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Natural fibre-reinforced composites for bioengineering and environmental engineering applications

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ABSTRACT

Recently, the mankind has realized that unless environment is protected, he himself will be threatened by the over consumption of natural resource as well as substantial reduction of fresh air produced in the world. Conservation of forests and optimal utilization of agricultural and other renewable resources like solar and wind energies, and recently, tidal energy have become important topics worldwide. In such concern, the use of renewable resources such as plant and animal based fibre-reinforce polymeric composites, has been becoming an important design criterion for designing and manufacturing components for all industrial products. Research on biodegradable polymeric composites, can contribute for green and safe environment to some extent. In the biomedical and bioengineered field, the use of natural fibre mixed with biodegradable and bioresorbable polymers can produce joints and bone fixtures to alleviate pain for patients. In this paper, a comprehensive review on different kinds of natural fibre composites will be given. Their potential in future development of different kinds of engineering and domestic products will also be discussed in detail.

1. Introduction

1.1. Environmental concern

Since the past few decades, research and engineering interest has been shifting from traditional monolithic materials to fibre-reinforced polymer-based materials due to their unique advantages of high strength to weight ratio, non-corrosive property and high fracture toughness. These composite materials consisted of high strength fibres such as carbon, glass and aramid, and low strength polymeric matrix, now have dominated the aerospace, leisure, automotive, construction and sporting industries. Unfortunately, these fibres have serious drawbacks such as (i) non-renewable, (ii) non-recyclable, (iii) high energy consumption in the manufacturing process, (iv) health risk when inhaled and (v) non-biodegradable. Biodegradation is the chemical breakdown of materials by the action of living organisms which leads to changes in physical properties. It is a concept of vast scope, ringing form decomposition of environmental wastes involving micro-organisms to host-induced of biomaterials.

Although glass fibre-reinforced composites have been widely used due to its advantages of low cost and moderate strength, for many years to provide solutions to many structural problems, the use of these materials, in turn would induce a serious environmental problem that most Western countries are now concerning. Recently, due to a strong emphasis on environmental awareness worldwide, it has brought much attention in the development of recyclable and environmentally sustainable composite materials. Environmental legislation as well as consumer demand in many countries is increasing the pressure on manufacturers of materials and end-products to consider the environmental impact of their products at all stages of their life cycle, including recycling and ultimate disposal [31]. In the United State, it encourages manufacturers to produce materials and products by practicing the 4Rs, which are (i) *Reduce* the amount and toxicity of trash to be discard (sourced reduction); (ii) *Reuse* containers and products; (iii) *Repair* what is broken (iv) *Recycle* as much as possible, which includes buying products with recycled content. After these processes are gone, the materials finally are entitled to be disposed to the landfill.

1.2. Bio-engineering concern

Bioengineering refers to the application of concepts and methods of the physical science and mathematics in an engineering approach towards solving problems in repair and reconstructions of lost, damaged or deceased (or "non-functional") tissues. Any material that is used for this purpose can be regarded as a biomaterial.

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According to Williams [1], a biomaterial is a material used in implants or medical devices, intended to interact with biological systems. Those common types of medical devices include the substitute heart valves and artificial hearts, artificial hip and knee joints, dental implants, internal and external fracture fixators and skin repair templates, and etc. One of the major features of biocomposite materials is that they can be tailor made to meet different applications' requirements. The most common types of conventional composites are usually composed of epoxy, unsaturated polyester resin, polyurethanes (PU) or phenolic reinforced by glass, carbon or aramid fibres. These composite structures lead to the problem of conventional removal after the end of life time, as the components are closely interconnected, relatively stable and thus difficult to separate and recycle.

In bone repair, stainless steel and titanium have been used as bone plates for many years due to their unique biocompatible properties. However, the use of these materials for bone repair requires several times of subsequent surgical operations to remove the plates and other fasteners, which may cause unnecessary pain or inconvenience to the patients. Uneven growth of bone cells underneath and surrounding the plates may cause porosis. Refracture of the bones may also occur after the plate removal due to the differential stiffness between the bone and the plates. Table 1 shows the mechanical properties of different types of biomaterials used for the implant application. The comparison of the bone and cartilage properties, with other materials is also given. Therefore, less stiff materials should be used for making plates for bone fixation. Poly(lactic acid) (PLA) has been well recognized as a good material with both biodegradable and bioresorbable properties that can be inserted into the human body without inducing any harmful effect. However, since a degradable implant does not have to be removed surgically once it is no longer needed, degradable polymers are of value in short-term applications that require only the temporary presence of a device. The most concern relating to the use of degradable implants is the toxicity of the implant's degradation products. Besides, the low strength of the degradable polymers also limits the range of their applications.

2. Natural fibre

Within the past few years, there has been a dramatic increase in the use of natural fibres such as leaves from flax, jute, hemp, pineapple and sisal for making a new type of environmentally–friendly composites. Recent advances in natural fibre development, genetic engineering, and composites science offer significant opportunities for improved materials from renewable resources with enhanced support for global sustainability. Table 2 shows the mechanical properties of different types of potential natural fibres for composite applications. A material that can be used for medical application must possess a lot of specific characteristics, which are different with that for the general domestic-used plastic products. The most fundamental requirements are related to biocompatibility, without having any adverse effect to the host tissues. Therefore, those traditional composite structures with non-biocompatible matrix or reinforcement are substituted by bio-engineered composites. Table 3 summarizes several important factors that are needed to be considered in selecting a material for the biomedical applications.

Table 1
Mechanical properties of typical implant materials and tissues.

	Elastic modulus (GPa)	Yield strength (MPa)	Tensile Strength (MPa)	Elongation to failure (%)
Al_2O_3	350	–	1000–10,000	0
CoCr Alloy	225	525	735	10
Stainless steel	210	240	600	55
316	120	830	900	18
Ti–6Al–4V	15–30	30–70	70–150	0–8
Bone (Cortical)	3.0	–	35–50	0.5
PMMA	0.6–1.8	–	23–40	200–400
HDPE Cartilage	Strong viscoelastic	–	7–1	20

Table 2
Mechanical properties of different types of potential natural fibres for composite applications.

	Tensile strength (MPa)	Elongation at break (%)	Young modulus (GPa)
Natural fibres			
Flax	300–1500	1.3–10	24–80
Jute	200–800	1.16–8	10–55
Sisal	80–840	2–25	9–38
Kenaf	295–1191	3.5	2.86
Pineapple	170–1627	2.4	60-82
Banana	529–914	3	27–32
Coir	106–175	14.21–49	4–6
Oil palm (empty fruit)	130–248	9.7–14	3.58
Oil palm (fruit)	80	17	
Ramie	348–938	1.2–8	44–128
Hemp	310–900	1.6–6	30–70
Wool	120–174	25–35	2.3–3.4
Spider silk	875–972	17–18	11–13
Cotton	264–800	3–8	5–12.6
Human tissues			
Hard tissue (tooth, bone, human compact bone, longitudinal direction)	130–160	1–3	17–20
Skin	7.6	78	
Tendon	53–150	9.4–12	1.5
Elastic cartilage	3	30	
Heart valves	0.45–2.6	10–15.3	
Aorta	0.07–1.1	77–81	

In the environmental concern, the use of these fibre mixed with biodegradable polymers, like PLA and poly(glycolide) (PGA) can produce biodegradable composites (somehow, they are called "Green composites" or "Environmentally–friendly plastics"). These composites should possess moderate strength and/or thermal stability while they must be recyclable after being used. However, the currently problems of using pure biodegradable polymers are their low strength and low service temperature, such as their glass transition temperature (T_g). In general, T_g of these polymers is around 40–60 °C, which is much below other synthetic polymers. In Tables 4 and 5, the characteristics of synthetic polymers, bioresorbable and biocompatible polymers are shown. Fig. 1 also shows the chemical structures of widely investigated biocompatible and biodegradable polymers. Polyethylene (PE) is normally in its high-density (HDPE) form in biomedical application because low-density material cannot withstand sterilization temperatures. Polypropylene (PP) is an isotactic crystalline polymer with high rigidity, good chemical resistance and moderate strength, which has a higher fracture toughness than HDPE. Polytetrafluoro-ethylene (PTFE) is a very high melting (T_m = 312 °C) and high crystallinity polymer. PLA and PGA and their co-polymers are used in resorbable surgical sutures. The degradation products are endogenous compounds and such are non-toxic. For the tables, it is obvious that those synthetic polymers for being used in biomedical devices have better thermal properties than that of bioresorbable polymers. However, the mechanical properties of all these polymers are relatively low as compared with traditional metallic materials for implants and other fixation applications.

Bio-composites consist of biodegradable polymer as matrix and usually bio-fibres as reinforcing elements which are generally low cost, low density, high toughness, acceptable specific strength properties, good thermal properties, ease of separation, enhanced

Table 3
Key factors for the selection of materials for biomedical applications [2].

Factors	Description		
	Chemical/biological characteristics	Physical characteristics	Mechanical/structural characteristics
1st Level material properties	• Chemical composition (bulk and surface)	• Density	• Elastic modulus • Shear modulus • Poisson's ratio • Yield strength • Compressive strength
2nd Level material properties	• Adhesion	• Surface topology • Texture • Roughness	• Hardness • Flexural modulus • Flexural strength
Specific functional requirements (based on applications)	• Biofunctionality • Bioinert • Bioactive • Biostability • Biodegradation behavior	• Form and geometry • Coefficient of thermal expansion • Electrical conductivity • Color, aesthetics • Refractive index • Opacity or translucency	• Stiffness or rigidity • Fracture toughness • Fatigue strength • Creep resistance • Friction and wear resistance • Adhesion strength • Impact strength • Proof stress • Abrasion resistance
Processing & Fabrication	• Reproducibility, quality, sterilizability, packaging, secondary processability		
Characteristics of host: tissue, organ, species, age, sex, race, health condition, activity, systemic response			
Medical/surgical procedure, period of application/usage			
Cost			

Table 4
Characteristics of different kinds of polymers.

Type	Thermal properties	Construction/useful form
Synthetic polymers		
Polyethylene (PE)	T_m = 203–208 °C	Heath care products, light weight orthopaedic casts, ligament prostheses
Polypropylene (PP)	T_m = 223–233 °C	Surgical drapes, and gowns
Poly(tetrafluoro-ethylene) (PTFE)	T_m = 313–315 °C	Vascular fabrics, heart valve sewing rings, orthopaedic ligaments
Nylon 6	T_g = 45 °C, T_m = 220 °C	Sutures
Poly(ethylene terephthalate (PET))	T_g = 65–105 °C, T_m = 265 °C	Sutures
Bio-absorbable synthetic polymers		
Poly(glycolide) (PGA)	T_g = 40–45 °C, T_m = 225 °C	Absorbable sutures and meshes
Poly(*p*-dioxanone) (PDS)	T_g = 10 °C, T_m = 205 °C	Sutures, intramedullary pins
Poly(lactic-acid) (PLA)	T_g = 60 °C and T_m = 166 °C	Bone fixture

energy recovery and biodegradability. Bio-fibres are chosen as reinforcements since they can reduce the chance of tool wear when processing, dermal and respiratory irritation. Conversely, these fibres are usually small in cross-sections and cannot be directly used in engineering applications; they are embedded in matrix materials to form biocomposites. The matrix serves as binder to bind the fibres together and transfer loads to the fibres. In order to develop and promote these natural fibres and their composites, it is necessary to understand their physico-mechanical properties.

3. Plant-based fibres

As aforementioned, with the increasing global energy crisis and ecology risk, plant-based fibre-reinforced polymer composites have attracted much interest owing to their potential of serving as alternatives for artificial fibre composites, like glass and carbon. Although the strength of such fibres (more than one type) are general lower than that of the traditional advanced composites, in certain extent, the strength of plant-based fibre-reinforced composites is sufficient enough for domestic or household plastic products. Many attempts have been done in the past few years on using jute, bamboo, sisal, coir, hemp, flax, pineapple leaves, etc., for reinforcing different kinds of thermoplastic and thermoset polymers to form green composites.

Wambua et al. [3] conducted a series of experiments to comprehensively study the strength enhancement of different plant-based fibre polymeric composites. They have reported that the successful production of good quality of plant-based composites is highly re-

Table 5
Mechanical properties of biomedical polymers.

Polymer	Water absorption (%)	Bulk modulus (GPa)	Tensile strength (MPa)	Elongation at break (%)	T_g (°C)	T_m (°C)
Polyethylene (PE)	0.001–0.02	0.8–2.2	30–40	130–500	71–76	203–208
Polypropylene (PP)	0.01–0.035	1.6–2.5	21–40	50–800	117–132	222–233
Polyurethane (PU)	0.1–0.9	1.5–2	28–40	600–720	93–121	233–272
Polytetrafluoro-ethylene (PTFE)	0.01–0.05	1–2	15–40	250–550	145–146	312–315
Polyvinyl-chloride	0.04–0.75	3.4	10–75	10–400	121–183	217
Polyamides	0.25–3.5	2.4–3.3	44–90	40–250	145–185	256–282
Polycarbonate (PC)	0.15–0.7	2.8–4.6	56–75	8–130	214	258–273
Bombyx mori silk	0.2–1	0.015–0.017	610–690	4–16	–	–

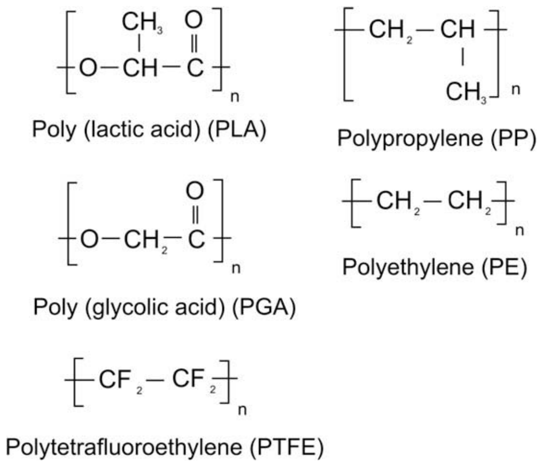


Fig. 1. Chemical structures of widely investigated biocompatible and bio-degradable polymers.

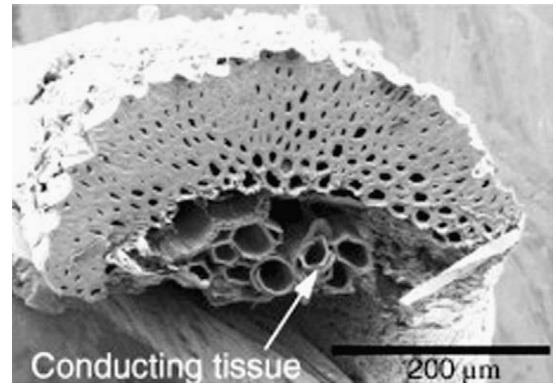


Fig. 2. Hollow section of a sisal fibre [5].

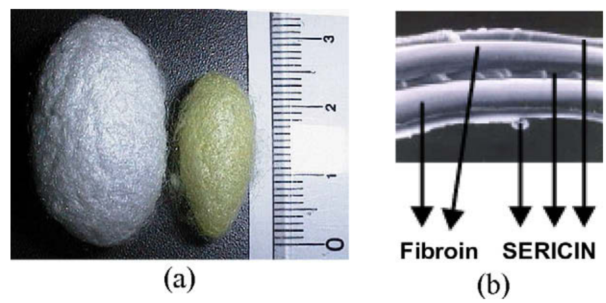


Fig. 3. Raw Cocoon silks (a) and side view of the silk fibre (b).

lied on their bonding properties between the fibre and matrix. This also governs their biodegradability in real life applications. It was found that a hemp fibre composite with 30% fibre volume fraction (V_f), demonstrated 73% better tensile strength than that of kenaf, sisal and jute fibre composites. The use of coir fibre showed the lowest strength (only 20% of that of the hemp fibre composite). According to the information shown in Table 6, the attribution to strength increment may be due to the strength of fibres, as well as their interfacial bonding properties. It was found that the use of hemp, jute and flax fibres bore better strength as compared with jute, ramie and cotton fibres. Coir fibre possesses the lowest tensile strength as compared with others. Rahman et al. [4] have indicated that the tensile strength decreased while the Young modulus, flexural strength and charpy impact strength increased with increasing the content of jute fibre for jute/polypropylene (PP) composites. Silva et al. [5] have shown that hollow structure inside the sisal fibre was found (Figs. 2 and 3) that would ultimately weaken the strength of its composites, as the net cross-sectional area decreases, and thus stress taken by the fibre is then increased. Selection of right portion with higher density of the fibre is needed to maximize the load bearing capability of the fibre is achieved.

A notable remark for making high strength biodegradable composites found in the table is the water absorbability of different types of plane-based fibre. Cotton and ramie demonstrate high possibility in absorbing moisture during composite manufacturing process, while hemp and flax are relatively less. This would seriously affect the integrity and complete chemical reaction due to excessive water molecules exist of a resultant composite. Such moisture penetration into composite materials occurs by three different conditions, they would diffuse inside the micro-gaps between (i) the polymer chains; (ii) the interface between the fibre and matrix and (iii) existing cracks. The control of manufacturing environment, such as low humidity or under vacuum, is essential to minimize any moisture molecules trapped inside the composite. Drying process is normally required to remove excessive moisture. As compared with the modulus of the hemp, flax and E-glass, they are almost the same but the weight of the plant-based fibres is much lower than the E-glass.

Wood fibre, due to its low density, high specific strength and Young modulus, non-abrasive to processing equipment, low cost and importantly biodegradable, has also attracted much attention in the last decade [6]. However, the main disadvantage of wood fibre is its hydrophilic nature as compared with carbon and glass fibres. Besides, the use of bamboo, a million tonne of wastes from this material produced every year, for producing polymer-based composites have been found in the past few years. The microstructure of bamboo is different from wood, for example, the different content of lignin and different polarity of materials. Wang et al. [7] studied the use of bamboo particles to reinforce Polyvinylchloride (PVC) to form a new type of composites. However, the results, in terms of their mechanical properties were not consistent with their moisture contents and mesh granule size.

4. Animal-based fibres

4.1. Silk-based fibre

Apart from the plant-based fibres, animal-based fibres become other alternatives for producing biodegradable, biomedical and bio-resorbable composite materials for bioengineering and orthopaedic applications. The content of these fibres are mainly made

Table 6
Material and mechanical properties of E-glass and other plant-based fibres.

Properties/fibres	E-glass	Hemp	Jute	Ramie	Coir	Sisal	Flax	Cotton
Density (g/m^3)	2.55	1.48	1.46	1.5	1.25	1.33	1.4	1.51
Tensile strength (MPa)	2400	550–900	400–800	500	220	600–700	800–1500	400
Tensile modulus (GPa)	73	70	10–30	44	6	38	60–80	12
Elongation at break (%)	3	1.6	1.8	2	15–25	2–3	1.2–1.6	3–10
Moisture absorption (%)	–	8	12	12–17	10	11	7	8–25

by proteins, like wool, spider and silkworm silk. The enhanced environmental stability of silk fibres in comparison to globular proteins is due to the extensive hydrogen bonding, the hydrophobic nature of much of the protein, and the significant crystallinity.

Silk proteins – known as silk fibroins are stored in the glands of insects and spiders as an aqueous solution. During the spinning process, silkworm accelerates and decelerates its head in arcs to each change of direction, and the concentration of silk in the solution is gradually increased and finally, elongation stress is applied to produce a partly crystalline, insoluble fibrous thread in which the bulk of the polymer chains in the crystalline regions are oriented parallel to the fibre axis. Faster spinning speed leads to stronger but more brittle fibres where slower speed leads to weaker and more extensible fibres. At even greater speed, silk toughness decreased, mainly due to the loss of extensibility [8].

Cocoons are natural polymeric composite shells made of a single continuous silk strand with length in the range of 1000–1500 m and conglutinated by sericin [9]. This protein layer resists oxidation, is antibacterial, ultra-violet (UV) resistant, and absorbs and releases moisture easily. Since this protein layer can be cross-linked, copolymerized, and blended with other macromolecular materials, especially artificial polymers, to produce materials with improved properties. In average, the cocoon production is about 1 million tonnes worldwide, and this is equivalent to 400,000 tonnes of dry cocoon (see Fig. 2). In the tissue engineering area, silks have been identified as a kind of biomaterials, used for healing process for bone, tendons or ligament repairs. Slowly degrading biomaterials which can maintain tissue integrity following implantation, while continually transferring the load-bearing burden to the developing biological functional tissue are desired. In such phenomena, the gradual transfer of the load-bearing burden to the developing and/or remodelling tissue should support the restoration and maintenance of tissue function over the life of the patient.

Silk fibre spun out from silkworm cocoons are consisted of fibroin in the inner layer and sericin in the outer layer, all are protein based. From outside to inside layers of cocoon, the volume fractions of sericin decreases while the relative content of fibroin increases. Also, it is known that silk fibroin consists of both hydrophilic and hydrophobic regions which is a block-like polymeric system. These fibres have a highly non-uniform cross-sectional geometry with respect to both shape and absolute dimensions. By changing the reeling conditions, silkworm silk can be stronger, stiffer and more extensible, approaching to the properties of spider dragline silk [10]. Each raw silk thread has a lengthwise striation, consisting of two separate but irregularly entwined fibroin filaments embedded in sericin. Silk sericin is a minor protein that envelops silk fibroin fibre and glues them together to form cocoon shape. Fibroin and sericin in silk account for about 75 and 25 wt%, respectively. Silk fibre is biodegradable and highly crystalline with a well-aligned structure.

Composition, structure and material properties of silk fibre produced by spiders, silkworms, scorpions, mites and flies may differ widely depending on the specific source and the uncontrollable reeling conditions of those insects. Spinning under controlled conditions will have more uniform cross-sectional area of silk fibre, reproducible molecular alignment and fewer micro-structural flaws. The size and weight of cocoons decrease with an increase in temperature and cocoons can bear efficiently both external static forces and dynamic impact loadings [10]. Normal compact cocoon exhibits a high ability of elastic deformation with an elastic strain limit higher than 20% in both longitudinal and transverse directions. Anisotropic properties mainly due to the non-uniform distribution and orientations of silk segments and the inner layer of cocoon has low porosity (higher silk density) and smaller average diameter of silk, therefore, there is an increase in elastic modulus and strength from outside to inside layers. That is, the thinner the silk, the higher the elastic modulus and tensile strength and the maximum values at the innermost layer. On the other hand, temperature above the glass transition temperature, the cocoon and its layers become softer and softer and behave similar to a rubber-like material. Silk fibre have higher tensile strength than glass fibre or synthetic organic fibre, good elasticity, and excellent resilience [11]. They resist failure in compression, stable at physiological temperatures and sericin coating is water-soluble proteinaceous glue.

Fibroin is a semi-crystalline polymer of natural fibrous protein mainly consists of two phases [12]: namely β–sheet crystals and non-crystalline including micro-voids and amorphous structure, by which the structure of sericin coating is amorphous acting as an adhesive binder to maintain the fibroin core and the overall structural integrity of the cocoon. Degumming is a key process during which sericin is removed by thermo-chemical treatment of the cocoon. Although this surface modification can affect the tensile behavior and the mechanical properties of silk significantly, it is normally done to enhance interfacial adhesion between fibre and matrix.

In addition, according to Altman [13], silks are insoluble in most solvents, including water, dilute acid and alkali. Reactivity of silk fibre with chemical agents is positively correlated to the largeness of internal and external surface areas [14]. When fabricating silk-based composites, the amount of resin gained by fibre is strongly related to the degree of swelling of the non-crystalline regions, that is, the amorphous regions and the micro-voids inside the fibre.

4.2. Chicken feather fibre

Chicken feather fibre (CFF) has attracted much attention to different product design and engineering industries recently, so as the use of CFF as reinforcements for polymer-based biodegradable materials has emerged gradually. The advantages of using this natural fibre over traditional reinforcing fibres in biocomposites are low cost, low density, acceptable specific strength, recycability, bio-degradability etc. CFF, because of its renewable and recyclable characteristics, have been appreciated as a new class of reinforcements for polymer-based biocomposites. However, the full understanding of their mechanical properties, surface morphologies, environmental influences due to moisture and chemical attacks, bonding characteristics between silk fibroin and surrounding matrix and manufacturing process is essential.

According to the survey conducted recently, a chicken processing plant produces about 4000 pounds of chicken feathers every hour. In most western countries, these feathers are used as (i) feather fibre feed; (ii) air filter elements that replaces traditional wood pulps (retarding the trees cut down rate) and (iii) lightweight feather composites. Chicken feathers are approximately 91% protein (keratin), 1% lipids, and 8% water. The amino acid sequence of a chicken feather is very similar to that of other feathers and also has a great deal in common with reptilian keratins from claws [15]. The sequence is largely composed of cystine, glycine, proline, and serine, and contains almost no histidine, lysine, or methionine. In fact, a CFF is made up of two parts, the fibres and the quills (see Fig. 4). The fibre is thin filamentous materials that merge from the middle core material called quills. In simple terms, the quill is hard, central axis off which soft, interlocking fibres branch. Smaller feathers have a greater proportion of fibre, which has a higher aspect ratio than the quill. The presence of quill among fibres results in a more granular, lightweight, and bulky material. A typical quill has dimensions on the order of centimeters (length) by millimeters (diameter). Fibre diameters were found to be in the range of 5–50 μm. The density of CFF is lighter than other synthetic and natural reinforcements, thus, CFF inclusion in a composite could potentially lower composite density, whereas the den-

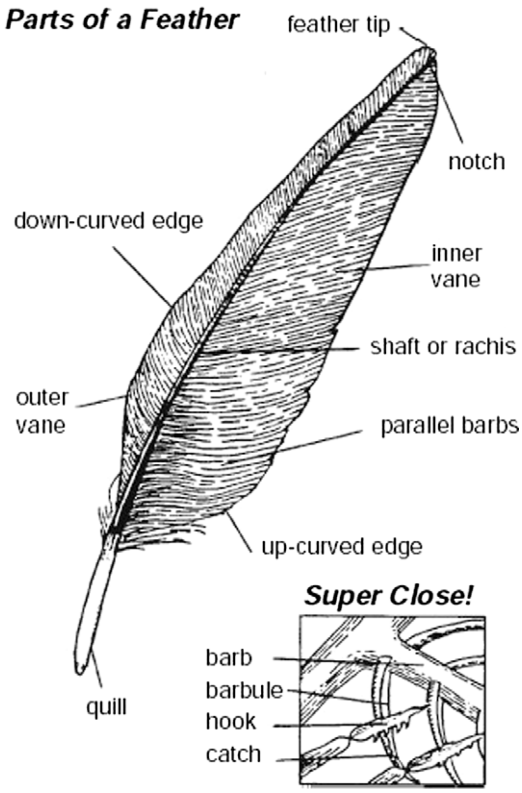


Fig. 4. A typical chicken feather fibre.

sity of a typical composite with synthetic reinforcing increases as fibre content increases. Hence, light weight composite materials can be produced by inclusion of CFF to plastics which even reduces the transportation cost. The barbs at the upper portion of the feather are firm, compact, and closely knit, while those at the lower portion are downy, i.e. soft, loose, and fluffy. The down feather provides insulation, and the flight feather provides an airfoil, protects the body from moisture, the skin from injury, and colors and shapes for displays. Fig. 5 shows the cross-sectional views of the flight and down feather fibres. It is obvious that flight feather fibre exited in a hollow form while down fibre is in solid. In terms of the purpose of fibre-reinforcement, the use of down fibre appears much better than that the use of flight fibre.

The moisture content of CFF is an important factor that can highly influence their weight and mechanical properties. The moisture content of processed CFFs can vary depending upon processing and environmental conditions. The glass transition temperature (T_g) of the feather fibres and inner quills is approximately 235 °C while an outer quills is 225 °C. High T_g represents that a tighter keratin structure is formed to which water is more strongly bonded. Fibres and inner quills do not begin to lose water below 100 °C. The moisture evolution temperature of the CFF and quill occurs in the range of 100–110 °C [16]. This suggests that it may be possible to have a fully dry fibres and inner quills at 110 °C.

The length and diameter (sometime in the form of bundles) of CFF would highly affect their properties and impregnatability of resin into a resultant composite. Therefore, the control of resin temperature (thus, its viscosity), while at the same time to manage the sonication (ultrasonic vibration) time to facilitate the resin penetration rate into the fibres are essential. Short or longer fibres would highly affect the stress transferability as well as shear strength of the composites. The fibres, themselves also would be a barrier to the movement of polymer chains inside the composites and it may result in increasing their strength and thermal properties, but reduce their fracture toughness. These properties will be studied in detail, in this project. Fig. 6 shows the SEM image of the down chicken feather fibre and its strength compared with other type of feathers. It was found that the development of chicken feather fibre biocomposites have been increasing in recent years, and the outcome are expected to be able to alleviate the global waste problem.

5. Applications

5.1. Wound sutures

Silk fibre has been used in biomedical applications particularly as sutures by which the silk fibroin fibre is usually coated with waxes or silicone to enhance material properties and reduce fraying. But in fact, there are lots of confusing questions about the usage of this kind of fibre as there is the absence of detailed characterization of the fibre used including the extent of extraction of the sericin coating, the chemical nature of wax-like coatings sometimes used, and many related processing factors. For example, the sericin glue-like proteins are the major causes of adverse problems with biocompatibility and hypersensitivity to silk. The variability of source materials has raised the potential concerns with this class of fibrous protein. Yet, silk's knot strength, handling characteristics and ability to lay low to the tissue surface make it popular suture in cardiovascular applications where bland tissue reactions are desirable for the coherence of the sutured structures [17].

5.2. Scaffolds tissue engineering

A three-dimensional scaffold permits the in vitro cultivation of cell–polymer constructs that can be readily manipulated, shaped, and fixed to the defect site [18]. The matrix acts as the translator

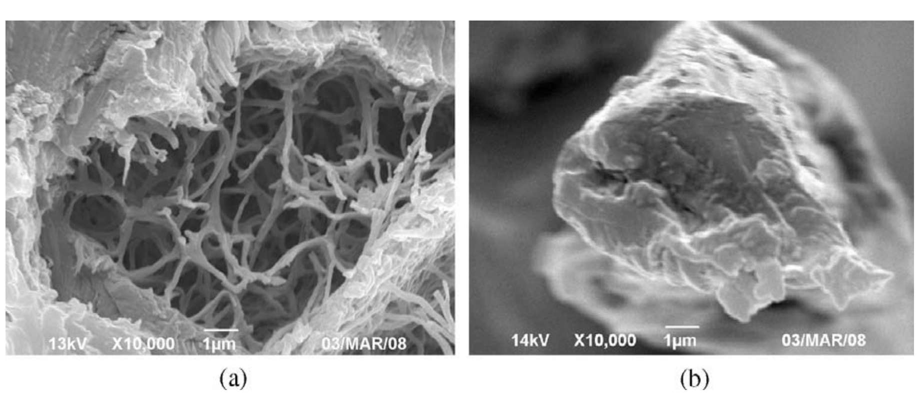
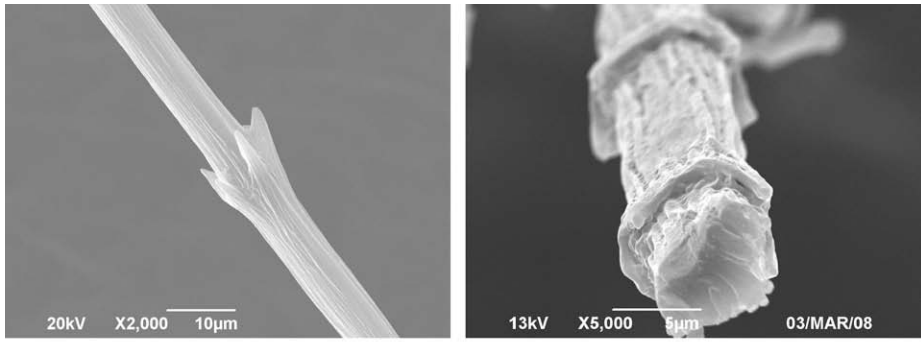


Fig. 5. Flight chicken feather fibre (a) and down chicken feather fibre (b).



(a) (b)

Fig. 6. SEM images of the chicken feather fibre.

between the local environment (either in vitro or in vivo) and the developing tissue, aiding in the development of biologically viable functional tissue. However, during the 1960s to the early 1980s, the use of virgin silk negatively impacted the general acceptance of this biomaterial from the surgical practitioner perspective, for examples, the reaction of silk to the host tissue and the inflammatory potential of silk. Recently, silk matrices are being rediscovered and reconsidered as potentially useful biomaterials for a range of applications in clinical repairs and as scaffolds for tissue engineering.

Silk, as a protein, is susceptible to proteolytic degradation in vivo and over longer period of time in vivo will slowly be absorbed. Degradation rate of implants mainly depend on health and physiological status of patient, mechanical environment of the implantation site, and types and dimensions of the silk fibres. Slow rate of degradation of silk in vitro and in vivo makes it useful in biodegradable scaffolds for slow tissue ingrowths since the biodegradable scaffolds must be able to retain at the implantation site, including maintains their mechanical properties and supports the growth of cells, until the regenerated tissue is capable to fulfill the desired functions. The degradation rate should be matched with the rate of neo-tissue formation so as to compromise the load-bearing capabilities of the tissue.

Additionally, scaffold structures including size and connective of pores determine the transport of nutrients, metabolites and regulatory molecules to and from cells. The matrix must support cell attachment, spreading, growth and differentiation. Meinel et al. [19] concentrated on cartilage tissue engineering with the use of silk protein scaffold and the authors identified and reported that silk scaffolds are particularly suitable for tissue engineering of cartilage starting from human mesenchymal stem cells (hMSC), which are derived from bone marrow, mainly due to their high porosity, slow degradation, and structural integrity.

Recent research with silk has focused on the development of a wire rope matrix for the development of autologous tissue engineered anterior cruciate ligaments (ACL) using a patient's own adult stem cells [20]. Silk fibroin offers versatility in matrix scaffold design for a number of tissue engineering needs in which mechanical performance and biological interactions are major factors for success, including bone, ligaments, tendons, blood vessels and cartilage. Silk fibroin can also be processed into foams, films, fibres and meshes.

5.3. Silk-based biocomposites

Annamaria et al. [21] discovered in the studies that environmentally–friendly biodegradable polymers can be produced by blending silk sericin with other resins. Nomura et al. [22] identified that polyurethane foams incorporating sericin are said to have excellent moisture-absorbing and –desorbing properties. Hatakeyama [23] also reported for producing sericin-containing polyurethane with excellent mechanical and thermal properties. Sericin blends well with water-soluble polymers, especially with polyvinyl alcohol (PVA). Ishikawa et al. [24] investigated the fine structure and the physical properties of blended films made of sericin and PVA. Moreover, a recent patent reported on a PVA/sericin cross-linked hydrogel membrane produced by using dimethyl urea as the cross-linking agent had a high strength, high moisture content and durability for usage as a functional film [25].

Silk fibroin film has good dissolved oxygen permeability in wet state but it is too brittle to be used on its own when in dry state; whereas for chitosan, it is a biocompatible and biodegradable material which can be easily shaped into films and fibres. Park et al. and Kweon et al. [26,27] introduced an idea of silk fibroin/chitosan blends as potential biomedical composites as the crystallinity and mechanical properties of silk fibroin are greatly enhanced with increasing chitosan content.

Another type of bio-composites is the silk fibroin/alginate blend sponges. For biotechnological and biomedical fields, silk fibroin's reproducibility, environmental and biological compatibility, and non-toxicity benefit in many different clinical applications. As the collective properties, especially mechanical properties of silk fibroin sponges in dry state are too weak to handle as wound dressing, thus, they can be enhanced by blending silk fibroin films with other synthetic or natural polymers, for examples, polysaccharide–sodium alginate.

Furthermore, Katori and Kimura [28] and Lee et al. [29] examined the effect of silk/poly(butylenes succinate) (PBS) bio-composites. They found that the mechanical properties including tensile strength, fracture toughness and impact resistance, and thermal stability of biocomposites would be greatly affected by their manufacturing processes. Moreover, a good adhesion between the silk fibre and PBS matrix was found through the observation and analysis by Scanning Electron Microscope (SEM) imaging.

The mechanical properties of Bombyx mori, twisted Bombyx mori and Tussah silk fibres were also investigated through tensile property tests. It was found that Tussah silk fibre exhibited better tensile strength and extensibility as compared with others, and the stiffness of all samples was almost the same. This may be due to the distinction of silkworm raising process, cocoon producing and spinning conditions. Based on the Weibull analysis, it was shown that the Bombyx mori silk fibre has a better reproducibility in terms of experimental measurement, than that of the Tussah silk fibre.

By using silk fibre as reinforcement for biodegradable polymer, the mechanical properties do have a substantial change. Cheung et al. [30] have demonstrated that the use of silk fibre to reinforce PLA can significantly increase its elastic modulus and ductility to

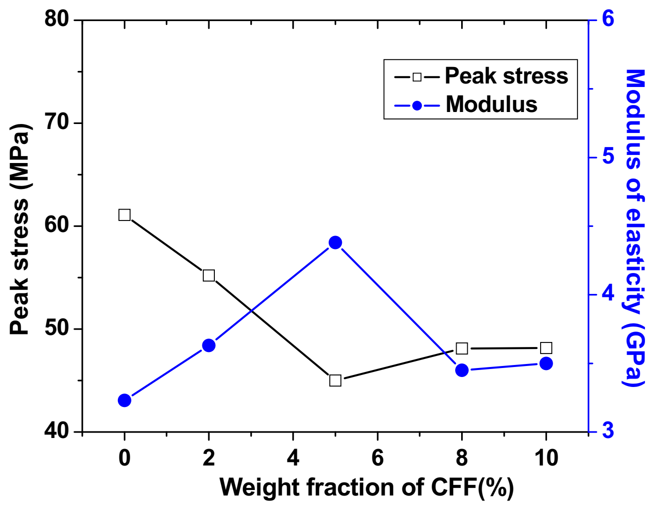


Fig. 7. Relationship between tensile properties and CFF content of CFF/PLA composite.

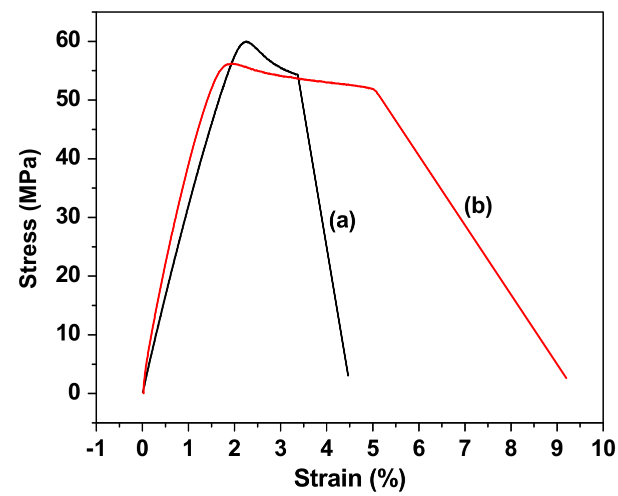


Fig. 8. Stress–strain curves of (a) pure PLA sample; (b) 5 wt% CFF/PLA composite.

40% and 53%, respectively, as compared with a pristine sample. It was also found that the bio-degradability of silk/PLA biocomposites was altered with the content of the silk fibre in the composites. It reflects that the resorbability of the biocomposites used inside the human body can be controlled, in which this is the key parameter of using this new type of materials for bone plate development.

5.4. CFF/PLA biocomposites

By mixing CFF with biopolymers, like PLA can form a biodegradable composites used for plastic products and implant applications. In preparation of the composites, chicken feather was immersed in alcohol for 24 h, then washed in a water soluble organic solvent, and dried under 60 °C for 24 h [32]. CFF with a diameter of about 5 μm and length of 10–30 mm were separated from the quill and then used. Fig. 5 shows an SEM photograph of a CFF. Fig. 7 also shows the relations between CFF content and peak stress and modulus of elasticity, respectively. The modulus of elasticity of CFF/PLA composite increases with the CFF content and reaches the maximum modulus of 4.38 GPa (increment up to 35.6%) at the CFF content of 5 wt%. These reveal that the incorporation of CFF into the matrix is quite effective for reinforcement. The decrease of modulus for the composite with the CFF content above 5 wt% will be due to the insufficient filling of the matrix resin, designating 5 wt% CFF is the critical content.

It also can be found from the peak stress that the tensile strength of PLA after the addition of CFF is lower than that of pure PLA. This phenomenon, also reported by other researchers [15,32], is an indication of poor adhesion between the CFF and the matrix. Although the CFF surface is rough (Fig. 6a) and the hydrophobic consistency of CFF and PLA, the adhesion between them is a problem to solve. And the stress could not be transferred from the matrix to the stronger fibres. Another possible explanation of this phenomenon can be that the CFFs were randomly oriented inside the composite; the failure of the composite might be initiated by the failure of the matrix and then followed by fibre breakage. Fig. 8 shows the stress–strain curves of the pure PLA and 5 wt% CFF/PLA composite. It is observed that a much longer plateau is located between a strain where the peak stress is reached and the strain at break. It can be concluded that the proper content addition of CFF shows a positive effect on elongation to break for PLA, which was expected because of CFFs acting as bridges to prolong the fracture process of the CFF/PLA composite and that the failure of the composite was controlled by the bridging effect of CFF inside the composite. These conclusions could be proved by the fractured morphology of the microstructures observed by SEM. The thermal properties such as glass transition temperature (T_g), crystallization temperature (T_c), melting temperature (T_m), crystallization enthalpy (ΔH_c) and melting enthalpy (ΔH_m) obtained from the DSC studies are plotted in Fig. 9.

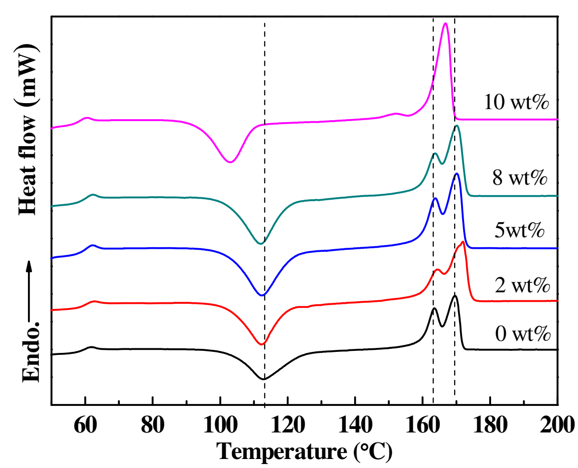


Fig. 9. DSC curves of pure PLA and CFF/PLA composites.

6. Conclusion

The potential use of natural fibre composites is discussed in this paper. Their mechanical and thermal properties have been in depth investigated since the last decade. The mechanical properties in terms of the elastic modulus and ductility of these biocomposites increased substantially compared to the neat polymers. The mechanical properties of most of plant-based fibre composites increased with increasing the amount of fibre into polymer matrix. However, the ultimate strength decreased as expected. From those experimental results, incorporation of the fibres gave rise to a considerable increase of the storage modulus (stiffness) and to a decrease of the tan delta values. These results demonstrate the reinforcing effect of animal-based fibre on PLA matrix. It also reveals that biocomposite with small amount of animal fibre provided also better thermal properties as compared with pristine polymers.

Although plant- and animal-based fibres were attracted much attention to the product design and engineering, and bioengineering industries and undergone comprehensive researches in the past few years, many works such as their interfacial bonding and stress transfer properties have not yet been solved to date. To wider the applications of these fibres in solving environmental problems, more studies have to be continued in the future.

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