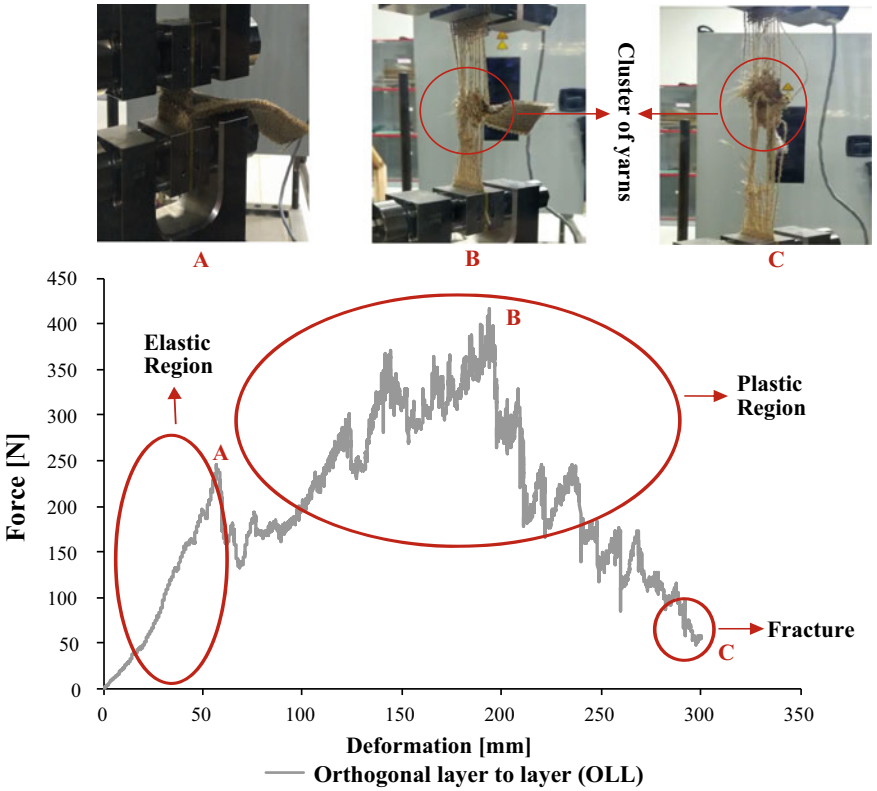

Fig. 32 Force versus deformation curve of OLL shapes [19]

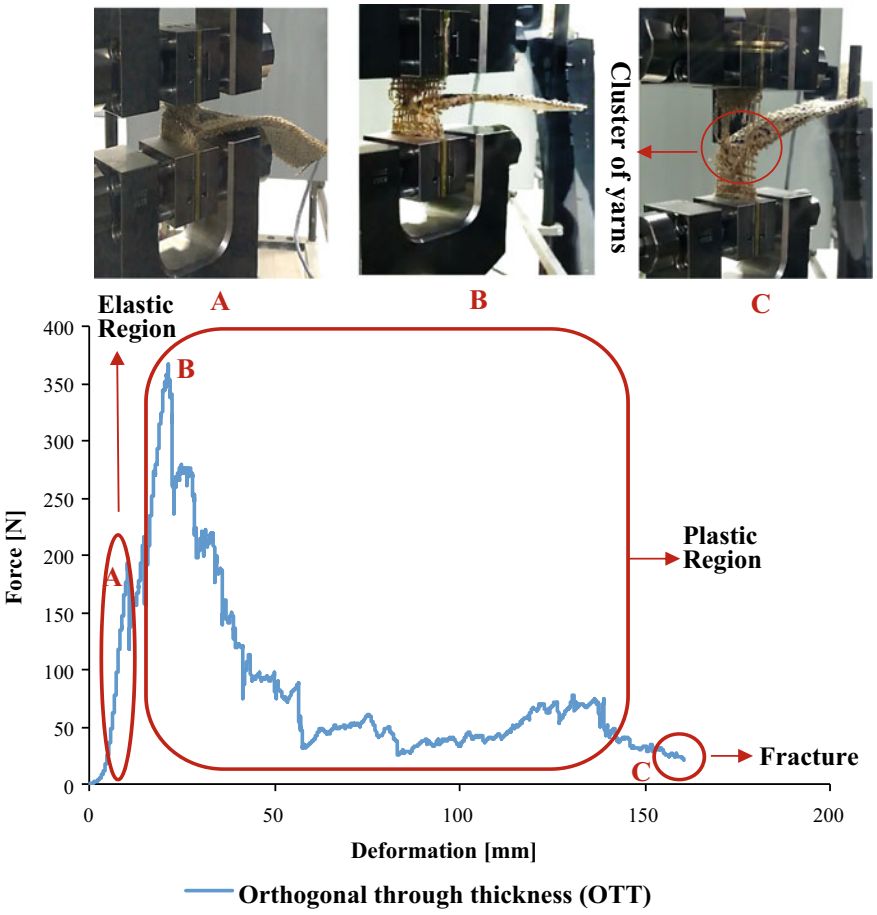

Fig. 33 Force versus deformation curve of OTT shapes [19]

region. In plastic region the maximum resistance against the applied force is observed which is called peak force, after that the force reduces due to breakage or pull out of interlocking yarns from the structure. In ALL structure, the interlocking yarns stitched the consecutive layers at certain angle to achieve this angle some weft yarns were skipped, therefore during applied force the stitching yarn can easily be pulled out instead of breakage. Consequently, it showed high deformation up to 300 mm. The fracture zone showed the complete failure of the structure and the force was rapidly reduced [19].

Figure 35 showed the force versus deformation curve of angle interlock through the thickness structure during peel test. ATT curve indicates that in elastic region, sharp curve behaviour was observed in the elastic region between force and deformation. It is due to the high resistance offered by the interlocking yarns in the structure.

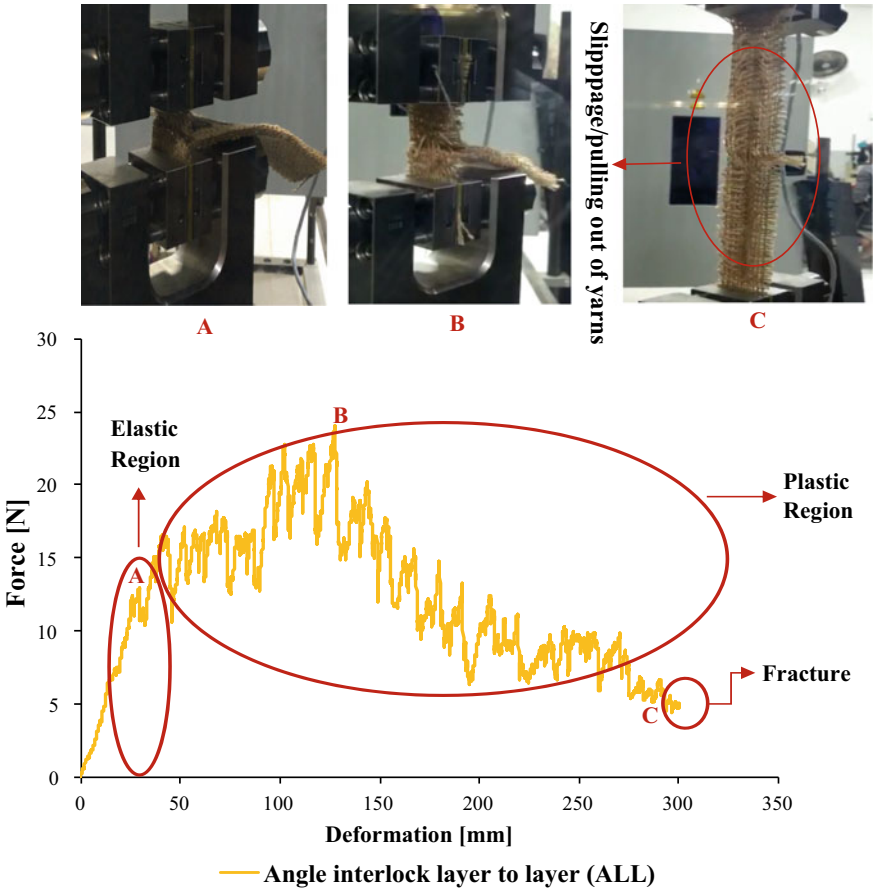

Fig. 34 Force versus deformation curve of ALL shapes [19]

Further increase in the force starts deformation in the structure which is due to the straightening of stitching yarns. In the elastic region some small peaks were observed, which indicates the fiber failure in the interlocking yarns.

Plastic region of the curve indicates that structures start to deform with the increase in the applied force. When the interlocking yarns become straight, peak resistance is observed to the force, after that instead of straightening of yarn failure starts. Since, resistance is only showed by the interlocking yarn, therefore the ground weave warp yarns form a cluster in the middle of the joint T. After peak force, the applied force reduces while deformation in the structure increases which is due to the slippage and breaking of interlocking yarns. In the fracture zone, complete failure of the structure was observed [19]. Figure 36 showed the relation between work done and deformation of different 3D woven shapes. It can be concluded that OLL structure showed the highest work done before failure, while Structure OTT showed the minimum work done before complete failure [19].

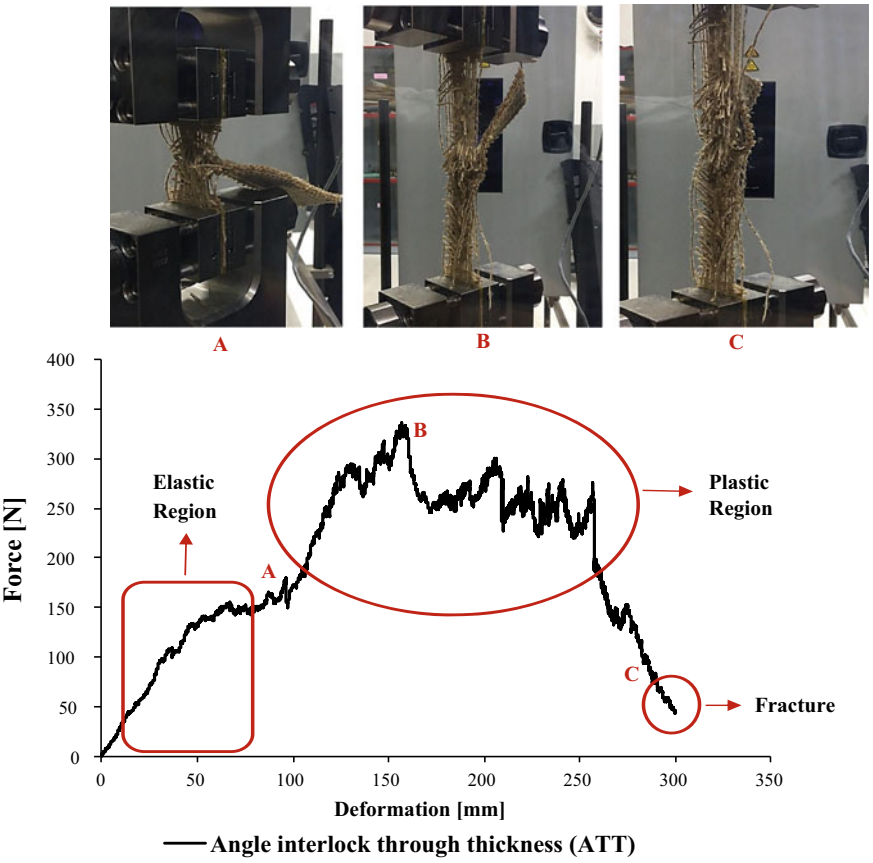

Fig. 35 Force versus deformation curve of ATT shapes [19]

4.4 3D Woven-Shaped Preforms and Their Associated Composites

Six layered shaped structures were developed from Jute yarn having linear density of 47 tex. The structures were developed on dobby loom with same warp and weft density. The interlocking patterns used to develop shaped structures were through the thickness and layer to layer. Hand layup technique was used to developed shaped composites. Unsaturated polyester resin was used as resin with 0.1% concentration of cobalt as initiator, and 1% concentration of methyl ethyl ketone per oxide as accelerator [14]. For comparison purpose 2D laminated shaped composite was also developed. Figure 37 showed the highlighted portions of the developed samples for testing and compared its results with laminated composite. Table 3 showed the peel off test results of developed samples [14].

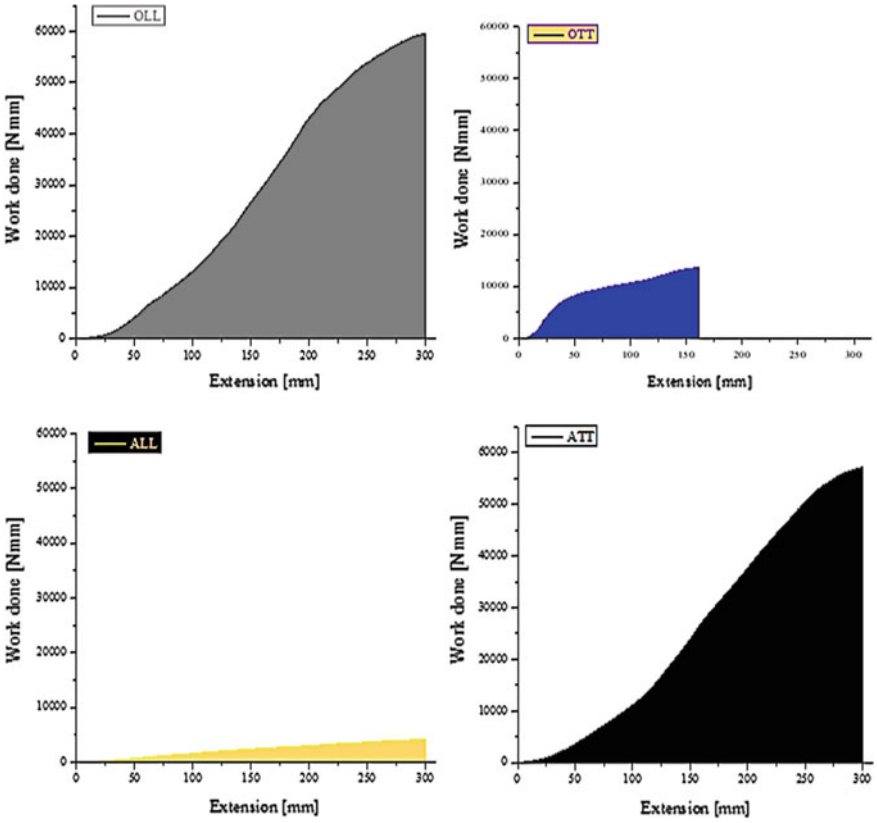

Fig. 36 Work done during failure of different 3D multilayer shapes [19]

Table 3 showed that orthogonal layer to layer interlock T shaped (S1C) and orthogonal layer to layer interlock H shaped (S3C) have the same mechanical performance. Because both samples have same type of 3D reinforcement, fiber volume friction and curing cycle. Same trend was observed in the case of S2C (T) and S4C (H) samples. Because both samples have orthogonal through the thickness reinforcement with T and H shapes, respectively. While sample S5C was developed with 2D laminated reinforcement. At reinforcement level, all structures showed higher elongation, but after conversion into composite the resin restricts the elongation of the samples. In sample S5C the applied force generate cracks in the matrix and delamination occurs in the composite sample, because in reinforcement there is no stitching point in the structure, therefore it showed minimum resistance [14]. Figure 38 showed the elastic modulus and breaking load results of developed shaped composite samples.

The results showed that 3D shaped composite has a higher value of modulus, maximum load and stress at peak load as compared to laminated composite at equal

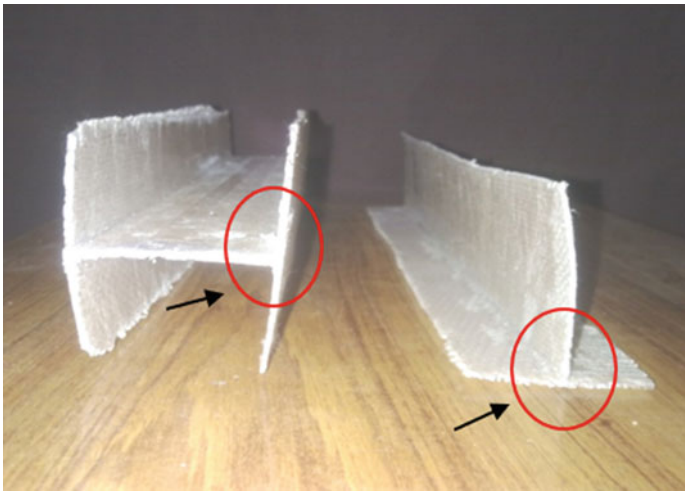

Fig. 37 T and H shaped composite samples produced using 3D woven preforms [19]

Table 3 Mechanical properties of T and H shaped composites

Type of structure	Reinforcement weave design	Breaking load (N)	Elongation % at max. force	Extension at max. load (mm)	Strain at max load	Stress at maximum load (N/mm^2)	Young's modulus (MPa)
S1C, S3C	LL	1739 ± 7.3	13.65 ± 2.3	6.81 ± 0.7	0.07 ± 0.01	69.5 6 ± 2.3	993.7 ± 15.3
S2C, S4C	TT	1067 ± 5.5	7.16 ± 1.5	2.36 ± 0.4	0.09 ± 0.02	42.68 ± 1.3	474.22 ± 3.3
S5C	Laminated (weave design of laminated)	990 ± 4.5	7.90 ± 0.9	2.78 ± 0.3	0.15 ± 0.1	39.6 ± 1.3	264 ± 3.2

fiber volume friction. Because the laminated structures have no stitching points within layers, therefore layers can easily separate from each other can causes failure in the whole structure. In 3D shaped structures LL showed better results as compared to TT shaped composites [14].

5 Challenges

Although natural fibers are derived from renewable sources, and polymer composites based on natural fibers are sustainable as compared to synthetic fiber reinforced polymer composites. But there are several drawbacks associated with the use of natural fibers as reinforcement in the composites. These drawbacks include raw

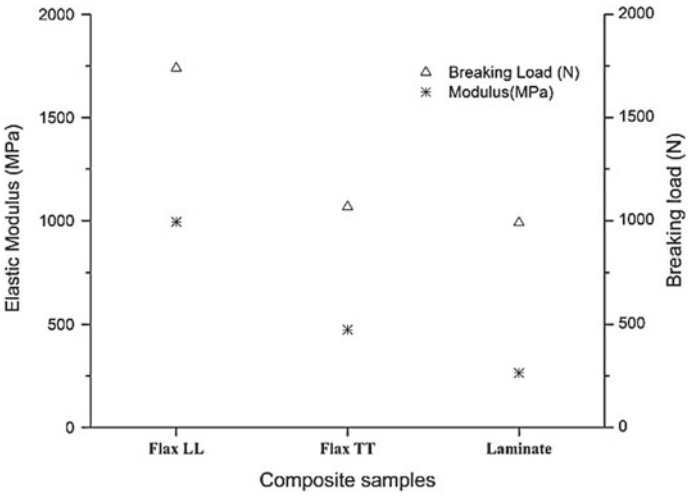

Fig. 38 Comparison of young's moduli and breaking loads of T and H shaped composites [19]

fiber quality variation, excessive moisture absorption, and thermal stability [30, 31]. Natural fibers have high moisture absorption rate, which is their main disadvantage. This decreases the interfacial connection between the polymer matrix and the fiber causing deterioration of mechanical properties. The high moisture sensitivity of lingo-cellulosic fiber results in dimensional instability [32, 33] and limiting their usage as reinforcement in composite materials. In many circumstances pre-treatment of the fiber surface or the introduction of a surface modification during composite preparation is necessary to overcome this difficulty and to increase the fiber-matrix adhesion. The impact of various types of chemical treatment on the physical and mechanical properties of natural fiber reinforced composite has been studied extensively in the literature [34–36]. Different fiber treatments result in considerable increase in tensile and flexural, strength and modulus. The chemical treatment of fibers can significantly minimize moisture absorption. Chemicals such as alkali (sodium hydroxide), isocyanate, KMnO4 (permanganate), CTDIC (cardanol derivative of toluene diisocyanate), peroxide, enzyme, and others have been used for the treatment showed significant changes in the mechanical and physical properties [12].

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Commingling Technique for Thermoplastic Composites

Muzzamal Hussain, Adeel Abbas, and Muhammad Ayub Asghar

Abstract Thermoplastic composites are an area of growing interest due to increasing environmental awareness as they are recyclable. However, high melt viscosities and poor fiber matrix adhesions lead to quality and performance issues. The commingling technique is a considerable solution to this problem; hence, textile preforms are engineered by utilizing both fibers and thermoplastic matrix simultaneously in the dry state. Such dry prepregs are capable of being converted into composites through the application of heat and pressure. Commingled prepregs provide close fiber-matrix physical contacts resulting in better interfaces and wetting characteristics. The chapter discusses possible techniques for prepregs manufacturing at fiber, yarn, and fabric levels. Thermoplastic commingled composites fabrication techniques have also been elaborated, including thermoforming and pultrusion. The impact of commingling on mechanical performance parameters is highlighted, and possible commingling routes are predicted to achieve endurable characteristics.

Keywords Commingling · Commingled Yarn · Weaving · Thermoforming · Mechanical properties

1 Introduction

Composite materials are the combination of two or more materials (reinforcement and matrix), providing unique characteristics that are not exhibited by the individual component. Matrix may be thermoplastic or thermoset; however, the reinforcements could be particles, fibers, yarns, and fabrics. Thermoset matrices are of low viscosity and hence are capable to provide better impregnations, while thermoplastic matrices are of high viscosity and are thus difficult to impregnate within reinforcement materials. On the other hand, reinforcement materials have porosity issues at micro, meso, and macro levels which require viable impregnations to provide better mechanical

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and physical characteristics to the composites. The conventional thermoforming route isn't capable to provide the proper impregnations and desired characteristics; hence the commingling technique is used. Commingling involves the fabrication of dry prepregs, comprising a thermoplastic matrix and reinforcement materials. Natural fibers reinforced composite materials are an area of growing interest due to environmental burdens. Natural fibers are usually hydrophilic and synthetic thermoplastic matrices are hydrophobic. Such affinity differences do not allow proper adhesion and bonding.

Commingling: expounded as converting into a single mass/entity, is a chiefly preferred technique to obtain better reinforcement wetting/impregnation characteristics resulting in increased fiber-matrix adhesion. Natural fiber composites (NFCs) offer poor integrities governed by improper fiber matrix interactions; hence commingled NFCs are an area of interest. Textile preforms offer commingling opportunities at each possible stage i.e., fibers, yarns, nonwovens, woven, and knitted fabrics. Commingling technique for thermoplastic composites creates an effective route allowing to achieve better fiber wettings. Conventional thermoplastic composites manufacturing follows a route where reinforcement materials and thermoplastic sheets are stacked alternatively for composite thermoforming. However, the commingling technique allows dry prepreg formation, the prepreg consists of both thermoplastic matrix and reinforcement in desired proportions. The prepregs are directly thermoformed for composite formation [1]. Figure 1 shows the described fabrication routes.

Fiber reinforced composites are used in different applications including sports, packaging, automobile, aerospace, agriculture, etc. Fiber reinforced composites have two components: reinforcement and matrix. Depending upon the nature of applications, the material of the components may vary. The reinforcement material determines the properties of composites material being the main load-bearing component. The use of Natural Fiber Composites (NFCs) has increased in recent years due to many environmental reasons [2–8].

The properties of NFCs depend on many factors e.g., fiber orientation, fiber length, fiber diameter, fiber distribution, fiber volume fraction, and porosity [9]. Natural fibers used for reinforcement are jute, banana, flex hemp, banana coir, etc. [10–15]. NFCs are more preferred because, at the end of their life cycle, the amount of CO_2 emitted is the same as consumed by these fibers during their growth [16]. NFCs

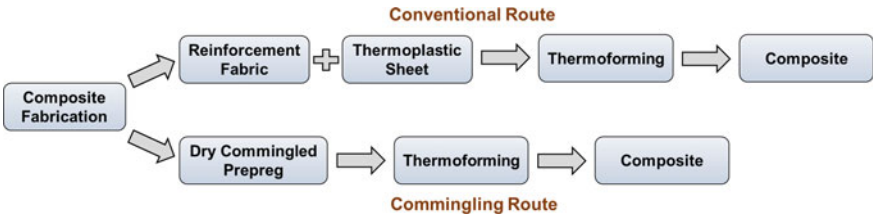

Fig. 1 Conventional and commingled composite fabrication routes

can have either one or both components made with natural material, However, the use of natural fibers as a reinforcement is most convenient due to its availability in abundance, easy processing, and cost-effectiveness [6–8, 17–19]. The NFCs can be made using both thermoset and thermoplastic composites however thermoplastics are more preferred due to recyclability and processing. There is a certain issue while processing the thermoplastic matrix due to high viscosity, which makes fabrication quite difficult. Apart from the high viscosity of the matrix, the natural fibers have a rough surface, protruding fibers on the surface, and poor wetting properties, these lead to low fiber volume fraction and low fiber matrix interface [2]. The fiber matrix interface can be improved through chemical treatment of fibers, but while dealing with the spun fibers the yarn-packing density is more difficult to achieve due to a higher amount of hairiness. These issues can be reduced by commingling the natural fibers with the matrix. The commingling in thermoplastic provides readymade dry prepreg which can be converted to composite by hot press.

2 Techniques of Commingling

The commingling of natural fibers with the matrix can be done at three levels i.e., fiber, yarn, and fabric. However, the integrity and desired mechanical characteristics may vary depending on the adopted technique.

2.1 Fiber Level Commingling

The fiber level mixing is done either to make a non-woven mat or mixed at the blow room stage to later make spun yarn. The fiber–fiber mixing is shown in Fig. 2. The natural fibers and thermoplastic matrix in the form of short fibers are mixed in the blow room to form the blow-room stage commingled yarn and then the commingled fabric can be woven from such yarns. However, this blend of fibers and matrix yarn can be subjected to hot compression molding to form non-woven composite matts. Literature comprises novel studies on fiber level commingled composites. Ameer et al. [20] developed jute and polypropylene fibers commingled nonwoven prepregs with different blend ratios of jute and polypropylene, the prepregs were then converted into composites via thermoforming. The characterizations show maximum mechanical performance at 40% jute and 60% polypropylene ratio by volume in nonwoven commingled preforms. Increasing or decreasing the percentage of both polypropylene and jute causes the properties of composite material to change. However, the key concern is also to reduce the composites-moisture interactions as natural fibers are hydrophilic. Moisture absorption decreases linearly via increasing thermoplastic fibers percentage in nonwoven mats. Karaduman et al. [21] architected nonwoven commingled composite sandwich structures having polyester foam, polypropylene honeycomb, and balsa wood inside the core. Flexural behaviors of

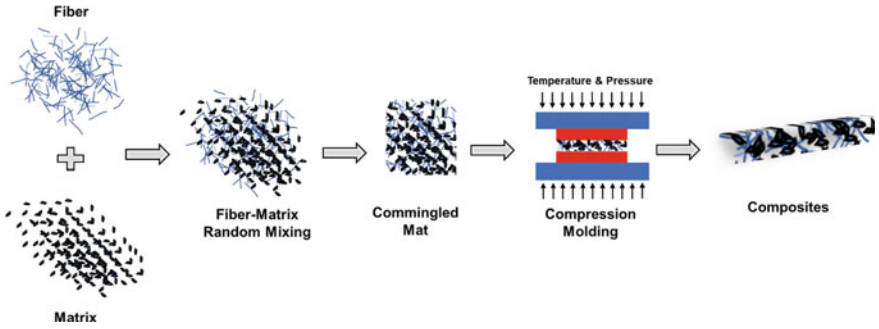

Fig. 2 Flow chart of fiber–fiber commingling [9]

nonwoven commingled sandwich composites show an enhancement with increasing fiber volume fractions.

Increasing environmental concerns have compelled the industry to find more more sustainable composite solutions. Although polyolefins i.e., polypropylene-based natural fiber reinforced thermoplastic composites are considered environment friendly due to recyclability. A significant share of such composites is of natural materials, and also these reduce environmental burdens via reducing non-biodegradable synthetic materials consumption. Polylactic acid (PLA) is a biodegradable thermoplastic material and is a suitable alternative to polyolefins. Lingansio et al. [22] fabricated nonwoven sandwich commingled prepregs through the pultrusion technique. Prepreg comprised flax/PLA commingled yarns as outer layers and nonwoven sheets as inner sandwich materials (Fig. 3). Commingled yarns providing better matrix distribution and good fiber matrix adhesion provide superior mechanical attributes over conventionally fabricated nonwoven sandwich composites (Fig. 3). Different surface treatments i.e., alkali, benzoyl chloride, $KMnO_4$, and saline treatments also led to good fiber wetting characteristics, governing better fiber-matrix adhesion [23]. Nonwoven fabrication methods offer a wide range of opportunities to engineer commingled composites. Matrix/reinforcement percentage critically defines composite mechanical characteristics.

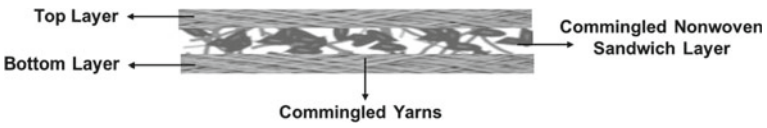

Fig. 3 Commingled nonwoven sandwich prepreg [22]

2.2 Yarn Level Commingling

Yarn level commingling involves the spinning of reinforcement fibers with thermoplastic matrix fibers. Variable mixing ratios can be set w.r.t required characteristics i.e., mechanical properties, hydrophobicity, etc. Yarn produced by this process is used to engineer fabrics that are capable of being converted into composites after thermoforming. Yarn level commingling comprises three basic techniques including core-spun commingled, co-wrapped commingled, and co-twisted Commingled yarns (Fig. 4).

Core-spun commingled yarns comprise reinforcement core and thermoplastic fibers wrapping. This is carried out at the spinning stage. However, core-wrapped yarns have matrix yarn wrapped on reinforcement core [24], and the process is carried out at the doubling stage of spinning. Described architectures have been shown in Fig. 5. Thermoplastic matrix and reinforcement yarns can also be co-twisted. The process is carried out at the doubling stage of spinning. Figure 5 shows the co-twisting setup of jute and polypropylene yarns [2]. Roving machine modifications can be performed for the purpose. Four polypropylene yarns are fed with one jute yarn from the input side of the simplex machine instead of roving. The yarns are initially passed over two guide rollers which help them to behave as a single toe. In the simplex drafting frame, the top assembly is disabled, hence no draft occurs. Afterward, the flyer inserts the required twist (1–2 twists per inch) into yarns.

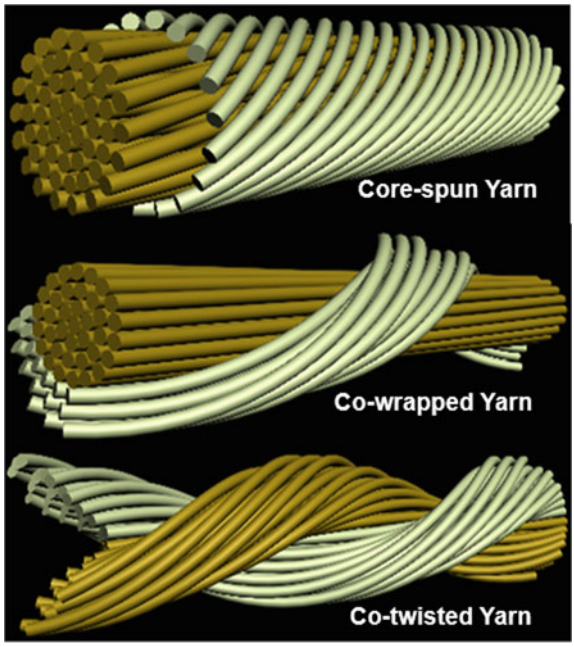

Fig. 4 Yarn level commingling technique [24]

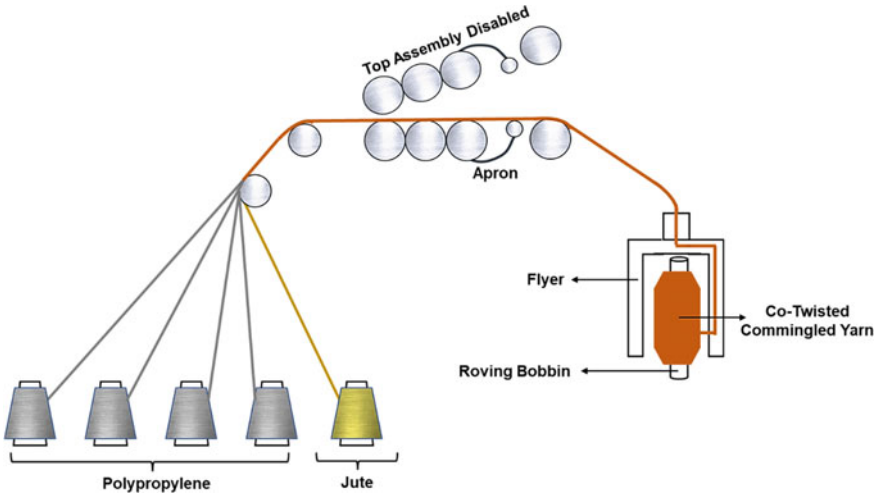

Fig. 5 Co-twisting setup on roving machine

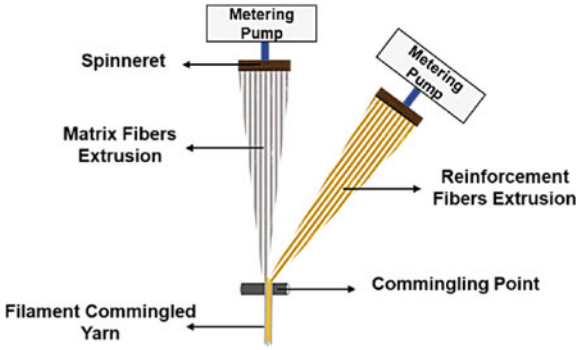

Fig. 6 Commingled filaments extrusion setup [25]

For yarn level commingling two different extrusion setups could also be installed having a common ending point where newly extruded filaments are mixed as shown in Fig. 6. Both filaments form a dry prepreg filament yarn capable of being converted into cured composite upon thermoforming i.e., polypropylene and glass fibers [25].

2.3 Woven and Knitted Commingled Composites

Woven commingled composites can be architected using different warp and weft yarns, the reinforcement and matrix yarns can be used in either warp/weft or both directions simultaneously. Knitted commingled composites are engineered with specialty inlaid structures (Fig. 7). Thermoplastic yarns are utilized in base

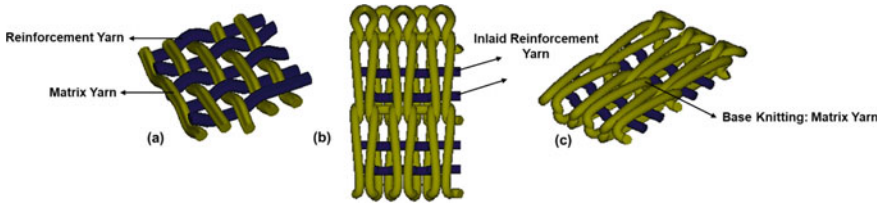

Fig. 7 **a** Woven commingled prepreg **b** knitted commingled prepreg front view **c** knitted commingled composite prepreg (top view) [26]

knitting; however, reinforcement yarns are inlaid. Asghar et al. [2] compared the mechanical characteristics of commingled and non-commingled woven thermoplastic composites using jute and polypropylene. Results elaborated that commingled composite performed better than non-commingled composites fabricated through thermoforming. Moreover, fiber wetting also showed an improvement during SEM analysis of woven commingled composites.

Awais et al. [26] compared the knitted inlaid unidirectional commingled composites and woven commingled composites, the results showed superior mechanical performance of knitted commingled composites. Woven commingled composites, architected through interlacement of thermoplastic yarns and reinforcement yarns, possess crimps in structure. The crimps are retained by reinforcement yarns during thermoforming; hence the yarns are not 100% unidirectional which leads to a negative contribution to mechanical characteristics. However, knitted inlaid structures comprise straight inlaid reinforcement yarns with base knitting thermoplastic material, the inlaid yarns do not have any crimp, so the unidirectional mechanical characteristics are better. However, fabric architectures do not influence the inherent mechanical properties of the material i.e., flax being the strongest natural fiber showing higher mechanical characteristics [27].

3 Fabrication of Commingled Composites

3.1 Thermoforming

Thermoplastic materials have the capability of being molded into newer shapes upon melting. Thermoforming composites fabrication technique involves the application of heat and pressure for converting dry commingled prepregs into a composite form. The thermoforming machine comprises of two heavy metal platens whose working temperatures can be adjusted according to requirements as shown in Fig. 8. The plates are moveable and can be set into a close position after specimen placement between the plates. Teflon sheets are utilized to avoid direct contact of composite with platens. Along with temperature setup, the thermoforming machine also comprises of hydraulic pressure setup which helps to keep specimens under constant pressure.

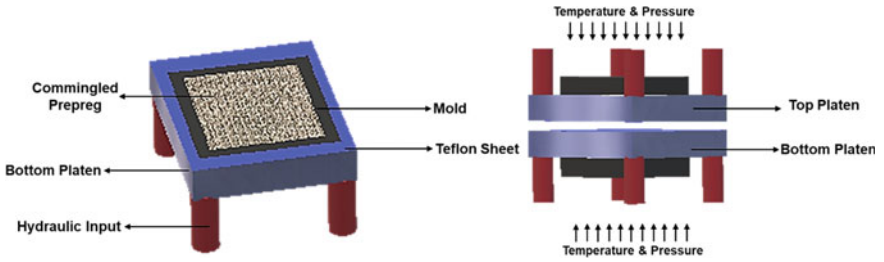

Fig. 8 Thermoforming setup used for thermoplastic composite fabrication

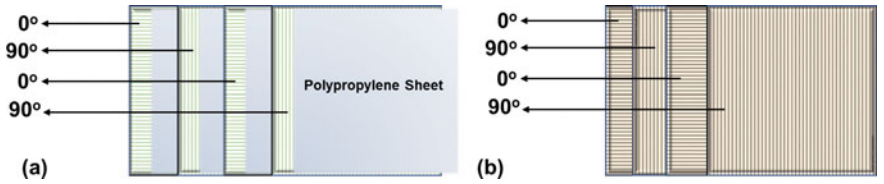

Fig. 9 **a** Non-commingled stacking sequence **b** commingled prepregs stacking sequence [26]

Temperature and pressure cycle times could be set on thermoforming machines and automatic appropriate processing could be achieved.

Stacking sequence of reinforcement materials during thermoforming helps achieve the isotropic mechanical properties. Usually, the cross-ply laminates of 0°–90° are used for both commingled and non-commingled composites thermoforming (Fig. 9). For non-commingled thermoplastic composites after each reinforcement ply a thermoplastic matrix e.g., polypropylene sheet is to be placed. However, commingled dry prepregs are directly placed upon each other in a 0°–90° sequence.

3.2 Pultrusion

Pultrusion is a well-known manufacturing technique for thermoset composites, where the product of a constant cross-section is required up to a certain length. However, for commingled thermoplastics, the pultrusion can be used as a preparatory process before thermoforming, where the thermoplastic composites are fabricated i.e., sandwich commingled structures. Straight commingled yarns are passed through the respective guides of the pultrusion machine (with no heating arrangement) in the form of two warp sheets having a predetermined distance between them. The gap can be utilized for incorporating nonwoven sheet or any other high-performance fibers governing a sandwich commingled structure, which are then subjected to thermoforming to make the sandwich composite [22].

3.3 Effect of Different Factors

Temperature and pressure are the two basic factors influencing thermoplastic commingled composites fabrication. Temperature is necessary to heat the prepreg till the melting point of the thermoplastic matrix. However, pressure is likely to be applied to overcoming the voids and required free spaces in composites which can be a place for moisture and other environmental factors settlement in the future. Pressure also allows the viable impregnation governing a better fiber-matrix adhesion and better fiber volume fraction.

However, optimization of parameters is necessary during any product engineering, and the same is for thermoplastic commingled composites. Compression pressure could be a parameter during thermoforming. While fabricating a thermoset composite by thermoforming technique, the phenomenon is different as compared to the thermoplastic composites. In thermoset composites, fibers are in a solid state while the matrix is liquid. As the compression is applied matrix being in a liquid state tends to bleed, hence there is less stress on fibers due to matrix stress dissipation. On the other hand, in thermoplastic composites both fiber and matrix are in the solid state. When temperature and pressure are applied in thermoforming, the solid matrix fibers cannot bleed, and hence remain in place until they reach their melting point. In this case, there is higher stress on fibers, which can tend fibers to group closer to each other until the matrix does not come into a molten state (Fig. 11).

Natural fibers used in commingling are usually lignocellulosic, such fibers have their unique fibrils architecture. Lumen being the sensitive portion of fibril can quench and fiber can be damaged due to instant stresses. Damaged fiber structures can lead to compromised mechanical characteristics. Different behaviors of natural fibers reinforced commingled prepregs have been shown in Fig. 10. When there is no compression, the commingled yarns are in their original round cross-sectional and relaxed state. The yarn cross-section turns elliptical while subjected to the instant compression loading, such a phenomenon compresses the reinforcement fibers and makes them closer in bundles. Lumen inside fibers also starts being compressed and fiber performance is compromised. However, in Fig. 10c during gradual compression, jute fibers remain in their original round state and are not compressed like instant loading. Hence the gradual compression loading helps natural fibers to keep their original shape retained, providing a positive share of the composite performance. Pressure increments are also crucial, short increments help to bleed out the thermoplastic matrix slowly and steadily imparting minimum stress on the reinforcement fibers.

4 Effect on Physical Properties

Physical properties of composites include fibers/yarns orientations within commingled composites, fibers wetting, and moisture management behaviors. Woven commingled composites are engineered by using matrix and reinforcement yarns

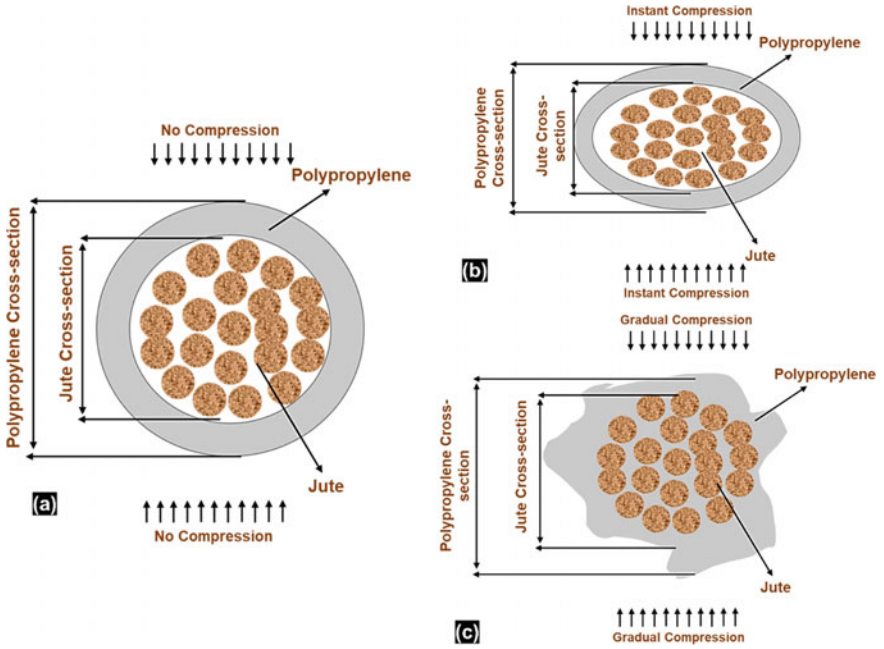

Fig. 10 Fiber structure **a** with no compression, **b** instant compression **c** gradual compression

in warp and weft respectively. Woven fabrics have crimps in their architectures, and the crimps are retained in reinforcement yarns even after composites fabrication. Such crimps contribute to directional mechanical properties reduction, as yarns are not straight. However, knitted commingled composites consist of straight inlaid yarns without having any crimp, hence the mechanical properties are superior to woven commingled thermoplastic composites [27]. Commingled composites possess better fiber wetting properties, leading to enhanced fiber-matrix adhesion. The commingling technique distributes the matrix more evenly with the reinforcement fibers, hence there are more chances of impregnation. Woven composites fabricated by thermoforming of fabrics and polypropylene sheets exhibit poor wetting compared to the woven commingled composites. Figure 11 shows the microscopic analysis of both woven and woven commingled composites. It can be seen that there is improved wetting in woven commingled composites as compared to woven composites [27].

Moisture regain identifies the amount of moisture absorbed in the dry weight of a composite. Polypropylene has the least moisture regain of 0.4%, hence is nearly hydrophobic. Increasing the amount of polypropylene in jute reinforced nonwoven composites decrease the affinity of composite towards moisture. However, the increase above a certain limit causes fiber volume fraction to decrease, which ultimately tends the mechanical performance of composites to be compromised [20].

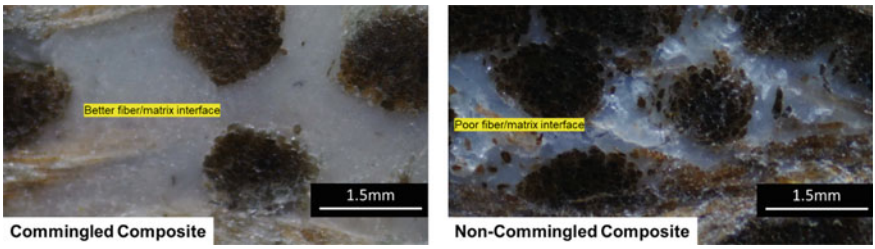

Fig. 11 Microscopic analysis woven commingled and non-commingled composites [2]

5 Mechanical Properties of Commingled Composites

Composite processing parameters are also influential in the mechanical characteristics of commingled yarns composites. Consolidation pressure and holding time directly relate with mechanical properties up to a certain limit [28].

5.1 Tensile Properties

Tensile strength describes the maximum load bear by the materials in directional extensions. Composite materials being a macroscopic combination of matrix and reinforcement exhibit the unique characteristics of both materials. Fiber matrix interface/adhesion is crucial in defining the tensile properties of composites. Natural fiber reinforced non-commingled composites exhibit poor tensile characteristics due to poor fiber wetting and low adhesion [2]. Due to this factor, the material cannot behave as a single entity compromising its strength. Fabric architectures also play an important role in tensile characteristics even within commingled composites. Stain rates during tensile testing are also influential. Tensile strength and modulus of E-glass and polypropylene woven composites increase with increasing strain rates [29]. Woven thermoplastic composites having multiple layers show better mechanical characteristics; however multiple layered composites having commingled prepregs show superior tensile strength over the same number of layers comprising non-commingled composites, as shown in Fig. 12.

5.2 Flexural and Impact Behaviors

Flexural characteristics determine the maximum bending loads and strains of composite materials. Non-commingled composites possessing poor fiber matrix adhesion have poor flexural properties. However, commingled composites have superior characteristics. Similarly, the knitted commingled UD composites have better

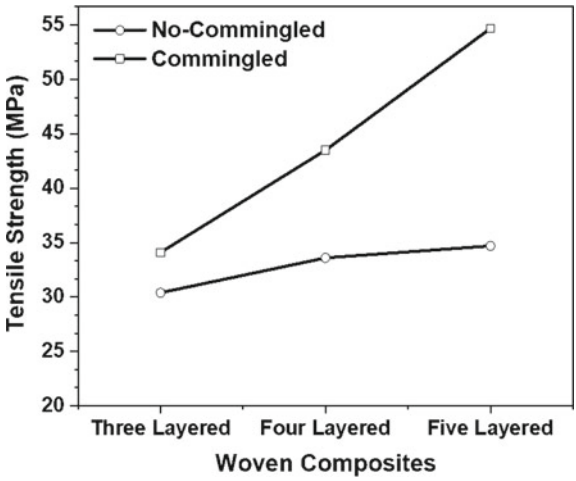

Fig. 12 Tensile strength of multiple layered woven commingled and non-commingled composites [2]

shear behaviors than woven commingled composites [27]. Shear characteristics of woven commingled composites seem to be compromised by increasing strain rates in testing [29]. Commingling level can also be influential on flexural performance i.e., fiber and yarn level commingling offer better fiber matrix interface compared to the fabric level commingling. Similarly, an increasing number of reinforcement layers enhance the composite strength. However, the overall flexural performance of commingled composites is better than non-commingled composites as shown in Fig. 13.

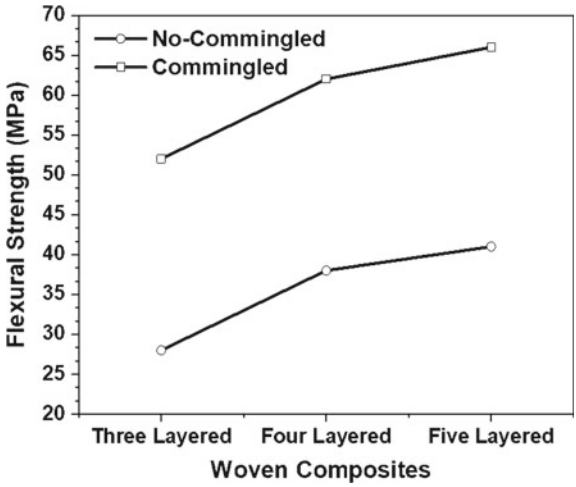

Fig. 13 Flexural strength of multiple layered woven commingled and non-commingled composites [2]

6 Conclusion

Commingling is a promising method for thermoplastic composites engineering with matrix and reinforcements having different affinities. Different commingling levels, i.e., fibers, yarns, and fabric offer a variable range of properties. Nonwoven commingled thermoplastic composites show an increasing trend of mechanical properties with an increase in matrix content up to a certain limit; however, moisture affinities have shown a linearly decreased trend. Commingled yarns can be manufactured using core-spun, core-wrapping, and co-twisting methods with some machine modifications. Both knitting and weaving can be employed in UD thermoplastic prepregs manufacturing. Knitted commingled UD composites show better mechanical performance i.e., tensile, flexural, impact, etc. as compared to woven commingled composites. Knitted and woven structural modifications are also vital in property enhancement. Though, the commingling techniques are an influential initiative toward natural fiber reinforced composites manufacturing with more effective material utilization and properties attainment.

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Process Induced Residual Stresses

Yasir Nawab, Khubab Shaker, and Abdelghani Saouab

Abstract Fiber reinforced polymer composites (FRPC) offer advantages including lightweight, flexibility of design, ease of fabrication, long service life, superior mechanical performance, etc., over conventional materials such as steel, aluminum, wood, etc. Their market has grown significantly ($101.6 billion by 2026) in automotive, aerospace, sports, and construction in the last decade. In spite of numerous benefits, these materials are complex and heterogenous in nature and have certain issues as well. Residual stress is one of these problems because two or more heterogeneous materials are present in a composite. This stress leads to deformation, fracture of the matrix, delamination, reduced strength, buckling of the fibers, etc. in a composite part. In this chapter, we discussed the factors that induce residual stress in composite material parts and its related issues. Methods for evaluation and simulation of residual stress in such materials are also discussed briefly.

Keywords Fiber reinforced composites · Residual stress · Shape distortion · Spring-in · Warpage

1 Introduction

Controlling the shape distortion induced by the manufacturing process in composite parts has been a concern for the composites industry and worried researchers over the past years. Residual stress is one of the reasons for shape distortion because two or more heterogeneous materials are present in a composite [1, 2]. Residual stress can be defined as "a stress that persists in a material that is free of external forces

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or temperature gradients". Residual stress not only causes the shape to distort, but also has a significant effect on the physical as well as structural properties of the composite parts [3].

Residual stress is inherent to the fiber reinforced polymer composites. The amount and type of this stress depends on the following factors: the stacking sequence of the reinforcement plies, curing cycle, cure dependent chemical shrinkage due to cross linking of polymer molecules in thermoset and alignment of chains in thermoplastic polymers, and mismatch of coefficients of thermal expansion. It is important that the residual stresses generated in the composites during the manufacturing process are well understood and quantified to ensure manufacturing a composite part within safety limits and reliable design [4].

The thermoset resins are mostly used in fiber reinforced polymer composites. While fabrication, the viscosity of thermoset resin is low, and it is not able to withstand an important amount of stress during the early stages of curing. However, with proceeding of curing the resin transforms to the rubbery state. In this state, its bulk modulus has a significant value so that hydrostatic stress may occur in 3D stress areas because of tool-part interaction.

The generated residual stress in the early stages of curing may cause occurrence of cracks in thick composites [5]. Further curing of polymer leads to transformation to rubbery state. The mechanical properties, such as elastic and shear moduli, of resin in the glassy state glassy state are significantly higher than those in the rubbery state. The resin in this state behaves like an elastic solid. Since curing process has completed, the major cause of strain in glassy state is thermal expansion/contraction due to temperature change. Its value is quite higher than the strain caused by chemical shrinkage due to cross-linking of polymer chains.

Residual stress is generated in a composite generally due to the offset of curing related chemical shrinkage, temperature dependent thermal shrinkage/expansion, and hygral behavior of fibers, matrix, and tool [6, 7]. The process induced residual stress in composites may be categorized into chemical, thermal and hygral stress depending on the factor creating this stress [1]. However, there is possibility that these stresses are created by more than once source. Chemical and thermal residual stresses are related to the different response of the fibers and resin during fabrication. While the hygral residual stress is the result of uneven absorption of moisture by the constituents of the composite part.

2 Mechanical Levels of Residual Stress

For better understanding, the generation of residual stresses in composite part can be studied at three mechanical levels [3].

2.1 Micromechanical Level

This stress is generated in a composite ply, at micro level, mainly due to mismatch of generated strains at the level of fibers and nearby matrix, due to application of mechanical, thermal, chemical or hygral stresses. Since, the mismatch in properties (except hygral properties) of fibers and matrix is not significant in case of natural fiber reinforced polymer composites, the amount of process induced stresses is not very high. However, due to a significant different of moisture absorption properties and consequent swelling of fibers, the effect of hygral residual stresses is significant. These stresses may cause micro cracks in the matrix and fiber—matrix debonding.

2.2 Macro-mechanical Residual Stress

This stress is generated within composite plies due to occurrence of different strains in response to process induced stresses. To understand this stress, a composite ply (layer) is considered as a unit. Depending upon reinforcement, the behavior of a ply is generally anisotropic. For example, there is a significant difference of strain response, if the ply is made using unidirectional fibers. In case of ply made from balance woven fabrics, the transverse and longitudinal properties are almost similar, however, diagonal properties are different. If nonwoven fibrous reinforcement is used, the quasi-isotropic properties can be achieved.

If a composite is made using stacking serval plies at known orientations, the difference of properties such as chemical shrinkage, thermal expansion/contraction etc. leads to generation of the residual stresses within plies.

During the fabrication process, in unidirectional plies (fibers are aligned in one direction within the ply) made from high modulus fibers, the contraction in longitudinal (fiber's direction) direction is negligible due to fiber dominant behavior, whereas the contraction is significant in transverse direction due to resin dominant behavior. This contraction is mainly result of chemical shrinkage occurs during curing process or thermal contraction occurs during cooling after curing.

In cross ply composites made using unidirectional plies the strain, caused by curing and/or cooling after curing, in 90° ply is significantly greater than the strain in 0° ply. This leads to formation of macro-mechanical stress within the plies.

In natural fiber composites, since the difference between mechanical properties of fibers and matrix is not very large, the strain response of plies can be different than as stated above. The residual stress is generally analyzed using *Classical Laminate Theory* which is also called classical laminate plate theory.

2.3 Global Residual Stress

This stress generally generates at the level of composite part due to mismatch of its global behavior. This may be due to formation of thermal gradients in the thick composites during curing and cooling [7]. Polymers are bad conductors of heat. Thermal gradients in thick polymer composites are generated as heat is entrapped in the core during fabrication process. This leads to formation of gradient of temperature, mechanical properties, chemical shrinkage and thermal expansion contraction. Similarly, due to dissimilar materials, the shrinkage and expansion behavior of mold/tool and adjacent surface of composite is different. This often results in an aparabolic distribution of residual compression stresses in the surface layers and tensile stresses in the center layers of a composite [1].

The natural fiber composites are very sensitive to the formation of thermal gradients. Most of the natural/cellulosic fibers start degrading from 200 °C, therefore, any rise in temperature due to thermal gradients may lead to deterioration of fibers [8, 9].

3 Parameters Contributing to Residual Stress Formation

Several factors have been reported in the literature [2], that contribute to the generation of residual stresses in composites. Some of the major factors are discussed below.

3.1 Coefficient for Thermal Expansion

The coefficient of expansion (CTE) is defined as the tensile or compressive strain generated in a material per one Kelvin change in temperature. Its units are K^{-1}. This is a material property. In general, the materials with higher tensile modulus have a lower value of CTE. Coefficients of thermal expansion of some of the fibers is given in Table 1.

Table 1 Coefficient of thermal expansions of some manmade and natural fibers [10]

	E-glass	Carbon	Aramid	Flax	Sisal
E_{f1} (GPa)	77	220	152	62.5	21.9
E_{f2} (GPa)	68	14	4.2	1.0	1.6
G_{f12} (GPa)	30	14	2.9	1.4	1.1
α_{f1} (μm/m.°C)	5	−0.4	3.6	−8.0	−3.9
α_{f2} (μm/m.°C)	5	18	77	83	80

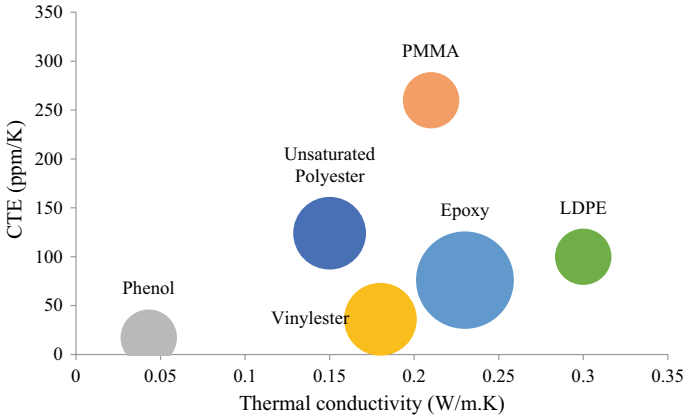

Fig. 1 Chart showing coefficient of thermal expansion and thermal conductivity of several polymer matrices

Talking about high performance composites such as Carbon/epoxy, Glass/Polyester, Kevlar/epoxy etc., the CTE of resin is generally significantly higher than CTE of the fibers. In addition, the CTE of many such fibers are orthotropic. For example, CTE of carbon fiber in the direction of the fiber is very small or slightly negative, but in the transverse direction, its value is positive.

Figure 1 shows the thermal conductivity and coefficients of thermal expansions of several polymer matrices, commonly used for composite materials. This data may be very helpful in designing composites.

The difference of the thermal expansion coefficient at micro, macro and global level is the main cause of residual stress generation, which occurs particularly during cooling once the curing of the composite is done. In case of natural fiber composites, the difference of polymer matrix and fibers in not very big and so is the amount of generated residual stresses. This stress causes warpage in the composite plates, spring-in or spring-out in the angled composite parts, micro cracks in the matrix, and fiber buckling.

3.2 Cure Shrinkage

Complex 3D crosslinking network is formed in thermosetting polymers during curing, which transform polymer from liquid to gel and finally solid state. The volume of thermoset resins shrinks during these transitions as the free volume in the polymer reduces. This reduction is called cure shrinkage or chemical shrinkage. This phenomenon is irreversible and associated with curing of thermoset polymers only. The volumetric cure shrinkage in the polymer is globally in the range of 5–8%. Depending upon the type of resin, fibers and reinforcement type, the cure shrinkage of resin may cause a global cure shrinkage in composites in the range 2–3% [11, 12].

In the composites, the residual stress is generated due to mismatch of shrinkage/expansion of fibers and polymers bonded together. During the curing process, a significant amount of chemical shrinkage occurs in resin before the gel point of resin. Since in this phase, bond between resin and fibers is not very strong, and resin being liquid is allowed to flow, the contribution of chemical shrinkage is not significant. However, after gelation, cure shrinkage can cause similar issues as done by thermal expansion/contraction. This includes formation of internal stresses at the micro and macro mechanical level, warpage of plates and spring-in of the laminated composite parts, etc. [5, 13]. The measurement and simulation of these stresses is still not fully understood. However, measurement of cure shrinkage is used to calculate internal stresses. Coefficient of chemical or cure shrinkage (CS) is a material property that is used to define shrinkage behavior of polymer. The coefficient of chemical shrinkage (CS) can be defined as 'change in dimension (Δe) due to chemical shrinkage per unit initial dimension (e_i) per change in degree of cure ($\Delta\alpha$)'. It has no unit and can be written mathematically as:

$$CS = \frac{\Delta e_{shrinkage}}{e_i \Delta\alpha}$$

In general, dilatometric techniques are used to measure volumetric chemical shrinkage of resin and composite [14, 15].

3.3 Moisture Absorption

Moisture absorption may cause swelling of either matrix or fibers. In case of manmade fiber reinforced composites, the moisture absorption of fibers is negligible whereas absorption in the matrix is relatively higher which produces similar effects to thermal or chemical volume changes. The moisture absorption of thermosetting polymers such as epoxy, phenolic and unsaturated polyester is generally less than 1% [9] in the saturation state, causing a swelling in the matrix. On the other hand, the moisture absorption of natural fibers (Table 2) is very high as compared to polymer matrices. This leads to swelling of fibers especially in the lateral direction causing fiber-matrix debonding and reduction in strength of composite material.

3.4 Tool/Part Interaction

The interaction between mold (also called tool) and composite is also a source of generation of residual stresses. Molds are generally made using Aluminum or steel. These metallic tools have significantly high CTEs in comparison to molded composite parts. During heating, due to their higher expansion, metallic molds tend to stretch

Table 2 Moisture content of different natural fibers used as reinforcement in composites [16]

Sr. #	Fiber	Equilibrium moisture content (%)
1	Hemp	11
2	Jute	9
3	Flax	12
4	Abaca	7
5	Ramie	15
6	Pineapple	9
7	Coir	13
8	Bagasse	10
9	Bamboo	8.8
10	Sisal	8.9

surface of composite parts laid towards mold. In case of shrinkage or thermal contraction, the phenomena are opposite. This may lead to generation of residual stresses in the composite part, gradient of stresses in the thickness direction and may cause bending when the stresses are released [17]. Furthermore, this tool-part interaction mechanism may lead to the locking of composite part with mold, which creates problems while releasing and can result into a damaged part.

3.5 Other Mechanisms

The resin bleed or accumulation in the composite part during fabrication may lead to variation of volume fraction along surface and through the thickness of composite part. This variation can result into distortion, even in flat unidirectional composites parts [18]. Fiber displacement during the fabrication by resin injection can cause resin rich areas, changes in geometry and properties that may result in generation of residual stresses and shape distortion. Wrinkles can also be observed at composite corners, especially in case of use of convex tooling [19].

4 Problems Generated by Residual Stress

The presence of residual stresses in the composites leads to the generation of several problems/defects in the composites including shape distortion of the final composite part, cracking of matrix, delamination of plies, reduction in strength of composite, fiber buckling in the plies, etc. There are two type of shape distortions are commonly observed in the composites known as warpage and spring-in. Warpage is the distortion of shape of thin composite laminate after fabrication which was designed as a thin plate [20]. On the other hand, spring-in can be defined as the change of corner angle

of a shaped composite part. If the part angle is decreased than the designed angle it is called spring-in, whereas if the part angle is increased, it is called spring-out. For a thermoset composite part, change in the angle of a perpendicular corner is in the order of 1°–3° [21].

5 Strategies to Reduce Residual Stresses

Following remedies are recommended to improve the fabrication process and avoid the stresses generation and deformation in the final composite part.

5.1 Countering the Effect of Chemical Shrinkage

Residual stresses arise because of constrained thermal expansion and chemical shrinkage. The fundamental way to manage the generation of stresses is therefore to match the thermal expansion and chemical shrinkage so as to minimize changes in volume early in the cure. It will be accomplished by Cure kinetics/Chemical shrinkage information.

5.2 Stress Relaxation

It can be accomplished by the following ways:

(a) Heating above Tg (This mitigates the effect of chemical shrinkage)
(b) Slow cooling rate (This reduces the effect of thermal contraction)

5.3 Modification of Product

The residual stresses are minimized effectively by modifying the composite using one of the following measures:

- Lowering the thermal and chemical shrinkage of resin
- Modifying the formulation of resin used
- Adding fillers to the part during fabrication

Adopting the first two options as a potential measure of reducing residual stress will certainly affect the performance of the part either by deteriorating the properties or not exhibiting the required properties. The product modification by the addition of fillers is currently found to be most effective means of reduction in the shape distortion [22]. The addition of fillers not only reduces the residual stresses and

shape distortion in the composite part, but also have a positive influence on the properties of composite part.

6 Brief Literature on Reduction in Process Induced Residual Stress

Several studies were reported on residual stresses and shape distortion in thermosetting composites, and its reduction. Shaker et al. [21, 22] studied the reduction in the shape distortion in flat and angled composite parts. The influence of incorporating silica microparticles on the mechanical characteristics and shape of cured glass/vinyl ester composites was the main focus. This approach used both numerical and experimental techniques for the investigations. The schematic of that study is shown in Fig. 2.

Unidirectional glass fiber, silica particles and vinyl ester resin were the constituents used for composite fabrication. For fabrication of silica particle loaded composites, the silica particles were dispersed in the matrix to prepare the suspension. The UD glass was impregnated with this suspension to fabricated composite plates and angled brackets. Shape distortion in each composite was recorded in terms of warpage and spring-in.

For numerical approach, thermal and mechanical properties of particle loaded cured resin were investigated. The incorporation of silica microparticles was found to cause reduction in thermal expansion coefficients (CTE) and enhance the resin modulus.

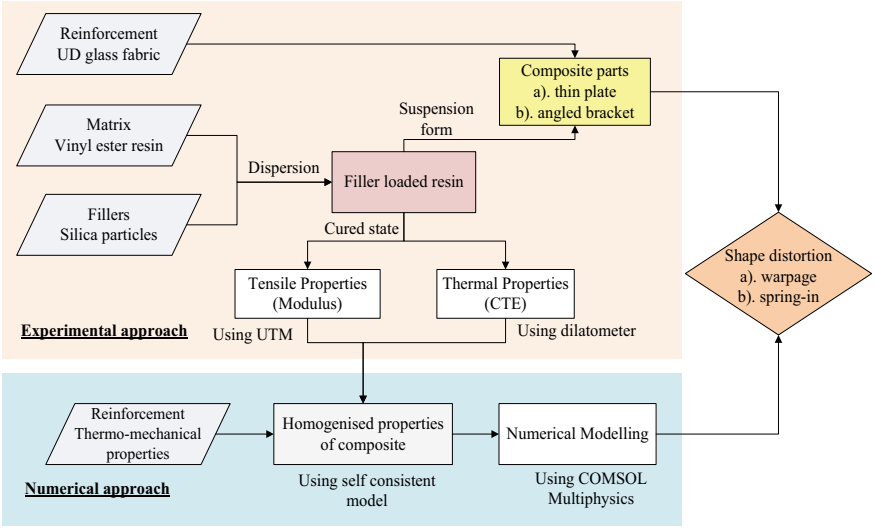

Fig. 2 Schematic of numerical and experimental technique adopted for study by Shaker et al.

The thermal and mechanical properties of particle loaded cured resin and UD glass reinforcement were used to determine the homogenized properties of composite material using self-consistent model. The numerical modeling of warpage and spring-in in flat and angled composite parts was done using COMSOL Multiphysics® (v5.4).

The experimental results showed that spring-in in angled parts was reduced from 1.807° to 0.632° with 5% fillers only. Similarly, the curvature in flat parts was found to reduce from 4.3243 to 2.0973 mm with same filler concentration. Figure 3 shows an impression of curvature in flat composite plates with and without fillers, and spring-in in angled composite parts.

The comparison of experimental and modelled results is given in Fig. 4. Both the results are in accordance. The addition of fillers also improved the mechanical performance of the resulting composites [23].

Raza et al. [24], investigated shape distortion due to process induced residual stresses in C-shaped composite parts, using silica microparticles (Fig. 5). The shape distortion was determined in terms of reduction in the part diameter. Contrary to warpage in flat plates and spring-in in angled brackets, process induced residual

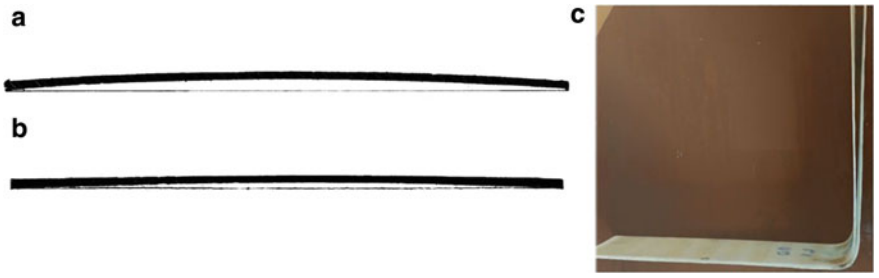

Fig. 3 Curvature in flat composite plates **a** without fillers, **b** with fillers, **c** spring-in in angled composite parts

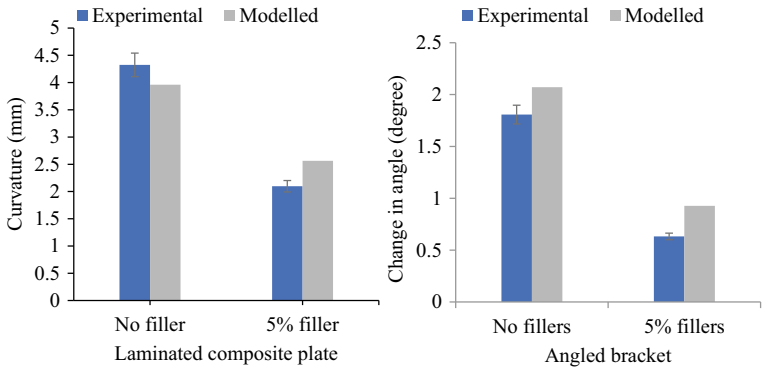

Fig. 4 Experimental and numerical results of shape distortion

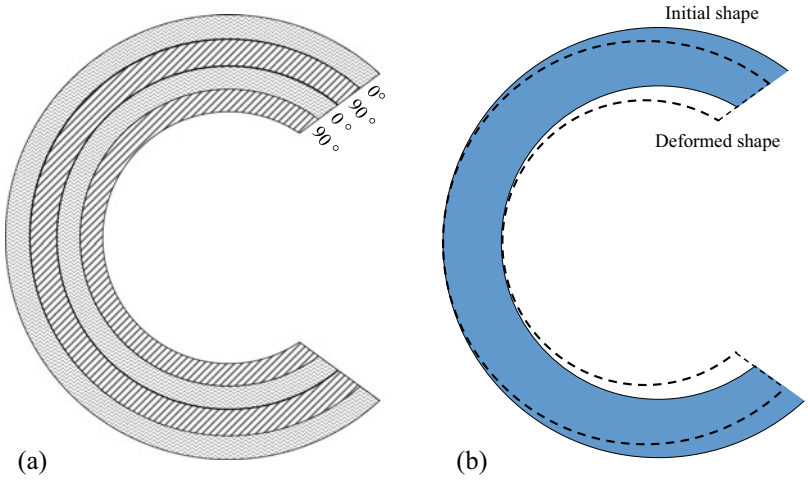

Fig. 5 **a** Stacking sequence in cylinder, **b** shape distortion in cross ply cylinder

stresses result in change of part radius for circular/C-shaped parts. The C-shaped composite parts with an enclosed angle of 240° were fabricated for this study using glass fabric and UP resin. Thermal kinetics of the resin system were investigated using differential scanning calorimetry to examine curing behavior. According to the experimental findings, adding 5% silica particles caused the diameter reduction value to drop from 6 to 3 mm. Additionally, an analytical model was used to forecast the distortion of the composite part and validate it by comparing it to the experimental values.

Ali et al. [25], studied the fabrication induced spring-back in woven composite parts with variable thickness. Reported literature focusses on shape distortion in uniform thickness plates, angled brackets or rings, but the applications of composites do not always require a uniform thickness structure, e.g., wind turbine blade, conical structures, etc. They investigated the variable part thickness (Fig. 6), angle of the bracket, gradient of resin content, and convex and concave tooling on the process induced deformation in angled brackets. The behavior of parts having a variable thickness was quite different as compared to the uniform thickness parts. Composite parts with thicker base and flanges showed up to 10% reduction in distortion. However, this effect is applicable only at large flange angles. Angled composite parts having sharper angles show an increase in distortion, when thickness is increased.

7 Conclusion

Process induced residual stress creates several issues in composite parts including shape distortion, fiber buckling, delamination and reduction in strength. There are

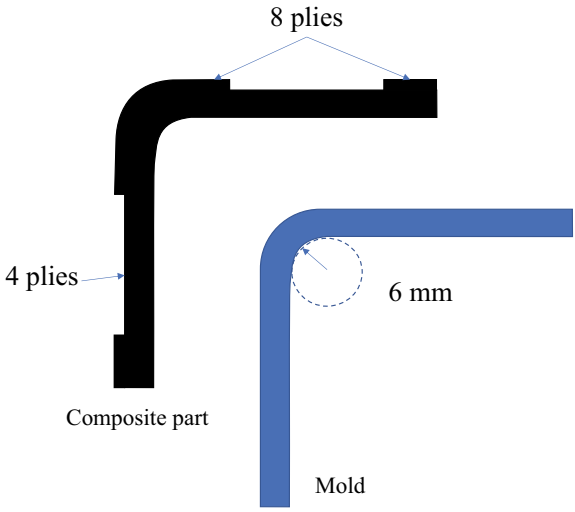

Fig. 6 Variable thickness composite parts, with increased thickness on flanges and corner

several studies available in the literature to model residual stresses considering one or all of factors including thermal expansion/contraction, chemical shrinkage, moisture absorption and tool-part interaction. However, the available literature on the residual stresses in natural fiber composites in not a lot. For natural fiber composites, it becomes difficult to understand the problem of residual stresses due to diversity in properties of natural fibers. It is almost impossible to eliminate residual stresses in composites, however, there are different techniques to lower their effect. These include the use of resin with lower chemical shrinkage, and reducing the thermal expansion coefficient of resins. The CTE can be reduced by adding nano and/or micro reinforcements in the resins while fabricating the composite parts.

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Performance of Filler Reinforced Composites

Habib Awais, Adeel Abbas, and Madeha Jabbar

Abstract Natural fiber-reinforced composites are the continuously growing class of sustainable materials for automotive, sports, construction and aerospace applications owing to their low weight, biodegradability, ease of availability and economic nature. However, fillers are mainly incorporated in natural fiber-reinforced composites to accelerate the performance characteristics of composites. The chapter describes the fillers (natural and synthetic), filler incorporation and composite fabrication techniques and the effect of fillers on performance characteristics such as tensile, flexural and impact. Finally, the application areas of filler reinforced composites are also highlighted.

Keywords Natural fiber composites · Fillers · Composite fabrication techniques · Performance characteristics

1 Introduction

Natural fiber-reinforced composites are gaining the attention of researchers as a green replacement for conventional composites, but their performance is quite low. Performance parameters are usually characterized in terms of mechanical properties, i.e., tensile, flexural, shear, impact behaviors, etc. [1, 2]. Natural fibers also lag the high-performance synthetic materials due to their porosity issues at micro, meso, and macro levels. Such pores create places for external environmental factors to influence [3]. Natural fibers polymer chains comprise hydrophilic functional groups, which are the key concern as affinities of natural fibers towards water lead to deterioration of the composites. Adhesion issues are also significantly involved in natural fiber-reinforced composites, where the hydrophilic fibers are not capable of making bonds with the hydrophobic matrix materials [4].

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Several techniques are utilized to overcome these issues to improve the fiber matrix adhesion and increase the wetting characteristics. Surface treatments of natural fibers are carried out, i.e., alkyne, saline, and acetylation treatments which remove the water-loving groups from fiber surfaces; hence the fiber's nature shifts towards hydrophobic functional groups [5]. Similarly, resins having low flow viscosities are preferred to improve impregnation. Commingling techniques: preparing dry textile prepregs are also introduced for properties enhancement. However, the fact remains that natural fibers are of low strength due to the incoming variations and polymeric structures. Hence there comes a need for third material to enhance characteristics while keeping natural fibers in use [6]. Fillers are commonly used third materials into natural fibers reinforced polymeric composites. Fillers/particle-loaded composites are capable of providing more isotropic mechanical and physical characteristics. Fillers are the nano, micro, or meso level reinforcements that are evenly distributed inside composites providing structural integrity. Fillers can be conventional or special. Conventional fillers are used only to enhance the mechanical properties; however, the specialty fillers induce tailored characteristics, e.g., anti-bacterial fillers and conductive fillers for structural health monitoring [7].

2 Natural Fillers

Natural fiber-reinforced composites are preferred due to their environment-friendly nature; hence, they are an area of growing interest [8, 9]. Synthetic fillers incorporate significant mechanical and physical characteristics into composites but are not in favor of the environment. Some of the commonly used natural filler materials have been described below.

2.1 Microcrystalline Cellulose Filler

Microcrystalline cellulose (MCC) is a very fine white powder obtained from refined wood. Due to inert nature, it has been widely used in the medical sector to create food bulk in medicines. MCC also finds applications in fiber-reinforced composites for performance enhancement [10]. A major building block of the filler is cellulose. Due to its very fine size and good dispersion, the filler offers enhanced mechanical properties at the least concentrations also. However, the dispersion factor is still crucial in order to obtain the optimum properties. Several techniques are utilized to obtain homogeneous dispersion, i.e., ultrasonic agitation, mechanical stirring, and solvent-based dispersion [11]. Using MCC with polylactic acid (PLA) matrix purposes a hundred percent green approach to composites manufacturing. The studies show that the storage modulus of MCC reinforced PLA-based composites increases

using more concentrations of MCC [12]. MCC comprises both amorphous and crystalline regions in its physical structure. The amorphous regions contribute negatively towards properties determination; hence chemical treatments are carried out to remove amorphous regions, i.e., acid hydrolysis. Such amorphous regions free MCC serves as a promising filler for polymer composites. Machining of natural fiber-reinforced composites is a crucial issue. MCC filler addition also reduces the pullouts and flush-offs during machining of flax and pineapple reinforced epoxy composites [13].

2.2 Rice Husk

Agricultural wastes keep the potential of being converted into textile materials, i.e., significant research is being carried out to engineer agro-waste-based fibers and yarns [14]. Similarly, the waste can also be utilized as filler materials for the natural fiber-reinforced composites to enhance the performance parameters of composites [15]. Rice husk is a cellulose-based natural filler obtained from the rice crop waste (Table 1). Rice husk can comprise the silica contents up to 94%; hence it is being used as an alternative to synthetic silica fillers [16]. However, the rice husk fillers to composites boost the mechanical properties related to the isotropic filler distribution in the composite. However, the physical properties, especially the moisture management characteristics, are compromised. Rice husk fillers have micro spaces in their structure, and such spaces provide the place for moisture adsorption leading to micro-level cracks into the composites. The waste settlement also increases the composite weight, hence stress failures of composites can also occur. Moisture affinities of rice husk fillers are related to the hydrophilic functional groups present in the polymer chains, i.e., hydroxyl. Since the increased percentage of rice husk fillers also increase the number of hydroxyl groups present in overall composites, more fillers addition leads to more moisture affinities [17]. Rice husk fillers only lose their 7% of weight above 500 °C temperature; however, the high-density polyethylene degrades above 550 °C. Hence the rice husk fillers could be a better alternative [18].

Table 1 Rice husk contents [14]

Rice husk content	Amount (%)
Cellulose	25–35
Hemicellulose	18–21
Lignin	26–31
Silica	15–17
Soluble	2–5
Moisture content	7–10

2.3 Wood Sawdust Fillers

Wood processing industries are among the significant waste-generating industries. Wood sawdust is a cellulose-based biodegradable material and can be potentially used as filler in fiber-reinforced composites [19]. Wood sawdust filler is also of hydrophilic nature, and the moisture management issues are always there. An increased percentage of the wood fillers in the composite materials increase mechanical characteristics [20]. More precisely, the tensile strength of wood fillers-loaded composites increases linearly with an increasing number of fillers; however, the flexural characteristics may show fluctuations somehow. Glass transition temperature indicates the softening of polymer chains and it is directly related to the amount and strength of bonding. The increased amount of wood saw fillers enhances the glass transition temperature of composites, and the phenomenon is related to the increased amount of reconstructed hydrogen bonds between the matrix, reinforcement, and fillers functional groups [21]. Different types of wood saw fillers provided different characteristics depending on the processed wood origin. Chemical treatments of the fillers are performed to increase the adhesions; however, some studies reveal that NaOH-treated wood fillers loaded composites show poor mechanical properties than untreated composites. Chemical treatments remove the lignin content from the wood; microvoids are created inside fillers which are the prior places for moisture settlement. Similarly, the wood fillers incorporated with composites tend to increase the thermal conductivities of composites. The fillers naturally comprise thermal releasing agents; hence the phenomena occur [21].

2.4 Coconut Shell Fillers

Coconut shell fillers are among the agricultural waste that comprises around 36–43% cellulose, 41–45% hemicellulose, 0.15–0.25% lignin, and 3–4% pectin. Cellulose being the significant part creates similar hydrophilicity issues as are discussed for earlier natural filler materials. Hence an optimized percentage of the coconut shell filler is required to fulfill the end requirements [22]. Coconut fiber reinforced epoxy composites having coconut fillers exhibit better performance by 8% filler concentration. Below 8% concentration, the coconut fibers are insufficient to support the composite's performance; however, above 8%, the hydrophilicity issues evolve, affecting the physical and mechanical characteristics [23]. Before moisture settlements, the coconut shell fillers-loaded composites can show enhanced mechanical performance with increasing filler concentration. However, the possible agglomerations of the coconut shell fillers with matrices can lead to decreased moisture affinities even at higher concentrations of fillers [24].

2.5 Peanut Shell Fillers

Peanut is the most existing dry fruit and is consumed in massive amounts. There is a considerable peanut shell wastage that can potentially be utilized as filler materials in fiber-reinforced composites. Like other plant-based materials, peanut shells are composed of cellulose, hemicellulose, and lignin constituent [25]. Peanut shell powders have been utilized as fillers in rubber materials and offer a noticeable improvement in torque behaviors. Alkali treatments of peanut shell powders are carried out to create better adhesions with the matrix. FTIR analysis of epoxy-based composites reveals better intensities of NaOH-treated fillers within composites than untread ones. NaOH treatment removes impurities and lignin content from the shells, enhancing mechanical characteristics [26]. Polarities difference is crucial in filler-matrix adhesions. Such polarity difference is due to filler and matrix hydrophilic and hydrophobic natures, respectively. If the filler concentration increases to a certain level, the polarities difference may lead to decreased mechanical performance compared to the fillers-free composites [27].

2.6 Egg Shell Fillers

Eggs are used in daily life for food purposes; hence a substantial amount of egg-shell waste is generated from housing communities to restaurants. Using the egg-shell as a filler material in fiber-reinforced composites can reduce the egg-shell wastage from the environment and enhance the performance characteristics of fiber-reinforced composites [28]. Eggs are protein-rich products, so the egg shell is majorly composed of proteins, unlike cellulose-based rice husk, wood, and coconut shell fillers. Egg shells are fine ground and converted into a valuable fillers state. Egg-shell fillers loaded glass/epoxy composites exhibit a noticeable improvement in mechanical performance parameters even at the 6% concentration [29]. Glass fiber reinforced epoxy composites are susceptible to flammability issues; however, the addition of egg-shell fillers into the glass/epoxy composite generates an excess amount of $CaCO_3$ which liberates the CaO_2 and CO_2; hence the flammability is reduced. Moreover, the smaller the size more the egg-shell fillers will contribute to the mechanical strength of composites [30].

2.7 Wheat Straw Filler

Wheat straws are the waste of wheat crop after harvesting and are usually burned, creating climatic issues in several countries. However, the waste can be effectively utilized as filler materials in fiber-reinforced composites. Such tactics reduce not only environmental burdens but also servers for the waste value enhancement along with

composites mechanical properties improvement [31]. Adding wheat straw fillers enhances mechanical properties; however, there are chances of poor fiber matrix adhesion due to polarity differences between hydrophilic filler and hydrophobic matrix [32].

2.8 Fish Bone/Fish Scale Fillers

Hydroxyapatite (HAP) enriched collagen waste of fish scales is widely found in plywood wastages. Fish scales are a considerable choice for being used as filler materials due to their chemical composition and usage in the defense and electronic industry [33]. Fish scale addition as a filler in the fiber-reinforced composites serves for mechanical properties, i.e., tensile strength enhancement up to 22% even at the 4% filler concentrations [34]. Fish scale fillers also work for the reduction of the pore in the fiber-reinforced composites, and the tendency to rust is reduced. Smaller filler grains have low specific gravity values, which contribute toward more even distribution of fillers inside the composite, leading to more isotropic characteristics [35]. Tensile strength of fish scales loaded composites increases linearly with increasing filler concentrations; however, flexural behaviors are different for different materials, and specific studies are necessary to evaluate the trend [36].

2.9 Clay Filler

Clay is a natural mineral material composed of fine soil grains. Chemical constituents of clay are magnesia, silica, alumina, and water., there are also some variable portions of potassium, calcium, sodium, etc. Clay has the capability of being soaked up and releasing ions; hence the bonding potential of clay is better [37]. Clay is used as filler in plastics and fiber-reinforced composites. Clay converted into nano-sized particles works tremendously for providing the isotropic characteristics in fiber-reinforced composites. Although the major use of clay fillers is in plastics and rubber materials, fiber-reinforced composites also have a significant scope [38].

2.10 Fly Ash Filler

Fly ash is the residual of coal burning. It comprises glassy particles of sphere shape mostly in grey color. Coal composition/grades define the chemical constituents of the fly ash; hence the amount of silica, alumina, and magnesium is different [39]. Fly ash-filled jute reinforced epoxy composites show enhanced hardness and wear resistance

compared to the unfilled composites. Fly ash fillers are influential in developing lightweight composites for marine applications; fly ash fillers increase modulus while keeping low densities [40].

3 Synthetic Fillers

Natural fillers are considered suitable due to their eco-friendly nature; however, physical properties-related issues exist. Natural fillers have polarity differences with the synthetic matrices, leading to poor adhesions and compromised performance. Hence synthetic fillers can be a better alternative regarding the composite performance and service lives. The section describes some common synthetic fillers used in fiber-reinforced composites.

3.1 Graphite Filler

Graphite: a crystalline form of carbon consisting of graphene stackings, is an anti-friction and wear-resistant material. The powder form of graphite can be used as a filler in composite materials. Graphite filler addition improves both surface and mechanical properties significantly. Due to its good tribological properties, it reduces the malleable nature of composites; hence increasing graphite filler percentage increases the brittleness of composites [41]. The degradation temperature of the graphite-loaded epoxy composites is observed to be higher than the simple epoxy composites. So, graphite fillers are solution to improve epoxy composites degradation temperatures. Both in-plane and through-plane thermal conductivities of the composite materials are enhanced using the graphite fillers [42]. The shape and layer proportion of graphene within graphite are influential in defining the composite performance. Slippage of graphene layers can sometimes lead towards the properties deterioration of composites; however, the issue could be overcome by using the modified graphite [43].

3.2 Zinc Oxide Filler

Zinc oxide (ZnO) is a white inorganic powder insoluble in water and has nonpolar nature. The ZnO powder has good adhesive and antimicrobial characteristics keeping its place in medical applications [44]. Such fillers can be used in fiber-reinforced composites to induce antimicrobial properties. Mechanical properties are also enhanced using the fillers due to isotropic distribution within the matrix. Tensile strength increases linearly with increasing concentration, while flexural behaviors are the base composite material-dependent [45]. However, increasing the ZnO filler

concentration to a specific limit causes the matrix percentage to decrease. Load distributions are not possible evenly due to the reduced percentage, and performance is compromised [46].

3.3 Calcium Carbonate Filler

Calcium carbonate ($CaCO_3$) is the carbonium salt commonly found in daily life applications. It occurs in abundance and could be in the form of aragonite, calcite, and vaterite. Calcite is more existing phase than other ones. The natural surface characteristics of the material are not satisfying, i.e., irregular. Hence surface modifications are carried out to improve properties creating better adhesion. Lower concentrations of $CaCO_3$ can also improve mechanical characteristics if the fillers are incorporated at the nano-level inside the fiber-reinforced composites. Nano-level particles have effective diffusion characteristics; hence the tensile and compressive properties increase. Degradation temperatures of the $CaCO_3$-filled composites, i.e., kenaf and HDPE, are enhanced compared to the unfilled composites governed by better adhesions due to $CaCO_3$.

3.4 Boron Carbide

Boron carbide is the boron–carbon ceramic material and is the hardest after diamond and cubic boron nitride. Boron carbide fillers have good abrasion and wear-resistant characteristics. Low density and hard materials are available in commercial and domestic lightweight products [47]. Increased concentrations of the filler increase the sample stiffness. Thermal degradation temperature is also increased; hence the filler addition leads to the composite thermal stability. Due to brittle nature, boron carbide composite bending properties are not significant; however, the tensile properties are higher than simple composites [48]. The fillers are of sharp polygonal edges; hence, increased concentrations can induce cracks in the composites. Combustion characteristics are increased by the boron carbide filler addition, as the overall heat release value is decreased by the fillers [49].

3.5 Aluminum Oxide Filler

Aluminium oxide is a chemical combination of aluminium and oxygen atoms and is used as filler materials in polymer matrix composites [50]. The aluminium oxide offers enhanced mechanical properties, making it a suitable choice for both metal and polymer matrix composites. The filler addition contributes positively to both

tensile and flexural characteristics. Being metallic oxides, the fillers possess good thermal conductivity and are acknowledged for heat dissipation applications [51].

3.6 Carbon Nano Reinforcements

Carbon nanomaterials, including carbon nanotubes, carbon nanoparticles, and carbon nanodots, are novel materials being focused as filler materials in composite applications. Carbon nano reinforcements in epoxy using carbon nanodiamond (CND), carbon nanotubes (CNT), carbon fibers (CF), and graphene oxide (GO) platelets have been incorporated. Surface area per unit volume and interface volume per unit particle influenced mechanical properties. Carbon nanotubes prove to be optimum for mechanical characteristics enhancement of both tensile and fracture toughness [52]. Ultrasonication energy and time factor correlate with carbon nanotubes (CNTs) and graphene oxide (GO) dispersion quality within paste and mechanical properties of cementitious composites. UV visible spectrum used for dispersion analysis shows an increase in absorbency w.r.t time and energy. Mechanical characterization consisting of flexural testing reveals an increase in Young modulus (E), flexural strength (σ_f), and fracture energy (G_F) by increasing ultrasonication time. At the same time, the increase in ultrasonication energy increases the properties to a certain limit, after which damages governed a sharp decrease in nanoparticles [53] Cylindrical (nanofiber) NF and (graphene oxide) GO platelets perform viably in mode I and mixed-mode loadings, where nano reinforcement act as a laminate barrier restricting further crack propagation. However, (nanodiamonds) ND reinforced composites show the highest fracture resistance in pure mode II failure [54]. Carbon nanofibers (CNF) and granulated graphitized graphene (GRP) reinforced polypropylene (PP) composites are usually first melt-extruded and then injection molded. Mechanical characterizations of carbon nanoparticles reinforced PP nanocomposites showed the highest mechanical responses on 45 wt% and 60 wt% of CNF and GRP, respectively. The weight percentages are optimal where maximum bonding occurs between polymer chains and fillers, creating a strong interface [55]. Liquid nano reinforcements can be easily dispersed into the polymer solution. Liquid nano reinforcements include carbon nanofiber (CNFs), carbon nanotubes (CNTs), and hybrid nanomaterials. Hybrid nano reinforcement exhibit higher flexural properties showing a broader fracture resistance [56].

4 Manufacturing Techniques: Filler Loaded Composites

4.1 Vacuum Infusion

Vacuum infusion is a composite fabrication technique where the resin/matrix is incorporated into reinforcement by creating an artificial vacuum through the compressed

air. The setup follows a procedure where the dry reinforcement is laminated on a platform using the standard protocols, i.e., Teflon sheet placement between laminate and platform. The vacuum infusion composite engineering offers better volume fractions, clean production, less matrix wastage, and better laminates consolidation benefits [8]. However, the setup is somehow complicated to install, and there as possible initial errors. For filler/particle-loaded composites, vacuum infusion is a preferable method. Regular vacuum resin infusion inserts resin directly after the infusion setup preparation in filler-loaded composites. The fillers are firstly dispersed in resin to prepare a solution comprising the required filler concentration. The stage is termed the suspension preparation phase. Dispersion can be performed through mechanical, chemical, and ultrasonic methods (Fig. 1). The mechanical approach involves stirring the matrix after fillers addition, and chemical dispersion uses a third binding/dispersing agent to distribute fillers evenly in the whole matrix. Ultrasonic fillers dispersion is carried out using the sonication technique. Filler and matrix are placed inside the ultrasonic agitation chamber, where the dispersion is carried out through ultrasonic vibratory waves [57–59].

After suspension preparation, there comes the suspension infusion stage. The filler dispersed resin/matrix pot is connected to the vacuum infusion setup. Dispersion is a tailored particle fluid system where the intention is to keep particles evenly distributed within the fluid during the infusion process rather than segregating them [60]. However, some issues are faced during suspension infusion. The flow of resin slows down due to a constantly applied injection pressure, and there arises a chance of gel formation before the infused resin completes its path. Dry spots can also occur with reinforcement due to the non-homogenous distribution of matrix, and specifically, the uneven distribution of fillers with matrix and reinforcement. Different filtration behaviors are also governed due to varying ratios between filler diameter and reinforcement pore sizes. When the filler size is larger than the pore size, particles form a cake-like stacking over the reinforcement, preventing even filler distribution.

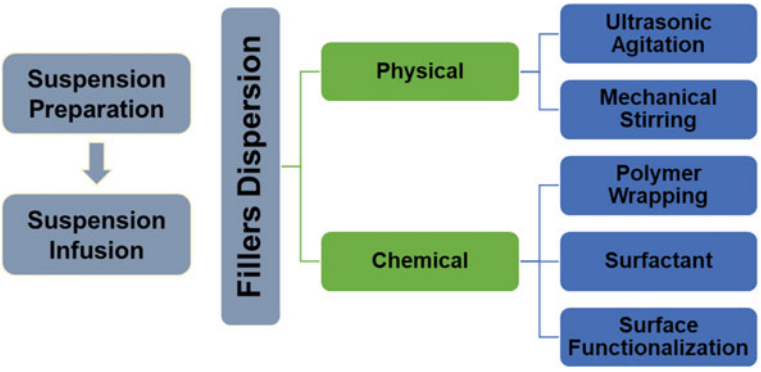

Fig. 1 Suspension preparation techniques

Smaller particle sizes than reinforcement pores lead toward settlement of fillers in the bottom bed after being filtered out from reinforcement. However, appropriate filler and pore size governs an even filler distribution, and the phenomenon is termed deep bed filtration [61].

4.2 Hand Layup Technique

Hand layup is the old and most conventional method of composites manufacturing [62]. For filler-loaded composites, the technique also has two phases. Fillers dispersion is carried out similarly by any of the methods described above. However, composite plies are stacked one by one through hands after the suspension preparation, and resin/matrix is spread by hand consolidation onto the layers [63]. The technique is simple compared to vacuum infusion, but the irregularities and non-homogeneity issues are more prominent due to more human factor involvement.

4.3 Thermoforming

The technique is mainly followed up for thermoplastic composites fabrication. Dry thermoplastic prepregs are subjected to high temperature and pressure, causing the matrix to flow and impregnate inside reinforcement [64, 65]. However, in filler-loaded thermoplastic composites, dry prepregs are first dipped in an emulsion consisting of fillers; the immersing lead fillers to be settled onto the dry prepregs. Such fillers embedded prepregs are then placed into a hot compression/thermoforming machine (Fig. 2). Before prepreg immersion, there is a similar process of suspension preparation either through mechanical or chemical techniques [66].

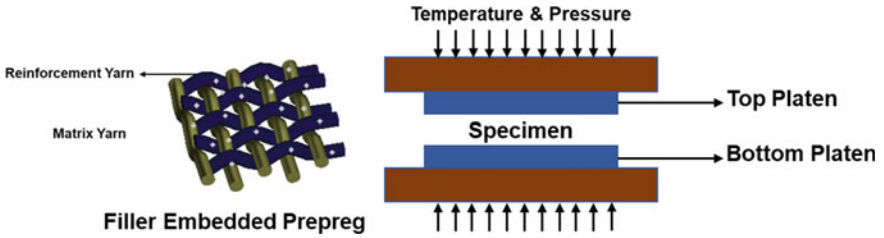

Fig. 2 Thermoforming

5 Effect of Fillers on Performance

Filler-loaded composites are engineered to enhance the performance of conventionally manufactured composites with anisotropic reinforcements. The performance parameters are characterized in terms of tensile, flexural, impact loading properties, etc. The section briefly describes the influence of different filler materials on the composite performance reported in the literature.

5.1 Tensile Characteristics

Tensile loading identifies the composite performance when stretched axially. The characterization determines the directional mechanical characteristics of materials. Fillers-loaded composites show better tensile performance over the simple two-system (matrix and reinforcement) composites. Fillers form the micro, meso, or macro-level interfaces depending on their size, and more interfacial points are responsible for enhanced tensile properties. Some of the literature-reported results are shown in Table 2.

5.2 Flexural Characteristics

Flexural loading helps to determine the bending behaviors of composites. The specimen is placed on two clamps, and a third vertical jaw tends to bend the specimen from the center. Flexural strength: maximum bending forced born by the specimen, flexural strain, and flexural modulus are the output parameters. Although the fillers loading enhances flexural performance, some of the results from the literature are shown in Table 3.

5.3 Impact Strength

Impact strength is a measure of material impact resistance and is quantified in terms of energy absorbed before fracture. Fillers addition enhances the impact performance through the more isotropic distribution of filler reinforcements inside composites. The impact properties of some literature reported filler-loaded composites are shown in Table 4.

Table 2 Effect of fillers on tensile characteristics

Sr. No.	Reinforcement	Matrix	Filler	Concentration (%)	Tensile strength (MPa)	Tensile modulus	References
1	Sisal fabric	Epoxy	Silicon carbide	0.72	31.853	0.2219	[67]
2	Hemp, and aloe vera	Epoxy	Barium sulphate	5	49 N/mm^2	–	[68]
3	Flax and hemp	Epoxy	Barium sulphate	5	55 N/mm^2	–	[68]
4	Jute woven fabric	Epoxy	Silicon carbide	2.5	55	12.5	[69]
5	Jute woven fabric	Epoxy	Silicon carbide	5	57	16	[69]
6	Jute woven fabric	Epoxy	Silicon carbide	7.5	62	14	[69]
7	Jute woven fabric	Epoxy	Aluminum oxide	2.5	60	12.3	[69]
8	Jute woven fabric	Epoxy	Aluminum oxide	5	60	16	[69]
9	Jute woven fabric	Epoxy	Aluminum oxide	7.5	33	8.5	[69]
10	Bamboo fiber	Polyester resin	Micro coconut shell powder	5	35.9	–	[70]
11	Bamboo fiber	Polyester resin	Nano coconut Shell powder	5	46.34	–	[70]
12	Jute fiber	Epoxy	Silica powder	5	80	–	[71]
13	Jute fiber	Epoxy	Silica powder	10	69	–	[71]
14	E-glass	Epoxy	Graphite powder	2.5	182.6	9.875	[72]
15	E-glass	Epoxy	Graphite powder	5	191	10.005	[72]
16	E-glass	Epoxy	Graphite powder	7.5	205.1	10.501	[72]
17	E-glass	Epoxy	Graphitic carbon nitride	1	175	–	[73]
18	E-glass	Epoxy	Graphitic carbon nitride	1.5	211	–	[73]

(continued)

Table 2 (continued)

Sr. No.	Reinforcement	Matrix	Filler	Concentration (%)	Tensile strength (MPa)	Tensile modulus	References
19	E-glass	Epoxy	Graphitic carbon nitride	2	245	–	[73]
20	E-glass	Epoxy	Graphitic carbon nitride	2.5	249	–	[73]
21	E-glass	Epoxy	Graphitic carbon nitride	3	214	–	[73]
22	Kenaf fiber	Epoxy	Oil palm nanofiller	3	35	5000	[74]
23	Kenaf fiber	Epoxy	Montmorillonite	3	45	5100	[74]
24	Kenaf fiber	Epoxy	Organically modified montmorillonite	3	58	5900	[74]
25	Jute fiber	Polyester	Fly ash	2	31.4	–	[40]
26	Jute fiber	Polyester	Fly ash	4	31.17	–	[40]
27	Jute fiber	Polyester	Fly ash	6	30.91	–	[40]
28	Jute fiber	Polyester	Fly ash	8	29.83	–	[40]
29	Jute fiber	Polyester	Fly ash	10	28.94	–	[40]
30	Jute fiber	Polyester	Wood charcoal	4	32	1150	[75]
31	Jute fiber	Polyester	Wood charcoal	8	28	1050	[75]
32	Jute fiber	Polyester	Wood charcoal	12	27.5	1150	[75]
33	Jute fiber	Polyester	Wood charcoal	16	25	1100	[75]
34	Jute fiber	Polyester	Wood charcoal	20	20	970	[75]

Table 3 Effect of fillers on flexural characteristics

Sr. No.	Reinforcement	Matrix	Filler	Concentration (%)	Flexural strength (MPa)	Flexural modulus	References
1	Sisal fabric	Epoxy	Silicon carbide	0.72	168.396	10.430	[67]
2	Hemp, and aloe vera	Epoxy	Barium sulphate	5	77 N/mm^2	–	[68]
3	Flax and hemp	Epoxy	Barium sulphate	5	87 N/mm^2	–	[68]
4	Jute woven fabric	Epoxy	Silicon carbide	2.5	103	5.5	[69]
5	Jute woven fabric	Epoxy	Silicon carbide	5	113	7.4	[69]
6	Jute woven fabric	Epoxy	Silicon carbide	7.5	107	6.2	[69]
7	Jute woven fabric	Epoxy	Aluminum oxide	2.5	105	4.8	[69]
8	Jute woven fabric	Epoxy	Aluminum oxide	5	106	5.6	[69]
9	Jute woven fabric	Epoxy	Aluminum oxide	7.5	102	5.2	[69]
10	Glass woven fabric	Vinyl ester	Calcium carbonate	1	75 GPa	6.8	[76]
11	Glass woven fabric	vinyl ester	Calcium carbonate	3	90 GPa	7	[76]
12	Glass woven fabric	Vinyl ester	Calcium carbonate	5	91 GPa	7.1	[76]
13	Glass woven fabric	Vinyl ester	Phenolic hollow microspheres	1	128 GPa	6.5	[76]
14	Glass woven fabric	Vinyl ester	Phenolic hollow microspheres	3	110 GPa	6	[76]
15	Glass woven fabric	Vinyl ester	Phenolic hollow microspheres	5	117 GPa	6	[76]
16	E-glass	Epoxy	Graphitic carbon nitride	1	229	–	[73]
17	E-glass	Epoxy	Graphitic carbon nitride	1.5	245	–	[73]

(continued)

Table 3 (continued)

Sr. No.	Reinforcement	Matrix	Filler	Concentration (%)	Flexural strength (MPa)	Flexural modulus	References
18	E-glass	Epoxy	Graphitic carbon nitride	2	270	–	[73]
19	E-glass	Epoxy	Graphitic carbon nitride	2.5	253	–	[73]
20	E-glass	Epoxy	Graphitic carbon nitride	3	220	–	[73]

6 Application Areas

Filler-loaded composites find wide application areas due to their isotropic and enhanced mechanical characteristics. Thermoset epoxy composites are preferred in aerospace applications, and adhesive bonding techniques are used for the joining of components. Due to their inferior features than heavy aerospace metals, natural fiber composites are not preferred for outdoor applications. Hence are primarily used in the interior parts, i.e., seats, luggage racks, etc. Fillers embedded natural fiber composites show enhanced mechanical performance, allowing natural fiber composites to be used in some useful aerospace articles [77]. Airbus 230, 310, 320, and 340 have attained up to 400 kg weight reduction using filler-loaded natural fiber composites [78]. However, synthetic high-performance fibers reinforced composites are still superior to natural fibers due to their polymer chain structures. Filler-loaded polymer composites are also used for aerospace antistatic applications [79]. The automotive sector utilizes a significant portion of filler-loaded fiber composites. About 31% of wood plastic composites are utilized in automotive interiors. In contrast, there is a 1% contribution towards the aerospace sector. While there is, 12% of usage in marine interiors [80]. Hybrid cellulose-glass fiber composites are desired in automotives [81]. Filler embedded composites also have usage in medical applications. Both implantable and non-implantable medical products can be engineered using filler-loaded fiber-reinforced composites. Ceramic fillers and polylactic acid (PLA) composites are used in bone fracture fixations [82]. Biomaterials containing composites are used in dental replacements [83]. Silver and zinc oxide-filled composites are useful in anti-bacterial medical applications [84]. Similarly, filler-loaded composites have a variety of applications that will be challenging to describe under one flag; however, advancements are being carried out in each field, enhancing technology day by day.

Table 4 Effect of fillers on impact strength

Sr. No.	Reinforcement	Matrix	Filler	Concentration (%)	Impact strength (J/m)	References
1	Sisal fabric	Epoxy	Silicon carbide	0.72	168.396	[67]
2	Hemp, and aloe vera	Epoxy	Barium sulphate	5	10.5	[68]
3	Flax and hemp	Epoxy	Barium sulphate	5	35.5	[68]
4	Jute woven fabric	Epoxy	silicon carbide	2.5	39	[69]
5	Jute woven fabric	Epoxy	Silicon carbide	5	42	[69]
6	Jute woven fabric	Epoxy	Silicon carbide	7.5	50	[69]
7	Jute woven fabric	Epoxy	Aluminum oxide	2.5	38	[69]
8	Jute woven fabric	Epoxy	Aluminum oxide	5	40	[69]
9	Jute woven fabric	Epoxy	Aluminum oxide	7.5	43	[69]
10	Glass woven fabric	Vinyl ester	Calcium carbonate	1	75	[76]
11	Glass woven fabric	Vinyl ester	Calcium carbonate	3	78	[76]
12	Glass woven fabric	Vinyl ester	Calcium carbonate	5	70	[76]
13	Glass woven fabric	Vinyl ester	Phenolic hollow microspheres	1	90	[76]
14	Glass woven fabric	Vinyl ester	Phenolic hollow microspheres	3	89	[76]
15	Glass woven fabric	Vinyl ester	Phenolic hollow microspheres	5	97	[76]
16	Kenaf fiber	Epoxy	Oil palm nanofiller	3	24	[74]
17	Kenaf fiber	Epoxy	Montmorillonite	3	31	[74]

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Testing of Natural Fiber Composites

Muhammad Imran Khan and Mehmet Karahan

Abstract Efforts have been made to develop biodegradable and environment-friendly materials due to environmental issues and increased dependency on fossil fuels. The usage of natural materials in composites has increased, resulting in lower greenhouse gas emissions and a lower carbon footprint for composites. Green composites have the potential to be a viable replacement for petroleum-based materials. However, it cannot be accomplished without addressing a number of concerns, including but not limited to the adhesion of the matrix with the natural fibers, reduced fire resistance, hydrophilic nature, less impact strength, durability, etc. Researchers have explored the performance of the composites reinforced with natural fibers in terms of different characterizations like physical testing (Surface morphology, thermal testing, moisture absorption, etc.), mechanical testing (tensile, compression, flexural, impact, hardness, etc.), and nondestructive testing (ultrasonic, radiography, etc.). This chapter includes the standard test methods used for these tests and the properties of natural fiber composites obtained after testing.

Keywords Characterizations · Eco-friendly · Mechanical · Thermal · Physical

1 Introduction

Composites are made up of inherently diverse materials that, when combined, generate a material with qualities that are superior to the constituent materials. A typical composite is made up of a matrix that holds reinforcing materials in place. The Fibers, which are the most essential reinforcing components, provide the composite's basic strength. Reinforcing materials, on the other hand, can provide much more than

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just strength. It can conduct heat/electricity and withstand chemical corrosion. Reinforcement can be chosen based on stiffness (modulus of elasticity) or a variety of other characteristics [1].

Natural fiber-reinforced composites (NFRC) can be tested for physical, thermal, and mechanical properties. The investigation of NFRC mechanical properties is critical in determining their final applications. Mechanical properties of NFRC are governed by the constituent including fibers, matrix, and the fiber/matrix interfacial bonding. Fiber strength is directly dependent on numerous elements such as fiber length, fiber shape, fiber bonding, voids, and moisture in NFRC. In addition, different natural Fibers require different surface treatments to prevent moisture from affecting the properties of natural Fiber reinforced composites. The matrix, on the other hand, is in charge of preventing cracks from propagating between Fibers, keeping Fibers in appropriate orientation, protecting Fibers from the environment, and, most critically, transferring weight from broken Fibers to surrounding unbroken Fibers across the interface [2–4].

Natural Fibers have gained popularity as a replacement for glass Fibers in composite components over the last decade, particularly in the housing industry. Jute, sisal, coconut Fiber (coir), ramie, banana, flax, hemp, and other natural Fibers are inexpensive, have a higher stiffness per unit weight, and have a lesser environmental impact. Structural applications are uncommon due to the inapplicability of present production procedures to such NFRC products and the lack of suitable semi-finished materials. Natural Fibers' intermediate mechanical qualities preclude them from being employed in high-performance applications, such as where carbon reinforced composites would be used, although they can compete with glass fibers for a variety of reasons [5].

However the properties of NFRCs can be improved by the treatment of the natural fiber before the fabrication of the composite. The choice of their consideration is determined by their advantages and disadvantages. When compared to glass fiber, NFRCs are lightweight, which results in higher specific strength and stiffness, making them a better choice for parts that require bending stiffness. Many components are now made as composites, with polyester or polypropylene as the primary materials. Fibers such as flax, jute, sisal, banana, or ramie can be used instead. It can be shaped into sheets, boards, and gratings, among other things [6].

Natural fibers are classified according to the source, whether they come from plants, animals, or minerals. Natural Fibers are presently used in a wide range of industries, including building, automotive, plumbing, and furnishings. Bio-based materials, a new field, promises to produce eco-friendly, high-performance NFRC that have potential to replace synthetic materials [7, 8].

2 Natural Fiber as Biodegradable Materials

Due to ecologically beneficial qualities like as biodegradability, which can lead to a large reduction in carbon footprint, NFRCs have been extensively explored by scientists and engineers in several areas. NFRC have been used in applications in sophisticated industry sectors such as automotive and aerospace to promote sustainable technology [9]. Natural fibers have been used in the fabrication of green composites by many researchers. Jute fiber has a low density, improved elongation, and higher tensile strength. According to the literature, jute fiber has a higher tensile strength than other natural fibers [10].

3 Natural Fibers Properties

Natural fibers are classified as vegetable, animal, or mineral based on their origins. Vegetable fibers can be found in cereals and fodder stems, fruit, seed, leaves, straw, or bagasse. Fibers have a complicated chemical composition and structure due to their organization, which is similar to that of a composite material. Nature created it, generating a hard matrix with crystalline cellulose micro fibrils fused with lignin and/or hemicellulose. Except for cotton, most vegetable fibers are mostly made up of cellulose, hemicellulose, lignin, waxes, and certain water-soluble components [11]. Cellulose, hemicelluloses, pectin, and lignin are the main components of natural fibers. Depending on the fiber type, the percentage of each of these components varies. This difference can also be influenced by growing and harvesting conditions. Natural fibers' hydrophilic qualities are due to cellulose, a semi-crystalline carbohydrate. Because of their amorphous form, hemicelluloses are relatively soluble in water and alkaline solutions. Pectin is a polysaccharide that holds the fiber together, similar to cellulose and hemicellulose. Unlike hemicellulose, lignin is an amorphous polymer that is primarily aromatic in nature and has little effect on water absorption [12, 13].

The mechanical properties of natural fibers represents a cornerstone especially in compost manufacturing and usage. A lot of attention has been given to the mechanical properties before deciding which fiber to use for any application the properties of different natural fibers are shown in Table 1.

It is mentioned earlier that the properties of natural fibers is more essential issue in the composite material so it important to decide upon the tests need for each fiber before assigning the fiber for any stage. The natural fibers like coir, banana, almond shell, sugarcane leaves, bagasse, pineapple leaf fibers and hemp can be utilized to manufacture different products like wall panels, PVC pipes, composite boards, bricks, fiber reinforced concrete etc.

Jute fiber reinforced composites are still in the early stages of development, and their practical uses require further investigation. Some studies, however, looked at their viability for certain applications. Gowda et al. [14] used hand lay-up

Table 1 Mechanical properties of natural fibers [12, 13]

Fiber	Density (g/cm^3)	Tensile strength (MPa)	Young's modulus (GPa)	Elongation at break (%)
Cotton	1.5–1.6	287–800	5.5–12.6	7–8
Flax	1.50	345–1100	27.6	2.7–3.2
Jute	1.3–1.45	393–773	13–26.5	1.16–1.5
Hemp	–	690	–	1.6
Vakka	0.810	549	15.8	3.45
Date A	0.990	309	11.3	2.73
Date B	0.960	459	1.9	2.4
Bamboo M	0.910	503	35.9	1.4
Bamboo C	0.890	341	19.7	1.73
Palm	1.030	377	2.7	13.71
Coconut	1.150	500	2.5	20
Banana	1.350	600	17.8	3.36
Sisal	1.450	567	10.4	5.45
Hemp	–	690	–	1.6
Ramie	1.5	400–938	61.4–128	1.2–3.8
*PALF	–	413–1627	34.5–82.5	1.6

* Pineapple leaf fibers

procedures to investigate the mechanical characteristics. Sheng et al. [15] investigated the effect of varying jute fiber content and length on composite attributes such as tensile strength, flexural strength, and impact strength. Hong et al. [16] performed silane treatment of jute fiber to improve the interfacial adhesion with polymer matrix. It resulted in improved tensile and dynamic mechanical properties of the jute/polypropylene composites [17]. Flexural and impact characteristics were improved as a result of the treatments. Jabbar et al. [18, 19] studied the effects of chemical treatment and nanocellulose spray coating on the mechanical properties of nonwoven jute composites. Chemical treatments and nanocellulose coating improved mechanical qualities, according to the findings. The interrelation of moisture recovery, hydrophobic treatment, and mechanical characteristics of jute fiber-reinforced composite materials was investigated by Ameer et al. [20]. When compared to untreated fabric composites, composites with hydrophobic treatment reinforcement demonstrated less moisture recapture and improved tensile and flexural strengths. Karahan et al. studied the fabric structure, yarn physical characteristics, fiber cross-section, and fiber molecular structure parameters [21, 22].

Banana fiber composites have been the subject of extensive investigation in recent years. These studies reveal that banana fiber composites outperform conventional composites in terms of characteristics. Adding banana fiber reinforcement to a structure has been demonstrated in numerous studies to improve a variety of qualities, including stiffness and strength [23]. Because of these characteristics, the materials

are appropriate for a wide range of applications, including construction, automotive, electronics, and packaging. Banana fiber composites have been the subject of a number of researchers [24–26].

4 Applications of NFRC

Natural fiber research has seen a surge of attention in the last 20–25 years, particularly with reference to the replacement of glass fibers in composite material applications (Suddell et al. [27]). When moisture is present at the fiber/matrix interface and at the lamina level, the mechanical characteristics of many natural fiber reinforcement composites are negatively impacted. Natural fiber reinforcement composites have three main applications: automotive, civil engineering, and aerospace. It's been employed for primary as well as secondary structural components in some cases. Window, door, frame, and roof panels in automobiles, are examples of secondary structural applications, while front and rear bumper in automobiles are examples of major structural applications. The performance of NFRC is evaluated in terms of different standard testing, as given in Fig. 1.

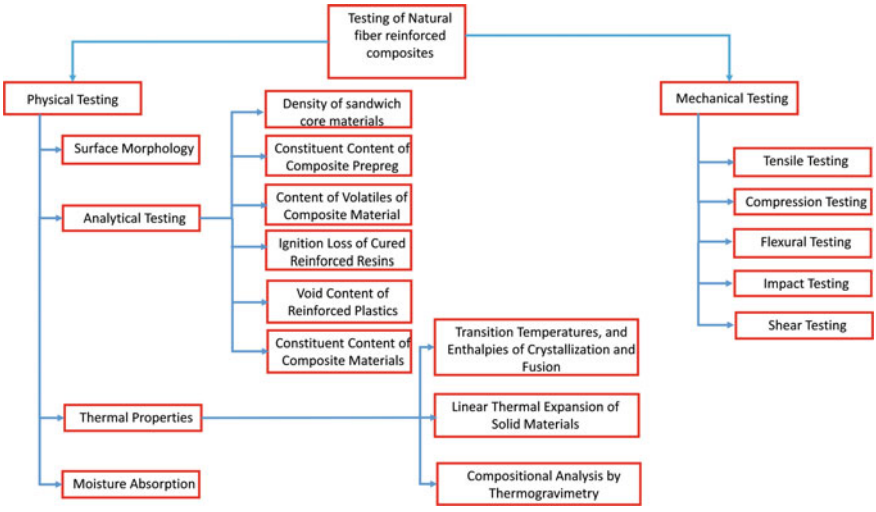

Fig. 1 Classification of testing commonly used for NFRC

5 Physical Testing

The effect of physical phenomena on the performance qualities of a composite material is investigated through physical testing. It uses a predetermined procedure to determine one or more properties of a given material. The shape, density, resin/fiber content, voids, moisture content, water absorption, thermal characteristics, etc. are all part of the physical testing.

5.1 Surface Morphology

The term morphology refers to the study of a part's structure or shape. Surface morphology in composite materials includes high-resolution 3D imaging with powerful microscopes. Details that cannot be seen with the naked eye are examined on the exposed surface of the sample or product. Even at the Nano metric scale, this approach produces extremely fine details of the specimen surface. The scanning electron microscope is the most powerful tool for studying composite material surface morphology.

5.2 Analytical Testing

5.2.1 Density of Sandwich Core Materials

Material density, along with other properties, is one of the fundamental physical properties that can be used to characterize sandwich core materials. The density of sandwich core materials influences the majority of their structural properties (e.g., stiffness and strength). The standard test for determining the density of sandwich core materials is ASTM C271/C271M-16 [28]. This test method entails conditioning a sandwich core specimen in the environment, weighing the specimen, measuring its length, breadth, and thickness, and estimating its density. The cross-section of the test specimen must be square or rectangular. 300 mm $\times$ 300 mm is the minimal specimen size (length by width). The thickness of the specimen is measured with the caliper. For conditioning the specimens to one of the following conditions, an air circulating oven or vacuum drying chamber capable of maintaining a temperature of ± 3 °C is used.

- Standard atmospheric conditions of 23 °C $\pm$ 3 °C and 50% $\pm$ 5% relative humidity
- In oven-dried equilibrium at a temperature of 105 °C $\pm$ 3 °C
- In oven-dried equilibrium at a temperature of 40 °C $\pm$ 3 °C.

After conditioning, the specimens are cooled at room temprature and and readings are noted.

5.2.2 Constituent Content of Composite Prepreg

Prepreg is a fibrous reinforcement that has been impregnated with a polymeric matrix system and is typically in the form of a sheet, tape, or tow. It is used to make composite materials without the use of additional resin by simply laying it in the mould. The ASTM D3529M-10 standard test method is used to determine the Fiber, matrix solids, and matrix content of the composite material prepreg [29]. The amount of matrix solids in a sample is determined by the amount of volatiles in the sample. The matrix is removed from the prepreg specimen of a specified location using the suitable method. One way involves immersing the specimen in a solution that affects the matrix (but not the reinforcement) and dissolves it. Following that, the residual reinforcement is dried and weighed. Acetone, methyl pyrrolidinone, methyl ethyl ketone, dimethylformamide, dichloromethane (methylene chloride), and methyl isobutyl ketone are some of the most often utilized matrix solvents for thermosetting matrices. The specimen is first weighed, then placed in a separate container with at least 100 mL of solvent for each specimen. Specimens must spend at least 3 min in the solvent. After that, the specimen is placed in a fritted glass crucible and dried in a circulating oven at a temperature and time that removes the solvent (at least 100 °C for 5 min). The crucible is then cooled to ambient temperature for at least 5 min while the specimen is kept in a desiccator. The remaining Fiber is next inspected for completeness. The Fibers should be easily separated and there should be no sign of binding between them. The specimen is then placed in a muffle furnace for a specific time and temperature (at which the reinforcement remains unchanged) until the matrix can be completely removed as ash residue. The specimen is exposed to 500 °C in an air environment for up to 6 h. The remainder of the reinforcement is weighed. The minimum specimen size is 80 mm by 80 mm, and the minimum weight is 1 g.

5.2.3 Volatiles in Composite Material Prepreg

When matrix solids content is needed, nearby samples are analyzed for volatiles content using ASTM D3530-97 (Reapproved 2015) [30]. The average mass loss owing to volatiles is removed from the average resin content result, and the result is reported as a percentage of the starting specimen mass as matrix solids content, which is mostly applicable to thermosetting matrices. The samples are placed on a rack before being placed in a preheated oven set to the nominal cure or consolidation temperature. The specimen is positioned so that as much surface area as possible is exposed to the circulating heat. The specimens are removed from the rack and placed in a desiccator after 15 min. Within 1 min of being removed from the desiccator, the specimens are weighed after cooling to room temperature.

5.2.4 Ignition Loss

According to ASTM D2584-11 [31], the test is performed. In a crucible, a 2.5 × 2.5 cm (about 5 g) specimen is inserted. The crucible is heated to 500–600 °C for 10 min while the specimen burns entirely, leaving just carbon and ash remaining. The residual carbon is then converted to ash in a muffle furnace at 565 and 28 °C, cooled in a desiccator, and weighed to determine the residue weight.

5.2.5 Void Content of Reinforced Plastics

The void content of reinforced polymers is determined using ASTM D2734-09 [32]. The density of reinforcement, resin, and composite material are all assessed independently in this procedure. To begin, the resin composition of the composite material is determined, followed by a theoretical composite density calculation. This composite material's theoretical density is compared to its measured density. The void content of the composite material is determined by the difference between the two values. A well-fabricated composite material should have a void content of less than 1%, whereas a badly produced composite should have a substantially larger void content. The density of resin in the composite material is expected to be the same as it is in a bigger mass in this test procedure. The density of bubble-free resin pieces cured under similar temperature and time circumstances as the composite is determined. Manufacturer-supplied density figures are also acceptable.

5.2.6 Constituent Content of Composite Materials

In ASTM D3171-15 [33], there are two test procedures for determining the contents of the composite material constituent. The matrix is physically destroyed by digestion, fire, or carbonization in the first technique, while the reinforcement is left unaltered. The reinforcement is then weighed, and the matrix or reinforcement content (by volume or weight) as well as the percentage of void volume are calculated. Only if the composite and reinforcement densities are known can the volume % be computed. If the density of the matrix is known or computed, an additional void volume computation can be performed. The other test method is for laminated composite materials with known areal density reinforcement. Based on the measured thickness of the laminate, it determines the matrix or reinforcement content (by volume or weight) and cured ply thickness. This approach cannot be used to determine the contents of voids. A caliper is used to measure the length, width, and thickness of the flat composite material.

The test techniques given here are intended for a two-part composite material system and cannot be utilized with a composite material that has a third ingredient, such as filler particles. To apply these test methodologies to filled composite material systems, certain provisions are required (having more than two constituents).

5.3 Thermal Properties

5.3.1 Enthalpies of Crystallization and Fusion

Differential scanning calorimetry is used to determine the enthalpies of crystallization and fusion, as well as transition temperatures of polymers and composites according to ASTM D3418-15 [34]. The test method incorporates a regulated flow rate of purge gas heating or chilling the test material at a specific pace. The test substance is continuously monitored using appropriate sensing equipment. A transition occurs when the test specimen absorbs or releases energy, resulting in an endothermic or exothermic peak. The test material's regions under a fusion endotherm or a crystallization exotherm are compared to those of a well-characterized standard material. Thermal analysis is a rapid method of determining material transitions caused by chemical or morphological changes. When a polymer is heated or cooled across a specific temperature range, these changes occur. Changes in material parameters such as heat flow, temperature, and specific heat capacity are determined throughout these transitions. Differential scanning calorimetry is a valuable method for identifying thermal transitions in polymers, polymeric alloys, and polymeric additives. This technique is useful for determining the chemical reactions that affect/cause specific transitions (oxidation, thermoset resin curing, heat degradation, and so on).

5.3.2 Thermal Expansion Coefficient

The standard test technique for determining linear thermal expansion using a thermomechanical analyzer is ASTM E831-14 [35]. The specimen is heated at a steady rate in order to complete this test. The electronic recording of the change in specimen length as a function of temperature is then used to calculate the coefficient of linear thermal expansion. The initial length of the specimen is measured in the direction of expansion to 25 μm at 20–25 °C and documented. After that, the specimen is placed in a specimen holder beneath the probe, with a temperature sensor in contact with the specimen. The furnace is adjusted to enclose the specimen holder, and the sensing probe is given a suitable contact load to ensure that it is in touch with the specimen. The specimen is subsequently heated to the required temperature at a consistent rate of 5 °C/min, and the length change with temperature is recorded. Meanwhile, without the specimen, the length of the specimen holder is measured and rectified, especially for low expansion specimens.

5.3.3 Compositional Analysis by Thermogravimetry

A general method for determining the amount of medium and highly volatile matter, flammable material, and ash content in compounds [36] is described in ASTM E1131-08 (2014). The thermogravimetric technique is used in this process. This method is

useful for determining the composite material's composition. The 10–30 mg specimen is carefully inserted in the specimen container, and the mass is recorded at the start. The specimen is heated to the required temperature range, and the mass change is recorded continuously throughout the temperature interval. The weight loss is measured in milligrams or as a percentage of the original specimen weight. The whole thing happens in a vacuum (nitrogen). The atmosphere is changed from inert to reactive after mass loss is established in the range 600–950 °C (air or oxygen). Following the introduction of the reactive gas, the analysis is complete when a mass loss plateau is reached. The mass loss between the beginning temperature and temperature X represents the highly volatile stuff.

5.4 Moisture Absorption

The standard test technique for water absorption of core materials for sandwich composites is ASTM C272/C272M-12 [37]. A square or rectangular cross-section with dimensions of 75 × 75 × 13 mm (length, breadth, and thickness) was tested. Depending on the material's water absorption behavior, the specimen is oven dried at 50 °C ± 3 °C for 24 h or at 105 °C ± 3 °C for 2 h. The specimen is then exposed to one of the following conditions:

- Twenty-four-hour immersion: The specimens are immersed in a container horizontally for 24 + 1/0 h at a temperature of 23 °C ± 3 °C under a 25 mm head of water. After that, the specimens are removed and all surface water is wiped away with a dry cloth until there is no visible water left, and their weight is recorded.
- Increased temperature humidity: For 30 days, the usual conditioning environment is 70 °C ± 3 °C and 85% relative humidity. The specimen is dried using a dry cloth before being weighed.
- Maximum percentage mass gain: The specimen is immersed in a container horizontally for 48 + 1/–0 h at a temperature of 23 °C ± 3 °C under a 25 mm head of water. After drying, the weight is tallied. The specimens are then returned to the water and the process is repeated until the mass increase after the last 48 h is less than 2% of the total mass gain across all intervals.

Another test method for determining moisture absorption properties of polymeric composite materials and their moisture equilibrium conditioning is ASTM D5229/D5229M-14 [38]. If the change in moisture content of a specimen is less than 0.02% across two consecutive measurements, the material is considered to be in a state of moisture equilibrium. Before concluding that the specimen has attained wetness equilibrium, it is a good idea to plot and evaluate the mass change vs time curve. To reduce the impact of the edges, this approach requires a length to thickness ratio of 100:1. However, different length-to-thickness proportions are used, such as 50:1 (for 1 mm thick), 25:1 (for 2 mm thick), and 12.5:1 (for 4 mm thick) proportions. Existing composite specimens have a thickness of 5 mm and a length of 100 mm, resulting in a length-to-thickness ratio of 20:1. (5 mm thickness). The specimen

should weigh at least 5 g and its thickness should not vary by more than 5% across its surface. Following cutting and cleaning, the specimens' surfaces must be cleaned with methanol to remove any machining-related filth and oil, and then dried in a stove at 60 °C. During the drying process, the masses of the composite instances are monitored on a regular basis until no more weight change occurs. The initial weights of the specimens are recorded once they have dried, and they are quickly placed in a standard environmental system. The atmosphere is set at 20 °C ± 2 °C and 65% relative stickiness. A mechanical logical parity with a precision of 0.1 mg is used to determine the mass adjustment. The weight of the specimen is measured at various intervals until equilibrium is achieved or a specific behavior is defined. The weight rise vs the square root of time is then plotted on a graph.

6 Mechanical Characterization

6.1 Tensile Testing

In plane tensile characteristics of polymer composites are determined using the ASTM D3039/3039 M standard test procedure [39]. A test specimen is made out of a rectangular thin strip of composite material with a homogeneous rectangular cross-sectional area. This strip is clamped in a tensile testing machine's jaws, and a predetermined load is applied. At regular intervals of time, the force and elongation values are determined, and a curve is drawn. The composite material's tensile strength is responsible for the maximum tensile load that the specimen can bear before mechanical failure. Meanwhile, the stress–strain curve plotted during the test allows us to acquire additional information such as the composite material's ultimate tensile strain, transition strain, tensile modulus, and Poisson's ratio. Each sample is examined five times, with the average value and standard deviation recorded. As indicated in Fig. 2, the specimen dimensions (thickness, width, and length) are chosen to encourage gauge section failure. A statistical representation of the bulk material is required for the test specimen. This can be ensured by selecting a specimen width that has a sufficient number of yarns/fibers in its cross section. Without tabs, materials including 2D laminates, fabric-based composites, and randomly reinforced materials are effectively tested. Tabs, on the other hand, are.

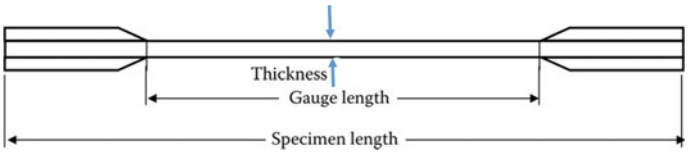

Fig. 2 Test specimen cross-section

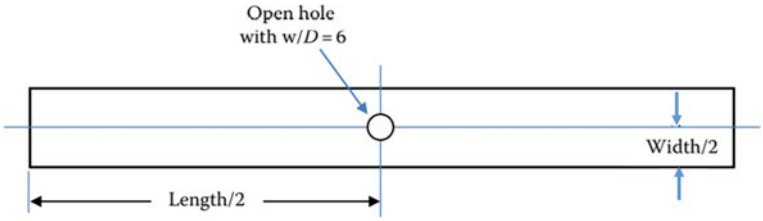

Fig. 3 The open-hole test specimen is seen in cross-section

Strongly advised for testing unidirectional materials in order to trigger Fiber direction failure. Prior to the test, three locations in the gauge section define the specimen area (as A = w * h, in mm^2) after milling and conditioning. The strain rate is set to generate failure within 1–10 min of the test, and the head displacement rate is set to 2 mm/min in constant head speed tests. The specimen's ultimate tensile strength, F_{ut} (MPa), is derived by dividing the highest force, Pmax (N), upon failure by the beginning area, A (mm^2).

When composite material is reinforced with high modulus fibers, open-hole tensile strength is determined using ASTM D5766/D5766M-11 [40]. A centrally positioned hole is used to perform a uniaxial tensile test on the laminate in line with ASTM D3039. Only failure modes that pass through the hole in the test specimen are considered. The proportion of specimen width to hole diameter (w/D) has a significant impact on test outcomes. The current ratio is 6. The ratio of hole diameter to thickness (D/h) has an impact on the results. A ratio of 1.5–3 is ideal. Figure 3 depicts the specimen's layout. All calculations follow ASTM D 3039 guidelines. The specimen's area is the gross cross-sectional area, ignoring the hole, when doing the computations.

6.2 Compression Testing

The in-plane compressive characteristics of polymer composites can be determined using a variety of test methods. The type of load transfer, that is, loading or shearing, differs between these standards. In test methods ASTM D695 [41], the compressive force is transmitted into the specimen by end-loading. In case of ASTM D3410M [42], the load is applied by shear at the wedge grip contact, while in ASTM D6641M [43], a combination of shear and end-loading is used for load transfer. The specimen for compression strength testing should be in the shape of a right cylinder or a prism, with its length twice its major width or diameter, according to ASTM D695. The recommended cylindrical specimen has a diameter of 12.7 mm and a height of 25.4 mm, whereas the preferred prism specimen has 12.7 × 12.7 × 25.4 mm. The testing speed should be 1.3 ± 0.3 mm. Alternative specimen dimensions may be used when it is difficult to get the standard specimen dimensions due to the material

limitations. A compress meter or equivalent device is used for materials that are less than 3.2 mm thick. The specimen is supported by the supporting jigs during the testing.

6.3 Flexural Testing

A three-point bending test (ASTM D7264) [44] or a four-point bending test (ASTM D6272) [45] are used to determine the flexural properties. Depending on the test type, a rectangular bar rests on two supports and is loaded at one or two spots (through loading noses) at an equal distance from the adjacent support point. As indicated on Fig. 4, the distance between the loading noses (the load span) is one-third or one-half of the support span. The composite material's span-to-thickness ratio affects the specimen dimensions. The recommended width of the specimen is 13 mm, Thickness 4 mm and Span to thickness ratio is 32:1.

The ratio of span to thickness is 32:1. The support span is the length of the specimen computed using the span-to-thickness ratio; the entire specimen length is approximately 20% longer than the support span (Fig. 5). While employing the usual span-to-thickness ratio, the standard allows for any alternative specimen thickness (32:1). There are other possible span-to-thickness ratios of 16:1, 20:1, 40:1, and 60:1 that must be stated in the report. A crosshead rate of 1.0 mm/min is used to set the test speed. The maximum applied force and deflection during the flexural strength test are given. As a sample of the composite behavior, the load–deflection curve is also derived. Using these data, the flexural strength, maximum strain, and flexural modulus of elasticity are calculated.

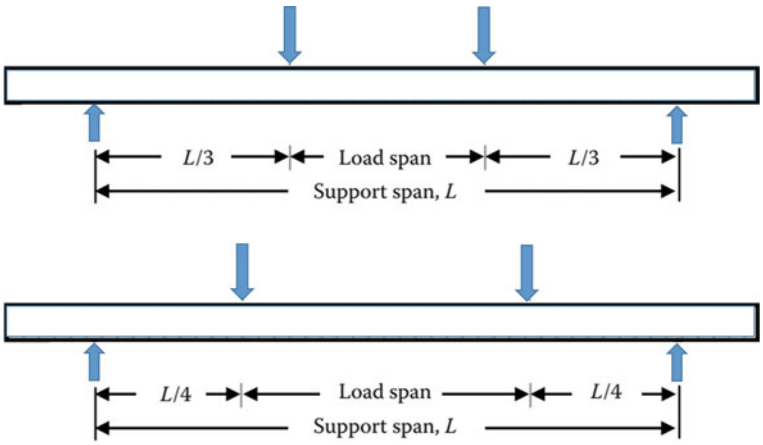

Fig. 4 One-third of support span, and one-half of support span

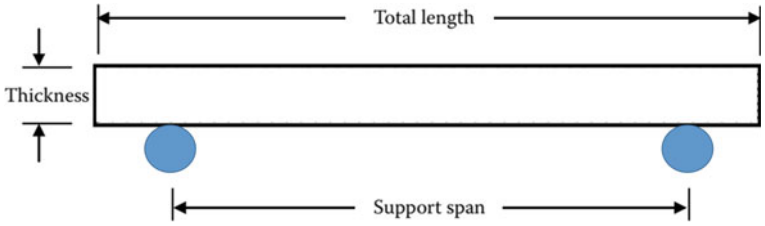

Fig. 5 Sample specifications for flexural testing

6.4 Impact Testing

The out of plane impact resistance of NFRC is determined according to ASTM D7136/D7136M-15 [46] standard using a drop weight impact tester. A hemispherical impactor is connected to the dead weight and creates a concentrated impact on flat composite plate when allowed to fall freely. The mass and drop height of the impactor determine the potential energy of the drop-weight, which is determined before the test. Damage resistance is measured by the size and type of damage that results in the specimen. The impact response is affected by a number of parameters, including laminate and ply thickness, stacking sequence, impactor shape (geometry, tip, etc.) and mass, impact velocity, and impact energy. Before testing, the specimen's width and length are measured twice, and its thickness is measured four times near the impact point. The impactor is dropped toward the specimen from the calculated height. During contact, force versus time data is captured continuously or at predetermined intervals.

EN-ISO 179 (Charpy impact test) and EN-ISO 180 (Izod impact test) [47, 48] are the other two procedures for determining impact performance. Both these approaches use a pendulum impact tester foe testing. The test specimen is supported near its ends as a horizontal beam in Charpy impact testing and is impacted by a single striker blow. While in case in Izod test, the specimen is positioned vertical, and its lower end is clamped in the fixtures. The upper end of specimen is free, and pendulum strikes the free end.

6.5 Shear Testing

The tensile test utilizing a 45° laminate in the x direction according to ASTM D3518/D3518M-13 is used to determine the in-plane shear properties of NFRC [49]. Except for the stacking order of the laminate, the test procedure is the same as ASTM D3039. A force of 1–1000 g is applied. 10 mm sphere, for example following the removal of the load, measure the size of the indent. A notched specimen is compressed. The shear between two centrally positioned notches drilled halfway

Table 2 Shear properties of jute and hemp fiber composites

Specimen	Hemp fiber composite		Jute fiber composite	
	Deflection (mm)	Shear stress (MPa)	Deflection (mm)	Shear stress (MPa)
1	1.69	28.31	1.98	25.41
2	1.65	29.90	2.13	23.96
3	1.77	28.94	1.67	19.39
4	1.41	27.36	1.83	22.31
5	1.67	24.40	2.11	18.43

through the thickness of the specimen and at a predetermined distance apart on opposing faces causes the specimen to fail. The specimen's maximum load is kept track of. The length of the unsuccessful (sheared) area is determined. The specimen's in-plane shear strength is calculated by multiplying the highest shear load by the product of the specimen's breadth and length of the failed area.

Another testing standard ASTM D5379M [50] is also used to test in plane shear response of NFRC. In this method a rectangular beam that receives a shear force at the v-notch from a unique fixture. By using an alignment that refers to the fixture, the sample is introduced into the fixture with the notch located along the line of action of loading. While the load is being observed, the testing apparatus squeezed the fixture's two halves. The test, was carried up until the tested item fractured. The shear strength is defined as the shear stress borne by a material at failure under a pure shear situation per the reference standard ASTM D 5379 M [50]. The shear properties of hemp and jute fiber composites are given in Table 2.

7 Conclusions

Natural fiber reinforced composites (NFRC) have been extensively studied by the researchers and engineers for their potential application in a variety of industries due to their long-term sustainability. They also have a large carbon footprint decrease. Fiber length, fiber content, and chemical treatment are the key elements that determine the mechanical behavior of NFRCs natural composites. Banana fibers have a number of advantages as a reinforcing material, including being environmentally benign, having a low density, and being readily available. NFRC may find application in low-end applications as well as sophisticated applications including industrial sectors such as automotive and aerospace.

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Natural Fiber Metal Laminate and Its Joining

Muzzamal Hussain and Abdellatif Imad

Abstract The fiber metal laminates (FML) combine the properties of metal and composites. The new material shows better mechanical properties as compared to constituents. As the two different types of materials are joined together so the joining of metal and composites become very crucial. The quality of the adhesive bond determines the final mechanical properties of fiber metal laminate. Most of the time aluminum is used as a metal part. Different types of surface treatments have effect on the mechanical properties and different techniques are used to evaluate the quality of different surface treatments. This chapter covers all the details related to natural fiber metal laminates including fibers, metals, surface treatment of metals, fabrication techniques, methods to evaluate metal-composite bond, and mechanical characterization of natural fiber metal laminates. The reinforcement material for the composite is synthetic fiber but the natural fibers-based fiber metal laminates are also gaining popularity due to environmental issues. This chapter presents the detailed literature on different natural fibers used for FMLs fabrication. The FMLs made with both thermoplastic and thermoset matrix along with manufacturing techniques are also presented in this chapter. This chapter also presents a comprehensive literature review of the static and dynamic properties of natural fiber reinforced thermosets and thermoplastic fiber metal laminates.

Keywords Fiber metal laminate · Joining · Natural fiber · Mechanical properties

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1 Introduction

Fiber Metal Laminates (FML) are multilayered materials in which a composite layer is sandwiched between metal layers. The resultant material has properties of both constituents. Due to this coupled effect, these materials are in high demand in a variety of applications including aerospace [1–3], blast protection [4], automotive industry, and fire barriers [5]. The Natural Fiber Metal Laminates (NFML) can be used in automobile applications due to their low mass, high strength, high impact, and fatigue strength [6, 7].

The properties of FMLs are linked with the type of constituents used to make them, the main raw materials used for the FML fabrication are:

1. Reinforcement
2. Matrix
3. Metal.

The reinforcement can be synthetic, natural, or hybrid. The synthetic reinforcement includes aramid, carbon, and glass [8, 9]. "Natural reinforcements include jute, kenaf, Empty Fruit Bunches, banana, coir, sisal, bamboo, flax, basalt and hemp fibers" [10–16]. The hybrid reinforcement is made with a combination of different natural and synthetic fibers to overcome the problems associated with one of the constituents [17]. The synthetic fiber-based FMLs are used abundantly in different applications while natural and hybrid reinforcements are investigated to find its use in different applications.

Natural fibers are used in the composites industry due to more and more pressure to use materials from renewable resources. Synthetic fibers are hazardous to the environment and non-recyclable most of the time.

Natural fibers due to sustainability, renewability, low density, high specific mechanical properties, and biodegradability have become the material of choice [18]. They have gained popularity in recent years to replace synthetic fibers in different applications e.g., automobile interior and non-load bearing parts, sports equipment, Electrical and Electronics (E&E), and civil engineering [17]. The composite part of the FMLs can be replaced with the Natural Fiber Reinforced Composites (NFRCs), it can increase its area of applications other than aerospace applications e.g., automobile, transport, and cargo, etc.

The metal part is most of the time aluminum, and most popular grades of aluminum being used to fabricate FMLs are 2000 (2024-T3), 6000 (6061-T6), and 7000 (7075-T6,7475-T6). Aluminum is the preferred alloy due to its low cost, easy handling, and ease of fabrication [19]. Table 1 shows the various kind of alloys which are used to fabricate the FMLs other than aluminum.

Both thermoplastic and thermoset matrix are used for the FMLs fabrication however epoxy is used mostly due to its easy processability and high mechanical properties [2, 23]. Epoxy is also used in commercially available FMLs e.g., Aramid fiber Reinforced Aluminum Laminate (ARALL), Carbon Reinforced Aluminum Laminate (CARAL), and Glass fiber Reinforced Aluminum Laminate (GLARE).

Table 1 Properties of different metals used for the FMLs manufacturing [19–22]

Metal type	Preferred properties
Aluminum	Low density and easy processing
Magnesium	Low density, high corrosion resistance, and better electromagnetic shielding (EMI)
Titanium	Low ductility and strength
Steel	High density and seldom used

Despite the advantages of thermoset matrix, the growing urge to use recycled materials has paved way to use the thermoplastic matrix [24]. Thermoplastic FMLs are tough, and easy to process due to the direct manufacturing route with the advantage of recyclability [23].

Combining distinct materials required special preparations. To achieve the optimum properties of FMLs the surface of the metal is prepared to increase surface roughness and surface free energy. Similarly, different types of matrices also required different manufacturing techniques. The type of application also derives the use of manufacturing techniques, e.g., the autoclave is used to achieve highest mechanical properties. FMLs can be classified on the bases of constituent materials, lay-up configurations, and direction of lay-up. FMLs categorization is shown in Fig. 1. FMLs have high impact properties as compared to both the constituents. They have easy repairability like metal, high fatigue strength and crack arrest mechanism [23]. Composite layer act as a barrier layer and render the crack propagation of FMLs [25].

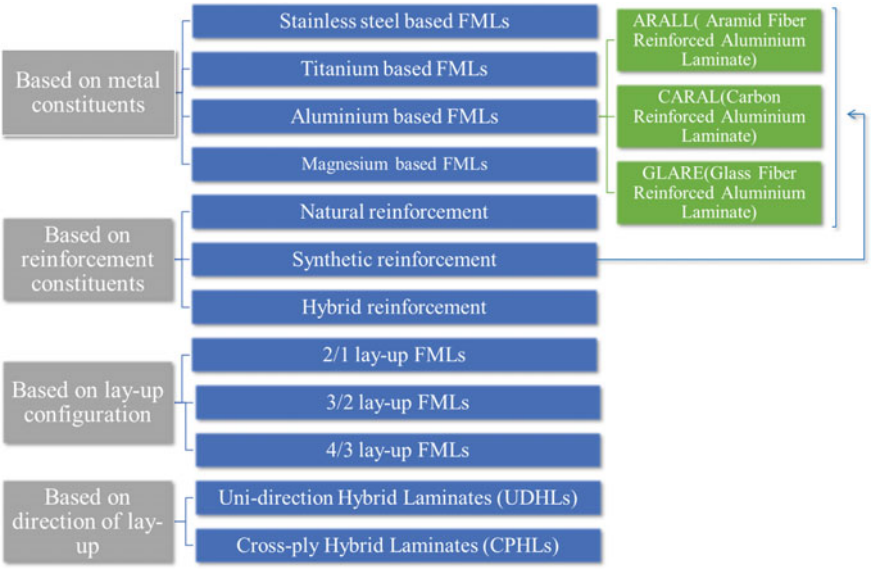

Fig. 1 Classification of fiber metal laminates [8, 26]

2 Fabrication of Fiber Metal Laminates

In this part the FMLs fabrication procedure will be discussed in detail. The manufacturing process include surface preparation of metal part which is most of the time aluminum and plays an important part in the final properties of NFML. Then different type of reinforcement and matrix will be discussed which are used to make the NFMLs and at the end the fabrication processes will be discussed which are used to consolidate all the component to make the final part. The fabrication processes depend upon the type of matrix being used and intended applications.

2.1 Preparation of Metal Surface

The surface preparation of metal is very important step in the FMLs fabrication as the adhesion of metal and composite part depend on this. The surface preparation is done to increase the roughness of surface at micro or meso level depending upon which type of technique being used. Furthermore, the type of metal all decides that which type of surface treatment is suitable for fabrication of FMLs. Table 2 shows the commonly used surface treatments to prepare the surface of aluminum and other metals.

Before any subsequent process the metal is first degreased to remove the dust, dirt and foreign impurities to increase its ability to bond. Degreasing is not sufficient to

Table 2 Different surface preparation techniques used for aluminum [27]

Mechanical treatment	Grit blasting Abrasion with sandpaper Excimer laser texturing
Chemical treatment	Alkaline etch Forest product laboratory Chromic-sulfuric acid etch Sulfo-ferric acid etch
Anodizing AC—Alternating Current DC—Direct Current	Phosphoric Acid Anodizing (PAA) (AC&DC) Sulphuric Acid Anodizing (SAA) (AC&DC) Boric-Sulphuric Acid Anodizing (BSAA) Chromic Acid Anodizing (CAA) (DC)
Coupling agent	Sol–Gel Silane
Dry surface treatment	Plasma-sprayed coating Ion Beam Enhanced Deposition (IBED) Excimer laser texturing

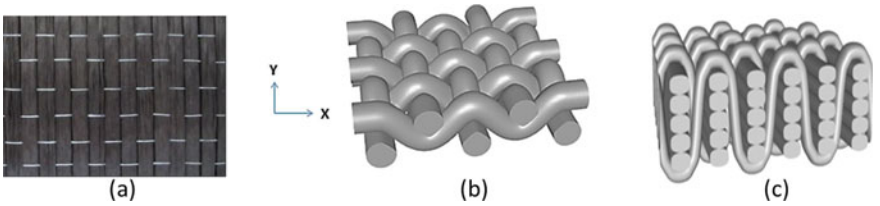

Fig. 2 Different type of reinforcements used for the FMLs fabrication **a** Uni-Directional (UD), **b** two-dimensional (2D) and **c** three-Dimensional (3D)

make the metal acceptable for FML fabrication, further surface preparation processes are adopted to make the metal ready to fabricate FMLs. The degreasing is either done with chlorinated or non-chlorinated solvents [26].

After degreasing the metal surface is ready for subsequent processes. It is done through removal of unfixed oxide layer by mechanical means with grit blasting/sandpaper. After the mechanical preparation the metal is further process to enhance its roughness, it is done through different chemical, electrochemical, coupling or dry surface treatment. The most used and effective method of surface treatments are electrochemical process e.g., CAA and PAA. These processes create the porous oxide layer which leads to the best bonding. The PPA is more preferred due to porous oxide layer of PAA as compared to CAA which produce barrier oxide layer. The porous-oxide layer results in better interfacial bond [28].

3 Reinforcements Used for NFMLs

The reinforcement of different type is used for the NFMLs. The direction of reinforcement determines the final properties of FMLs. The reinforcement can either be woven or non-woven. The woven reinforcement is further characterized as Uni-Directional (UD), Two-Dimensional (2D) and three-Dimensional (3D) as shown in Fig. 2 [29–31].

4 Matrices Used for NFMLS

Thermoset matrix is most widely used resin system for FML fabrication, especially epoxy. As epoxy is easy to process and have good mechanical properties. Epoxy is used in all available current commercial solutions i.e. ARALL, CARAL and GLARE [2, 23]. However, there are certain drawbacks associated with the thermoset matrices, e.g., brittle, non-recyclable, high curing temperature and pressure (which cause thermal residual stresses when used for FMLs fabrication). In spite of good mechanical properties and stability, the thermoset matrices are considered

environmental concern due to non-recyclability and end of life disposal [19, 24]. Thermoplastic matrices are now being used by many researchers as a replacement of thermoset matrices for NFML. Thermoplastic matrix has distinct advantages over thermoset matrices e.g. short processing cycle, recyclability and high toughness [23]. Polypropylene (PP) is most reported matrix system for NFML due to its low processing temperature. NFML has most of the time limitations as they start losing its mechanical performance at high temperature [32–34].

5 Fabrication Process of FMLs

The fabrication of FMLs as discussed earlier depends mainly on the intended applications. If the mechanical properties of highest level are required than the autoclave manufacturing process is used, while other fabrication processes such as Vacuum Infusion and compression hot press are also investigated. The details of different processes adopted for FMLs fabrication are given below.

5.1 Autoclave Process

The autoclave manufacturing process is most effective and oldest process used for the fabrication of FMLs. The parts with the best quality are achieved with the autoclave process. Dry prepreg and metal sheets are used for the manufacturing of FMLs (Fig. 3).

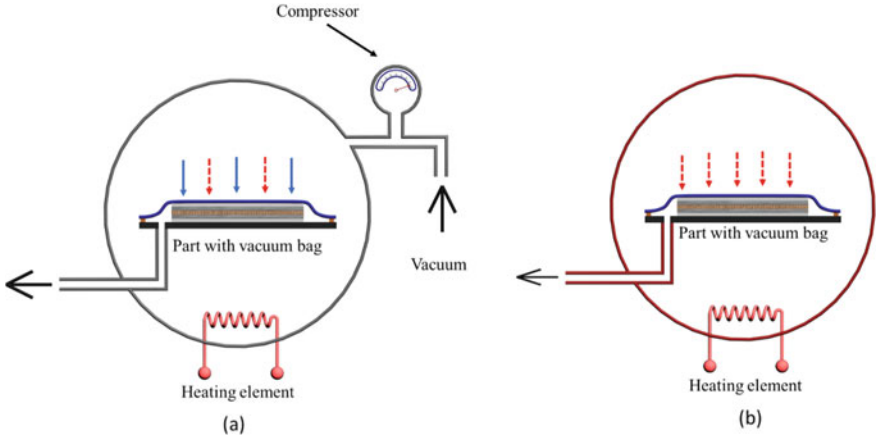

Fig. 3 Schematic of FML consolidation process in the autoclave **a** autoclave consolidation process (Red dashed arrow-heat and Blue solid arrow-consolidation pressure) and **b** oven post-curing (Red dashed arrow-heat) [37]

Following four steps are performed to fabricate FMLs through autoclave [5, 35, 36].

1. Tool and material preparation e.g., aluminum surface treatment
2. Placement and arrangement of material for the consolidation which include cutting of metal sheet, lay-up, reinforcement cutting etc.
3. Consolidation in autoclave to cure the FMLs
4. Final testing and inspection of cured part.

The autoclave process has issue when FMLs are made, it causes high internal stresses due to mismatch of composite and metal part. The issue of high internal stresses is resolved, and different methods are employed to overcome this issue. A number of researchers have worked to resolve this issue. In one such study Khan et al. [38] reported a method in which part is stretched-up after autoclave processes during post curing. The part is stretched up to plastic region to remove the problem of high internal residual stresses. Other researchers are also proposed different solution though not very common. Xue et al. [39] used the clamps to control the internal thermal residual stresses. Park et al. [37] compared the physical and thermal properties of FMLs for three different manufacturing processes a. autoclaves, b. oven and c. consolidated autoclave. They reported that consolidated autoclave and oven post-cured FMLs shown the less voids, higher fiber-fraction and even thickness of plies.

Autoclave is preferred process for the high quality and uniform quality parts but the issue with this process is that it is lengthier and expensive process further there is limitation of part size. Distortion and high internal residual stresses are also caused which sometimes compromise the quality of FMLs. Due to these issues the researchers have tried to explore other manufacturing methods for the FMLs fabrication including Vacuum Assisted Resin Transfer Moulding (VARTM) and compression hot press [40, 41].

5.2 Vacuum Infusion Process for FMLs Fabrication

"National Aeronautics and Space Administration (NASA), Langley Research Center LaRC developed the FMLs with VARTM process, which is low cost and faster with comparable properties with FMLs prepared with autoclave process". NASA also claimed that they used the FMLs manufactured by the VARTM in aircraft and aerospace structure [5]. The VARTM process used for FMLs fabrication is modified version of VARTM process used for the composite fabrication since the FMLs have metal and composite part involved in manufacturing so made certain perforation in the metal layers to provide passage to the resin flow through the thickness (Fig. 4).

Since the perforation pathways are made to assist the resin flow the method of drilling also effect the properties of final composite part [42].

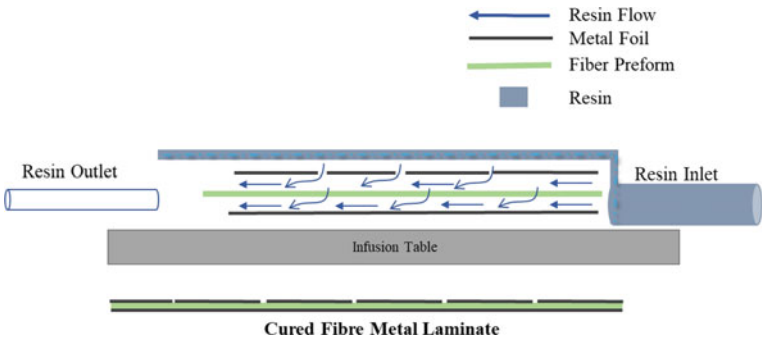

Fig. 4 Illustration of VARTM process used for FMLs fabrication with pathways [40]

A lot of researchers later on investigated the effect of perforations and resin flow in different directions. The resin flow in length wise and through the thickness direction. Since the flow in the through the the thickness direction is slow so the perforations assist to make the resin distribution even.

The VARTM process are quite effective and the generally can be employed to replace the FMLs made with the autoclave process [42–44]. Even though the FMLs made with the VARTM has comparable properties with the one prepared with the autoclave, the small perforations serve as potential crack initiation points, which need to be addressed [45, 46].

When the part size increases, the localized variations also start to increase e.g., thickness variations. Hergan et al. [47] developed the FMLs with vacuum infusion and used a rigid mold to control the issue of thickness variation. To assist the resin flow through rigid mold a high injection pressure was used, as shown in Fig. 5.

Most of the researchers reported the use of VARTM process for the synthetic fibers and the dynamics of synthetic fibers are different than the natural fibers, since natural fiber has an inherent porosity which lies both at fiber and fabric level. Hussain

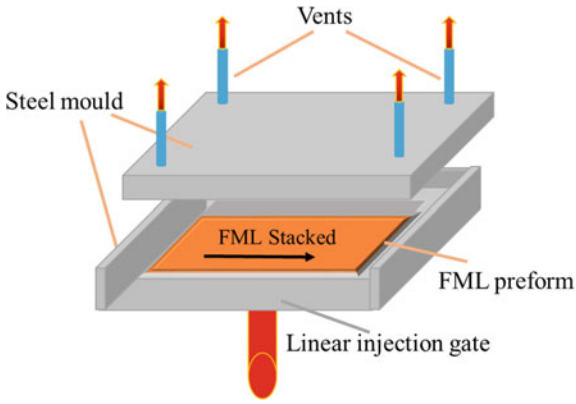

Fig. 5 Modified VARTM to control the thickness of sample during FMLs fabrication [47]

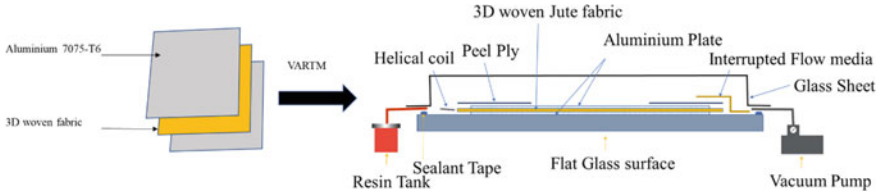

Fig. 6 NFML fabrication using vacuum infusion process [31]

et al. [31] developed the NFML with the vacuum infusion process as shown in Fig. 6, they used the 3D woven reinforcement made with the jute fibers. They didn't use the perforations as used by some previous researchers. The advantage with the 3D woven natural fiber reinforcement is that it is single structure and natural fibers inherently provide resin flow pathways due to uneven surface and porosity.

Once of evident advantage of VARTM process is that the process is short which make it less expensive. Overall the production time for VA(RTM) 35–40% less than autoclave and oven cure, while cure cycle required 90% less time which make it cost effective.

5.3 Compression Hot Press Molding

Thermoplastic matrix based FMLs are mostly developed using the compression hot press, this process is relatively short and direct process since the consolidation take place in one single step [23]. The two type of thermoplastic FMLs have been reported in literature: 1-Conventional thermoplastic FMLs which have thermoplastic matrix, 2-Self reinforced thermoplastic FMLs in which both the both constituents are of same material [33, 34]. Most of the work reported in the literature is related to synthetic fiber based thermoplastic FMLs [48, 49] but the natural fiber FMLs are also reported by some researchers [46]. Hussain et al. [30] developed the FMLs using compression hot press and used both the thermoset and thermoplastic matrix. Figure 7 shows the experimental set-up used by Hussain et al.

There are issues of variation in the FMLs developed using compression hot press most commonly are e.g., part thickness and plies orientation during fabrication. The problem of thickness variation is pronounced as the size of part of is increased. This issue can be addressed using the frame of pre-determined thickness and sample is placed inside it before applying pressure.

Fig. 7 FMLs fabrication process **a** stacking sequence of constituent material **b** constituents placed in the machine for the fabrication **c** compression hot press [46]

6 Mechanical Properties

The FMLs are made with metal and composite which are joined together with the some adhesive. There is cohesive layer between metal and composite, the properties and integrity of the FMLs are heavily determined by the characteristics of that cohesive layer. The characteristics of the adhesive joining the metal and composite are accessed through different methods e.g., mechanical and physical.

The mechanical characterization of adhesive layer is done through different methods. The performance of adhesive joints is determined through different tests, these tests are characterized in four different ways according to nature of tests: 1-Peel tests, 2-Shear test, 3-Tensile and cleavage tests and 4-Fracture tests. The details of tests are shown in Fig. 8. All these tests are done to check the different characteristics under different conditions. Although these tests are conducted for adhesive bond but these are also used to check the delamination characteristics of FMLs as there is involved adhesive bonding [50]. Figure 8 shows the different type of tests conducted to check the delamination characteristics of adhesive bond [51, 52]

The mechanical characterization of FMLs done through different static and dynamic tests. These tests mostly reported in the literature. These tests are performed as per different standards depending upon the final requirement. The tensile test is

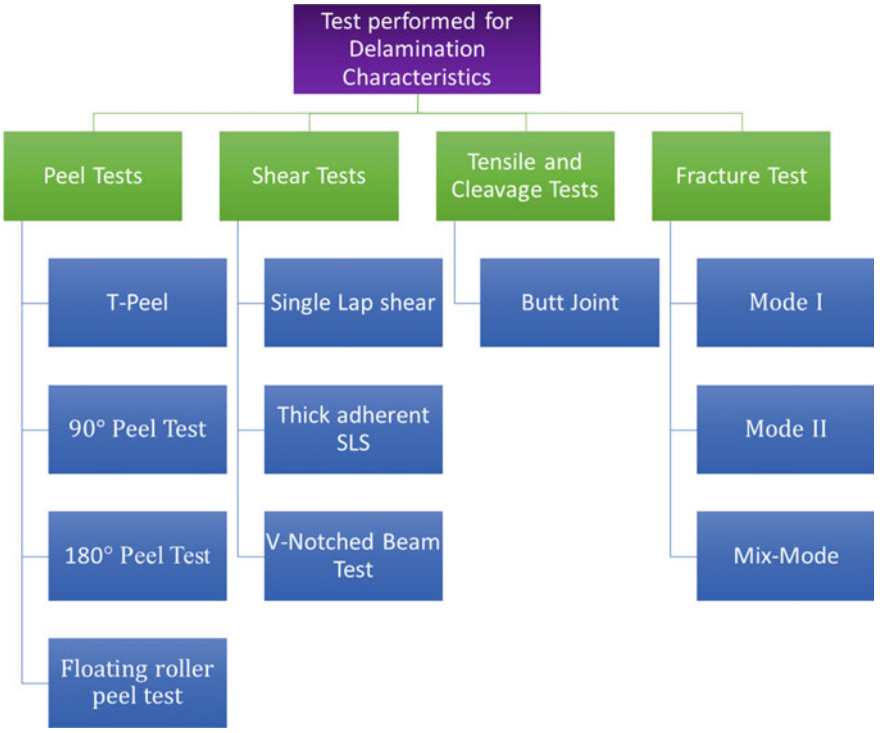

Fig. 8 Different test performed to check the quality of the adhesive bonding layer [51, 52]

performed either with dog bone or rectangular shape specimen. Flexural testing is also done with different type of standards and every standard has its own requirements. The Flexural testing for FMLs is performed in three ways: (a) Compression loading, (b) Three and four-point bending, and (c) Short beam shear.

Impact loading is one of key application area of FMLs beside fatigue loading. Impact properties are accessed by three different methods, low-velocity impact (less than 10 m/s), high-velocity impact (10–200 m/s), and blast loading (more than 200 m/s [26, 53]. Figure 9 show the different mechanical testing done for FMLs.

6.1 Characterization of Metal-Composite Bond

As already discussed, that the metal and composite are joined together with the help of an adhesive, this adhesive make an interface with both the metal and composites. These two interfaces are marked as adhesive zone 1 and 2 in the Fig. 10. Then there is cohesive zone which is purely matrix and composite (resin rich fiber layer).

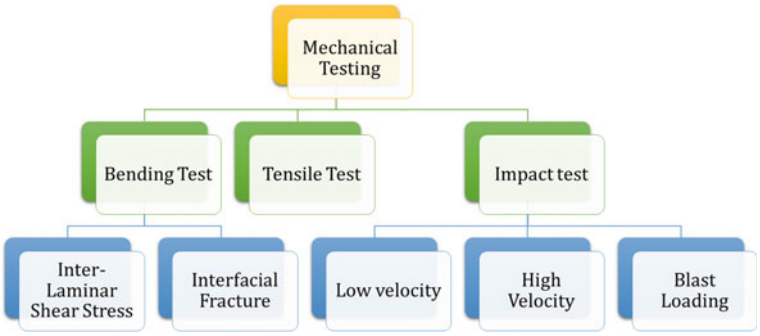

Fig. 9 Flow chart showing different tests conducted for mechanical characterization of FMLs

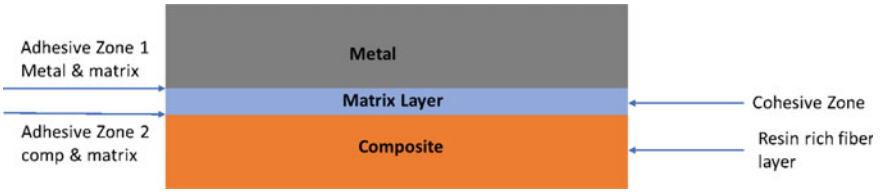

Fig. 10 Type of interface and component of FMLs

Based on these components and interfaces, the failure occurs in four different ways in FMLs, (a) adhesive failure, (b) cohesive failure, (c) intra-laminar failure and (d) mix mode failure. The adhesive failure happens at the interface point either on metal side or composite side, it indicates the poor quality of bonding. The cohesive failure occurs within cohesive zone; it normally shows that the interface points have good bonding. The intra-laminar failure is in the composite laminate; Some of fibers in top laminates in composites break instead of failure at interface point. It is also indicative of good adhesion. The mix-mode failure is when both the both adhesive and cohesive failure occurs together [54].

Different researchers have investigated the effect of different factors on the delamination characteristics of metal composite bonding. Since the metal surface treatment is an important component which contribute to the properties of FMLs. The different type of metal surface treatments has different effect. Agha Mohammadi et al. [14] investigated the effect of four different type of treatments, a-Mechanical, b-Alkaline Etching, c-Forest Products Laboratory Etching (FPL-Etching) and SAA on the surface of aluminum. FMLs were made with all four types of treated aluminum and basalt was used as a reinforcement. The flexural test was conducted to compare the properties of all four types of FMLs. The results showed poor that mechanical and alkaline etching had poor interfacial bonding with adhesive failure. Different metal surface treatments have different level of adhesion and mechanical performance. Wu et al. [55] compared the different surface treatments to optimize them, they investigated Inter-Laminar Shear Strength (ILSS) properties of developed FMLs. The used

different surface treatments including solvent degreasing, plasma treatment, mechanical abrasion with sandpaper and alkaline degreasing. Later on, after optimization they also used best above surface treatments in combination also to check its cumulative effect. It was concluded that alkaline cleaning with 10% NaOH solution, 180 grit sandpaper give the best results.

Abrading metal surface with the sand has different effect on the ILSS, when abraded with sandpaper of lower grit the higher ILSS is achieved. The reason of high ILSS is that higher roughness is achieved with the sandpaper of lower grit as compared to higher grit. The alkaline cleaning with NaOH produce an interface layer, this interface layer result in the higher ILSS up to 10% concentration, beyond that the aluminum plate thickness starts to decrease and increase thickness of interface layer to an extent which compromise the mechanical performance.

The Lee Hamill and Steven Nutt [56] compared the abrasion, silane surface treatment and Phosphorous Acid Anodizing (PAA). He concluded that PAA is the most effective surface treatment for aluminum. Khan et al. [57] evaluated the interlaminar bond strength using T-peel test. The result showed that PAA has the excellent bonding characteristics as shown in Fig. 11.

Although thermoset matrix (epoxy) is the most used matrix for the FML fabrication, but these days researchers are also extensively using thermoplastic matrix for the FML fabrication. Some researchers have also evaluated the metal-composite bond made with thermoplastic matrix. Cache et al. [58] evaluated the thermoplastic matrix based metal-composite and metal–metal bond. He used Single-Lap-Joint (SLJ) test to characterize the bond performance. The results presented optimum properties of bonding for both type of bonds and PP can be employed for FML fabrication.

Hussain et al. [32] developed FMLs with Aluminum and Oil-Palm Empty Fruit Bunch fiber (OPEFB) and used polypropylene (PP) as a matrix, two different types of surface treatment of aluminum was done to check interfacial shear strength; a-degreasing with ethanol, b-Alkaline cleaning in 0.1% NaOH solution for 10 min.

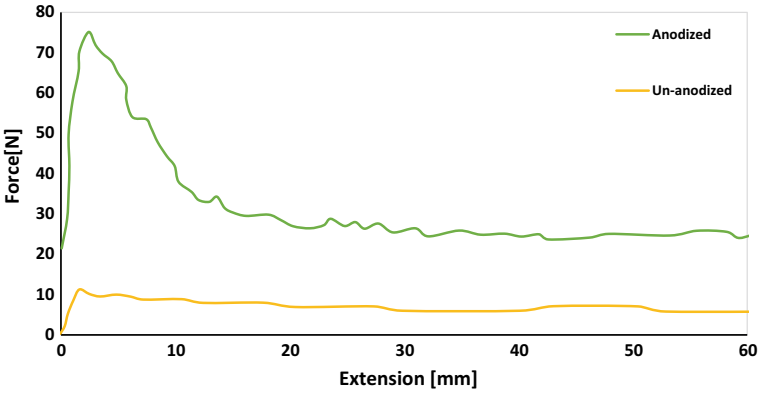

Fig. 11 Comparison of load-extension curve for T-peel test of anodized and un-anodized aluminum-carbon bond [57]

The alkaline cleaning removes the impurities from the surface of aluminum. The results showed similar outcomes for both type of surface treatments.

6.2 *Monotonic Properties of FMLs*

The monotonic properties of FMLs are basic properties which actually describe the material behavior. The properties of FMLs are combination of both the constituents (metal and composites). Due to the contribution of constituent materials the stress–strain curve of FMLs show highly non-linear behavior as compared to composite part. Figure 12 shows the stress–strain curve of Glass Laminate Aluminum Reinforced Epoxy (GLARE) which is showing high deviation from the hook's law, due to the high non-linearity imparted by the metal part. The modulus is also in between the both the constituents [48, 49].

The Glass Fiber Reinforced Epoxy (GFRE) has a marginally lower modulus than GLARE and GLARE has lower than aluminum [59–61]. The FMLs exhibit higher fatigue life as compared to monolithic metal. The reason of this high fatigue life is that the fiber layer act as a barrier and slow down the crack propagation thus extending the fatigue life [62]. The most critical part that determines the properties of FMLs is interface point, thus the delamination between metal-composite interface is often observed and that due to shear forces that is due to transfer of load from composite part to metal part [63].

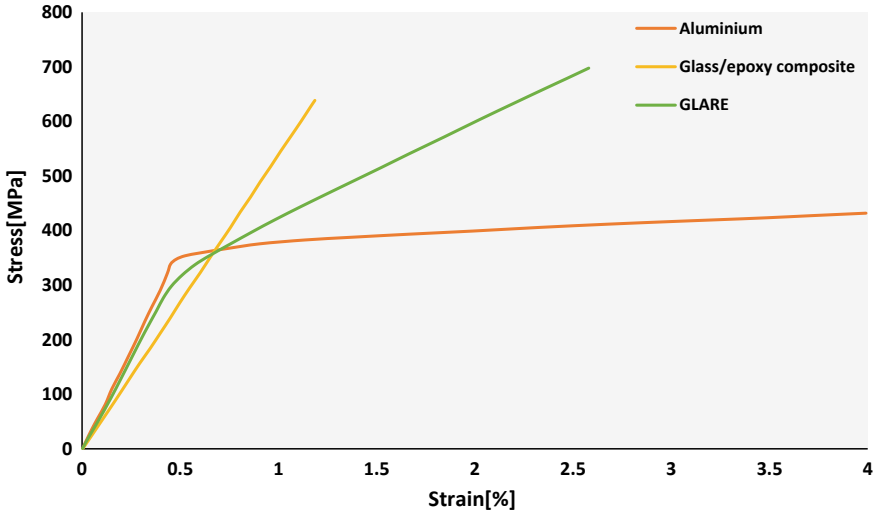

Fig. 12 Typical stress–strain curves showing comparison of tensile properties of FML, aluminum and composites [59]

Just like the composites, where fiber volume fraction is important to determine the final properties, the Metal Volume Fraction (MVF) govern the mechanical properties [64]. "The MVF is the ratio of the thickness of the metal and the total thickness of FML as shown in the equation below:

$$MVF = \frac{\sum_1^n t_{metal}}{t_{lam}} \tag{1}$$

In Eq. 1, t_{metal} = thickness of each aluminum layer, n is the number of aluminum layers, and t_{lam} is the thickness of the total laminate. The value of MVF varies between 0 and 1. 1 shows the 100% metal part, while 0 indicates composite part" [65].

6.3 Monotonic Properties of NFMLs

Natural Fiber Reinforced FMLs are being developed extensively to replace the synthetic counterparts. Since the properties of FMLs are much better than constituents so they are getting a lot of attention. They will also lessen some drawbacks associated with natural fiber-based composites. There are two types of FMLs based on natural fibers which mostly investigated by researchers: a-100% natural fiber-based FMLs (same fiber) and hybrid NFMLs (synthetic-natural, natural-natural fiber reinforcement). NFMLs are developed with both type of matrix systems (thermoplastic and thermoset) [66]. The researchers are studying the properties of NFMLs since long to check its commercial viability. Li et al. [67] compared the mechanical properties of FMLs made with two type of bamboo fibers, simple bamboo fiber and reformed bamboo fibers. The results showed better mechanical properties for FMLs made with reformed bamboo fibers. Sui et al. [68] developed the bamboo based FMLs by extending the work of Li et al. [67], they used cross-ply and unidirectional bamboo fibers instead of randomly oriented and developed NFML. The static mechanical properties were investigated to check the failure mechanism. The results showed that the large size parts have more delamination and defects as compared to small parts this cause lowering its mechanical properties [68].

Malingam et al. [69] investigated the fatigue properties of NFMLs developed with OPEFB. Four different fiber weight percentages (10, 20, 30 and 40%) were used to compare the fatigue life performance. The results showed that NFML made with 30% OPEFB had the highest properties. Sisal is another natural fibers which found in abundance in different countries especially in south America and Africa. Vieira et al. [23] developed the NFMLs with the woven sisal fabric. They compared the mechanical properties of FMLs, and composites made with sisal woven fabrics, the results showed the significantly better tensile and flexural properties of NFML as compared to composite.

Many researchers have developed FMLs with hybrid reinforcement. The hybridization of two different natural fibers is an interesting area as the different natural fibers have different physical and mechanical properties. In one such study

Zareei et al. [12] used basalt and jute to develop a hybrid FML. They used two combinations of reinforcement, one in which basalt was sandwiched with jute and in other combination jute was placed inside. They investigated the tensile and Inter Laminar Shear Strength (ILSS) of NFMLs. The NFML in which jute was in core and basalt fibers in the skin showed higher tensile properties, While the other showed high energy absorption. The NFML with basalt fiber on the outer side showed better adhesion with aluminum as compared to jute fibers.

Hybridization of natural fibers is done with another natural fiber or synthetic fiber to decrease the problems associated with natural fibers e.g., higher moisture regains, low mechanical properties etc. Thirumurugan et al. [70] developed the hybrid FMLs named Glass-Aluminum-Banana-Glass hybrid composite (GABGRP) using a novel method, they sandwiched aluminum and banana fibers strand in Glass Fiber Reinforced Plastic (GFRP). The aluminum was in the form of foils and wire meshes. The GABGRP FML showed higher tensile and impact properties due to its high ductility. In recent years the kenaf fiber is extensively studied to use in natural fiber reinforced composite due to its good characteristics. Notching significantly affect the properties of FMLs especially fatigue life. Feng et al. [6] investigated the hybrid FMLs developed with kenaf/glass hybrid reinforcement and compared with FMLs made with glass and kenaf only. They used two different configurations of plies (0°/90° and ± 45°). The results showed the notched FMLs has relatively lower tensile properties. The hybrid FMLs has also lower tensile properties as compared to glass reinforced FMLs.

Feng et al. [71] extended the earlier research [6] to further investigate the influence of ply-orientation and stress-ratio on the fatigue life of hybrid Thermoplastic Natural Fiber Metal Laminates (T-NFML). The mechanical properties were lowest for kenaf fibers reinforcement and as the content of glass was increased the mechanical properties starts to improve. The tensile test result showed that the tensile properties increase with the hybridization kenaf as shown in Fig. 13.

Subramaniam et al. [72] also made FMLs using glass/kenaf fibers, they used different ply configuration angles and stacking sequence. The tensile and quasi-static indentation properties were investigated with two different types of indenters.

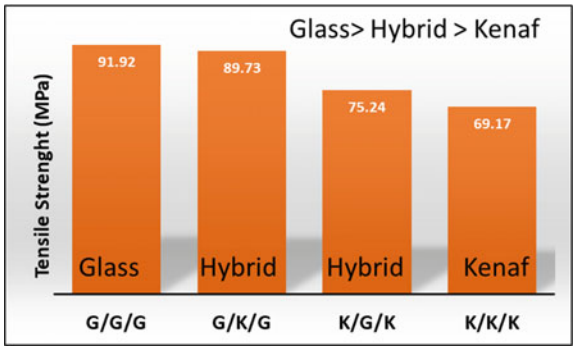

Fig. 13 Tensile stress–strain curve of hybrid FMLs made with 0°/90° plies configuration of glass/kenaf [71]

The results showed that the hybrid FML in which kenaf was sandwiched with glass fibers presented interestingly better mechanical properties. As the content of kenaf fiber increases mechanical properties also decreased.

Sivakumar et al. [73] explored the impact and tensile properties of glass-kenaf based FMLs using the similar ply configuration as used by Feng et al. [71]. The results showed that with hybridization of kenaf with glass fibers improve its mechanical properties as compared to 100% kenaf fibers composite.

Some other researchers also used other natural-hybrid fiber reinforcement to make natural fibers-based hybrid FMLs. In one such study, Muthukumar et al. [74] made hybrid FMLs with stacking of flax and carbon fibers reinforcement in two different ways: 1-flax in core and carbon plies in outer skin (CFC) and carbon in core and flax in outer skin (FCF), the results showed that the better tensile and bending properties were achieved with Carbon-Flax-Carbon configuration. The addition of synthetic fibers improves the mechanical performance of hybrid FMLs in general, similar results were achieved in a study conducted by El-Baky et al. [13] they developed jute/glass fiber based FMLs. The results showed improved mechanical properties for the FMLs in which jute was in core and glass in skin. Another factor which influences the properties of hybrid FMLs is nature of fibers which will be used on outer skin, as glass fibers have better interfacial adhesion so show improved mechanical performance. Chandrasekar et al. [75] explored the effect hybridization of CARAL by sandwiching carbon with flax and sugar palm fibers. The results showed better mechanical properties of flax based FMLs as compared to sugar palm based FMLs. The failure analysis showed that there was crack bridging in the flax fiber based FMLs which make it suitable for high performance applications. Aluminum is most used metal in FMLs fabrication due to its easy processing and handling, but researchers are also exploring other replacements as well.

6.4 Impact Properties of FML

The impact properties of FMLs are very tricky since the composite part is sandwiched between metal layers. The low, high and blast impact have different effect on the FMLs. The damage is normally visible and can be detected with the high velocity impacts but for low velocity impacts normally it is invisible and can keep on propagating inside the composite layer. So that's why the dynamics of low velocity impacts are very important to understand. Since the application area of NFMLs is in objects which are mostly subject to low velocity impacts so it is very important to discuss it low velocity impact properties [76–78].

6.5 Impact Performance of NFMLs

The low velocity impact properties are very important in context of NFMLs, since their potential application area will be in products which will subject to low velocity impact more often. Only few studies are reported in literature in which researchers has explored the impact properties of NFMLs. Zhang et al. [29] used bamboo-aluminum with different configurations to develop NFML and investigated its static indentation and impact properties. Sivakumar [79] prepared NFMLs with kenaf fibers with three variations: 1-fibre length 2-treatment with caustic (NaOH) and 3-Fibre weight percentage. The results showed that the sample absorbed the highest energy made with longer fibers, treated with caustic and higher fibers weight percentage. The NFMLs can pave way for many technical applications due to their unique characteristics and use of natural fiber-based reinforcement. The reinforcement, which is not hybridized is much preferred. Pang et al. [80] also investigated the static indentation behavior of kenaf fibers-based NFML made with aluminum and epoxy. They used 2/1 and 3/2 configuration of plies for FML fabrication. The samples were tested at three different loading rates. The results showed that for the 2/1 configuration, the maximum force and absorbed energy increase with increase in loading rate, whereas, in the 3/2 configuration interestingly, the delamination begins at a low loading, so the due to this reason the maximum-force (F_{max}) was higher at low loading as compared to higher loading rate. Kuan et al. [66] conducted a detailed experimental study in which he developed thermoplastic composites and FMLs using basalt, hemp, flax and Polypropylene (PP) fibers. The Self Reinforced Polypropylene (SRPP) due to their plastic deformation show better impact characteristics. The oil palm fibers can also be used as a reinforcement to make FMLs. Savikumar et al. [81] investigated the LVI properties of thermoplastic NFML developed with Oil-Palm Empty Fruit Bunches (OPEFB).

The impact properties of hybrid FMLs are also investigated by different researchers. Hybridization of different fibers is done to reduce the problems associated with different fibers. Malingam et al. [82] investigated the Charpy impact performance of PP based thermoplastic FMLs. They used glass-kenaf woven reinforcement. They used two type of plies orientations (0°/90° and ± 45°). The ± 45° shown better Charpy impact performance. Hussain et al. [30] also investigated the LVI performance of hybrid FMLs made epoxy and poly Vinyl Butryl (PVB) matrix. They used 3D woven jute reinforcement as core and 2D jute, 2D aramid and 2D carbon as a skin. The 3D woven jute reinforcement was sandwiched with 2D woven skin and later it was used for FML fabrication. The results showed significantly better impact properties for PVB based FMLs even for 100% jute reinforcement. Figure 14 shows the comparison of FMLs made with epoxy and PVB matrix. The more is close to one (01) shows a sample is more damaged.

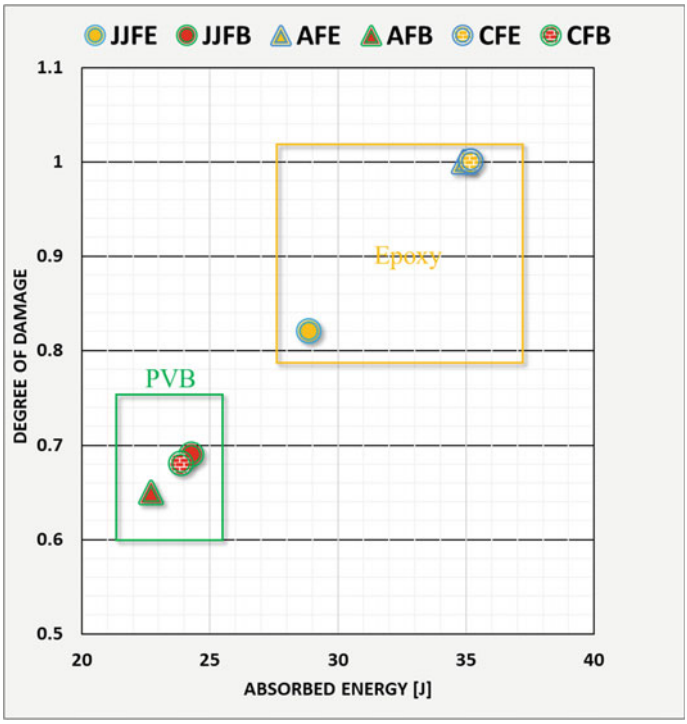

Fig. 14 Comparison of performance characteristics of FMLs made with hybrid reinforcement and two different matrices (epoxy and PVB) [30]

7 Conclusions

This chapter presents the natural fiber metal laminates, its manufacturing, mechanical characterization and joining techniques. The natural fiber metal laminates are emerging due to its better mechanical properties as compared to composites, further these are also being considered as a low-cost replacement of commercially available fiber metal laminates. Different type of natural fibers can be used for the manufacturing of fiber metal laminates in pure and hybrid form. The reinforcement ranges from UD to 3D and researchers has thoroughly studied the effect of different type of reinforcement on properties of NFML. The different types of surface treatments are also used to improve the adhesion of metal with composites. The electrochemical (anodizing) give the best results as far as the adhesion is concerned. The natural fiber metal laminates can be made with different type of manufacturing methods depending upon which type of matrix is being used including autoclave, vacuum infusion and compression hot press. Both thermoset and thermoplastic matrix can be used for the NFML fabrication. The static and dynamic properties evaluation suggests that the properties of NFML are superior to composites and can be used

for different type of applications including automobile, aerospace, trains and cargo containers.

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