

Quest for Environmentally Sustainable Materials: A Case for Animal-based Fillers and Fibers in Polymeric Biocomposites

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Abstract

This review explores the potential of animal-based fillers and fibers as eco-friendly alternatives to conventional synthetic ones. Examining materials such as wool, silk, feather, hair and beak, the review elucidates their chemistry, structure, properties and sources, emphasizing biodegradability and renewability. It also discusses the compatibility of these materials with polymer matrices and their mechanical, acoustic and thermal performances. The review critically analyzes environmental and ethical implications, presenting challenges and opportunities in the emerging field. By addressing ecological and performance aspects, it contributes to global efforts in fostering sustainability in materials science. Future research to address gaps and enhance the design, manufacture and application of animal-based reinforcements in various industries are clearly outlined at the end of the review.

Keywords: Biocomposites; Animal-based fillers/fibers; Sustainability; Biodegradability.

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1. Introduction

The society is gradually becoming aware of the need to protect our planet from environmental disaster. The past few decades have witnessed policies and research efforts towards environmental sustainability. Polymer matrix composites (PMCs) materials are gradually replacing metals and ceramic materials because of the huge environmental effects and energy consumption associated with the production and use of metallic and ceramic products. They represent a dynamic class of materials, characterized by their organic polymer matrices, which can be either thermoset (such as unsaturated polyester or epoxy) or thermoplastic (including polycarbonate, polyvinyl chloride, nylon or polystyrene). These matrices serve as the foundation upon which strong, reinforcing materials such as glass, carbon, steel, or Kevlar fibers are integrated, forming the dispersed phase. PMCs combine the advantages of both fillers and polymers to produce materials with extended properties, which cannot be realized, by the separate materials.

PMCs are known for their excellent specific strength, heat resistance, lightweight, corrosion resistance, durability, specific stiffness, and fatigue resistance. Such excellent property profile qualifies the innovative class of materials to be applied in areas such as chemical vessels, building construction, aerospace (airplanes, helicopters, spacecraft), marine, sport, civil infrastructure (including bridges), automobile, biomedical and household appliance [1-4]. An important aspect of this class of materials is the possibility of tailoring their performance for specific applications through appropriate design. PMCs have gained considerable attention in various industries, including automotive, aerospace, electronics, medical, civil construction, chemical, and consumer applications [5,6]. Their appeal lies in their exceptional properties, such as a high strength-to-weight ratio, excellent electrical insulation, efficient load transfer, and cost-effective processing. In polymer composites, the matrix phase serves as the primary component, typically providing greater ductility and holding the reinforcement phase, which acts as the secondary component. The reinforcements are typically stronger than the polymer matrix, enhancing the mechanical and thermal properties of the composite. Manufacturing PMCs involves various methods, such as injection molding, compression molding, transfer molding, pultrusion, and filament winding. Furthermore, the advent of additive manufacturing technology has enabled the production of these composites, offering design flexibility and the ability to create complex shapes that would be challenging to achieve using traditional manufacturing processes.

Fiber-reinforced PMCs present a spectrum of possibilities, with the choice between short and continuous fibers dictating key characteristics. Short fibers offer distinct advantages in terms of

processability and an increase in part stiffness, although they may be susceptible to fiber pull-out and relatively lower strength. In contrast, the utilization of continuous fibers, while potentially constraining design flexibility, ushers in a substantial enhancement in mechanical performance.

Moreover, the incorporation of particle-reinforced PMCs further expands the horizon of composite materials, adding another layer of versatility and functionality. This pivotal selection between short and continuous fibers, as well as the integration of particle reinforcements, underscores the multifaceted nature of PMCs, which are poised to revolutionize material engineering [7].

Basalt, glass, graphene, carbon, graphite, aramid, Kevlar, wool, silk, shells, natural and wood have been used as fillers in the form of fibers, particulates, and particles. Among these, carbon, glass, and natural plant fibers are the most frequently used and commercialized fillers. Recently, attention has been directed to developing polymer matrix composites with less environmental footprints and cost. Natural fillers have been identified as suitable alternative to some synthetic fillers such as carbon and glass fibers. Natural fillers are generally classified into two groups according to their source, namely, natural plant and animal-based fillers. Over the past decades, attention has been paid to research and development of natural plant filler. Natural fillers are lightweight, carbon neutral, low cost, mostly biodegradable, renewable, and relatively available. They also possess attractive properties such as high specific strength and modulus, vibrational damping, toughness, and elasticity. Polymer composites with reinforcements such as sisal, abaca, coir, cotton, flax, hemp, Kenaf, ramie, jute and palm have been used to produce materials such as seat covers, back seats, package trays, rear parcels, and other useful materials [5,6].

In contrast, animal-based fillers have attracted less attention. These fillers, which are generally waste products from animals include wool, silk, spider web, feather, chicken, animal skin, hair, sinews, positional tendons, sinews, bone, claw, shells, teeth, beak, and toucan, among others. They are mostly protein containing fillers with extremely varying structure, chemistry, and properties. Some of these fillers possess resilience, elasticity, fire-retardance, among others. Fascinatingly, many of these fillers are waste materials produced directly by animals or by-products of food processing. In addition, unlike synthetic and natural plant fillers, most animal-based fibers do not produce greenhouse gases at their end-of-life. Moreover, their end-of-life degradation products are not harmful to health. Essentially, some these fibers possess multiple functional groups such as –OH, –SO₃H, –COOH, –C₆H₅OH, –NH₂, –CONH–, =CO, and –NH. These functional groups afford the possibility of cross-linking and functionalization with other polymers without strenuous

modifications. Furthermore, because these fillers are mostly waste materials, their usage in composite materials will not compete with food production. Summarily, the use of animal-based fillers in reinforced polymer production do not pose adverse environmental effects (Fig. 1).

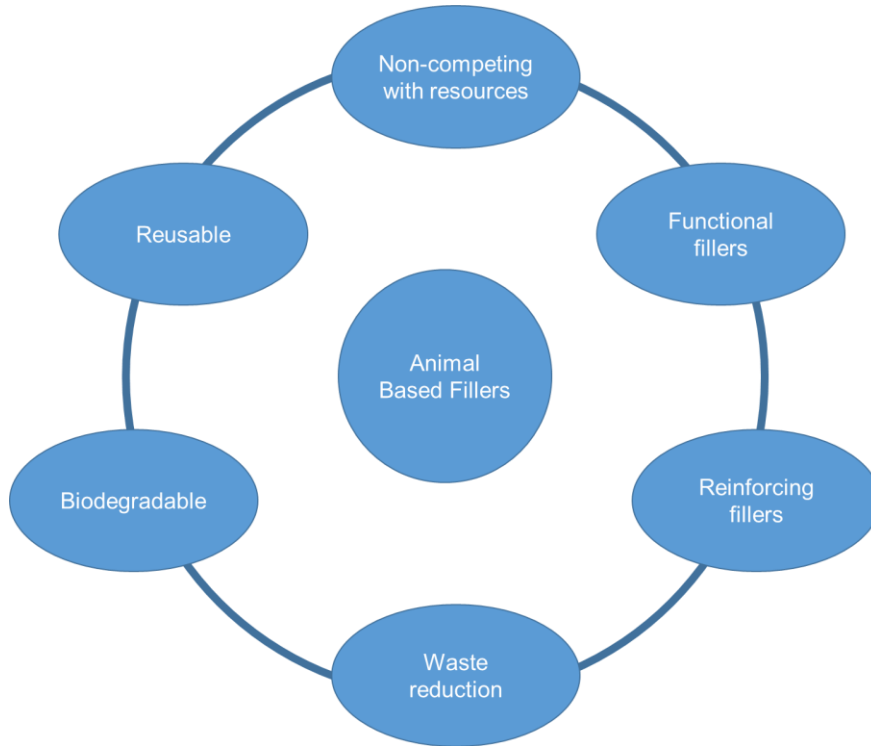


Fig. 1. The sustainability aspects of animal-based fillers.

To increase knowledge in natural fiber reinforced composites, several review articles have been published on natural plant fibers and their hybrids [1,5,6,8,9] . To the best of our knowledge, no research article focusing on animal-based fibers have been published. In this review, we set out to critically discuss the current state-of-knowledge in the development of animal-based fiber reinforced polymer composites. First, we present an extensive theoretical perspective relating to the structure, chemistry, and properties of various animal-based fibers. Thereafter, a detailed presentation of treatment methods and resulting effects on the fiber quality are presented. Polymer composites with animal-based fibers are extensively reviewed to show their properties in relation to composites with natural plant fibers. Finally, future perspectives and research gaps are presented.

2. Chemistry, structure, and properties of animal-based fillers

There is a growing interest in using animal and plant-based fillers, such as wool, silk, basalt, and flax fibers, in the development of PMCs as alternatives to expensive synthetic fillers. There is also a need to produce sustainable and environmentally friendly engineering materials. These natural fibers/fillers offer advantages such as acceptable physical, mechanical, corrosive, and thermal properties [10]. However, the composite properties are influenced by the chemical composition of the natural fibers. For instance, due to the hydrophilic nature of fibers, composites using these materials tend to have a high water absorption rate, which can reduce their mechanical properties [11,12]. This limitation is often addressed through chemical modification of the fibers. Natural fiber-reinforced polymer composites are primarily used for internal engineering applications, such as in automotive interiors [13]. By-products from agricultural processing activities can also be utilized as filler materials in the development of PMCs, further reducing production costs [12,14-17].

Natural fibers used to reinforce bio-based polymers can be categorized as plant/vegetable fibers (cellulosic materials) or animal fibers (mainly proteins, such as keratins and collagens), as depicted in Fig. 2 [18,19]. Collagen, the most abundant structural extracellular matrix protein in mammals, is composed of amino acids bound together to form a triple helix structure and is predominantly found in tissues, skin, tendons, and ligaments [20]. Keratins are proteins stabilized by disulphide cross-linkages and can have ordered α -helix or β -sheet structures, as well as some disordered structures [21].

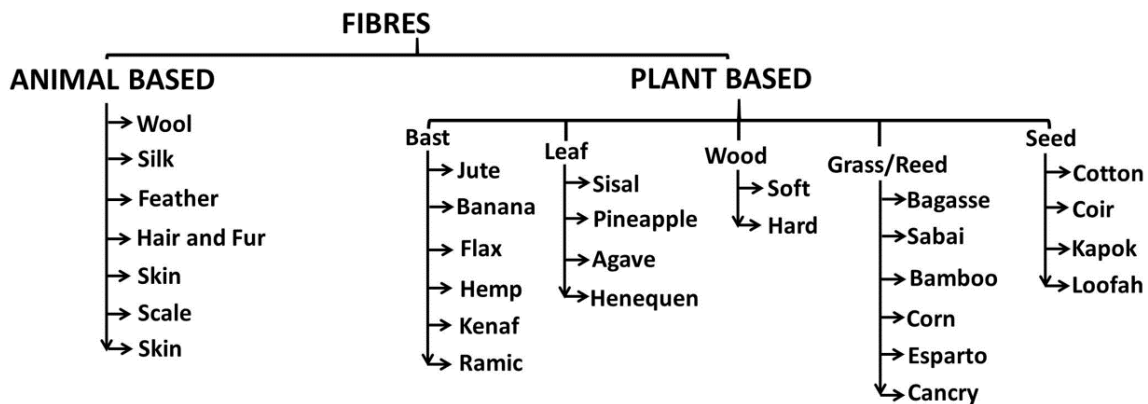


Fig. 2. Natural fibers classification [18,19].

The utilization of animal-based fillers in the development of biobased polymer matrix composites is gaining traction due to their ecological friendliness, sustainability, biodegradability, lightweight nature, high-specific strength, abundance, and cost-effectiveness. However, the mechanical properties of these composites can be limited by the poor compatibility of natural fillers with polymer matrices and their hydrophilic characteristics. The chemistry, structure, and properties of these filler materials are closely interconnected.

To address these limitations, various chemical treatments have been proposed to modify the surface morphology of natural fillers and fibers. Hydrolysis and plasma treatment are examples of treatments that have been suggested for animal-based fibers [22]. Hydrolysis can produce more polar hydrophilic groups such as hydroxyl- and carboxyl groups on the fiber surface while plasma treatment create a rough surface topology on the fiber. Similarly, chemical treatment such as acetylation, benzylation, and those involving the use of alkali, silane, maleated coupling agents, isocyanates and permanganates have been found suitable for animal-based fillers [23-26]. These methods modify the hydroxyl and carboxyl functional group on their surface. Such treatments are aimed at decreasing filler moisture absorption, enhance compatibility with polymer matrices, and improve strength. Additionally, optimization techniques such as fiber stretching, calendaring, thermo-treatment, and electric discharge can be employed to enhance fiber-matrix interaction without altering the filler's chemical composition [27]. Table 1 presents a summary of the physicochemical properties of some of these fillers/fibers, facilitating a quick comparison for reference.

Table 1. Overview of animal-based material physicochemical properties.

Materials	Density (g/cm ³)	Modulus of elasticity (GPa)	Tensile strength (MPa)	Strain (%)	References
Wool	1.3	0.5	200	0.5	[28-30]
Silk, spider web	1.3	10	1100		[31-33]
Feather, chicken	0.8	2.50			[34]
Skin		1-10			[35]
Hair, Human		0.002–7.5			[36-38]
Sinews, positional tendons		0.7-1		6-8	[39]

Sinews, energy-storing tendons	0.1-0.15		12-15	[40]
Bone				
Claw, ostrich	2.7	90	5.7	[34]
Shell				
Teeth, dentin	13-17	70-80		[29]
Teeth, enamel	60			[29]
Horn, oryx	6.1	137		[41]
Beak, Toucan	1.5	30	0.1	[42]

The properties of biobased materials are predominantly influenced by relative humidity and moisture content, making it crucial to understand the characteristics of these natural materials for the advancement of enhanced biobased polymer matrix composites. In this review, animal-based fillers are categorized based on their suitability for use as fibers or in particulate form.

2.1 *Fibers*

An insight into the chemical characteristics of natural materials is required for the development of improved composite materials. This section focuses on the chemistry, structure, and properties of some animal-based fillers.

2.1.1 *Wool fibers*

Wool fibers are sourced from the follicle of sheep, rabbits, camels, and goats. Keratin (α -keratin), which has polypeptide and amino acid chains is the main component of wools [43]. Wool fiber has an α -helical arrangement which can be transformed to β -configuration by mechanical stretching or plasma modification [44]. The fiber offers advantages such as eco-friendliness, sustainability, resistance to bacteria attack, lightweight, recyclability and good flame resistance [45,46]. They are generally classified as either fine (merino, 20-24 μm), coarse (karakul) or medium wool (24-30 μm) and very coarse ($\geq 35 \mu\text{m}$) wool, which has limited applications [47]. Wools are composed of carbon (50-52 wt%), hydrogen (6.5-7.5 wt%), nitrogen (16-17 wt%), oxygen (22-25 wt%) and sulphur (3-4 wt%) [48]. They consist of three distinct morphological

components, which are cuticle (outer layer), spindle-shaped cortex cells (middle layer) and medulla (inner layer), as shown in Fig. 3 [49-51].

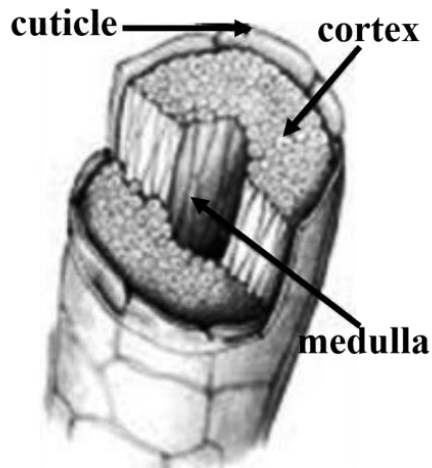


Fig. 3. Wool fiber components [52].

The wool fiber structure and sub-components are captured in Fig. 4 and 5. The outer layer, which constitutes about 10-15% (for fine wool fiber) is made up of lipid (epicuticle) and cystine residue (exocuticle) [52]. The epicuticle bonds with the cuticle cell via covalent isopeptide crosslinks, providing good resistance against enzyme, alkali, acid as well as reducing and oxidizing substances. Its removal by alcoholic alkaline or chlorine solution treatment can improve wettability. The exocuticle is characterized by peptide and isodipeptide bonds. These bonds can be solubilized by oxidation or reduction treatment after initial enzyme treatment. Generally, wool cuticle act as a diffusion barrier to chemicals due to the presence of -S-S bonds (disulphide bonds), which are the secondary primary linkages in the structure [53]. Pre-treatments involving the use of chemicals (organic phosphonic compound) [54], hydrothermal [55], oxidation [56], which can modify the cuticle cells disulphide bonds can improve wool fibers reactivity by changing their composition and morphology.

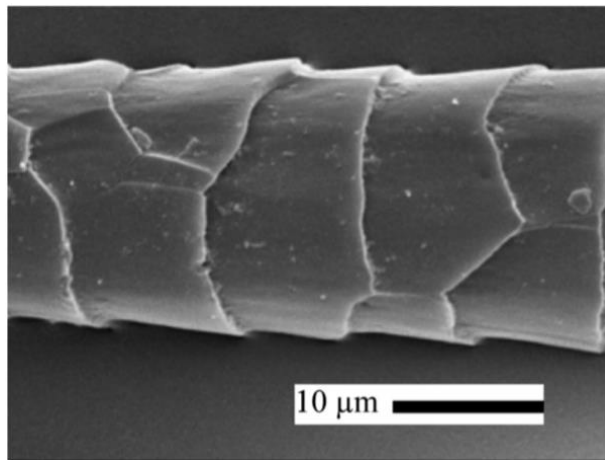


Fig. 4. Wool fiber structure [57].

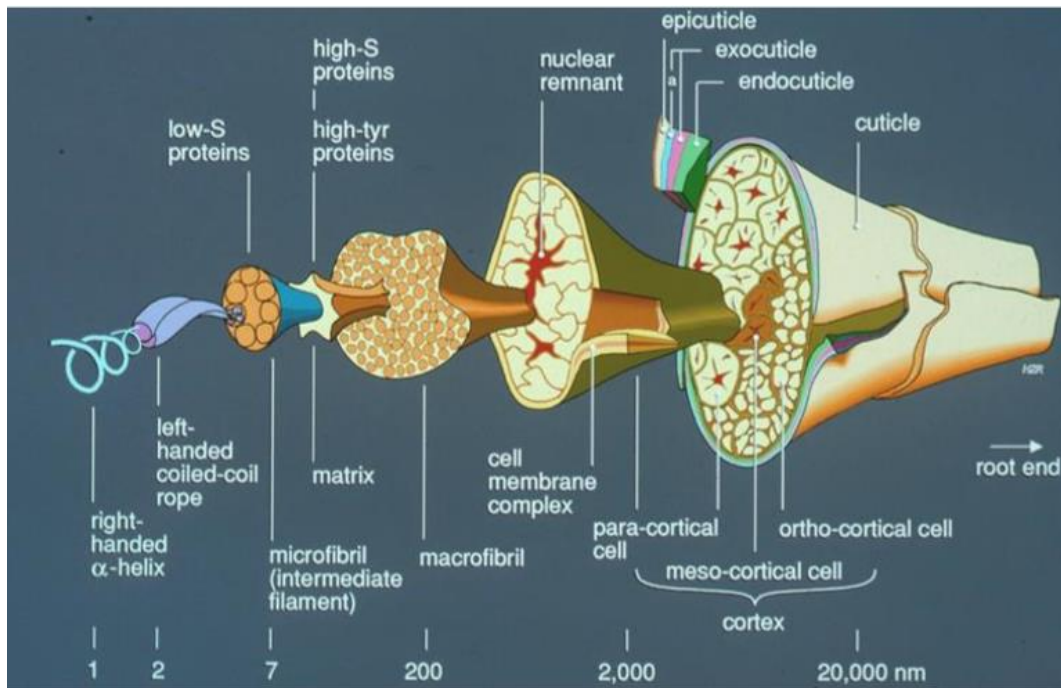


Fig. 5. Schematic representation of wool fiber structure [58]

The cortex layers, which are separated from each other by a cell-membrane complex constitute about 80-90% of wool fiber. The cortex is divided into ortho (60-90%), para (4-10%) and meso cortex (that is present only in low-crimp merino wool) with distinct and different chemical and physical characteristics [52,59]. For example, para cortex is less reactive. The presence and behavior of this layer provide important characteristics such as resilience, brittleness and non-lustre that are important properties that are considered for the application of bio-based polymeric composites [52,60].

Medulla is not present in fine wool and only a few fractions of coarse wool are medullated. This inner layer is formed due to an inadequate supply of nutrition during wool growth [52]. The medullation process can be classified as hetero medullation (fragmented), hairy medullation (unbroken) and heavily medullated (kemp). Fig. 6 captured the various components and sub-components of wool fiber.

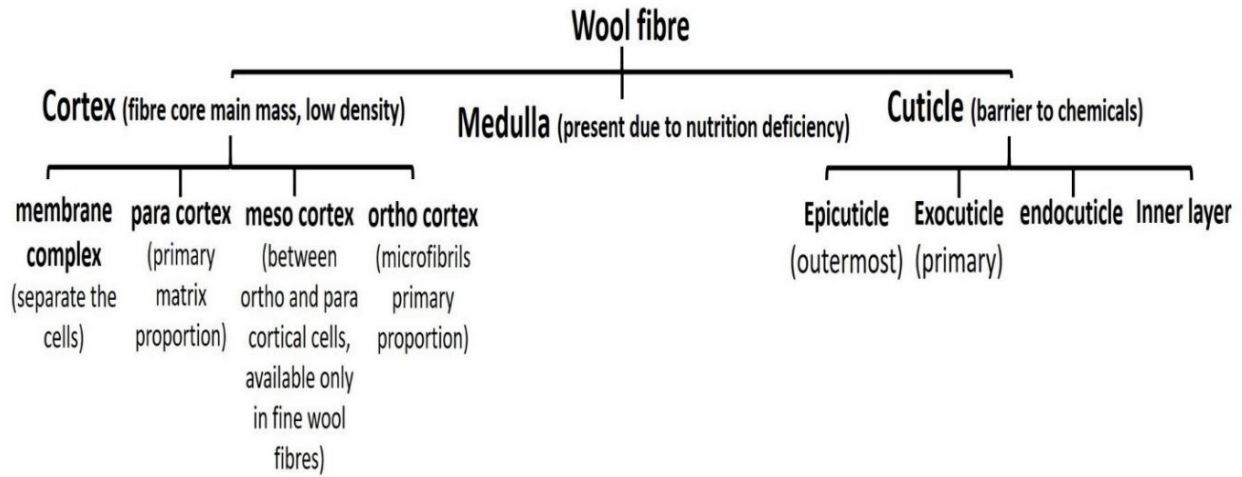


Fig. 6. Illustration of the components of wool fibers.

Wools are used as fibers for polymer matrix composites because of benefits such as relatively low weight and density, low cost, less damage to processing equipment and high flame retardancy, which is the highest among natural fibers [61]. The stiffness, strength, extensibility and toughness of wool are 0.5 GPa, 0.2 GPa, 0.5 and 60 MJm⁻³, respectively [28,29]. The major demerit of this fiber is its shrinkage (relaxation, consolidation, swelling, felting, hygral) due to the presence of cuticle cells and the absorption of moisture [52,62]. Wool fibers, up to 60%, have been used to reinforced polypropylene, using melt blending technique with poor fiber-matrix adhesion. Nevertheless, the fiber-matrix interaction become better with the use of a compatibilizer [63]. Wool fibers have also been combined with epoxy resin with improved tensile and bending properties [48]. Based on the articles reviewed in this section, there is still need to carry out extensive studies to determine the mechanical properties of single wool fiber derived from diverse animal sources and correlate same with the material structure. This information can aid in the development of improved bio-based polymeric composites.

Another interesting aspect is the limited degradability of keratin component of wools, which can make it constitute a long-term environmental pollution [64]. As an organic waste, wools can

be used in the development of composites to mitigate against their improper disposal. Nevertheless, they are still underutilized in PMC production, which may be attributed to their poor interfacial bonding with resins [48]. However, this drawback may be resolved by surface functionalization using chemicals (enzymatic treatment and chlorite treatment) or physical (radiation treatment) methods.

Govindaraju *et al.* [65] utilized a carding process (based on Box-Behnken design of experiments and response surface methodology) to incorporate 50% by mass of scoured wool fibers, each 45 mm in length, along with polypropylene (PP) into a web, achieving fiber orientation. This web was then subjected to compression molding under varying process parameters. The optimal parameter combination yielded a tensile strength of 31 MPa and flexural strength of 37 MPa [65].

Furthermore, Conzatti *et al.* [66] conducted a study to assess the impact of fiber length and interfacial strength on the mechanical properties of wool fiber-reinforced PP compounds, focusing on attributes such as low flammability [66]. Short wool fibers were compounded with both neat PP and PP grafted with 5% maleic anhydride. The wool fibers, with a thickness of 18 μm , were cut into approximately 20 mm lengths and blended via melt blending, resulting to compound mass fractions of 20 and 40% for both types of matrices. An additional compound with a fiber mass fraction of 60% was created using the maleic anhydride grafted PP matrix. Compression molding was employed to produce thin sheets measuring 0.32 mm in thickness. The critical fiber length in the maleic anhydride grafted PP matrix was determined through a single fiber fragmentation test, revealing that intense shear stresses during blending shortened the fibers to an average length of less than 0.2 mm. Despite a decrease in tensile strength due to the shortened fiber length, the use of compatibilized PP resulted to higher strength values when compared with pure PP. In another study, Conzatti *et al.* [67] pursued a novel approach to enhance interfacial strength by grafting a silane-based coupling agent onto the surface of wool fibers, which reacted with the compounding PP-based matrix. Prior to silanization, the wool fibers underwent oxidation to heighten the reactivity of their surface. These silanized fibers were then blended via melt blending with the same maleic anhydride grafted PP (MAPP) matrix and under identical processing conditions as described in their previous work [67]. The effectiveness of the silanization process was confirmed by measuring the critical fiber length of untreated, oxidized and oxidized and silanized fibers within the MAPP matrix. Oxidation and silanization reduced the critical fiber length from 0.72 to 0.43 mm, enhancing its potential for reinforcement. Moreover, fiber treatment increased resistance to breakage during compounding, with the mass-weighted average fiber length rising from 0.31

mm in untreated fibers to 0.5 mm in oxidized and silanized fibers. Consequently, compounds containing oxidized and silanized fibers exhibited higher tensile strength. Furthermore, samples comprising uncut, unidirectionally oriented fibers were produced using the same methodology, as described in Conzatti *et al.* [66]. These samples utilized the MAPP matrix, with fibers ranging from untreated to oxidized and silanized. While all samples containing the MAPP matrix demonstrated increased strength and stiffness, the impact of fiber treatment on these properties remained inconclusive. The study concluded that in samples containing short, unaligned fibers, the influence of longer fibers overshadowed the effects of enhanced adhesion resulting from fiber treatment.

2.1.2 Silk fiber

Silks are natural fiber with elastomeric networks (viscid silk) that are crosslinked by β -sheet crystals and α -helical or collagen triple-helical crystals structure [68]. They are derived from animals such as cocoon, spider, bees, moth, and mite. They are generally composed of two proteins: fibroin (tough strands) and sericin (gummy glue). Animal silk's mechanical properties can vary extensively [29]. The modulus and strength of silks (for rigid frame silk from spider web) can be as high as 10 and 1.1 GPa, respectively [68,69]. The value of the latter property is close to that of engineering steel, which is 1.3 GPa [70]. With these good properties, silk density is only 1.3 g/cm³ when compared with engineering steel, which is 7.8 g/cm³ [70]. These characteristics make silk fiber suitable for the development of lightweight biobased polymeric composites for several engineering applications in the automotive, aerospace, construction, electrical, and electronics industries [69]. Depending on the spinning conditions (wet, dry, microfluidic spinning), animal species and size, the fiber diameter can range from 20 to 7000 nm [68]. Silk contains a significant crystalline β -sheet structure with a chain axis parallel to the fiber axis (Fig. 7). Nevertheless, the connecting protein chains have amorphous structure, which represents about 50% of the total structure [71,72]. Generally, their crystal can be represented as pseudo-orthorhombic β -pleated sheets with molecular chains of different orderliness degree [73]

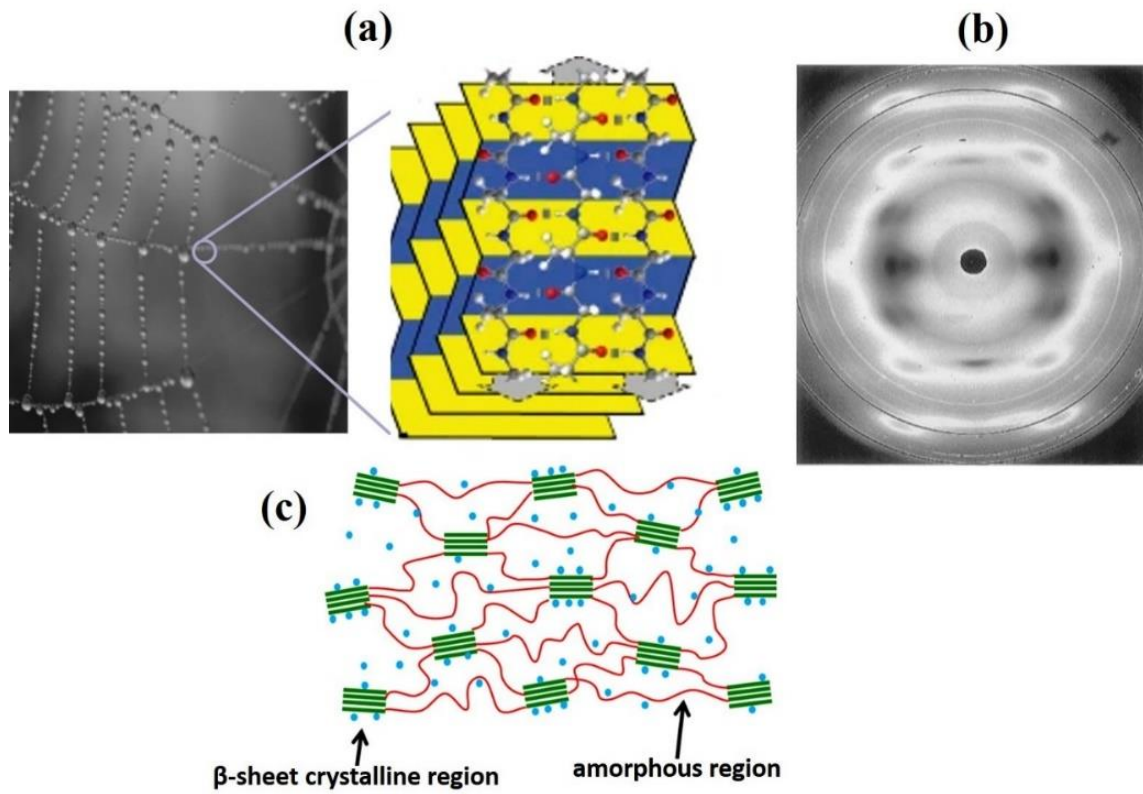


Fig. 7. Silk structure of (a) three-dimensional (3D) silk β -sheet crystals [74] and (b) dragline silk X-ray diffraction pattern [75].

When compared with plant-based fibers, silks reinforcement usually presents a better compact structure. The mechanical properties of silk when compared with some plant-based fibers with respect to their densities are shown in Fig. 8 [18,76]. This illustration provides an important consideration necessary for the development of biobased polymeric materials for engineering applications. As can be seen, silk compares favorably with other plant-based fibers in terms of strength against density although its modulus is relatively low.

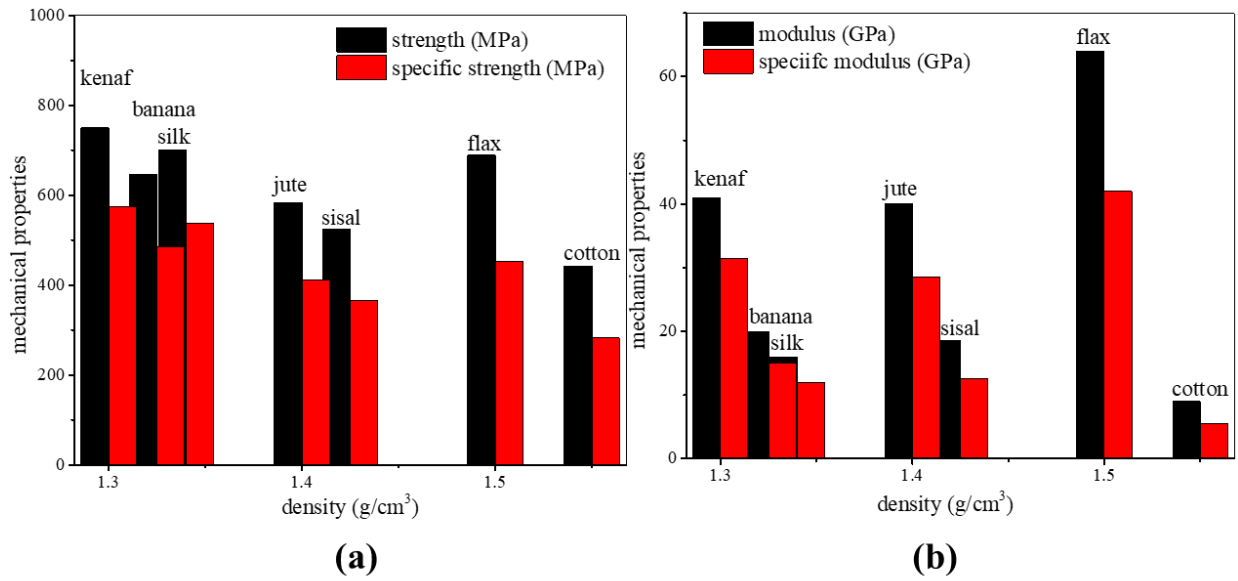


Fig. 8. Mechanical properties of some natural fibers against density (a) strength and specific strength and (b) modulus and specific modulus [18,76].

Silk is indeed one of the few animal-based fibers that can be replicated artificially [77,78]. The process of creating artificial silk involves using proteins found in natural silk fibers, typically derived from the cocoons of silkworms. These proteins are then dissolved and processed to create a solution, which can be extruded through fine nozzles to form fibers. This process mimics the natural spinning process of silkworms. Artificial silk, often referred to as "synthetic silk" can have properties like natural silk, such as a smooth texture, sheen and strength. Additionally, artificial silk can be engineered to have specific properties tailored to different applications, making it a versatile material in modern manufacturing. However, there can be substantial variability in the mechanical properties of silk fibers when compared with commercial carbon fibers. In the study by Greco *et al.* [79], the mechanical properties of various types of silk fibers, including *Bombyx mori cocoon* and degummed silk, native spider silk, and artificial spider silk, were determined and compared with classical commercial carbon fibers. The research aimed to address the challenge posed by the intrinsic variability of natural and artificial silk fibers, which has often been overlooked in previous studies relying on tensile tests. The study found variability in the mechanical properties of silk fibers with relative standard deviations for strength and strain at break ranging from 10 to 50%. Interestingly, increasing the number of tested fibers did not significantly decrease the variability, contrary to what has been frequently reported in the

literature. The study demonstrated that tensile testing of 10 fibers per type could be representative of a silk fiber population.

Rajkumar *et al.* [80] reinforced neat PP with a blend of silk (75 mm long) and scoured wool fibers (45 mm long), adjusting the relative proportions of the two fiber types across samples [80]. Among the produced samples, those with a total fiber mass fraction of 50% exhibited the highest mechanical strength when the silk/wool/PP ratio was 35/15/50, with tensile and flexural strengths of 30 and 20 MPa, respectively. However, samples with a 50% fiber mass fraction and a higher wool content showed lower strength, with tensile and flexural strengths of 23 and 10 MPa, respectively. Samples with a total fiber mass fraction of 70% exhibited even lower mechanical properties. This was attributed to inadequate fiber wetting, resulting to high porosity.

2.1.3 Feathers

Feathers (Fig. 9) waste generated from poultry industries is estimated to be up to 4 million tons per year [81,82]. They can be used to replace non-environmentally friendly raw materials for several engineering applications. Their fibers (Fig. 10) are composed of keratin (the second most abundant biopolymer after collagen) that is rich in hydrophobic residues with strength close to that of nylon and a fiber diameter lesser than wood [83]. Tamrat *et al.* (2017) analyzed chicken feather and reported the presence of crude lipid (0.83%), nitrogen-free extract (1.02%), ash (1.49%), crude fiber (2.15%), moisture content (12.33%) and crude protein (82.36%) [84]. The elemental compositional analysis shows that it contains 2.64% sulphur, 10.41% nitrogen, 22.34% oxygen and 64.47% carbon. FTIR analysis also revealed the presence of proteinous functional groups such as amide and carboxylic groups. The cross-sectional views of flight and down feather fibers are shown in Figure 10. Evidently, the flight feather fiber exists in hollow form, while the down fiber is solid. Considering the purpose of fiber reinforcement, the utilization of down fiber will be more advantageous [85]. The detailed information about the dimensions of the chicken feathers used in this study were not provided. However, the samples used for a portion were milled into powder.

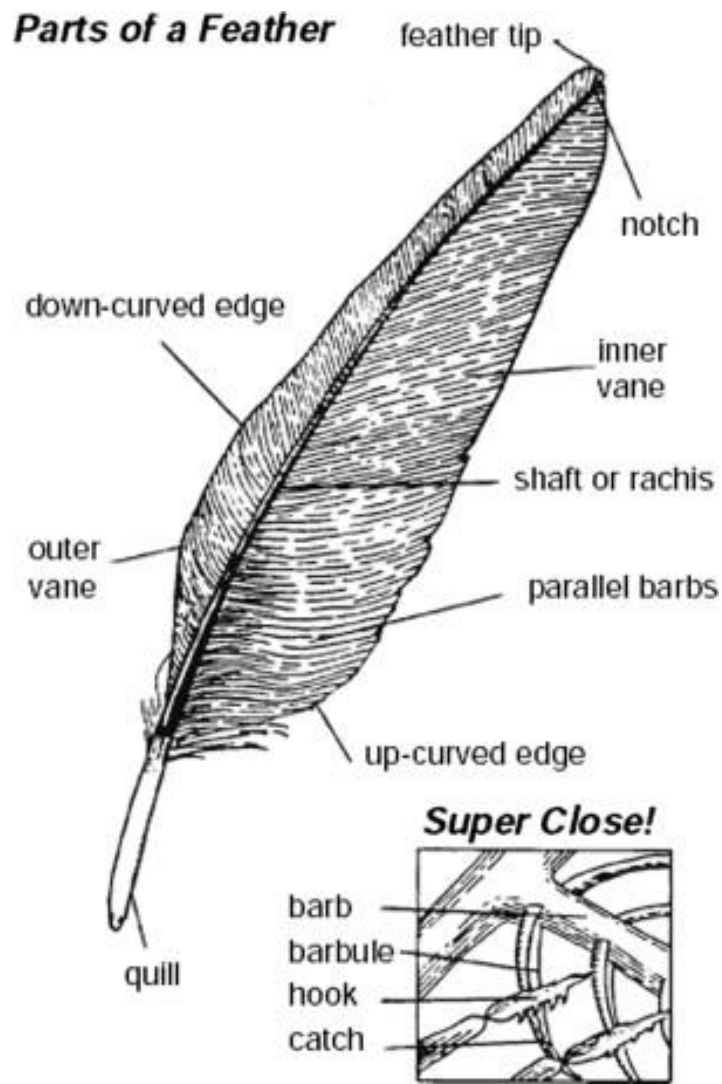


Fig. 9. Chicken feather fiber and its parts [86].

Polymer-based composites reinforced with chicken feather fibers have enhanced mechanical stress resistance (55 MPa) and a relatively high modulus of elasticity (4.2 GPa) due to the fillers' crosslinked structure and semicrystalline characteristic of the fiber [87]. Additionally, these fibers are readily available, cheap, biodegradable, resilient and light when compared with synthetic and some other reinforcing materials. For example, chicken feathers density is about 0.8 g/cm^3 whereas cellulose and wool fibers are about 1.5 g/cm^3 for and 1.3 g/cm^3 , respectively [30].

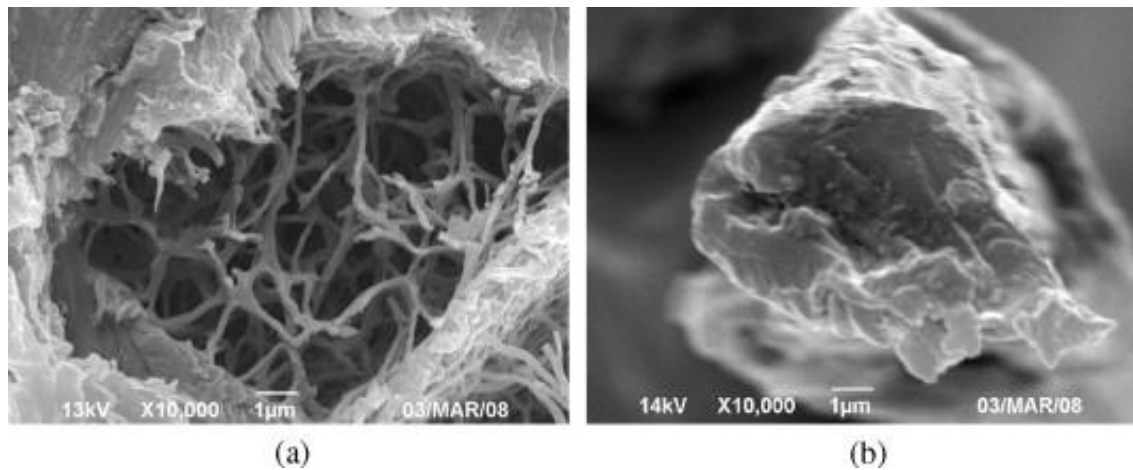


Fig. 10. Scanning electron microscopy (SEM) images of: (a) flight chicken fiber, (b) down chicken fiber [85].

Feathers are also used in composites development due to their low density coupled with good acoustic and thermal properties [88]. Carrillo *et al.* [88] reported that the Young's modulus of composites reinforced with chicken feathers increases slightly at low filler contents and levels off at a concentration of about 10%. This result is understandable considering the individual characteristic of feathers as alighted earlier. In another study by Aranberri *et al.* [89], the incorporation of 50 and 60 wt.% chicken feathers (1.0 mm) were found to decrease the thermal stability of biodegradable reinforced polylactic acid (PLA) and polybutyrate adipate terephthalate (PBAT) composites by 7-9% due to the higher content of the unstable keratin [89]. Also, there was only a slight improvement in the Young's modulus of the reinforced PLA composites with 50 wt.% fiber (2.1 GPa) when compared with the neat PLA (1.9 GPa). Nevertheless, there was substantial amount of improvement with PBAT composites reaching a value that is up to 1000% [89]. However, tensile strength and elongation at break were found to decrease with addition of the chicken fiber irrespective of the fiber loading or polymer type. This study is interesting as it highlights the benefits and drawbacks associated with the use of this material as reinforcement.

2.1.4 Hair and fur fibers

Human hair has been used as fiber for the preparation of bio-based polymer matrix composites. However, it is considered a waste in many societies and may constitute environmental pollution in streams due to its low degradability. Apart from discarding it, which is the common practice in most rural areas, high-quality hair is majorly used as wigs. Hair, which is a filamentous biomaterial consists of carbon (45%), oxygen (28%), nitrogen (15%), hydrogen (7%) and sulphur (5%). It is

composed essentially of hard, compact, and strong keratins in additions to lipids, minerals, and pigments [90,91]. The keratins are arranged helically to form α -keratins (a structural fibrous protein found in mammals, such as in wool fiber), which are linked together by strong disulphide bonds as well as relatively weak salt and hydrogen bonds. The -S-S- bonds are difficult to stretch or bend. The salt bridges are affected by water and pH changes, and they contribute to the overall structure stabilization. The hydrogen bonds located between the α -keratin helix are also influenced by the presence of water, which can penetrate the polypeptide chains and caused it to break [36]. Hair keratins are composed of three stratified layers: cuticle, cortex, and medulla (Fig. 1). The cuticle with a thickness between 1.5 and 5.0 μm consists of flat overlapping cells that protect the inner hair parts and is responsible for its rigidity [36]. The cortex complex and multi-scale fibrillar system, which is about 90% of the fiber weight plays a major role on hairs' mechanical properties [36,92] while the medullar (which is not always present) has a negligible effect. The cortex is composed of several micro-fibrils formed from protofibrils that are developed from the combination of two or three α -helixes. The microfibrils are linked together by covalent bonds inside an amorphous matrix [36,93]. Human hair's rigidity varied greatly between 2–7500 MPa [36-38] and its mechanical properties are affected by humidity, temperature, chemical treatments, and other factors such as ageing process and ethnicity [36,94].

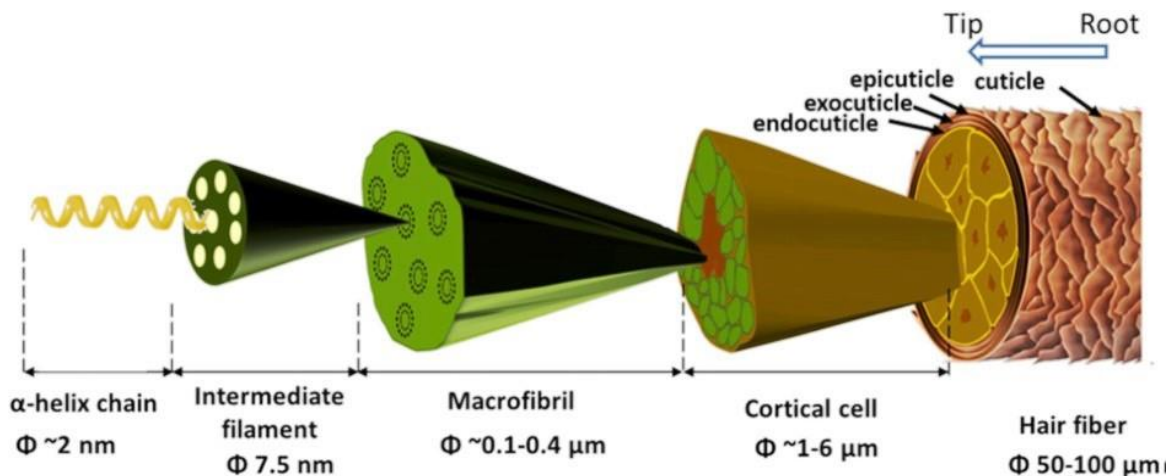


Fig. 1. Schematic representation of human hair structure from α -keratin helix [95].

Furs are also keratin-based animal fibers [96]. They differ in quality depending on the animal's health condition, age, and diet. They can be obtained from animals such as camels, alpacas, llamas, vicuñas, guanacos, reindeer, rabbits, Angora goats, and Kashmir. They are classified as long and coarser beard or guard hairs, and the short and finer under-fur, as shown in Fig. 2 [97,98]. Figure

12 shows the difference between the short undercoat fur hair and the straight shaft guard hairs that project the layer of the undercoat fur hair. The micrograph of alpaca hair fiber is shown in Fig. 3, and their properties are presented Table 1.

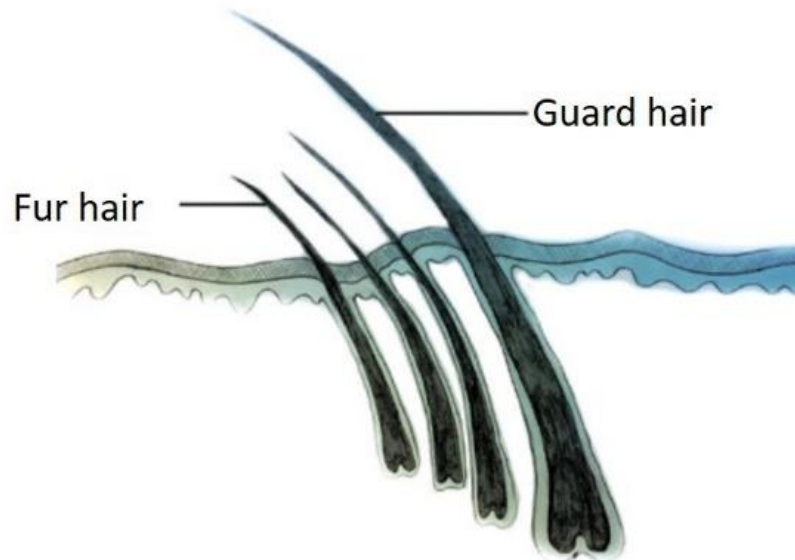


Fig. 2. Animal hairs classifications showing short fur and long guard hair [98].

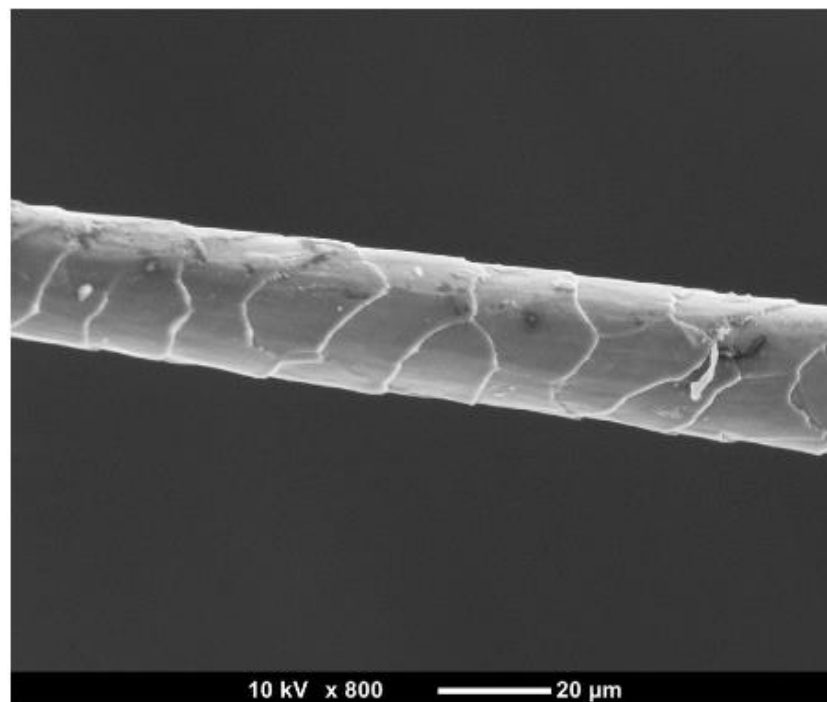


Fig. 3. SEM micrograph of alpaca hair fiber showing More radial distribution of para cells [99].

Table 1. Properties of alpaca hair fiber [99].

Fiber property	Huacaya	Alpaca
Scale pattern	Profile superior and harsher surface.	Even surface.
Cortical structure	Ortho/para construction. If fine fibers are medullated, the bilateral structure remains. At 25-35 μm , ortho/para cells spreading becomes variable.	No demarcation of ortho/para spreading. More radial distribution of para cells.
In air		
Elongation (%)	37.4 ± 2.4	33.8 ± 3.2
Stress at break (MPa)	$221.1 (\pm 24)$	$151 (\pm 24)$
In water		
Elongation (%)	51.2 ± 3.1	43.5 ± 3.0
Stress at break (MPa)	$137 (\pm 16)$	$86 (\pm 15)$
Friction coefficients		
μ_2 (with scale)	0.28 ± 0.09	0.29 ± 0.03
μ_1 (with anti-scale)	0.42 ± 0.07	0.40 ± 0.04
Directional friction coefficient	0.19 ± 0.06	0.16 ± 0.03

Ali *et al.* [100] investigated into the impact of incorporating human hair (4 mm) as a fiber on the physical and mechanical properties of polyester composites [100]. Tensile, flexural, impact strengths, and hardness were assessed to determine the viability of using human hair-based polyester composites in diverse engineering applications for extended durability. The addition of human hair fiber to polyester along with marble dust/rice husk/fly ash as a filler (40:40:20), were found to enhances the impact strength of the developed composite by about 20%. Nevertheless, as formulations were created without a specific one comprising solely polyester and human hair fiber, the actual influence of the human hair fiber remains unclear. Boyala *et al.*, 2023 explores the creation of 3D-printed human hair polymer composites using Stereolithography [101]. Surface modification techniques, including Methacryl Polyhedral Oligomeric Silsesquioxane (MA-POSS) grafting and cold air plasma treatment were applied to enhance interfacial adhesion between fibers

and the matrix. Single hair pull-out tests revealed that plasma-treated human hair composites exhibited the strongest interfacial strength. The mechanical properties of 3D-printed human hair composites were investigated, showing a 35% increase in ultimate tensile strength for plasma-treated composites when compared with the untreated ones and a 100% increase when compared with unreinforced composites. Nanda and Satapathy [102] utilized **short** human hair as a biofiber in fabrication of epoxy composites [102]. The composites were created through a simple hand layup technique, incorporating an epoxy matrix and varying proportions of hair fiber (0, 5, 10, 15 and 20% by weight). The incorporation of 20 wt.% fiber leads to a notable 21.4% reduction in the thermal conductivity of epoxy. As the hair fiber content increases, the composites exhibit a gradual rise in the glass transition temperature (T_g), progressing from 98 to 109.6°C at a 20 wt.% fiber loading. The compressive strength of neat epoxy starts at 84 MPa and, with a 5 wt.% fiber reinforcement, increases to 89 MPa. Further additions of 10, 15 and 20 wt.% of fiber result in composite compressive strengths of 98, 106, and 112 MPa, respectively. In terms of flexural strength, neat epoxy begins at 58 MPa, escalating to 92 MPa with a 5 wt.% fiber reinforcement. The introduction of 10, 15 and 20 wt.% of fiber yields flexural strengths of 108, 121, and 129 MPa, respectively. The tensile strength of neat epoxy stands at 65 MPa, rising to 96 MPa with a 5 wt.% fiber reinforcement. Further increments of 10, 15 and 20 wt.% of fiber result in composite tensile strengths of 116, 141 and 151 MPa, respectively. The studies showcased the potential for producing epoxy composites with enhanced mechanical strength, improved insulation, elevated glass transition temperature, and a reduced thermal expansion coefficient.

More also, Muktari *et al.* [103] investigated into the impact of fiber treatment on the performance of cow-hair/recycled low-density polyethylene (LDPE) composites derived from waste streams [103]. Various chemical treatments, including sodium hydroxide (NaOH), potassium hydroxide (KOH) and hydrogen peroxide (H_2O_2), were utilized to remove lipids from the cuticles' outer layer of the fibers. The treated fibers were then melt-blended with LDPE constituents, using a two-roll mill and subsequently compression molded into sheets. The fiber mass fraction ranged from 10 to 50% in increments of 10%. Results indicated that the incorporation of untreated cow-hair did not enhance tensile strength, whereas pretreatment showed improvements at specific fiber mass fractions for certain chemicals. Notably, the most significant enhancement was observed with H_2O_2 -treated fibers at a mass fraction of 30%, resulting to a tensile strength increase from 7.45 MPa in neat LDPE to 9.62 MPa in the reinforced composite. Tensile stiffness was enhanced across all samples, with H_2O_2 -treated fibers demonstrating the most

substantial improvement, achieving a sevenfold increase over the neat matrix at a 30% mass fraction.

Oladele *et al.* [104] incorporated locally sourced cow hair waste, a byproduct of the meat industry, into an HDPE matrix, with a specific focus on chemical treatment prior to compounding. To enhance interfacial adhesion by removing fatty acids from the cuticle, the fibers underwent washing with NaOH, followed by rinsing with deionized water multiple times. The addition of 2–10% by mass into the matrix generally resulted in decreased tensile strength and stiffness, albeit with a slight improvement in flexural properties at certain percentages. However, there was no clear trend regarding how the effect of prior NaOH washing could be discerned.

2.1.5 Scales

Scales are natural plate-like armor found on animals' skin such as fish, reptiles, and some mammals. Most fish have bony scales formed by dermal ossifications while reptiles are characterized by scaled integuments with horny scale and some mammals, such as rats and mice, have scales on the tail. They are composed of β -keratins, which behaves linearly elastically under mechanical loading. Such materials have higher stiffness when compared with those composed of α -helix [29]. In this section, emphasis has been given to fish scales, which can readily be used as filler for the development of biobased polymer matrix composites. The fish processing industries can generate up to 1.4 million fish scale annually and they are mostly dumped at sea or discarded in landfills [105].

Fish scales have a flexible laminated composite structure with good specific strength and toughness [106]. Based on their structure, fish scales are generally classified as placoid and non-placoid. Placoid scales are found in many cartilaginous fish with a structure like that of teeth. Non-placoid scales are classified as cosmoid, ganoid, and elasmoid [107]. Cosmoid scales have a thinner and harder outer layer relative to placoid scales but a slightly different crystallographic setting. Its inner layer is made up of lamellar bone, spongy or vascular bone, and a layer of non-cellular, hard, dentine-like material called cosmine. This scale can only be found in extinct Crossopterygians, but the modified version is found in Coelacanth without cosmine. Ganoid scales, which are found on gars are more rigid with a layered microstructure resembling that of teeth and cosmoid scales but contain a hard inorganic salt substance called ganoin [106]. Elasmoid scales on bony fishes (Osteichthyes) are thin and translucent without the dense enameloid and dentinal layers found in the other types. They evolved from ganoid scales to a partially calcified

composition and are of two types: ctenoid and cycloid [107]. They consist of hydroxyapatite (HA) (mineral component), type I collagen (organic component) and water. They are usually in stratified layers consisting of an external layer that is composed of calcium-deficient apatite or calcium carbonate [108] and an internal layer that is composed of minerals and type I collagen fibers organized in unique plies [107]. The collagen fibers (assembly of fibrils roughly 100 nm in diameter, which have a diameter approximately 1 μm) are coaligned in each layer and rotates by 36° in successive layers [109].

Babu *et al.* [110] used fish scales (10-40 wt%) to reinforce epoxy resin [110]. The incorporation of the fish scale filler was reported to significantly enhanced the tensile and flexural properties of the epoxy composites up to 30 wt%. However, the authors did not report the actual value obtained for the neat epoxy resin in their study. Bezerra *et al.* [111] investigated energy absorption and ballistic performance of epoxy composite reinforced with Arapaima scales [111]. Composite plates with varying volume fractions of arapaima scales (20, 30 and 40 vol.%) were tested, revealing an increase in energy absorption with higher scale content. The optimal volume fraction for enhanced energy absorption was found to be 30%. The increase in volume fraction from 20 to 40%, resulted in a notable improvement in impact strength from 29.75 to 292.36 J/m. This establishes a clear correlation between the quantity of reinforcement and the absorbed energy. Notably, in stand-alone tests, composite variants featuring 30 vol% of arapaima scales demonstrated the highest average absorbed energy, especially when subjected to medium velocity (0.22 caliber) and high velocity (7.62 mm caliber) ammunitions. The study concludes that arapaima scales have the potential to serve as effective reinforcement in polymeric composites, particularly in applications requiring energy absorption. Sekaran *et al.* [112] focused on enhancing polymer plastics properties by incorporating varying proportions of fish scale fiber powder (10-40 wt%) to prepare epoxy-based composites [112]. The composites were subjected to shear stress to analyze fracture behavior. The composite exhibits superior strengths, with a tensile strength of 24.2 N/mm² at 40%, a flexural strength of 63.6 N/mm² at 30%, and an impact strength of 5.5 J at 25%. The study emphasizes achieving a uniform distribution of fish scale in the epoxy matrix to enhance material structure and improve bonding strength between the fiber and matrix.

Generally, the mechanical properties of fish scales decrease with an increase in water content [113-115]. Besides, Young's modulus was found to vary across the scale of fish such as *Polypterus senegalus* and *Arapaima gigas* [116,117]. Fish scales have also been reported to have good puncture resistance, toughness, tensile properties, and exhibit anisotropic behavior due to their

composition and microstructure [114,118-120]. For example, the collagenous layer provides flexibility and can undergo substantial elastic deformation before failure thereby providing good toughness [118]. Fish scale fiber is mostly used as filler in polymer-based composites without any form of pre-treatment [121,122]. For example, in a study by Satapathy *et al.* [121] untreated dried fish scales measuring 6-8 mm in length and 1 mm width were used as reinforcement for epoxy resin (Epoxy LY556) [121]. Consequently, there may be need to explore how some initial treatments can improve their use as filler in polymeric composites. Presently, their usage in the development of biocomposites is yet to be fully explored.

2.1.6 Sinews

Sinews are tough fibrous tissue also known as tendon fibers. They are composed mainly of tenocytes, which has abundant densely packed collagen fibers, arranged parallel to each other into fascicles that contain collagen fibrils and elastic fibers, as shown in Fig. 14 [123]. The fascicles are linked together by a dense sheath of irregular connective tissue called epitenon. This material has unique properties that make it suitable for use as filler for the development of bio-based polymeric composites. They have a viscoelastic characteristic, are highly anisotropic and exhibit negative Poisson's ratio (became thicker when stretched) in some planes when stretched up to 2% in their length direction [124]. The latter property is the characteristic of materials with high-energy absorption and fracture resistance. Positional tendons are less elastic and fail under 6-8% strains and have moduli in the 700-1000 MPa region [39]. However, energy-storing tendons fail at 12-15% strains in the 100-150 MPa stress region, except for some tendons such as horse digital flexor [40]. The mechanical behavior of sinew depends significantly on the orientation and diameter of the collagen fiber component. Collagen is a family of proteins present in the extracellular matrices (60-85% collagen), which maintains the structural integrity of vertebrates and other multicellular organisms [125,126]. Despite the diverse applications and capabilities of sinew, it remains largely uncharted territory in the context of polymer reinforcement. Sinew, with its inherent strength and unique structural properties, presents an exciting opportunity for enhancing the mechanical properties of polymers. More so, considering the growing demand for advanced materials with improved strength and durability, delving into the use of sinew in polymer reinforcement holds promise for innovation.

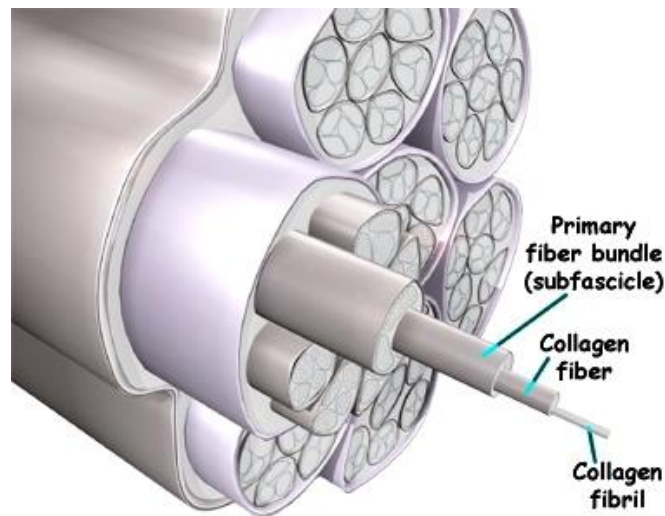


Fig. 4. Sinews structure showing collagen fiber and fibril [123].

2.1.7 Skins

Animal skin can be a good fiber source for the reinforcement of biobased polymers. Generally, there are primarily three types of skin: glabrous (in palms and soles), hairy (most of the human body) and mucocutaneous (orifices) [127]. The skin is composed of the epidermis (outer layer), the dermis (middle layer), and the subcutaneous fat tissues (inner layer), as shown in Fig. 4. Each layer has its mechanical responses although, the skin behaves as a homogeneous elastic material that can stretch and contract [128]. Skins exhibit anisotropic characteristic with its ability to stretch or contract in different directions, depending on the fiber position.

The outer layer, which is about 0.05 mm to 1.5 mm thick [35] can be subdivided into stratum corneum, granular cell layer, prickle cell layer and basal cell layer [129]. The stratum corneum acts as a barrier to water, viral and bacterial. About 95% of the epidermis is composed of keratinocyte [129]. The dermis tissues, which is about 0.6 mm to 3.0 mm thick, consists of the papillary layer, subpapillary layer and reticular layer. They are located under the epidermis and separated by the basement membrane. They are the most important mechanical and thermal component of the skin [35]. They are composed of the extracellular matrix, which has collagen as its main component in addition to elastin fiber, proteoglycan (hyaluronic acid, chondroitin sulphate). Collagen (at least sixteen types [128]), which constitutes about 70% of the dermis dry weight and accounts for about 33% of proteins in the human body provides firmness to the skin. It has a Young's modulus value of 1-10 GPa [35]. The increasing tension of collagen fibers is directly proportional to its stiffness as the fibers become uncrimped, producing an overall non-

linear response [35,130]. Elastin fiber, which accounts for 1-4% of the dermis dry weight is responsible for skin elasticity and resilience [35,129]. The highly stretchable elastin fibril is made from tropoelastin molecule with chains that are crosslinked in quadruplets [131]. Its degradation usually occurs between ages 30-70 years and become profound above 70 years [35,132]. Proteoglycan colloidal gel provides moisture to the skin. The subcutaneous tissues (hypodermis), which are located under the dermis play the role of energy storage, act as a shock absorber and provide thermal insulation [129,133]. Like sinews, skins are yet to be really explored as a potential material for the development of fiber reinforced composites.

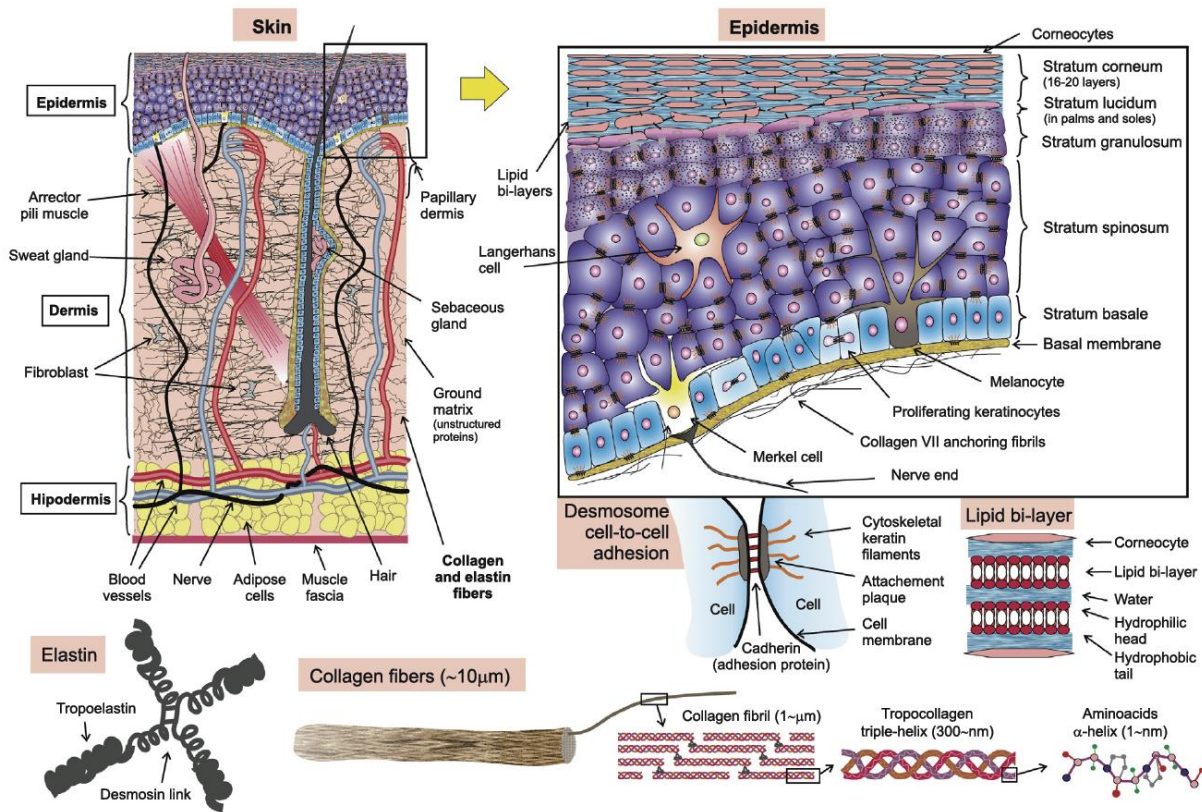


Fig. 5. Skin structures showing various biocomponents [35,129]

2.2 Particulate fillers

This section focused on animal-based materials that can be used in particulate form as reinforcers in the development of biobased polymer matrix composites. Particulate fillers are powdered materials with particles sizes that are usually less than 100 µm and a low aspect ratio [134]. Particle size, size distribution, specific surface area are important parameters that affect the

properties of particle reinforced polymeric composites. The term particle refers to the smallest unit that the filler is made up of and differs from aggregate and agglomerate [134].

2.2.1 *Bones*

Bone is an intercellular composite consists of both organic and inorganic materials. The organic components of bone include collagen, which is a fibrous protein arranged in bundles or long strands and small amounts of glycosaminoglycans and protein polysaccharides that are dispersed within and around the collagen fibers. These materials represent 50% of the volume and 30% of the dry weight of the intercellular composite. The collagen fibrils are highly elastic with substantial intrinsic tensile strength but low compressive strength. The inorganic materials are present as carbonates (calcium carbonate and carbonate apatite) and apatite (mostly hydroxyapatite, $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$) minerals in rod-shaped nanocrystals [135-137]. They are responsible for bone hardness, high compressive strength, and good rigidity at the expense of tensile strength as they allow stress concentration around defects and their propagation. Consequently, bone overall behavior and properties depend on the combined effect of collagen and the minerals component hierarchical structures organized at nanoscale level. This precise organization enables it to exhibit some degree of elasticity and nanomechanical heterogeneities necessary to withstand impact [135]. Besides the mineral (65 wt%) and organic constituents of bone, it contains about 10 wt.% water, which is present in mineralized and unmineralized bone and contributes to its structural and mechanical properties [135,136,138,139].

The benefits of bones have been explored in the production of polymer matrix composites. The incorporation of particles cow bones (106 μm) to polyethylene was found to have significant effects on wear behavior and mechanical properties of the developed composites [140]. The reinforced polymer composites produced from recycled polyethylene with up to 25 wt.% filler material showed improved tensile strength corresponding to about 300% increase. However, toughness decreased with increasing filler loading from 55 MPa for the neat polymer to about 31 MPa for sample containing 25 wt.% filler. In an experimental study, brake pads were produced by incorporating 30 wt.% of cow bone particles at different particle sizes (120 and ≤ 100) into epoxy resin containing palm kernel shell particles [122]. The material prepared with the 120-grade size exhibited optimum performance. The impact energy, crushing strength, hardness, water absorption, and oil absorption of the tested samples revealed values of 1.5J, 21 MPa, 46.0 HRB,

5.53%, and 4.16%, respectively. Thermogravimetric analysis (TGA) indicated the highest percentage weight loss as 63.24%, while the coefficient of friction and wear rate were 0.677 and 1.44×10^{-7} , respectively. The authors concluded that these values fell within an acceptable range for the brake pad's function. Carbonized and uncarbonized bone particles (5-25 wt%, 100 μm) have also been used to reinforce polypropylene [141]. The composites containing carbonized bone particles reinforcement presented superior properties than uncarbonized ones with respect to the compressive strength, hardness values, tensile strength and flexural strength corresponding to 53, 45 and 35%, respectively. For example, the compressive strength, hardness and tensile strength of sample containing 15 wt.% carbonized cow bone were 150 MPa, 70 HRF and 32 MPa, respectively. However, the percentage elongation and impact energy decreased by 75 and 32%, respectively, for the samples produced with 25 wt.% carbonized bone. Moreover, both the carbonized and uncarbonized filler material exhibited better properties than the neat polymer, which increases with increasing filler contents. The hardness of the material containing 25 wt.% was about 90 and 70 HRF for the sample containing carbonized and uncarbonized cow bone particles respectively, when compared with a value of 50 HRF for the unreinforced polymer. The improved properties (strength and hardness) were attributed to the distribution of hard phases of bone particles in the ductile polypropylene matrix.

2.2.2 *Claws*

Most mammals, birds and reptiles are sources of claws, which are usually present at the end of their toes or fingers for purposes such as catching and holding preys, climbing, digging, and defense. Avian or β -keratin claws have ultrastructure and composition similar to hair, feather and beak [29,142]. However, claw's molecular weight (13000 g/mol) is lower than that of beak (14500 g/mol) but higher than feather's (10500 g/mol) [143]. At different humidity levels, claws keratin is less stiff when compared with those from feathers due to the differences in their molecular orientation [34]. The structure and mechanical properties of claws usually depend on their composition, the amount of intercellular cementing substance and the arrangement of horn tubules, inter-tubular space keratin and filament-associated proteins [144,145]. Their mechanical properties are also a function of the animal species from which the claws are extracted [142]. Their quality and structural integrity depend on appropriate nutrients (minerals, vitamins, amino acids and fatty acids) and oxygen supply to the keratin-producing epidermal cells [144,146]. The claw size and shape affect the shock-absorbing characteristic associated with weight and movement

distribution [145]. Increasing hydration was found to decrease the modulus and strength of ostrich claw, which exhibit isotropic characteristic relative to its rigidity under compressive and tensile tests [34].

2.2.3 Beaks

Beaks (Fig. 5) is another potential material that can be used in preparing biobased particulate filler for development of polymer matrix composites. They are remarkable structural avian material that is composed of hard keratins (β -keratin layer), which are very rich in glycine and moderate levels of tyrosine and serine, inner bony layer, and cellular interface [147-149]. The keratins found in beaks are different from those of feathers, but like reptiles' keratins [148]. Besides, biological HA such as those found in chicken beaks can be extracted from them [150]. Generally, beaks consist of brittle minerals and fibrous protein. These materials are arranged hierarchically, interacting with each other [149]. Birds' beaks or rhamphotheca (keratinized sheath covering the upper and lower beaks) have good mechanical properties, which are suitable for use as a filler because of their low weight. For example, Young's modulus, stress plateau under compression and the mean nano hardness of toucan's rhamphotheca are approximately 1 GPa, 0.3 MPa and 0.5 GPa, respectively [147]. The density of toucan and hornbill beaks is 0.1 g/cm^3 and 0.3 g/cm^3 , respectively with mesostructure and microstructure that resembles sandwich structures of functionally graded materials (Fig. 6) [29].



Fig. 6. Chicken image showing beaks that can be used as filler for polymer composites.

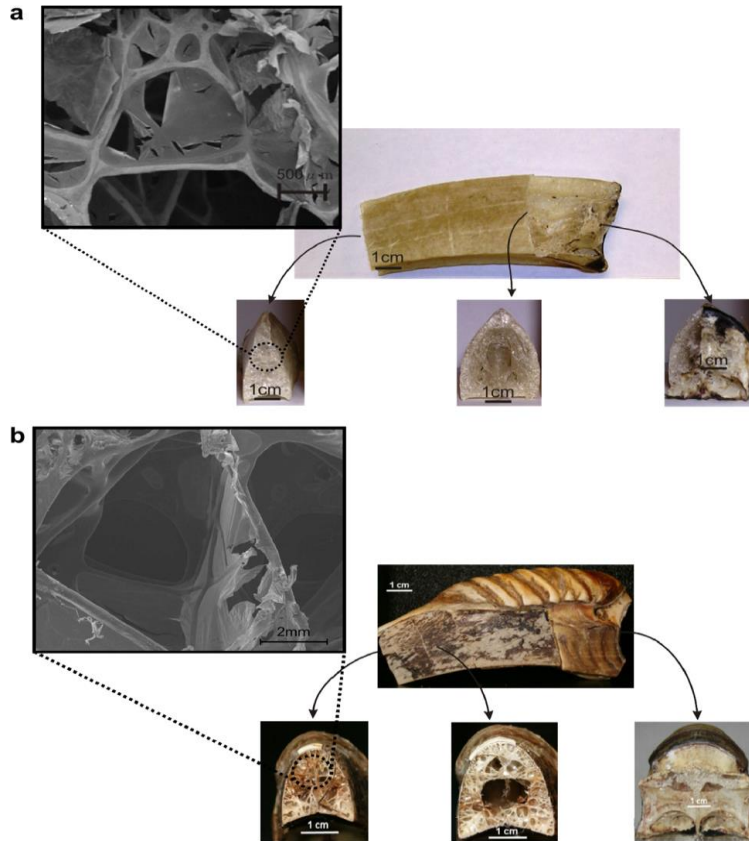


Fig. 7. SEM showing internal foam structure with three cross-sections of (a) toucan foam and (b) hornbill foam that resembles sandwich structures of functionally graded materials [29].

2.2.4 Horns

The world consumed about 60 million metric tons of beef annually. However, there is no reliable data on the specific amount of horns waste generated per year. Nevertheless, a large quantity of horns waste can be assumed based on the extent of beef consumption. Horns producing animals may be classified as bovid and antlers. Bovid horns are permanent, unlike antlers that are shed and regrown each year [151]. They are composed of a keratinous sheath (α -keratin) overlying a bony core (Fig. 7) without a mineralized component when compared with other structural biological materials, such as tusks, bones, and teeth [151,152]. Horns are produced through keratinization and subsequent cornification process. The hard keratin outside layer has a laminae structure arranged in a rippled tabular shape [153]. The lamellae flat sheets are held together by proteinaceous substances. There are primarily hydrogen and disulphide bonds between the keratin fibers and protein matrix as well as ionic interactions and van der Waals bonds, which are

intermolecular cross-links. These bonds play significant roles in the mechanical properties of this material [154]. Generally, horns have good toughness, stiffness, and strength to prevent breakage [151,152,155,156] and these properties depend on horns' water content [153,157]. Water molecules affect keratin matrix by acting as a swelling agent, replacing the secondary hydrogen bond and binding with the proteins matrix as a plasticizer [158]. These reduce the interaction between chains and increase their mobility, which decreases the horn's stiffness [158,159]. As a major defense weapon in bovid animals, it can also bear static and dynamic load [153]. The major differences in their microstructure relative to different species are in the number and shape of the tubules, the orientation of aligned lamellae and the keratin cells shape [159]. Pronghorn horn has also been reported to have the largest water-absorbing ability due to the presence of nanopores in their keratin cells [159].

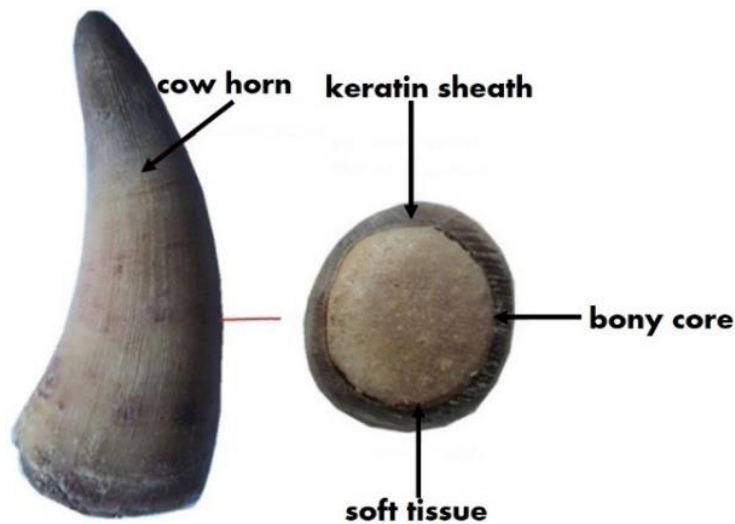


Fig. 8. Structure of cattle horn showing bony core, soft tissue and keratin sheath [152].

Composites produced with this material can have low density and favorable properties, which can hold a potential for applications in various fields such as automobiles, computers, construction, houseware, and more. For example, Kumar and Rajendra, 2014 developed a particle-reinforced composite by combining **500 μm particulate sized** defatted horn fiber (HF) and PP [160]. The physical, mechanical, thermal and microstructural attributes of HF/PP composites, featuring varying fiber weight percentages of 5, 10, 15 and 20% were characterized and compared with the properties of pure PP and pure HF. The mechanical properties of HF were found to be higher than those of both neat PP and HF/PP composites. In contrast to pure PP, HF/PP composites exhibited

a slight increase in tensile yield strength, a 15.74% rise in tensile modulus, a 16.95% improvement in flexural strength, and an impressive 59.69% improvement in flexural modulus. However, there was a 15.03% decrease in ultimate tensile strength, as well as notable reductions in percentage elongation at break and impact strength. The melt flow index for HF/PP composites decreased with an increase in fiber content. Thermogravimetric analysis indicated enhanced thermal stability in HF/PP composites with higher fiber content. The SEM micrographs also show that good compatibility existed between HF particles and PP. Among the HF/PP composites, the one with 15 wt.% of HF particles exhibited optimal results. Horn has also been combined with thermosetting resin. Kumar *et al.* (2017) investigated the mechanical properties of horn powder-filled epoxy composites [161]. The fabrication process involves a Taguchi L₉ (3×3) orthogonal array, with HP particles (125, 250 and 425 μm) and the matrix mixed in specific ratios, compression-molded, and cured at room temperature. Key properties, including ultimate tensile strength, tensile modulus, elongation percentage at break, flexural strength, flexural modulus, impact strength, and microstructure, were analyzed. SEM micrographs revealed good compatibility between HP particles and resin at 10% HP volume, with increased agglomeration at higher volume percentages. Grey relational analysis (GRA) and analysis of variance (ANOVA) were employed for optimization, determining the highest NaOH concentration (0.3 N), the smallest HP size (125 μm), and the lowest HP volume percentage (10%) as the optimum factors. The main contributors to property improvement were found to be HP volume percentage (90.87%), HP size (6.43%), and NaOH concentration (0.78%). The high R squared values (>95%) confirm a 95% confidence level. A comparison between the optimum specimen and untreated HP specimens was found to also showed superior properties in the optimum specimen.

2.2.5 Shells

Shells are exoskeletons of mollusks such as snails, clams, oysters, which are made up of three distinct layers consisting of outer proteinaceous periosteum (uncalcified), a prismatic layer (calcified) and an inner pearly layer of nacre (calcified). An exception to this description are turtles, which are classified as vertebrate animals with endoskeletons.

Shells are composed mostly of calcium carbonate (ceramic phase) and about 0.1-5% protein [29]. Due to inherent brittleness, the ceramic phase is not suitable for structural applications. However, as a natural structure, shells have unique mechanical properties due to their microstructural and macrostructural arrangement, which increases the toughness of what would

essentially have been brittle ceramic matrix. Mollusk shell structures are classified into nacreous (flat tablets), crossed lamellar (plywood-like), prismatic (polygonal columns), foliated (long thin crystals in overlapping layers) and homogeneous (fine-scale rubble) types. The first two types were discussed below.

The nacreous shells (Fig. 8) have an outer prismatic layer (calcite- rhombohedral) and an inner nacreous layer (aragonite- orthorhombic) [162]. The mesolayers of thickness of 300 μm , which are created because of periodic growth and separated from the inorganic phase by layers of viscoplastic material thickness of 20 μm usually influence the shell mechanical properties [163]. The fracture strength in bending of nacre from mollusks such as bivalves, gastropods and cephalopods was found to vary between 56 to 116 MPa [164]. Also, Jackson *et al.* (1988) reported a work of fracture varying from 350 to 1240 J/m² as a function of span-to-depth ratio and hydration degree using nacre from *Pinctada* [165]. The authors found that wet samples showed superior toughness, howbeit inferior tensile strength (approximately 140 MPa when compared with 170 MPa obtained for dry sample) and Young's modulus (approximately 60 MPa when compared with 70 MPa obtained for dry sample). They attributed these to plasticization and reduction of the shear modulus and shear strength of the organic phase (which constitute about 5 wt%), respectively. Several other authors [166-171] have equally investigated the mechanical properties of nacre shells and proposed models relative to improving or predicting these properties [172-175].

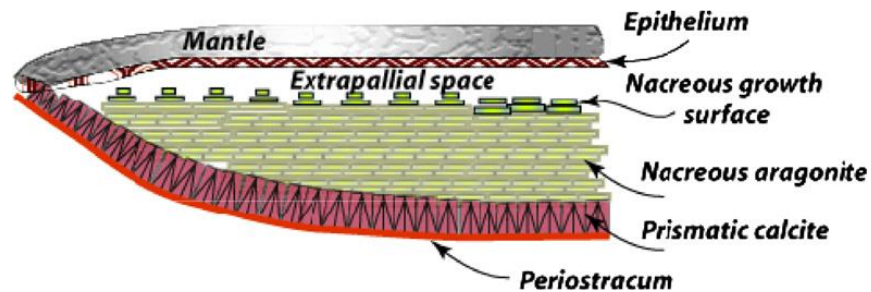


Fig. 9. Abalone shell structure showing its various components [176].

The crossed lamellar structure consists of 99.9 wt.-% lath-like aragonite crystals and 0.1 wt.-% tenuous organic layer, as shown in Fig. 9. The three layers are arranged in 0°/90°/0° direction and oriented at $\pm 35\text{--}45^\circ$ relative to each other [29]. Kuhn-Spearing *et al.* [117] investigated into the apparent flexural strength, work of fracture and crack-density evolution for wet and dry crossed lamellar *Strombus gigas* conch shell samples [177]. In the parallel orientation, the apparent flexural

strength of the wet and dry specimens were 156 ± 22 and 84 ± 49 MPa respectively, while it was 107 ± 38 MPa in the perpendicular direction. However, the disparity in the attained results is very significant. These values are within the range (70-200 MPa) of those obtained by Curry and Kohn [178] in a study involving crossed-lamellar crystal architecture of many kinds of mollusks. The fracture work (13 ± 7 J/m²) of the wet samples was found to be higher than the dry samples (4 ± 2 J/m²) due to decreased interfacial strength resulting to a more extensive cracking pattern. Crossed lamellar shells have higher toughness when compared with the nacreous structures [165] and can exceed that of the single crystal pure mineral [179]. Generally, as with nacre shell, structural hierarchies (complex layered architecture) are responsible for their mechanical responses.

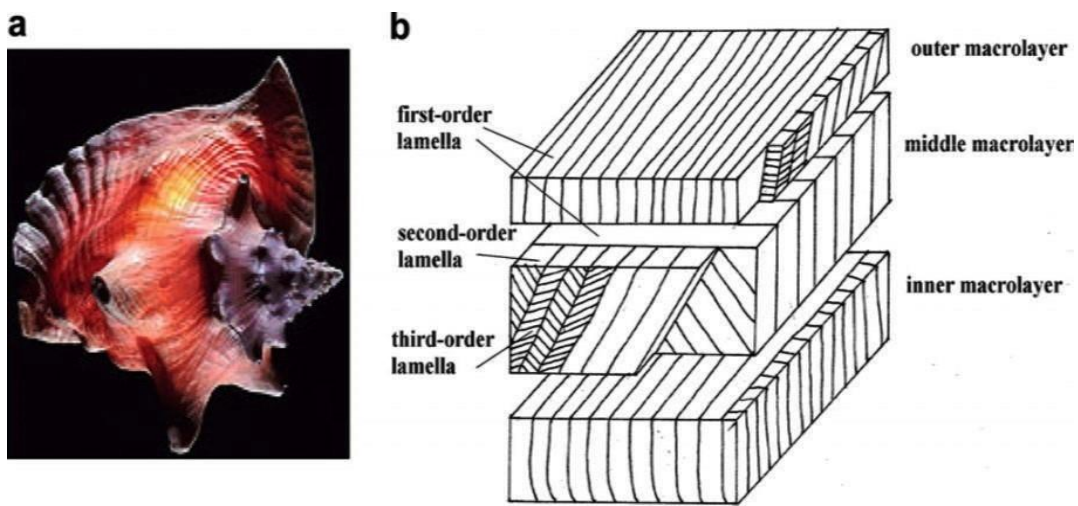


Fig. 10. (a) Overview and (b) schematic representation of crossed lamellar shell structure [29,180].

2.2.6 Teeth

Teeth have good hardness, fracture toughness, viscoelasticity, elasticity and radiodensity arising from the structural arrangement of the HA needles and its composition [181]. Although damage may be activated within the brittle enamel coat; however, its catastrophic spreading to the vulnerable tooth interior is prevented under sustained occlusal loading [182]. The entire structure is designed to possess an external hard layer with a high degree of mineralization and a tough internal core (dentin) [29]. Teeth consist of two parts, which are the crown that protrudes out of the jaw and the root that is usually buried inside the bony socket of the jaw [183]. Mammals' teeth are divided into four basic types, which are incisors, canines, premolars, and molars, which varied in both size and shape [184]. Si, P, Ca, Ti, Mn, Fe, Ag, Cd, Sn and Sb are elements commonly found in animals' teeth [185]. Bovine teeth have a higher number of dentin tubules next to the pulp

than human teeth with a smaller number of dentin tubules next to the enamel [186]. Bovine dentin tubules concentration per square millimeter is also slightly higher than that of human [186]. Nevertheless, mammalian teeth are essentially the same anatomically and histochemically although there are differences in the organic and inorganic content of ovine, porcine bovine, and human enamel and dentin [187,188]. The human tooth can be divided into the outer enamel, dentin, cementum, and dental pulp (Fig. 10) [181]. The initial three components represent the hard tissue of the tooth with unique and good mechanical properties. The enamel rod microstructure revealed a 'keyhole-like' structure, approximately 5 μm diameter, which is positioned perpendicular to the dentinal-enamel junction at sharp angles [181]. The micrograph of the dentin is characterized with dentinal tubules that radiate outward through the dentin while cementum showed a structure like that of bone tissue [181].

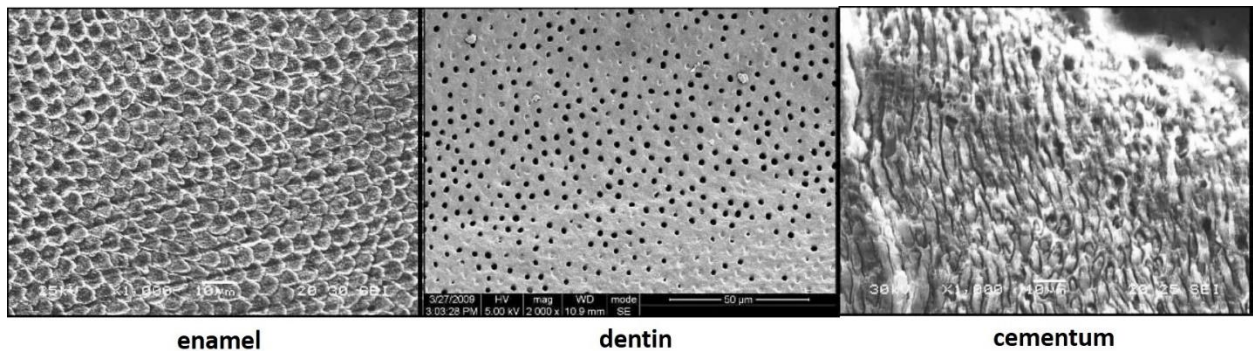


Fig. 11. Microstructure of enamel, dentin, and cementum components of human tooth [181].

The root part of most animals' teeth is coated with a layer of bone-like tissue called cement [183]. The cementum is made up of calcium ion in the form of apatites (45-50%), collagen and non-collagen proteins. It has a hardness value that is lower than dentin. It exists as cellular-cementum, acellular-cementum with a gradient structure that contributes to the anisotropic mechanical properties of teeth [181].

The dentin is a bone-like material composed of protein (mainly collagen), 70% HA and water. It is a very tough, resilient tissue [183]. The microstructure revealed dentinal tubules that point externally from the pulp to the cementum or enamel border [181]. The wall thickness, quantity and size of dentinal tubules varied from outside to inside. The dentin tensile strength, fracture toughness, hardness and Young's modulus are 70-80 MPa, 1-2 MPam^{1/2}, 0.5 GPa and 13-17 GPa (parallel to the tubules), respectively [29].

The enamel, which is the inorganic and mineral phase of human teeth is composed of 97% HA, $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ and is the hardest part of vertebrate body [189]. HA has a hexagonal crystallographic structure with a space group, P63/m which consisting of isolated PO_4^{3-} tetrahedra that are connected by distorted octahedra network and twisted trigonal prisms [188]. The HA rods ($\sim 5 \mu\text{m}$) are arranged in lamellae (a complex hierarchical organized microstructure) and bound to collagen. Its surface is usually covered by pellicle, which contains salivary proteins, carbohydrates, and lipids. The hardness and elastic modulus of the tooth enamel decreases with depth towards the dentin enamel junction as a function of calcium content and direction of the enamel rods exhibiting anisotropic mechanical properties [29]. The enamel has fracture toughness, hardness, and Young's modulus of 0.7-1.3 $\text{MPa}\cdot\text{m}^{1/2}$, 4 GPa and 60 GPa, respectively [29].

In contrast to the bone, enamel and dentin do not have a vascular structure, they are rather static and cannot be-repaired nor remodel [29].

2.2.7 Hoofs, sole and heel

Hoofs, sole and heel are made up of soft keratins composed of α -helical protein bundles [29,146]. Equine hoof wall consists of two structural elements in the form of tubules and intertubular material as well as some traces of cysteine conjugates, which can be found in their hydrolysates [142]. The tubules and direction of the intermediate filaments (a fibrous structure of keratin) control the crack propagation and enhance its fracture toughness [29]. Bovine hoof keratin behaves as hydrophilic gel with moisture-absorbing ability, which can increase its wearing rate by abrasion [190]. Its pretreatment with aldehydes can reduce moisture uptake by mechanically stabilizing the proteins through chemical fixation [190].

3. Modification of animal-based fillers

To achieve maximum reinforcement efficiency in fiber reinforced polymer composites, an effective stress transfer from the matrix to the fibers is required. This requires a strong adhesion at the fiber-matrix interface. Unlike natural plant fillers, most animal-based fibers do not possess extensive hydroxyl groups and are therefore not as hydrophilic as natural plant fibers. For this reason, a certain level of compatibility with polymeric resins, which are generally hydrophobic, is expected. However, in some cases, animal-based fibers such as keratin fibers contain up to 40% hydrophilic amino acids. To increase their compatibility with thermoplastic polymers, chemical

treatments are necessary. Moreover, for effective processing of some animal-based fibers into mats, treatment with oxidizing agents or enzymes are utilized to overcome the directional frictional effects. Some treatments are also required to remove dirt and foreign materials from the fiber surface before use.

3.1 Chemical Treatment

Treatment with silane coupling agents improves the compatibility between animal-based fillers (such as fibers or particles from animal sources) and polymer matrices [191]. Silane molecules create a chemical bridge between the filler surface and the polymer, enhancing adhesion [192]. Acetylation is another treatment that introduces acetyl groups to the filler surface to improve its compatibility with polar polymer matrices, enhancing the overall interaction and performance of the composite [193]. Alkaline treatments, often involving the use of solutions like sodium hydroxide (NaOH), modify the filler surface by removing impurities and enhancing its reactivity [194]. Alkaline treatment can increase the surface roughness of fillers, leading to improved mechanical interlocking with the polymer matrix. Subjecting biodegradable fiber to hydrolysis procedures can produce more polar hydrophilic groups such as hydroxyl- and carboxyl groups on the fiber surface [22]. Hydrolysis is an irreversible process associated with breaking of polymer chains and reducing bonding force between molecules that result in permanent material degradation [195]. Unlike cellulosic fibers, animal-based fibers are not easily hydrolyzed by chemicals because they contain highly crosslinked disulphide bonds, especially in the scale layers [196]. However, enzymes have been previously employed to modified animal-based fibers [197,198]. These enzymes can attack natural keratin and hydrolyzing some peptide linkages [198]. There is need to control the enzymatic process to avoid significant damage to the animal fiber structure because of the enzyme penetrating and attacking the fiber core [199]. Biosynthesized kerationlytic proteases has been reported as a green and sustainable way to modify the structure of wool via hydrolysis route [200]. Keratinases are important proteases for hydrolyzing feathers, hair, wool, horns and hooves [200-202]. Another method involving micro dissolution of the scale layers of animal fiber such as wool has been developed to cause minimal damage to its internal structure while improving surface-related properties [196]. This method made use of sodium hydrogen sulphite (NaHSO₃)/urea aqueous solution as a dissolving agent. Nevertheless, there is limited information to show that such modified fibers have been blended with polymers to develop polymer matrix composites.

3.2 *Plasma treatment*

Animal fibers such as wool and silk are susceptible to felting presumably because of the presence of scales on the surface of the fibers. To solve this problem plasma treatment has been widely studied to improve interfacial adhesion. Plasma treatment is an attractive environmentally friendly method of surface modification, which involves the exposure of the fiber surface to an electrically charged gas called plasma. Plasma radiations can create a rough surface topology on the fiber, which will increase mechanical interlocks between the fiber and the matrix [22,203]. The process improves the fiber–matrix adhesion by introducing polar or excited groups that create strong covalent bonds between the fiber and the matrix [25]. Plasma treatment offers advantages such as lower material consumption, significant bulk properties retention, lower processing time, zero waste, elimination of the need for further purification or materials drying after processing, minimization of thermal degradation and safety [22,203]. Moreover, oxygen and argon plasma gas treatments have been employed to introduce hydroxyl functional groups on materials surface to improve their interfacial adhesion [22]. The benefits of plasma treatment of animal-based fillers or fibers have been reported in studies such as [204].

3.3 *Blending with Compatibilizers*

The addition of compatibilizing agents, such as maleic anhydride-grafted polymers [63], can enhance the interfacial adhesion between animal-based fillers and polymer matrices, leading to improved mechanical properties. They can function by providing a chemical bridge between the filler and polymer matrix [205]. They also aid in the dispersion of animal-based fillers within the polymer matrix, which prevents filler agglomeration and ensures a more homogeneous distribution [206].

4. Techniques of improving properties of animal-based bio-composites

There are many advantages of using natural fibers, as previously discussed. However, there are some significant drawbacks, such as more absorption rate of moisture wetting properties, which result to a weak fiber-matrix bond, due to the disparity in the density of the materials [195,207,208]. These disadvantages have significant effects on the application of natural fibers in the aircraft industry. Numerous techniques have been developed to enhance the properties of polymer composites [209]. The techniques are highlighted in Fig. 11. The exploration of methods

such as weaving, stitching, braiding, etc., remains relatively limited in the realm of animal fiber-reinforced polymer composites. While these techniques have been widely employed in the fabrication of polymer composites using synthetic fibers, their application to composites reinforced with animal fibers has not been extensively investigated. Nevertheless, exploring these techniques in the context of animal fiber-reinforced polymer composites holds promise for creating composites with tailored properties. Weaving, for example, could provide a controlled arrangement of animal fibers such as silk and wool, enhancing overall strength and durability. Stitching might offer improved interlaminar strength, while braiding could contribute to increased impact resistance.



Fig. 12. Various **potential** techniques of enhancing properties of animal-based composites.

Traditionally, techniques such as weaving, stitching and braiding have predominantly been associated with synthetic fibers, leaving their exploration with animal fibers processing largely overlooked in composite applications. The limited exploration of these techniques in animal fiber composites may be attributed to a combination of factors, including the uniqueness of animal fibers and the need for specialized processing. For instance, some animal fibers might be more susceptible to breakage or require specialized needles or looms for weaving or stitching.

Nevertheless, animal fibers such as wool and silk hold tremendous potential for weaving, stitching or braiding, due to their innate flexibility, fine texture, strength and durability, despite their delicate appearance. Exploring this potential would require substantial research, which can ultimately unlock the full capabilities of animal fiber-reinforced polymer composites. This endeavor may lead to the development of materials with improved performance, environmental sustainability for a broader range of applications in various industries. Consequently, the discussion of these techniques is aimed at highlighting their untapped potential for future utilization in animal-based fiber reinforced polymer composites.

4.1 Braiding

Braiding (Fig. 12) is the process of building binding pattern in a direction like a helix around a tube or in flat form by intertwining three or more yarn systems together. The fibers are manufactured under a certain angle between about 20° and 70° (one direction) with a combination of standard braider or radial braider and mandrel-like handling device [210]. However, when the composites are to be produced in 0° and 90° orientations, additional equipment such as winders (90°), feeders (0°) as well as units for local thickenings are required [211].

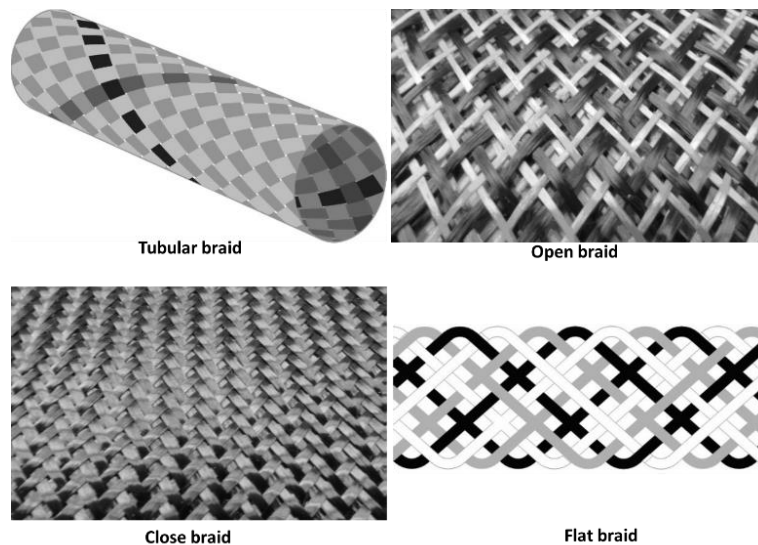


Fig. 13. Images showing different types of braids [210].

Braiding machine consists of a machine bed containing engraved groove that guide the bobbin carriers and a take-up mechanism that retrieve the completed braid, as depicted in Fig. 13 [210]. This preforming technology can be classified as circular braiding (conventional and radial

braiders), over-braiding and 3D braiding, producing either biaxial or triaxial braids based on Hercules, regular or diamond braid architecture [211,212].

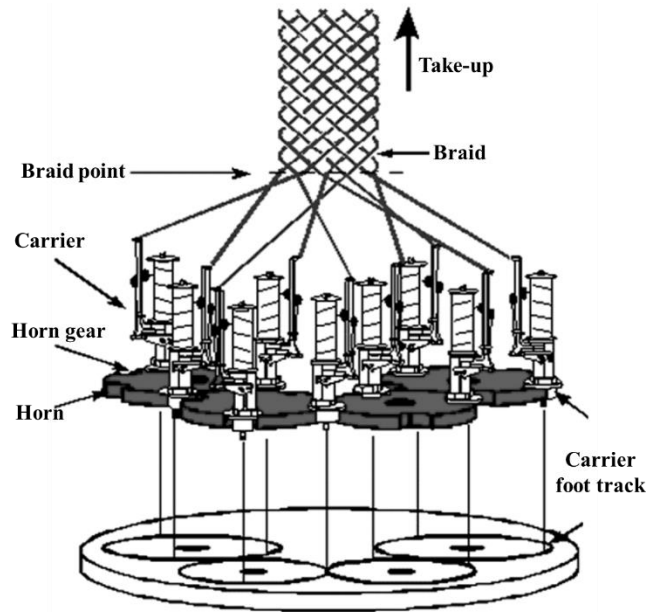


Fig. 14. The elements of a braiding machine [210].

The technique promotes a better interlock between the layers, which can support higher interlaminar shear strength and damage tolerance. It can also facilitate faster infiltration of the viscous polymer systems by injection molding or thermo-compression due to the gaps between yarns and layers [211]. The composites developed with technology involving braiding have unique characteristics such as high shear and torsional strength and stiffness, dimensional stability, and near net shape manufacturing capabilities, damage tolerance in static or low/high velocity, increased transverse moduli and transverse strength [212].

In addition, braiding is a versatile and cost-efficient process to produce fiber preforms for composites used as crash absorbers in sport cars, propeller blades in aircraft and for structural aircraft parts [210]. The process produces continuous, intertwined fiber tows as reinforcing braid architecture [213]. Nevertheless, it is difficult to find comprehensive calculation model to enable products criterion to be made based on materials qualification. Braiding technique is used to improve the fiber-matrix reinforcement. As the matrix and the reinforcement fibers are wound as one, matrix impregnation is more, and the fiber dispersion is also better. The tensile strength also increases. Additionally, a less energy is consumed as the thermal degradation of the fiber-matrix was prevented [214].

4.2 Knitting

Knitting (Fig. 14) is an effective technique during to automatically manufacture net-shape preforms fibers by reducing the time spent to lay up preform fiber in mold before casting. This technology facilitates development of intermeshed fiber yarns loops with better deformation ability compared with woven fabrics [215]. The method is suitable to produce thermoplastic [216], elastomeric [217] and thermosetting resin [215] composites. Depending on the knitting direction of two consecutive loops of the same yarn, knitting can be classified as warp and weft (flat or circular). The warp knitting process usually involves the same number of yarns and wales while weft knitting one requires a single yarn or more, knitted successive needles direction [211]. A wale is a set of stitches made by a single needle, aligned in the warp direction. Knits are suitable for processes such as liquid composite molding technique involving impregnation of the filler by a resin flow due to their high in-plane permeability [211]. The technique also offers the advantage of reduction in production time by avoiding cutting operation. The challenges with this process involved preventing rupture during knitting of large amount of fiber and the lower mechanical properties when compared with composite manufactured via woven technology [211].

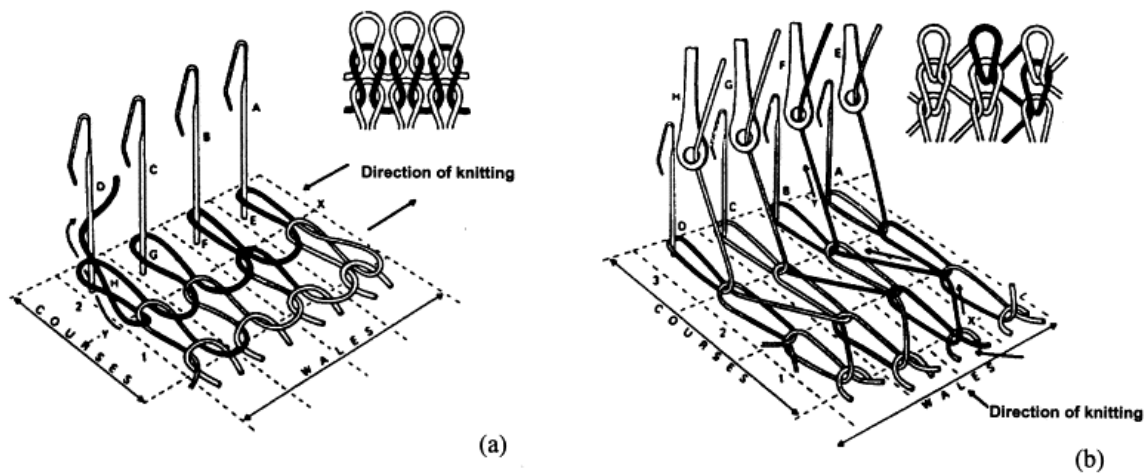


Fig. 15. Illustrations displaying the (a) weft and (b) warp knitting process of a knitted fabric [218].

4.3 Weaving

Weaving (Fig. 15) is a fiber preforming technique that involved warp yarns in the fabric direction interlacing with weft yarns at 90° [219]. The technique produces the majority of single layer, broadcloth fiber used as reinforcement in composites manufacturing and account for

significant amount of 2D fabric [219,220]. Although weaving is suitable for flat panels and woven fabric textiles, the resulting composites have poor impact resistance and delamination strength [220]. Nevertheless, these properties can be improved by angle-interlock weaves that weave adjacent fabric layers with fibers or weaved together all fabric layers. The former process produces layer-to-layer interlocks while the later results in through-the-thickness interlocks.

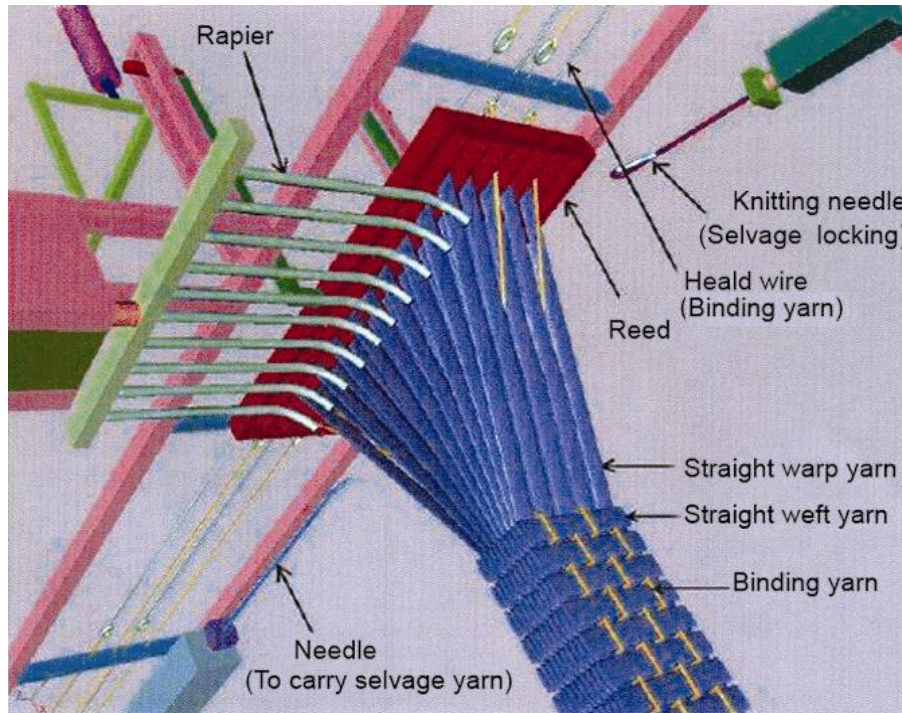


Fig. 16. Conceptual design of a mechatronic 3D weaving machine developed at the University of Manchester [221].

The technology offers the advantage of good drapability and production of complex shape with no gap at a low cost [219]. The process is generally designated as conventional 2D weaving that uses pile warp, ground warp and pile weft sets of yarn and 3D weaving that produces fully interlaced 3D fabric where all the three sets of yarns are interlaced. 3D weaving composites have good resistance to interlaminar shear, low shear stiffness and provide the structure with good formability [222]. Nevertheless, several specialized weaving machines that manufacture multidirectional preforms have been developed. Weaving machines can be used to produce different pattern (Fig. 16), such as plain-weave that resists shear deformation, satin weave that exhibits minimum interlacing and reduce resistance to distortion, basket weave with improved drapeability (when compared with plain weave) and twill weave that is easier to wet out with better

mechanical properties than plain woven fabrics [219]. The image of woven fabrics is shown in Fig. 17.

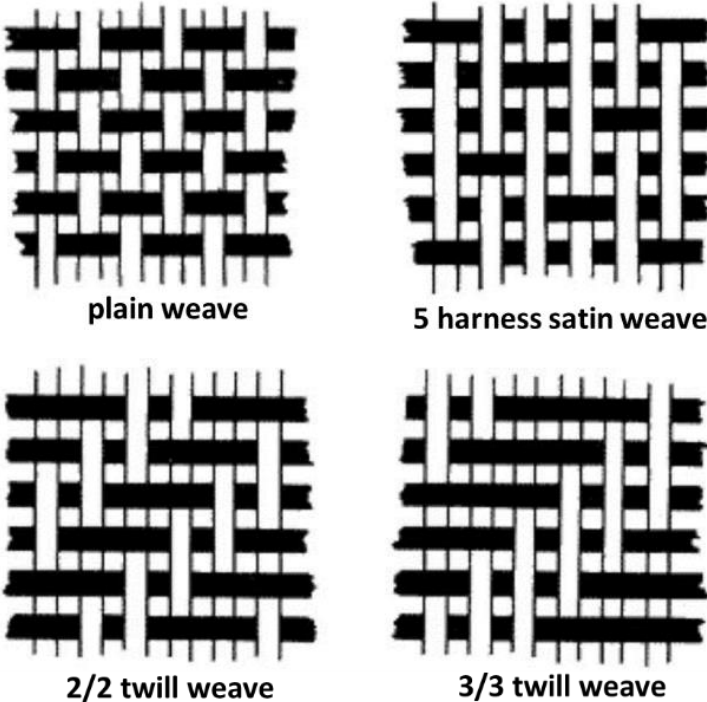


Fig. 17. Description of different types of biaxial woven constructions [219].

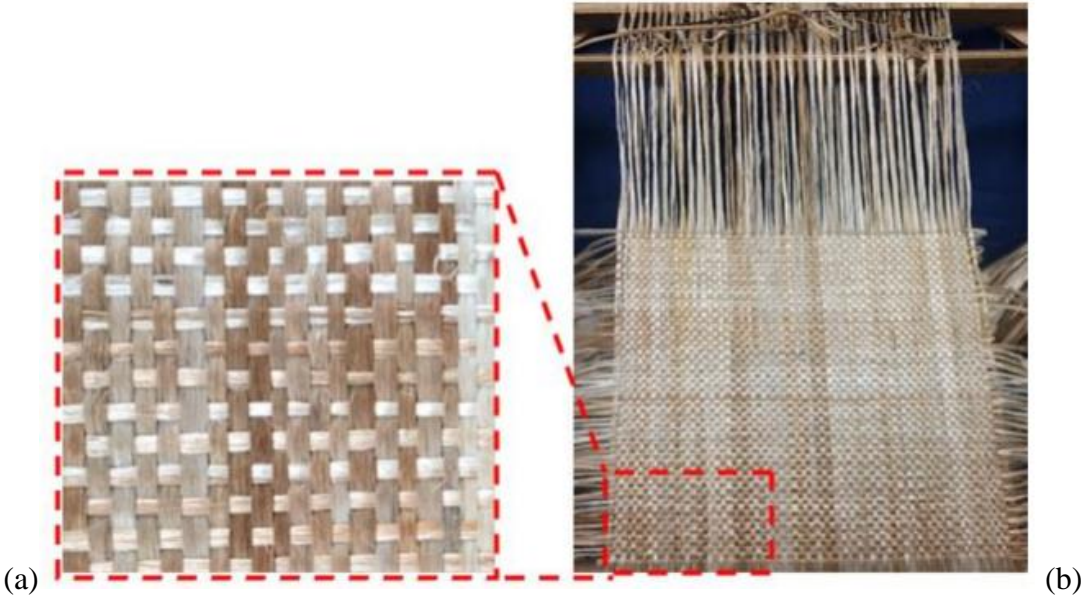


Fig. 18. (a) Woven fabric and (b) weaving process [223].

In 3D weaving, comparatively straight warp, and weft towers, which are usually limited to a 0°/90° alignment, low tensioned binder yarns are interlocked and can be introduced at a variable angle by the thickness [224]. Woven fiber composites show better mechanical, dynamic, and free vibration responses. There is an efficient transfer of stress from the matrix to the fiber, due to the presence of a strong fiber-matrix interface [225,226].

Hamidi *et al.* [227] employed a woven silk to produce epoxy-based composites laminates [227]. The composites exhibited a 23% improvement in specific flexural strength along the primary weave orientation in contrast to a reference glass/epoxy laminate. Moreover, the introduction of 300 kPa external pressure after resin infusion was found to possess the capacity to elevate the silk/epoxy interface, leading to a conspicuous upsurge in both breaking energy and interlaminar shear strength. For instance, the energy needed to fracture silk/epoxy laminates in the x direction witnessed a rise of approximately 17% from around 12 to about 14 MJ/m³ following the application of external pressure for the slow-filled silk/epoxy laminates infused along the longitudinal direction.

4.4 *Stitching*

Stitching is a method for bonding reinforcements to form several plies at the same time, especially non-crimp fabrics (NCF) preforms, which are unidirectional fibers with integrity and ease of handling. This approach creates what is commonly referred to as stitched tailored reinforcements, or stitched layups, which are ready-to-use batches of stacked plies joined with a seam [211]. In composites manufacturing, stitching technology offers advantages such as a variable selection of stitch types and sewing threads, waste reduction, manufacturing of complex components, pre-compaction of preforms and improved 3D properties [228]. It can be used for positioning, assembly, or structural purposes. Challenges such as void formation, fiber separation, formation of resin-rich zone are associated with the structural stitching [228]. There are different types of stitching, which include lock stitch, chain stitch, blind stitch, one-side stitch, and tufting. Mitschang [228] elaborates on the relationship between the stitching types in tabular format, as presented in Table 2.

Table 2. Stitch types and their influence on preforms characteristics [228].

Stitch type	Thread consumption	Handling	Fiber disorientation	Compression
Lock type	Hybridization and high efficiency possible High thread tensile force	Good shear strength of layer package Low tendency to warp	Different fiber displacement on the upper or lower surface of the material High thread tensile forces	Exactly adjustable Low fiber material content in the laminate
Blind stitch	Low efficiency Large thread demand No bobbin thread necessary	Lower layers are not fixed Possible stitching in a solid tool	Lower single layers are not influenced by the stitching Local thread concentration and high shear strength	Compaction of the layers not adjustable Slight deviation of needle possible due to pre-compaction

Stitching is a procedure that uses a needle and thread to fasten or fuse two parts. The parameters such as density and orientation of the stitch play a significant role in determining the overall mechanical property of the stitched biocomposite. Fig. 18 shows the basic mechanism of the stitching process [223].

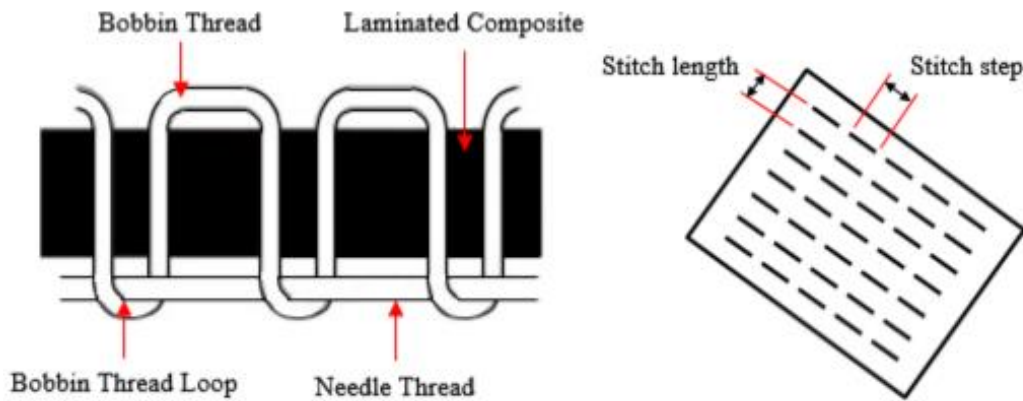


Fig. 19. Stitching process showing modified lock stitch and its application on woven fiber [223].

The stitching process involves inserting a stitching thread with the help of a needle in a laminated composite to create a 3D structure, as shown in Fig. 19. The stitching density can be calculated, using Eq. (1).

$$SD = \frac{1}{Sr * Sl} \quad [1]$$

where SD = stitch density,

Sr = distance between one stitching row and the other,

Sl = distance between two needle insertion points in the stitching row [209] .

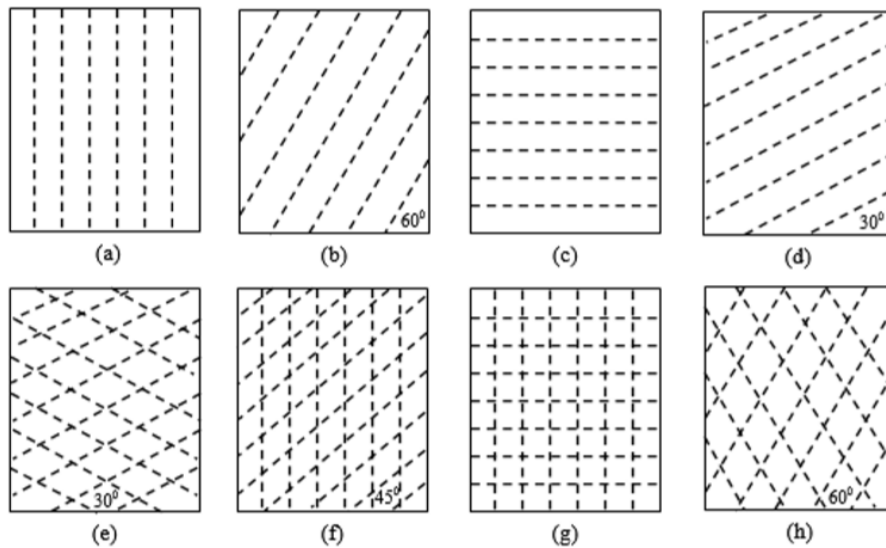


Fig. 20. Common stitch patterns, depicting (a) vertical, (b) tilt 60°, (c) horizontal, (d) tilt 30°, (e) tilt 30°/30°, (f) tilt 45°/90°, (g) box and (h) tilt 60°/60° [223].

Stitching prevents crack propagation during tensile loading and ensures equal stress distribution along the reinforcing fibers. The tensile strength and tensile modulus also increase, due to the increase in the volume fraction of the fibers, resulting from the stitching thread. However, the stitched biocomposites have less elongation when compared with unstitched counterpart, which can be attributed to more additional strength of stitch threads. Similarly, the stitched biocomposites have low flexural strength, but this difference is not as significant as the difference in impact strength of the stitched and unstitched biocomposites [223].

4.5 Pinning

The interlaminar properties and the delamination resistance of the fiber-reinforced composites can be effectively enhanced by the z-pinning technique. The z-pinned composites are effective in automotive and aerospace applications [229]. The process of z-pinning is shown in Fig. 20.

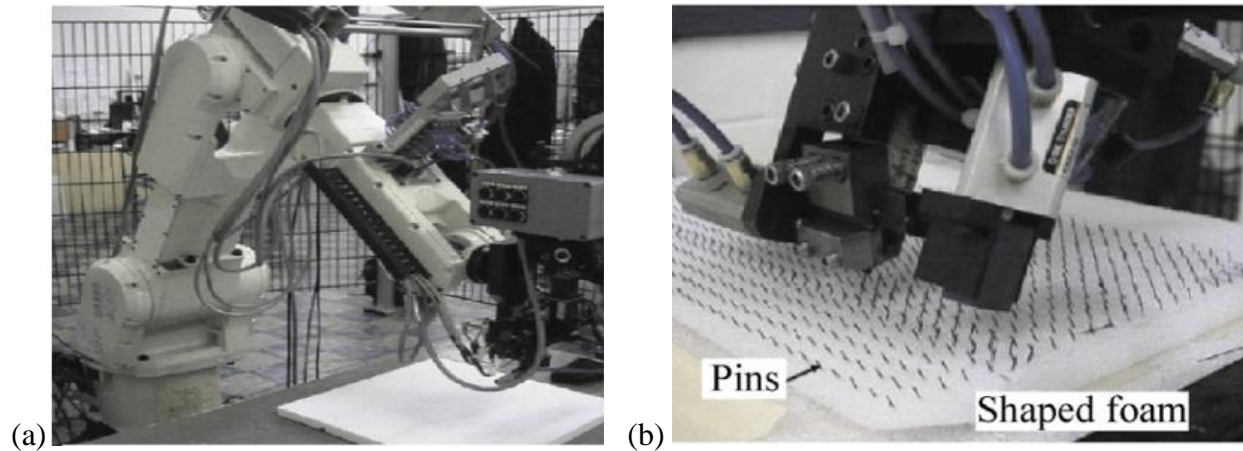


Fig. 21. Z-pinning process, showing (a) 6-axis robot with (b) Z-pin insertion head [229].

The z-pin insertion platform is comprised of an insertion head and a frame driving the plate. During the pinning process, the motors drive the linear guides to control the plate position. The insertion head comprises of a feeder, a directing, cutting and z-pin fixed depth cut-into devices. The direction, speed, feed, and the depth of insertion of the pins into the foam on the plate can be controlled, using specific input functions. The pins are inserted one at a time. Numerous technology advancements have also led to multi-pin feed in one press [229]. Z-pinning improves the toughness and the impact resistance of composites. Z-pinning is not capable of delaying the commencement of delamination damage, although it vastly enhances the resistance of laminates to the growth of established delamination. However, z-pinning effectively minimizes the size of the impact caused delamination only when they are wide enough to enable the joining action of the pins [230].

4.6 Tufting

Unlike traditional stitching methods, the tufting procedure entails only one-sided access to the dry fabric preform. Fig. 21 illustrates the tufting process. As observed in Fig. 32(a), specially crafted tufting needle inserts through-the-thickness reinforcement thread loops into a straight thread seam into the dry fabric pattern. After shifting to the specified tuft position (step 1), the

needle pierces the cloth by introducing a dry thread loop at comparatively low tension (step 2). As the needle is withdrawn (step 3), the dry thread loop is held in the preform by the tension of the surrounding tissue layers (Fig. 32b).

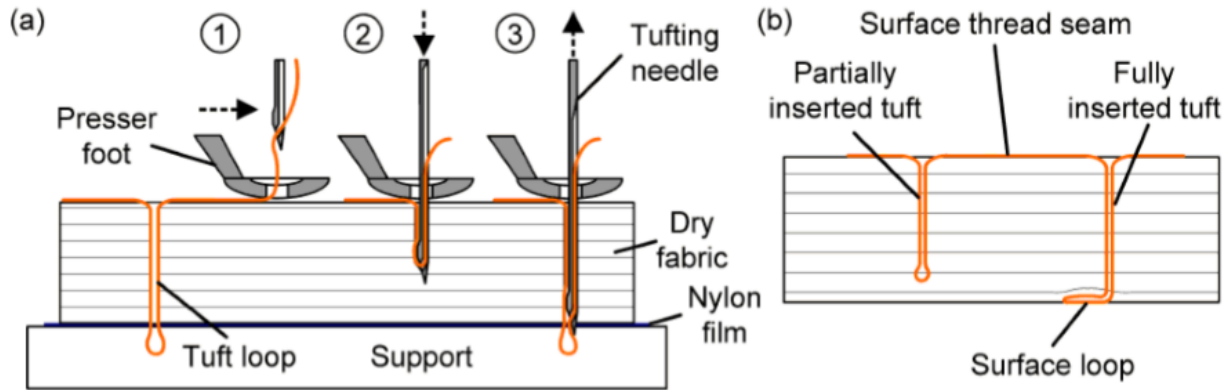


Fig. 22. Tufting process (a) Tufting insertion steps and (b) surface thread seam showing partially and fully inserted tuft [224]

Depending on the selected needle penetration depth, partial or full through-the-thickness reinforcing can be used. In the case of complete penetration, the free loop end is usually left to droop from the preformed back layer to create good tuft insertion efficiency [224]. Tufted composites exhibit a high load-carrying capacity and good impact resistance.

4.7 Hybridization

Fiber hybridization is a process of improving the properties of composite materials. When mixing two or more forms of fiber material, the hybrid composites provide a greater combination of mechanical behaviors than non-hybrid composites. Foreseeing their mechanical responses is difficult, due to the discordant influence between the two fibers. The key benefit of hybridization is that it combines the boons of all the fibers used in the hybrid and alleviates the disadvantages [231]. Fig. 22 shows the generally used hybrid arrangements of fibers. Increased dispersion tends to improve non-penetrative effect tolerance and residual behaviors in hybrid composites. Study has suggested that penetration effect tolerance often improves with dispersion. Positioning the fibers with the greatest energy absorbing potential on the exterior helps the hybrid composite to attract more energy [231].

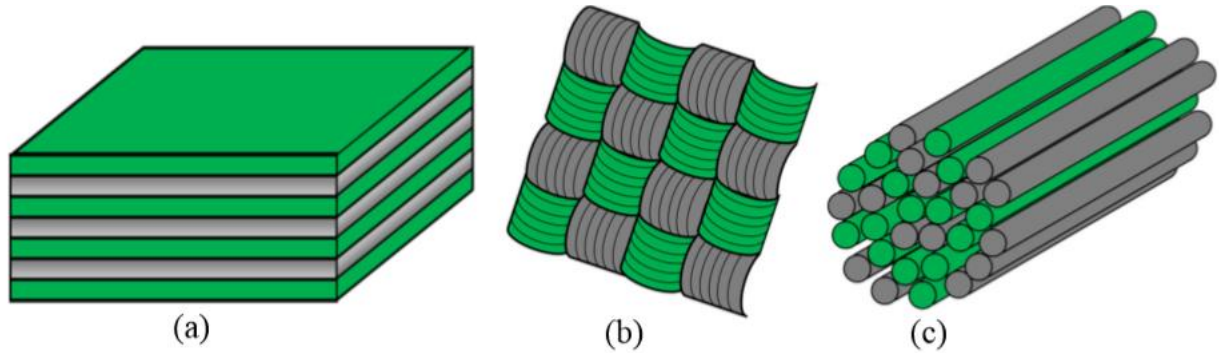


Fig. 23. Hybrid configurations of fibers, showing (a) interlayer or layer-by-layer, (b) intralayer or yarn-by-yarn, and (c) intra-yarn or fiber-by-fiber [231].

5. Advances and prospects of animal-based biocomposites

The utilization of animal fiber reinforcements in composites remains an area that requires further exploration and research. Additionally, there is currently no dedicated tool available to accurately classify and differentiate various types of animal fibers. Furthermore, animal fibers often undergo contamination during the marketing process. However, animal-based, or natural fibers can serve as viable substitutes in composite material manufacturing, offering a wide range of value-added applications beyond their established uses in bioengineering and medical fields. Animal fiber reinforced composite materials have already found applications in industries such as automotive, aviation, and sports. These composites exhibit a high strength-to-weight ratio and excellent corrosion resistance when compared with other metals and non-reinforced plastics. One key advantage of animal fiber composites is their ability to be customized to meet specific shape requirements. These biocomposites are expected to be cost-effective and offer an attractive alternative to traditional metal-based materials [232].

5.1 Present challenges/critical issues militating against the property enhancement

Animal-based fiber reinforcements have certain afforested advantages, but they are difficult to manufacture and require certain pre-processing before fabrication. The hydrophilic aspect of the animal fibers can be a drawback; making it more difficult to cope with. In wet environments, natural fibers accumulate water from the atmosphere (moisture), which in essence, creates fiber bumps inside the composite, causing a problem for the internal parts of the vehicle and aircraft. Moreover, these fibers have reduced fire resistance, which makes them unsuitable for aerospace

and automotive applications. Essentially, natural fibers are not thermoplastic, and their decomposition temperature is relatively low when compared with other thermo-physical behaviors, such as melting and glass temperatures. In addition, the mechanical behaviors of the animal-based fiber reinforced composites depend on the external environmental conditions and vary with time. In addition, many diseases are associated with different animals and insects, therefore sufficient care needs to be taken when processing these kinds of fibers [233].

5.2 *Future perspectives*

Due to numerous environmental issues, natural fiber has gained increasing interest from technologists, architects, industrialists, and manufacturers, because of its tremendous capacity of application to various engineering facilities in the fields of building, rail, automobile, packaging, defense, among other sectors. Furthermore, natural fiber composite is a recycled, environmentally friendly material, has a low density and decreases the evolution of carbon dioxide (CO₂). The use of animal fiber in composite manufacturing is an evolving field, as waste animal fiber can be used for suitable technological applications [232].

Although, animal-based fibers or reinforcements for polymeric biocomposites are gaining interest, the main issue arises when replacing these composites with the conventional counterparts, which exhibit similar properties. The commercialization and the market of the animal-based biocomposites are expected to grow in the nearest future, subject to the social and the environmental awareness of the customers, manufacturers, and the introduction of new technologies. However, one of the major issues is the lack of adequate and broad knowledge in this research field, as several studies on animal-based fiber-reinforced biocomposites are still ongoing [234]. Some animal fibers have not yet been discovered and those that are presently available have not been widely utilized and well experimentally characterized, despite of their potential capability of being used as a reinforcing medium for composite manufacturing. This poses a great challenge to the application of simulation technique to the animal-based fiber/filler reinforced polymeric biocomposite field, due to the availability of insufficient and/or unavailability of reliable, consistent, and robust data for accurate model prediction and numerical investigation. Therefore, composite production field is still limited, due to the scarcity of fibers, but the area of further research in this field is emerging. Also, the results of the increasing application of animal waste materials would contribute to the conversion of animal waste into material worth or wealth [232].

In addition, there are needs for innovative design and manufacturing techniques to be specifically tailored and introduced to the animal-based biocomposite technology. These include, but are not limited to, hybridization, pinning, weaving, knitting, and stacking sequences. Hybridization system could involve two or more compatible animal fibers or fillers and/or animal and plant fibers or fillers, after appropriate chemical characterization. Besides, the application of suitable animal fiber treatment and matrix modification to improve the fiber-matrix interfacial adhesion of their biocomposites is very germane. Both properties and performances of biocomposites are mostly dependent on the strength of their fiber-matrix interfacial bond. Therefore, the presence of some materials or organic groups in some animal-based fibers and fillers must be broken or completely removed, because they reduce the compatibility of animal fibers with matrices by forming a weak layer between the reinforcements and the matrices. Probably, the protein group present in some animal (wool and alpaca) fibers, such as keratin could be broken or removed by cooking and washing the fibers inside concentrated sodium hydroxide solution.

Generally, the conceptual design and development strategy for animal-based fibers/fillers reinforced polymeric biocomposites should be a balance of their performance requirements, sustainability, and cost. The research gaps are potential future studies towards advancing ever thriving biocomposite technology, as the quest for improved sustainable, environmentally friendly, and low-cost biocomposite products as well as their wide and specific application continues.

6. Concluding remarks

Traditionally, materials such as basalt, glass, carbon and graphite have been favored as reinforcement in PMCs, but recent emphasis has pivoted toward more environmentally friendly and cost-effective alternatives. These alternatives can be categorized into two groups: natural plant-based and animal-based fillers. While natural plant-based fillers have attracted significant research and development, animal-based fillers have been somewhat overlooked, despite their considerable potential. These materials, often originating from animal wastes or food by-products, encompassing a wide array of unique substances with diverse structures and properties, such as wool, silk, feathers and more. Notably, animal-based fillers present a sustainable choice, as they do not produce greenhouse gases at the end of their life cycle and their degradation products pose no harm to the environment or human health. Furthermore, the functional groups on these fillers offer opportunities for versatile applications without extensive modifications. [Consequently, the](#)

utilization of animal-based fillers or fibers in PMCs holds promise for significantly reducing adverse environmental effects when offering unique performance, benefits and cost considerations. Animal-based materials such as wool fibers, renowned for their resilience and flexibility, can enhance impact resistance and tensile strength, while also affecting the weight and density of the composites. Moreover, their distinct thermal and moisture properties, such as wool's inherent moisture-wicking capabilities, provide additional advantages, depending on the application. However, cost implications must be carefully weighed, as animal-based fillers may incur higher raw material costs and require specialized processing techniques, potentially increasing manufacturing expenses. Beyond economics and performance, environmental considerations are paramount, with animal-based reinforcements offering biodegradability and renewability advantages over synthetic counterparts. Nevertheless, the broader environmental impact, encompassing resource consumption associated with animal husbandry and processing, as well as ethical concerns regarding animal welfare and sourcing practices, must be rigorously evaluated.

Finally, the imperative need for environmental sustainability has driven a shift in the materials landscape, particularly in the domain of PMCs. As the world grapples with pressing environmental challenges, the adoption of these innovative materials presents an opportunity to strike a balance between technological advancement and ecological preservation. This review underscores the importance of embracing diverse and sustainable materials in materials science and engineering, ultimately paving the way for a more environmentally conscious and sustainable future.

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