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Review

A review of recent developments in natural fibre composites and their mechanical performance

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ABSTRACT

Recently, there has been a rapid growth in research and innovation in the natural fibre composite (NFC) area. Interest is warranted due to the advantages of these materials compared to others, such as synthetic fibre composites, including low environmental impact and low cost and support their potential across a wide range of applications. Much effort has gone into increasing their mechanical performance to extend the capabilities and applications of this group of materials. This review aims to provide an overview of the factors that affect the mechanical performance of NFCs and details achievements made with them.

Contents

1. Introduction

Interest in NFCs is growing for many reasons including their potential to replace synthetic fibre reinforced plastics at lower cost with improved sustainability; their advantages and disadvantages are summarised in Table 1 [1].

The main factors affecting mechanical performance of NFCs are:

- fibre selection – including type, harvest time, extraction method, aspect ratio, treatment and fibre content,
- matrix selection,
- interfacial strength,
- fibre dispersion,
- fibre orientation,
- composite manufacturing process and
- porosity.

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Table 1
Advantages and disadvantages of NFCs [2–5].

Advantages	Disadvantages
• Low density and high specific strength and stiffness • Fibres are a renewable resource, for which production requires little energy, involves CO_2 absorption, whilst returning oxygen to the environment • Fibres can be produced at lower cost than synthetic fibre • Low hazard manufacturing processes • Low emission of toxic fumes when subjected to heat and during incineration at end of life • Less abrasive damage to processing equipment compared with that for synthetic fibre composites	• Lower durability than for synthetic fibre composites, but can be improved considerably with treatment • High moisture absorption, which results in swelling • Lower strength, in particular impact strength compared to synthetic fibre composites • Greater variability of properties • Lower processing temperatures limiting matrix options

Details of the influence of these factors, the mechanical properties obtained and their applications form the basis for the rest of this paper.

2. Factors affecting mechanical performance of NFCs

2.1. Fibre selection

Fibre type is commonly categorised based on its origin: plant, animal or mineral. All plant fibres contain cellulose as their major structural component, whereas animal fibres mainly consist of protein. Although mineral-based natural fibres exist within the asbestos group of minerals and were once used extensively in composites, these are now avoided due to associated health issues (carcinogenic through inhalation/ingestion) and are banned in many countries. Generally, much higher strengths and stiffnesses are obtainable with the higher performance plant fibres than the readily available animal fibres. An exception to this is silk, which can have very high strength, but is relatively expensive, has lower stiffness and is less readily available [6]. This makes plant-based fibres the most suitable for use in composites with structural requirements and therefore the focus of this review. Furthermore, plant fibre can suitably be grown in many countries and can be harvested after short periods.

Table 2 shows properties of some natural fibres and the main type of glass fibre (E-glass). It can be seen that flax, hemp and ramie fibre are amongst the cellulose-based natural fibres having the highest specific Young's moduli and tensile strengths although it should be stated that much variability is seen within the literature. Commonly, geography relating to fibre availability plays a major role in fibre selection [7]. The focus, for example in Europe has been on flax fibre, whereas hemp, jute, ramie, kenaf and sisal have been of greater interest in Asia. Harakeke fibre, (*Phormium tenax* commonly known as New Zealand flax) is also being considered to be used in structural applications in New Zealand due to its good mechanical properties and its local availability there.

Generally, higher performance is achieved with varieties having higher cellulose content and with cellulose microfibrils aligned more in the fibre direction which tends to occur in bast fibres (e.g. flax, hemp, kenaf, jute and ramie) that have higher structural requirements in providing support for the stalk of the plant. The properties of natural fibres vary considerably depending on chemical composition and structure, which relate to fibre type as well as growing conditions, harvesting time, extraction method, treatment and storage procedures. Strength has been seen to reduce by 15% over 5 days after optimum harvest time [16] and manually extracted flax fibres have been found to have strength 20% higher than those extracted mechanically [12]. Strength and stiffness of natural fibres are generally lower than glass fibre, although stiffnesses can be achieved with natural fibres comparable to those achieved with glass fibre. However, the specific properties compare more favourably; specific Young's modulus can be higher for natural fibres and specific tensile strength can compare well with lower strength E-glass fibres.

When comparing data from different sources, it should be considered that a number of variables that are not always reported have an influence on fibre properties. These include testing speed, gauge length, moisture content and temperature. Generally strength increases with increasing moisture content and decreases as temperature increases; the Young's modulus decreases with moisture content [25]. Sometimes it is also unclear in the literature as to whether the tests have been conducted on single fibres (single cells) or on fibre bundles (sometimes referred to as technical fibres). Calculation of properties is generally based on the total cross-section of a fibre or fibre bundle, however, single fibres have a central hollow lumen which takes up a significant proportion of the cross-sectional area. The fraction of cross-sectional area taken up by the lumen has been found to be, for example, 27.2%, 6.8% and 34.0% for sisal, flax and jute respectively [26] and so it could be considered that measurements of strength and stiffness obtained not taking this into account are underestimations to the same degree. The lumen area fraction for harakeke has been found to be 41% which based on an apparent strength of 778 MPa obtained for the fibre neglecting the lumen, suggests a true fibre strength of 1308 MPa [14]. It should also be kept in mind when predicting composite properties that fibre properties vary with direction

Table 2
Mechanical properties of natural and synthetic fibre [6–24].

Fibre	Density (g/cm^3)	Length (mm)	Failure strain (%)	Tensile strength (MPa)	Stiffness/Young's modulus (GPa)	Specific tensile strength ($MPa/g\ cm^{-3}$)	Specific Young's modulus ($GPa/g\ cm^{-3}$)
Ramie	1.5	900–1200	2.0–3.8	400–938	44–128	270–620	29–85
Flax	1.5	5–900	1.2–3.2	345–1830	27–80	230–1220	18–53
Hemp	1.5	5–55	1.6	550–1110	58–70	370–740	39–47
Jute	1.3–1.5	1.5–120	1.5–1.8	393–800	10–55	300–610	7.1–39
Harakeke	1.3	4–5	4.2–5.8	440–990	14–33	338–761	11–25
Sisal	1.3–1.5	900	2.0–2.5	507–855	9.4–28	362–610	6.7–20
Alfa	1.4	350	1.5–2.4	188–308	18–25	134–220	13–18
Cotton	1.5–1.6	10–60	3.0–10	287–800	5.5–13	190–530	3.7–8.4
Coir	1.2	20–150	15–30	131–220	4–6	110–180	3.3–5
Silk-	1.3	Continuous	15–60	100–1500	5–25	100–1500	4–20
Feather	0.9	10–30	6.9	100–203	3–10	112–226	3.3–11
Wool	1.3	38–152	13.2–35	50–315	2.3–5	38–242	1.8–3.8
E-glass	2.5	Continuous	2.5	2000–3000	70	800–1400	29

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relative to the fibre axis, although not surprisingly given the experimental challenge, there is only limited information on transverse data available. The longitudinal Young's modulus for jute has been estimated to be seven times that for the transverse Young's modulus [27].

As would be expected given that the fibres are normally stronger and stiffer than the matrix, strength and stiffness of the composite are generally seen to increase with increased fibre content. However, this relies on having reasonable fibre/matrix interfacial strength, and strength can reduce with strongly hydrophobic matrices such as polypropylene (PP) with increasing fibre content unless coupling agents or some other interfacial engineering method is used; regardless, Young's modulus still generally increases with fibre content but more modestly than when the interface is not optimised [28].

When reasonable interfacial strength is established, composite strength commonly peaks with fibre contents of 40–55 m% for injection moulded thermoplastic matrix composites with reduction at higher contents explained as being due to poor wetting leading to reduced stress transfer across the fibre–matrix interface and increasing porosity (see section on porosity below). Stiffness has been found to increase up to higher fibre contents of around 55–65 m% with similar materials, possibly due to less dependency on interfacial strength than composite strength [14,28–30]. Further insight has been provided by work investigating the influence of fibre content in terms of weight fraction on porosity and volume fraction of fibre. This has shown that maximum volume fractions of fibre occur around fibre contents of 50–60 m% with further addition of fibre resulting in higher porosity rather than increased fibre volume fraction, the influence of which has been incorporated into rule of mixtures models and shown to improve accuracy of prediction for stiffness and strength [27,30,31].

As well as being an issue for short term composite properties, high fibre volume fractions are also of concern due to the potential for increased water uptake leading to degradation of longer term properties. It has been reported that hemp fibre reinforced PP composites with a fibre volume fraction of 0.7 absorbed almost 53 m% water and had not reached saturation after 19 days, whereas only 7 m% water uptake was observed in composites with a fibre volume fraction of 0.3 and saturation had been achieved in the same time period [32].

For composites containing fibres with failure strains lower than that of the matrix (commonly the case for NFCs), basic composite theory suggests that there should be a volume fraction of fibre below which composite strength will be lower than that of the matrix known as the critical volume fraction (V_{crit}). From a fracture mechanics perspective, below V_{crit}, when the fibres fail, the matrix can cope with load transferred from the failed fibres and the fibres are acting merely as holes within the matrix. Critical volume fractions of fibre have been found to be 8.1% and 9.3% for jute and flax respectively in unsaturated polyester (UP), much higher than values obtained for synthetic fibre composites, although lower than fibre contents commonly studied in the literature and so this effect is not observed often [33].

Fibre length, which can be incorporated into the aspect ratio for a fibre (length/diameter), is an important factor influencing the mechanical properties of composites. In a short fibre composite, tensile load is transferred into a fibre from the matrix through shear at the fibre/matrix interface. At the ends of the fibre, the tensile stress are zero and increase along the fibre length; therefore, a fibre needs to have a length of greater than a critical length (L_c) in order for the fibre to be able to be broken during tensile loading of a composite [34]. At the critical length, just prior to fracture, the fibre would theoretically only have been carrying half of the load compared to that of a continuous fibre at the same composite strain. Ideally, fibre length would be much greater than the critical fibre length to allow for efficient reinforcement of a composite such that the majority of the fibre could be loaded as if it were a continuous fibre. L_c can be expressed as follows:

$$\frac{L_c}{d} = \frac{\sigma_f}{2\tau_i} \quad (1)$$

where d is fibre diameter, σ_f is tensile strength of fibre and τ_i is the interfacial strength.

Not surprisingly, L_c has been found to vary with fibre, matrix, fibre treatment and fibre content. L_c values for hemp/PP composites with maleated polypropylene (MAPP) coupling agent were found using composite properties and the Kelly–Tyson model to be 0.49, 0.67, 0.67 and 0.62 mm for fibre contents of 20, 30, 40, and 50 m% respectively (τ_i = 14.5 MPa) [35]. A higher value of 0.83 mm obtained for similar materials was obtained using the fragmentation test (τ_i = 15.4 MPa) with alkali treated fibres [29] with an almost identical value of 0.82 mm (τ_i = 12 MPa) obtained for flax with the same matrix based on the Kelly–Tyson model [36]. Much larger values were obtained from other work using fragmentation for hemp, flax and cotton with PP and MAPP of 3.2, 3.2 and 5.0 mm (τ_i = 14.3, 12.0, 0.7 MPa) respectively and 2.3 mm (τ_i = 22.0 MPa) for sisal [37,38]. Relatively poor bonding between cotton and PP with MAPP was suggested to be influenced by lack of lignin which could potentially bond with PP containing MAPP, which also explains the best interfacial strength being found for hemp with more accessible lignin at the interface than flax. The adhesion between polylactic acid (PLA) and the hemp, flax and cotton was found to be insufficient for fragmentation testing analysis [37]. L_c for flax/thermoset matrix composites has been found to be generally at the lower end of the range. Values of 0.9, 0.5 and 0.4 mm (and τ_i = 13, 28 and 33 MPa) for UP, vinyl ester (VE) and epoxy resins respectively have been obtained using fibre fragmentation supporting their use in NFCs [39]. However, a large L_c of 3 mm (τ_i = 0.9 MPa) has been observed for jute fibre with UP, although this was noted as being an upper bound value [40].

Although increasing fibre length generally increases fibre load bearing efficiency, if fibre length is too long the fibres may get tangled during mixing resulting in poor fibre dispersion which can reduce the overall reinforcement efficiency [41–43].

2.2. Matrix selection

The matrix is an important part of a fibre-reinforced composite. It provides a barrier against adverse environments, protects the surface of the fibres from mechanical abrasion and it transfers load to fibres. The most common matrices currently used in NFCs are polymeric as they are light weight and can be processed at low temperature. Both thermoplastic and thermoset polymers have been used for matrices with natural fibres [44].

Matrix selection is limited by the temperature at which natural fibres degrade. Most of the natural fibres used for reinforcement in natural fibre composite are thermally unstable above 200 °C, although under some circumstances it is possible for them to be processed at higher temperature for a short period of time [45]. Due to this limitation, only thermoplastics that soften below this temperature such as polyethylene (PE), PP, polyolefin, polyvinyl chloride and polystyrene and thermosets (which can be cured below this temperature) are useable as a matrix [46]. However, it should be noted that the thermoplastics named constitute the most common thermoplastics consumed by the plastics industry and far outweighs the use of any other thermoplastic matrices generally used. Indeed, PP and PE are the two most commonly adopted thermoplastic matrices for NFCs. The main thermosets used are unsaturated polyester (UP), epoxy resin, phenol formaldehyde and VE resins. Thermoplastics are capable of being repeatedly softened by the application of heat and hardened by cooling and have

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the potential to be the most easily recycled, which has seen them most favoured in recent commercial uptake, whereas better realisation of the fibre properties are generally achieved using thermosets.

Replacement of petroleum-based with bioderived matrices has been explored. Of these, PLA is the clear front-runner from a mechanical property perspective, and has been shown to give higher strength and stiffness with natural fibres than PP [47].

2.3. Interface strength

Interfacial bonding between fibre and matrix plays a vital role in determining the mechanical properties of composites. Since stress is transferred between matrix and fibres across the interface, good interfacial bonding is required to achieve optimum reinforcement, although, it is possible to have an interface that is too strong, enabling crack propagation which can reduce toughness and strength. However, for plant based fibre composites there is usually limited interaction between the hydrophilic fibres and matrices which are commonly hydrophobic leading to poor interfacial bonding limiting mechanical performance as well as low moisture resistance affecting long term properties. For bonding to occur, fibre and matrix must be brought into intimate contact; wettability can be regarded as an essential precursor to bonding. Insufficient fibre wetting results in interfacial defects which can act as stress concentrators [48]. Fibre wettability has been shown to affect the toughness, tensile and flexural strength of composites [49]. Physical treatment and chemical treatment can improve the wettability of the fibre and thus improve the interfacial strength [50–52].

Interfacial bonding can occur by means of mechanisms of mechanical interlocking, electrostatic bonding, chemical bonding and inter-diffusion bonding [34]. Mechanical interlocking occurs to a greater extent when the fibre surface is rough and increases the interfacial shear strength, but has less influence on the transverse tensile strength. Electrostatic bonding only has significant influence for metallic interfaces. Chemical bonding occurs when there are chemical groups on the fibre surface and in the matrix that can react to form bonds and as a consequence the resulting interfacial strength depends on the type and density of the bonds. Chemical bonding can be achieved through the use of a coupling agent that acts as a bridge between the fibre and matrix. Interdiffusion bonding occurs when atoms and molecules of the fibre and matrix interact at the interface. For polymer interfaces this can involve polymer chains entanglement and depends on the length of chains entangled, the degree of entanglement and number of chains per unit area. It should be noted that it is possible for multiple types of bonding to occur at the same interface at the same time [29].

Extensive research has been carried out in order to achieve improved interfacial bonding in NFCs which can be largely divided into physical and chemical approaches. Physical approaches include corona, plasma, ultraviolet (UV), heat treatments electron radiation and fibre beating. Corona treatment uses plasma generated by the application of a high voltage to sharp electrode tips separated by quartz at low temperature and atmospheric pressure and commonly includes the use of oxygen-containing species [17]. It has been shown to bring about chemical and physical changes of fibres including increased surface polarity (thought to be due to increased carboxyl and hydroxyl groups) and increased fibre roughness but is known to be difficult to apply to three-dimensional surfaces including fibres [53,54]. Gassan and Gutowski have used corona plasma and UV to treat jute fibres which were both found to increase the polarity of fibres but decrease fibre strength leading to reduced composite strength with corona treatment, but improvement of up to 30% flexural strength of epoxy matrix composites with UV treatment [54]. Plasma treatment is similar to corona treatment but is performed using a vacuum chamber with gas continuously supplied to maintain the appropriate pressure and gas composition [17]. Plasma treatment has been shown to bring about hydrophobicity at fibre surfaces and increase fibre surface roughness increasing interfacial adhesion [51]. Using plasma treatment, improvement of interlaminar shear strength and flexural strength in NFCs have been increased up to 35% and 30% respectively [55]. Heat treatment involves heating the fibres at temperatures close to those that bring about fibre degradation and can affect physical, chemical and mechanical properties of the fibres including water content, chemistry, cellulose crystallinity, degree of polymerisation and strength. Specific chemical changes include chain scission, free radical production and formation of carbonyl, carboxyl and peroxide groups [17]. Similar to the corona and plasma treatment, the effect of heat treatment relies on time, temperature and composition of the gases involved during the treatment. Cao et al. obtained improvements of kenaf fibre tensile strength of over 60% using heat treatment explained as being due to increased fibre crystallinity [56]. Heat treatment has also been seen to give good improvement of sisal fibre strength (37%) [57]. This was associated with removal of aromatic impurities as well as increased fibre crystallinity. However, heat treatment resulted in more modest increases of composite properties in this work; tensile strength, Young's modulus, flexural strength and flexural modulus improvements for composites were 10%, 4%, 27% and 33% respectively. Electron radiation has been seen to improve interfacial bonding with natural fibres and PP by between 21% and 53% explained as due to producing free radicals that encourage crosslinking between the fibre and the matrix [58]. Fibre beating has brought about a 10% increase in strength of kraft fibre reinforced PP, which can be explained due to fibre defibrillation and the associated increased surface area and mechanical interlocking [59].

Chemical approaches are more represented within the literature than physical with better improvements obtained to date. Chemical treatments include alkali, acetyl, silane, benzyl, acryl, permanganate, peroxide, isocyanate, titanate, zirconate and acrylonitrile treatments and use of maleated anhydride grafted coupling agent [47,60]. The most popular are alkali, acetyl, silane and maleated anhydride grafted coupling agent, but enzyme treatment is becoming increasingly popular with particular benefit relating to environmental friendliness [47].

Alkali treatment removes fibre constituents including hemicellulose, lignin, pectin, fat and wax which exposes cellulose and increases surface roughness/area providing for improved interfacial bonding. Alkali treatment also modifies cellulose structure; modest treatments have been seen to bring about increased cellulose crystallinity considered to be due to removal of materials that could obstruct cellulose crystallinity, whereas at harsher treatments crystalline cellulose has been converted to amorphous material [61,62]. Improvement of fibre strength has also been obtained using alkali treatment [61,63]. Many studies have reported improvements in interfacial shear strength (IFSS) and improved tensile strength, Young's modulus, failure strain, impact strength, fracture toughness and flexural properties of composites as well as thermal stability and long term moisture resistance, the latter of which could be due to the reduced moisture uptake observed with alkali treated natural fibres [63–68]. For crystallisable matrices, fibre treatment with alkali has also been seen to influence the degree of matrix crystallinity, with exposed cellulose acting as a nucleation site for crystalline polymer [69].

During acetylation of natural fibres, esterification occurs by reaction of acetyl groups (CH_3CO–) with hydroxyl groups (–OH) on the fibres resulting in increased hydrophobicity (see Fig. 1) [70]. This has been shown to improve interfacial bonding, tensile

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and flexural strength and stiffness, as well as dimensional and thermal stability and resistance to fungal attack in NFCs [71–73]. However, over-treatment has been seen to be deleterious to mechanical properties, assumed to be due to degradation of cellulose and cracking of fibres known to occur with the catalysts used in this process [71]. Acetylation treatment has also been found to reduce impact strength of the composite. Commonly acetylation is preceded by an alkaline treatment.

Silanes used for treatment of fibres have different functional groups at either end such that interaction at one end can occur with hydrophilic groups of the fibre whilst the other end can interact with hydrophobic groups in the matrix to form a bridge between them. Initially, silane treatment of natural fibres involves hydrolysis of alkoxy groups on silane with water to form silanol (Si–OH) groups which can then react with hydroxyl groups on the fibre surface as shown in Fig. 2 [74,75]; hydrogen or covalent bonding can occur. The most commonly reported silanes used are amino, methacryl, glycidoxy and alkyl silanes. Silanes have been found to increase the hydrophobicity of natural fibres and strength of NFCs with larger increases occurring when covalent bonding occurs between silane and the matrix [75,76].

Maleic anhydride (MA) grafted polymers are widely used as coupling agents to improve composite properties. MA is commonly grafted to the same polymer as that used as the matrix to ensure compatibility between the matrix and the coupling agent. MAPP, produced by grafting MA to PP, is the most commonly seen in the literature. MAPP can react with the hydroxyl groups on fibre surfaces leading to covalent or hydrogen bonding Fig. 3. It can be used as an additive during processing or grafted to the fibre prior to processing. It has been shown to improve tensile and flexural strength and stiffness as well as impact strength of PP matrix composites [77]. Of all the methods of improving interfacial bonding, coupling with MAPP could be regarded as the most successful. It has been shown to give almost twice the composite strength as obtained with silane treatment [63]. Further to improved bonding, improvement of mechanical performance with MAPP has been explained as being due to its ability to wet fibre and enhance its dispersion [78]. MA grafted PLA has been shown similarly to increase properties of PLA matrix NFCs as well as thermal stability [79–81].

Improvement of composite properties has been seen with the application of enzyme treatment, with tensile and flexural strength of abaca/PP composites seen to improve by 45% and 35% respectively which was considered to be due to removal of fibre components and increased surface area leading to increased interfacial bonding [82]. Impact properties were also found to increase by 25%.

Further to improvements of instantaneous mechanical performance, chemical surface treatment has also been found to improve longer term mechanical performance of NFCs subjected to wet and humid conditions [60].

2.4. Fibre dispersion

Fibre dispersion has been identified as a major factor influencing the properties of short fibre composites and a particular challenge for NFCs, which commonly have hydrophilic fibres and hydrophobic matrices [83]. Use of longer fibres can further increase their tendency to agglomerate. Good fibre dispersion promotes good interfacial bonding, reducing voids by ensuring that fibres are fully surrounded by the matrix [84]. Dispersion can be influenced by processing parameters such as temperature and pressure; additives such as stearic acid have been used in PP and PE to modify dispersion as well as those used to increase interfacial bonding such as MAPP which encourage fibre matrix interaction [83]. Similarly, fibre modification through grafting can also be employed, but is more expensive [83]. Although the use of more intensive mixing processes such as using a twin-screw extruder rather than a single screw extruder leads to better fibre dispersion, this is generally at the cost of fibre damage and fibre lengths are found to reduce dramatically during such processing depending on temperature and screw configuration [61,83].

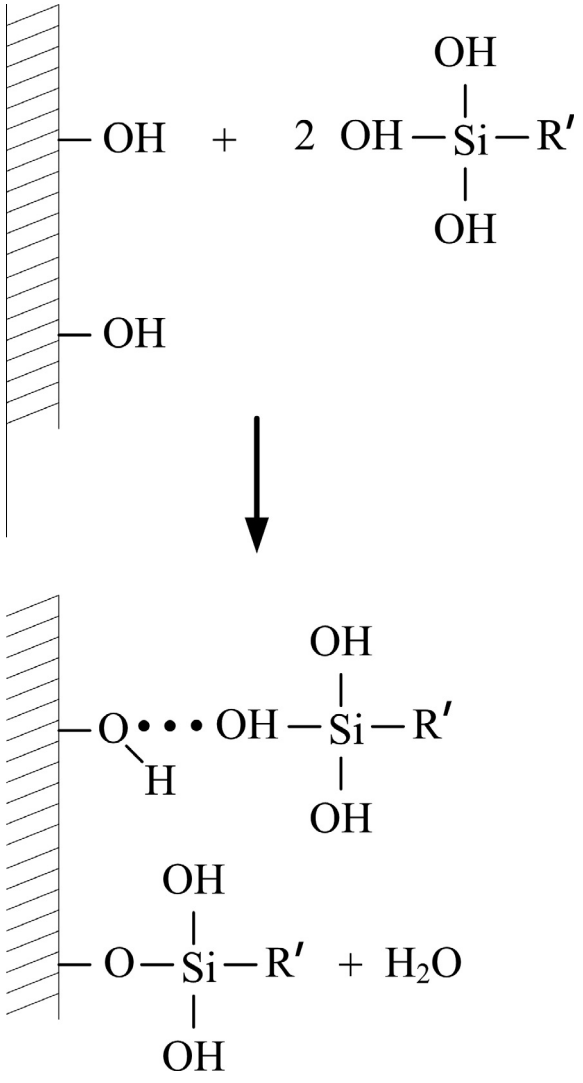

Fig. 2. Reaction of silane with natural fibre (R' representing organic group,... representing hydrogen bonding).

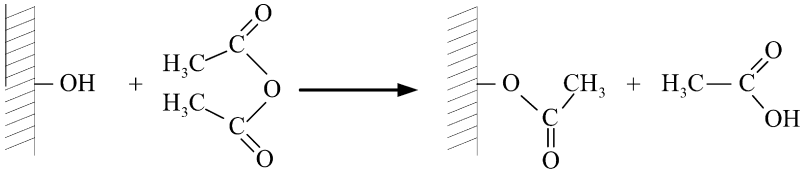

Fig. 1. Reaction of acetic anhydride with natural fibre.

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2.5. Fibre orientation

The best mechanical properties can generally be obtained for composites when the fibre is aligned parallel to the direction of the applied load [85–87]. However, it is more difficult to get alignment with natural fibres than for continuous synthetic fibres. Some alignment is achieved during injection moulding, dependent on matrix viscosity and mould design [88]. However, to get to higher degrees of fibre alignment, long natural fibre can be carded and placed manually in sheets prior to matrix impregnation. Alternatively, traditional textile processing of fibres can be employed including spinning to enable a continuous yarn to be produced. However, as the name suggests, this does involve a degree of fibre twist. Aligned fibre yarns can also be produced by wrap spinning, a method used since the 1970s in the textile industry; here short fibre can be converted to a continuous form through the use of a continuous strand wrapped around discontinuous fibre with sufficient frequency to provide the required integrity for subsequent processing. The continuous strand can be from the same type of fibre as the short fibre or can be thermoplastic and form the matrix material during compression moulding (CM). Thermoplastic fibre to be converted to the matrix can also be used aligned in the yarn direction to act as support for the natural fibre. Research comparing aligned fibre yarn with conventional twisted yarns for flax epoxy composites has demonstrated improved tensile and flexural strength and stiffness with unidirectional yarn [89]. More recently, continuous fibre tape has been produced through using the fibres own pectin as an adhesive using a water mist and then drying whilst stretched [90]. Continuous material can be employed in processing in much the same way as continuous synthetic fibre (e.g. filament winding, pultrusion) to give good degrees of fibre alignment within the composite, although it should not be forgotten that this is not the same as alignment of the cellulose chains as these tend to have an angle relative to the elemental fibre direction (microfibrillar angle).

A recently utilised alternative to the textile route with its extensive infrastructure requirements to bring about fibre alignment in composites is that of dynamic sheet forming (DSF). DSF is a method used to align fibre traditionally in paper production. Here, short fibres are suspended in water and sprayed through a nozzle onto a rotating drum covered with a wire mesh through which the water can be removed, which brings about alignment in the spray and rotation direction. This has provided improvements of mechanical performance compared to other short fibre processing techniques; recent work at the University of Waikato yet to be published has obtained strengths of over 100 MPa for short hemp and harakeke fibre aligned using DSF in PLA and epoxy matrices.

Regarding the degree of influence of orientation on mechanical performance of NFCs, similar large reductions of strength and Young's modulus to those seen with synthetic fibres have been obtained with increasing fibre orientation angle relative to the test direction. One study on aligned Alfa fibre reinforced UP showed strengths compared to those obtained in the fibre direction (0°) of 69%, 29%, 22% and 12% at angles of 10°, 30°, 45° and 90° respectively and corresponding Young's moduli of 93%, 66%, 52% and 41% of that in the fibre direction [11]. In another study on hemp/PLA composites, those with fibre aligned at 45° and 90° were found to have 48% and 30% of the strength and 53% and 42% of the Young's moduli of those with 0° fibre [91].

2.6. Manufacturing

The most common methods used for NFCs are extrusion, injection moulding (IM) and compression moulding. Resin transfer moulding (RTM) is also used with thermoset matrices and pultrusion has been successfully employed for combined flax /PP yarn composites and thermoset matrix composites [92,93]. Factors determining properties include temperature, pressure and speed of processing. It is possible for fibre degradation to occur if the temperature used is too high, which limits the thermoplastic matrices used to those with melting points lower than the temperature at which degradation will occur.

In extrusion, thermoplastic, usually in the form of beads or pellets, is softened and mixed with the fibre transported by means of a single or two rotating screws, compressed and forced out of the chamber at a steady rate through a die. High screw speed can result in air entrapment, excessive melt temperatures and fibre breakage. Low speeds, however, lead to poor mixing and insufficient wetting of the fibres. This method is used on its own or for producing pre-cursor for IM. Twin screw systems have been shown to give better dispersion of fibres and better mechanical performance than single screw extruders [94].

IM of composites can be carried out with thermoset or thermoplastic matrices, although is much more often used for thermoplastic matrices. Variation of fibre orientation occurs across the mould section with shear flow along the walls due to friction resulting in fibre aligned along the mould wall whilst a higher stretch rate at the centre produces fibre that is more transversely aligned to the flow direction, a structure referred to as skin core structure [95]. Alignment is more significant with higher fibre contents. Residual stress in thermoplastic matrix composites due to pressure gradients, non-uniform temperature profiles, polymer chain alignment and differences in fibre and matrix thermal expansion coefficients can reduce composite strength [96]. Due to the viscosity requirements, IM of such composites is generally limited to composites of less than 40 m% fibre content. Fibre attrition in IM as for extrusion reduces the length of the fibre during processing.

Compression moulding (CM) is generally used for thermoplastic matrices with loose chopped fibre or mats of short or long fibre either randomly oriented or aligned, but can also be used with thermoset matrices. The fibres are normally stacked alternately with thermoplastic matrix sheets before pressure and heat are applied. The viscosity of the matrix during pressing and heating needs to be carefully controlled, in particular for thick samples to make sure the matrix is impregnated fully into the space between fibres. Good quality composites can be produced by controlling viscosity, pressure, holding time, temperature taking account of the type of fibre and matrix [96]. Film stacking has been recommended as it limits natural fibre degradation due to involvement of only one temperature cycle [97]. Temperature still needs to be carefully controlled as commonly there is little difference in temperature between that at which a particular matrix can be processed and that at which fibre degradation will occur. Reduction of fibre strength has been shown to occur at temperatures as low as 150 °C and at 200 °C, with strength reducing by 10% in ten minutes [98]. Overall, there is a compromise between obtaining good wetting and avoiding fibre degradation that leads to an optimum temperature for a particular composite material/geometry. This has been explored with film stacking of flax reinforced poly(ester amide) composites, with an optimum temperature for composite tensile properties found to be 150 °C [99]. Flexural properties were found to be less dependent on temperature below 150 °C, but reduced significantly at higher temperatures. For composites made from jute yarn and the bacterial copolyester Biopol® the optimum compression temperature for a range of mechanical properties was found to be approximately 180 °C [1]. The highest strength was obtained at 200 °C for non-woven mat reinforced PP [100]. Alternatively to film stacking, sheet moulding compounds have been used in CM [101].

In RTM, liquid thermoset resin is injected into a mould containing a fibre preform. The main variables with this process are temperature, injection pressure, resin viscosity, preform

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architecture and mould configuration [96]. Advantages compared with other processes include lower temperature requirements and avoidance of thermomechanical degradation [102]. Compaction in this process is affected by the structure of natural fibres including the effect of lumen closing and due to lower degrees of fibre alignment, natural fibre composites are less compactable than glass fibre composites [102]. Good component strength can be achieved with this process which is suitable for low production runs [17].

Processing that is absent from the literature currently, but likely to be of significance in the future is that of additive fabrication or 3-D printing of NFCs. Early work at the University of Waikato, yet to be published, shows potential for NFC printing filament to improve the capabilities of components produced through fused deposition modelling printers. Here there is benefit of orientation that occurs when composite material is extruded through a fine nozzle. It is expected that this will be an area of large growth in the very near future.

2.7. Porosity

An often overlooked component of NFCs, porosity has long been known to have shown to have a large influence on mechanical properties of composites in general and much effort has gone into reducing it in synthetic fibre composites. It arises due to inclusion of air during processing, limited wettability of fibres, lumens and other hollow features within fibres/fibre bundles (which may become closed during processing at high pressure) and due to the low ability of fibres to compact [30]. Porosity in NFCs has been shown to increase with fibre content, more rapidly once the geometrical compaction limit has been exceeded, dependent on fibre type and orientation of fibre; flax/PP composites were found to have porosity increasing from 4 to 8 volume% as fibre content increased from 56 to 72 m% [27]. As mentioned earlier, its inclusion in models has been shown to give improved prediction of strength and stiffness.

3. Mechanical properties

There is a large amount of literature detailing the mechanical performance of NFCs. A graphical overview of the range of strength, stiffnesses, specific stiffnesses and specific strengths compared with those for glass fibre reinforced plastics produced recently by Shah [103] is shown in Fig. 4. More details of mechanical properties obtained including tensile and flexural strength and stiffnesses, as well as impact strength are provided in Table 3, arranged similarly to Shah's graph in that more aligned long fibre composites are given at the top followed by multiaxial, then short fibre aligned, with random aligned including IM composites further down, along with regenerated cellulose and glass fibre reinforced plastics (GFRPs) for comparison at the bottom. Within each section, thermoset matrix composites are detailed first. It would appear that similar composite optimisation is effective for strength and stiffness; Shah found strength as well as specific strength to be largely proportional to stiffness and specific stiffness respectively for NFCs as well as the trends for relative strength and stiffness maintained for specific strength and stiffness [103].

It can be seen that alignment of fibre is a major factor influencing composite properties, with the best tensile, flexural and impact properties achieved for aligned NFCs. The highest strength is achieved at approximately 73 m% fibre for aligned fibre composites, a higher fibre content than that from which the best strengths can be achieved with more randomly aligned/shorter fibre composites, presumably due to the higher compaction limit with more aligned fibre. Interestingly, the fibre with which the highest tensile strength was achieved is sisal, which from Table 2, would not be expected to bring about such high composite strengths as flax. This suggests that either the properties in Table 2 do not fully reflect the spectrum achievable with natural fibres, which would not be surprising given the limited data available and/or that the strength of the fibre is of less influence than other factors such as its aspect ratio, extraction method, treatment or ability to be aligned within the composite and manufacturing method. Comparison of composites reinforced using wrap spun yarn with conventionally twisted yarn have shown better flexural properties using discontinuous hemp with PP fibre as a carrier fibre, wrap spun by PP as well as flax sliver with PP fibre also wrap spun using PP for unidirectional composites [104]. Recent work using wrap spun hemp/PLA yarn from bleached hemp yarn and continuous PLA strand to produce oriented prepreg (pre-impregnated fibre) has given a reasonable combination of mechanical properties for biaxial laminates including relatively high Charpy impact energy (25 kJ/m^2) [105]. The same group extended wrap spinning of PLA to produce yarn from short hemp fibre with promising results for this technique [91]. Although it is early days in the development of such material for composites, it would be expected that this approach will produce high mechanical performance.

It should be realised that NFC composites with aligned fibres either are produced by hand if the fibre is in a reasonably raw form or require textile infrastructure or alternative processing to produce a continuous fibre form. Although yarn and sliver give good performance, processing is particularly intensive with many stages including skutching, carding or hackling and spinning for which each stage requiring specialist equipment [106,107].

Relatively high values (136 MPa with epoxy and 101 MPa with PLA) have been obtained at Waikato University with short fibres aligned using dynamic sheet forming; these overlap at the lower end of the strength range for those obtained using long aligned fibre composites, but not achieving the higher strengths obtained with longer fibre. Fibre length is shown to be important for more randomly arranged fibre composites with those having long fibre performing better than those with shorter fibre. Generally the highest tensile and flexural properties for NFCs are achieved with

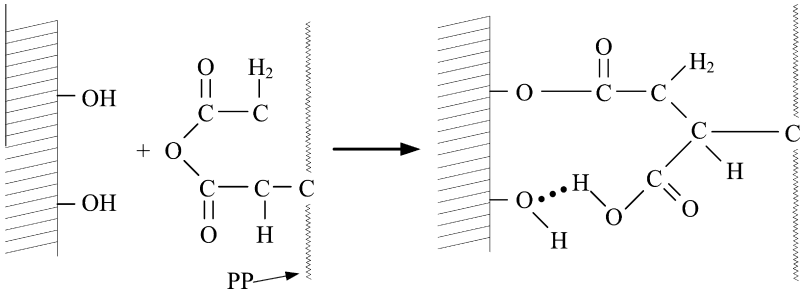

Fig. 3. Interaction of maleated polypropylene with natural fibre (. . .representing hydrogen bonding).

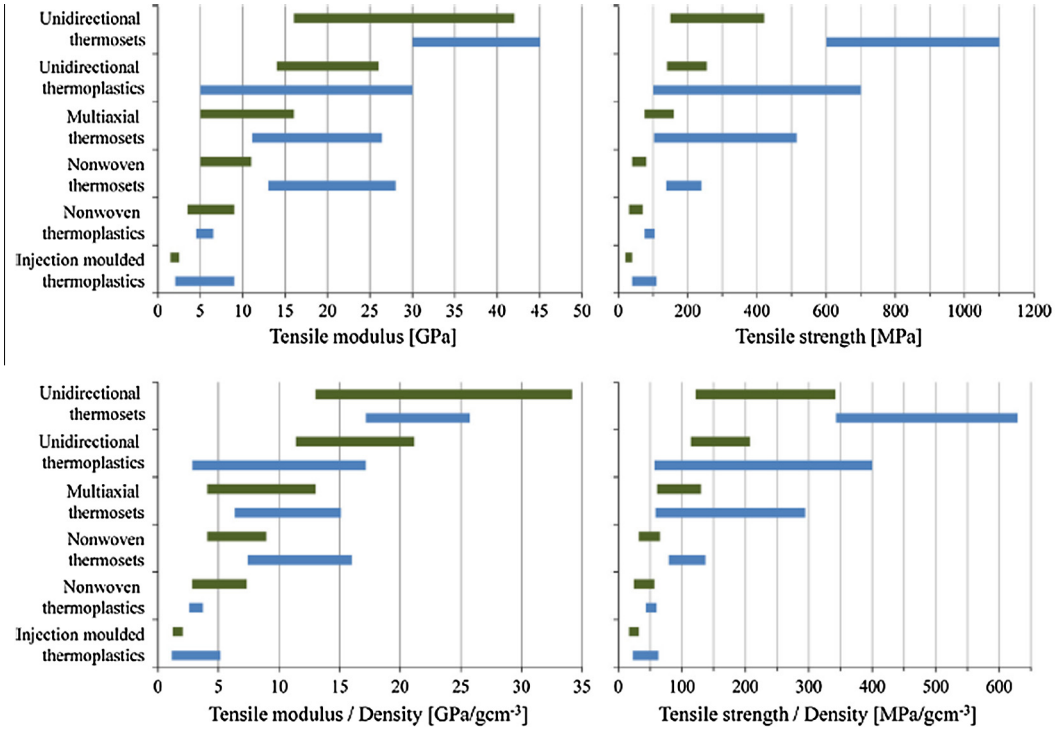

Fig. 4. Comparison of stiffness (tensile modulus), strength and specific stiffness (tensile modulus/density) and specific strength (strength/density) of NFCs (upper of paired bars) with glass fibre reinforced plastics (lower of paired bars) reprinted from Materials and Design, 62, Darshil U. Shah, Natural fibre composites: Comprehensive Ashby-type materials selection charts, 21–31, 2014, with permission from Elsevier. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

thermoset matrices for which the highest values in descending order are for epoxy, vinyl ester then unsaturated polyester, relating to the order of expected degree of adhesion with the fibre and hence stress transfer capability. However, it should not be overlooked that very high strengths are achieved with PP (321 MPa) and PLA (223 MPa) with flax and kenaf respectively, which appears to be due to good quality fibre and composite processing as well as a high fibre content. The best tensile strength for random/IM NFCs have a PLA matrix, however, it should be noted that thermosets are less commonly used for these composites due to their higher cost and the lower performance expected. PLA quite consistently outperforms PP as a matrix material, not surprisingly given its higher properties, supporting this bioderived material for application. Another bioderived matrix, shellac, can also be seen to compare favourably [98]. However, flexural strength as high as 188 MPa with randomly aligned alkali treated fibre can be seen in Table 3 for harakeke fibre with epoxy resin, supporting the use of vacuum bagging followed by CM for quality manufacture. RTM is also shown to be able to be used to produce high performing NFCs; the highest stiffness in Table 3 (39 GPa) is produced this way, however this work also supports the importance of fibre quality with biotechnical retting employed with fibre produced in a northern climate with long daylight growing hours noted for their contribution to performance [108]. It has been raised that the highest fibre and lowest void contents occur with pre-pregging of fibre followed by compression moulding, then RTM and hand-lay-up which matches a trend in reducing tensile properties [103]. Important processing details raised by Phillips et al. are the need for evacuation of moisture during cure of thermoset matrix composites and reduction of crimp [109]. High strength and stiffness for random film stacked flax fibre has been explained by high fibre strength (1339 MPa) along with processing facilitating good interfacial bonding whilst avoiding fibre degradation [97].

MAPP has been used as a coupling agent much more commonly with short rather than long fibre PP matrix composites to bring about improved mechanical performance, presumably due to the increased requirement for interfacial stress transfer with short fibre composites. The greatest impact resistant composites are those with PP matrices for which, as for other properties, long fibre NFCs have the highest values. Impact toughness has been found to reduce with addition of natural fibre to tough thermoplastic matrices such as PP, although less so with addition of MAPP, but has been seen to improve with addition of natural fibre to PLA and PHB [110,111]. Charpy impact has been seen to increase for UP almost linearly with increased fibre content [112]. Untreated sisal fibre has been shown to give a ten-fold increase in Charpy impact strength compared to UP alone with acrylamide and permanganate fibre treatments improving Charpy impact strength, but acetylation, alkali and silane treatments having a negative influence [113]. The impact strength of epoxy resin has been seen to reduce with addition of fibre up to 25 m% fibre, but then increase to give an overall improvement in impact strength of 40% at a fibre content of 40 m% [114].

Although impact toughness gives an indication of the tendency for a material to behave in a brittle manner, plane strain fracture toughness (k_{ic}) is required to prevent brittle fracture. Nonetheless, data is quite scarce for k_{ic} of NFCs. k_{ic} of injection moulded short hemp fibre composites was found to decrease with fibre content from 2 MPa $m^{1/2}$ for just PLA to 1.6 MPa $m^{1/2}$ with 30 m% fibre [69]. Alkali fibre treatment brought about further reduction of k_{ic} to 1 MPa $m^{1/2}$. Reduction of k_{ic} also occurred for long hemp fibre/PLA composites in the same work, but to less of a degree than with shorter fibre. However in contrast, k_{ic} values increased for PLA with addition of long hemp fibre from 2 MPa $m^{1/2}$ to 3.3 MPa $m^{1/2}$ for untreated fibre and 3.9 MPa $m^{1/2}$ for alkali treated long fibre [67]. From these contradictory results, it seems possible that the

Table 3
Mechanical properties of natural fibre composites compared with regenerated cellulose composites and GFRPs.

Fibre	Matrix	Fibre content (m%)	Tensile strength (MPa)	Stiffness/Young's modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	Charpy (c) or Izod (i) impact strength (kJ/m^2 or J/m)	Notes: Processing/length/ treatment	Reference
Sisal (aligned)	Epoxy	~73	410	6	320	27		Alkali treated bundles CM/leaky mould	[57]
Sisal (aligned)	Epoxy	~77	330	10	290	22		Untreated bundles CM/leaky mould	[57]
Flax (aligned)	Epoxy	46/54	280/279	35/39				Enzyme extracted RTM	[108]
Harakeke (aligned)	Epoxy	50/55	223	17	223	14		CM	[14]
Harakeke (aligned)	Epoxy	52	211	15				CM	[115]
Sisal (aligned)	Epoxy	48	211	20				RTM	[116]
Sisal (aligned)	Epoxy	37	183	15				RTM	[116]
Flax (yarn) (aligned)	Epoxy	45			311	25		Not stated	[106]
Hemp (aligned)	Epoxy	65	165	17	180	9	15 (c)	CM	[117]
Flax yarn (aligned)	Epoxy	~31	160	15	190	15		Hand lay–up (knitted yarn)	[118]
Flax yarn (aligned)	Epoxy	45	133	28	218	18		Autoclave	[106]
Flax (aligned)	Epoxy	37	132	15				RTM	[116]
Flax hackled (aligned)	Epoxy	~28			182	20		Pultruded	[118]
Flax yarn (aligned)	VE	~24	248	24				RTM	[118]
Flax (sliver) (aligned)	UP	~58	304	30				Soxhlet extracted Vacuum impregnated/CM	[107]
Flax yarn (aligned)	UP	~34	143	14	198	17		RTM (knitted yarn)	[118]
Alfa (aligned)	UP	48	149	12				Alkali treated then bleached	[11]
Flax yarn (aligned)	PP	72	321	29				Filament wound	[27]
Flax yarn (aligned)	PP	30	89/70	7/6			88/115 (c)	Pultruded flax/PP yarn	[92]
Flax (aligned)	PP	50	40	7			751 (i)	Needle punched flax/PP mats CM	[119]
Flax (aligned)	PP	39			212	23		Dew retted, boiled, MAA-PP coupled	[100]
Flax sliver (aligned)	PP	44			146	15		Wrap spun flax sliver/PP hybrid yarn, CM	[104]
Hemp (aligned)	PP	46			127	11		Wrap spun, short hemp/PP hybrid yarn, CM	[104]
Kenaf selected (aligned)	PLA	~80	223	23	254	22		Emulsion PLA Prepreg CM	[120]
Hemp (carded)	PLA	30	83	11	143	7	9 (c)	Alkali treated CM	[67]
Kenaf (aligned)	PLA	40	82	8	126	7	14 (c)	CM	[111]
Hemp (aligned)	PLA	30	77	10	101	7	19 (c)	Wrap spun alkali treated short hemp hybrid yarn, CM	[91]
Kenaf (aligned)	PHB	40	70	6	101	7	10 (c)	CM	[111]
Flax sliver biaxial/major axis	Epoxy	~46	200	17	194	13		Wrap spun sliver, woven, weft:warp strength 10:1	[89]
Flax (woven)	Epoxy	~50	104	10				Sized and dried prior to pre-preg	[109]
Flax yarn (woven)	VE	~35	111	10	128	10		RTM	[118]
Jute (woven)	UP	35	50	8	103	7	11 (c)	RTM	[93]
Hemp (biaxial)	PLA	45	62	7	124	9	25 (c)	Wrap spun bleached hemp hybrid yarn, CM	[105]
Harakeke (DSF)	Epoxy	45	136	11	155	10	10 (c)	Alkali treated CM	Waikato
Hemp (DSF)	Epoxy	50	105	9	126	8		Alkali treated CM	Waikato
Hemp (DSF)	Epoxy	65	113	18	145	10	11 (c)	CM	[117]
Harakeke (DSF)	PLA	30	102	8				Alkali treated CM	Waikato
Hemp (DSF)	PLA	25	87	9				Alkali treated CM	Waikato

(*continued on next page*)

Table 3 (*continued*)

Fibre	Matrix	Fibre content (m%)	Tensile strength (MPa)	Stiffness/Young's modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	Charpy (c) or Izod (i) impact strength (kJ/m^2 or J/m)	Notes: Processing/length/ treatment	Reference
Flax (short-non woven)	Shellac	~49	109	10					[98]
Harakeke (random)	Epoxy	45			188	9		Alkali treated Vacuum bagged CM	[121]
Flax (random)	UP	39	61	6	91	5	13 (c)	RTM	[93]
PALF (random)	UP	30	53	2	80	3	24 (c)	CM	[43]
Wood BKP	PP	40	50	3	78	3	40 (i)	MAPP coupled IM	[122]
Flax	PP	30			74[b]		22[c] (c)	MAPP coupled IM	[123]
Jute	PP	60	74	11	112	12	195 (i)	MAPP coupled IM	[124]
Newsprint	PP	40	53	3	94	4	200 (i)	MAPP coupled IM	[125]
Kraft	PP	40	52	3	90	4	235 (i)	MAPP coupled IM	[125]
Hemp	PP	40	52	4	86	4	210 (i)	MAPP coupled IM	[125]
Kenaf (random)	PP	30	46	5	58	4	39 (i)	CM	[126]
Flax	PP	30	52	5	60	5	18 (c)	IM	[71]
Flax (random)	PLA	30	100	8				Dew retted Stripped/combed (strength 1339 MPa) Film stacking	[97]
Flax (random)	PLLA	30	99	9				Dew retted, stripped, combed (strength 1339 MPa) Film stacking	[97]
Hemp (random)	PLA	47	55	9	113				[127]
Cellulose (continuous)	Bio-Epoxy		92	9	727	27	26 (c)	RTM	[128]
Cordenka[a]	PA	30	120	6				IM	[129]
Cordenka[a]	PP	42	90	4	–	–	87 (c)	MAPP coupled IM	[130]
Lyocell[a] (carded)	PLA	30	89	9	148	6	52 (c)	CM	[111]
Cordenka[a]	PLA	25	108	4	–	–	70 (c)	IM	[130]
Lyocell[a] (carded)	PHB	30	66	5	105	5	70 (c)	CM	[111]
E-glass (unidirectional)	VE	~60	905	39					[118]
E-glass (aligned)	UP	~60	695	31				CM	[107]
E-glass (aligned)	Epoxy	~41			450	21		Pultruded	[118]
E-glass (woven)	VE	~59	483	33					[118]
E-glass (CSM)	Epoxy	38–40			250	9			[121]
E-glass (CSM)	UP	~47	201	13	278	11	107 (c)	RTM	[93]

BKP = bleached kraft pulp, PALF = pineapple leaf fibre.
PHB = poly(3-hydroxybutyrate), PLLA = L-polylactide acid, PA = Polyamide.
MAA-PP = maleic acid anhydride modified PP.
CSM = chopped strand mat.
[a] Lyocell/Cordenka = regenerated cellulose fibre.
[b] High molecular weight MAPP
[c] Rubbery MAPP.

influence of treatment and fibre length are large influences on the fracture mechanics and plane strain fracture toughness.

Regenerated cellulose fibre composites (cellulose, Lyocell and Cordenka) can be seen in Table 3 to have useful properties. Indeed, properties for IM material generally surpass values seen with IM NFCs. Impact strength of 72 kJ/m^2 obtained for 30 m% cordenka fibre reinforced PLA is reported as the highest value seen in the literature for any bio-degradable composite [131]. Improvements of impact strength and tensile strength were also obtained by Bledzki et al. [132] for composites made from PLA with abaca and synthetic cellulose using IM; notched Charpy impact strength of composites with 30 m% abaca and cellulose fibres were improved by 120% (5.7 kJ/m^2) and 360% (7.9 kJ/m^2) compared to PLA alone. Comparison of hybrid hemp/Lyocell fibre composites with hemp or Lyocell only fibre composites showed improvement of tensile and impact strengths but reduction of stiffness [133]. The flexural strength of 727 MPa obtained with RTM cellulose bio-epoxy is outstanding, however, the tensile properties are more modest. Use of regenerated fibres has the benefit of consistent quality.

Due to the advantages of NFC given in Table 1, there is a strong driving force to use them to replace GFRPs. From Fig. 4, it can be seen that stiffness (tensile modulus) values for NFCs approach the upper values obtained for GFRPs, although strength falls well short. Taking account of density provides a more favourable comparison; higher specific stiffnesses can be achieved for NFCs than GFRPs, but the highest specific strengths are obtained with GRRPs. There is though good overlap of specific strength for NFCs and GFRPs with thermoplastic matrices. A number of studies have involved comparing GFRPs with NFCs made using the same procedures. Specific stiffness of flax fibre reinforced PLLA composites were found to be similar to those for glass fibre reinforced UP for random aligned film stacked composites, but specific strength was 20% higher for GFRPs [97]. Unidirectional (yarn or sliver) hemp fibre reinforced SCONACELL A (starch based matrix) laminates have been found to have 143% of the specific stiffness of glass fibre reinforced epoxy composites at similar fibre volume fraction (0.5) although only 59% of their specific strength [98]. Flexural modulus was found to be almost similar for flax/epoxy composites to epoxy reinforced with glass CSM at the same weight fraction but the flexural strength was only about two-thirds that for the GFRPs [121].

Impact strength is considered to be one of the weaknesses of NFCs [47]. The highest Charpy impact energy found in the literature is for unidirectional pultruded flax/PP composites of 115 kJ/m^2 [92]. However, based on relatively low flexural properties (strength/modulus 70 MPa/6 GPa) for the same composites, it was assumed that this was due to poor impregnation of fibre. Processing aimed at improving impregnation improved the flexural properties by around 15%, but reduced the Charpy impact energy to around 35 kJ/m^2. Higher die temperature combined with faster pulling speed lead to a composite more rounded in terms of mechanical properties with flexural strength, flexural modulus and Charpy impact strength of 89 MPa, 7 GPa and 88 kJ/m^2 respectively. The highest Izod impact strength (751 J/m) has been achieved with needle punched flax/PP compression moulded mat in the 0° direction [119].

Overall, it has been stated that composites reinforced with natural fibres compare well with GFRPs with respect to stiffness and cost, but not for impact and tensile strength or water absorption [93].

Development of nanocellulose fibre reinforced composites has been inspired by the high predicted mechanical properties of nanocellulose. Although strength and stiffness of nanocellulose have not been directly measured, predictions for strength range from 300 MPa to 23 GPa (fragmentation experiments support strengths between 1.3 and 1.6 GPa) and stiffness from 58 to 300 GPa [134]. Sources of nanocellulose are plentiful including the same sources from which macroscopic cellulose based fibres described already are obtained, as well as animals, algae and bacteria. However, processing is a challenge for these fibres with extraction generally involving multiple stages resulting in fibre suspensions, the use of which needs to avoid fibre aggregation and generally limits the composite matrices used [17]. Nanocellulose composite strengths and stiffnesses of up to 420 MPa and 21 GPa respectively have been reported based on use of bacterial nanocellulose, which puts them at the upper end of what can be achieved with NFCs [134].

4. Hybridisation

Recent studies have yielded promising results with hybridization of natural fibres for reinforcement. The variation in mechanical properties such as tensile, flexural and impact strength of hybridized kenaf and pineapple leaf fibre reinforced high-density polyethylene (HDPE) composites has been studied [135]; it was found that pineapple leaf fibre increased the tensile and flexural strength whilst kenaf improved impact strength and reduced water absorption. Evaluation of the effect of hybridization on mechanical performance of short banana/sisal hybrid fibre reinforced polyester composites found that tensile properties of NFCs were improved with addition of banana fibres [136]. The maximum tensile strength (58 MPa) was obtained for composites having a ratio of banana:sisal of 3:1 at 67 vol% fibre content. The results were explained as being due to the smaller diameters of banana fibres compared to sisal fibres and better stress transfer in unit area of banana/polyester composite.

5. Influence of moisture/weathering

As mentioned previously, moisture absorption is one of the main disadvantages experienced with NFCs. It has been shown to increase with increased fibre content and temperature as well as being influenced by fibre treatment/coupling agent and fibre arrangement. It is commonly associated with swelling of NFCs and reduced mechanical performance with the exception of impact energy which is commonly seen to increase.

Even with hydrophobic matrices such as PP, tensile and flexural properties of NFCs have been found to reduce considerably over a few weeks submersion in water with degradation rate increasing at higher temperatures [32,137]. MAPP used as a coupling agent has been seen to slow moisture uptake, reduce saturation moisture content and provide better properties after exposure to moisture for reinforced PP matrix composites [59,137]; however, large reductions in properties even with MAPP are found to occur, with for example reductions of strength for wood/PP composites after 238 days, during which time saturation moisture content occurred, of 32%, 45% and 59% for 30, 50 and 70 °C respectively.

In work on hemp fibre reinforced PLA, non-woven fibre composites have been found to be more absorbent than aligned fibre composites; this was considered to be due to increased porosity due to the increased complexity of the matrix flow path during processing [91]. Alkali treatment was found to increase resistance to moisture in the same work. Saturation of moisture has been found to occur after about two months for aligned hemp reinforced PLA with reductions of strength (13%), stiffness (22%) and fracture toughness (25%) but a large increase of impact energy of 530% at 25 °C with larger reductions of strength (30%), stiffness (30%) and fracture toughness (68%) and increase of impact energy (550%) at 50 °C all for alkali treated fibre [138].

Moisture absorption has been found to be different depending on its salt content. Absorption rates for composites containing curaua and E-glass fibres with an UP matrix for over 330 h of exposure

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to sea water have been seen to be slower than for exposure to distilled water, suggested to be due to NaCl ions migrating to fibre surfaces [139]. Reduced diffusion of sea water relative to distilled water in composites is further supported by work with jute and glass UP composites [140].

Although the presence of natural fibres is generally considered to increase moisture absorption of polymers, benefit of inclusion of natural fibres in glass fibre reinforced thermoset composites has been found for prolonged moisture exposure; although degradation rate with jute fibres was found to be faster for times of less than 70 h, above this time, jute was found to reduce degradation rate. This has been explained as the swollen jute fibre layers being able to accommodate the resin swelling strain or just acting as protection to a central glass fibre layer [141]. In this same research, although silane coupling was found to improve flexural strength, the benefit was lost after nearly 12 days in boiling water.

Weathering has, however, highlighted the benefit of natural fibres. Assessment of mechanical and thermal stability of date palm leaf fibres in PP composites containing UV stabiliser exposed to natural weathering conditions in Saudi Arabia and accelerated weathering conditions utilising UV wavelengths between 300 and 400 nm at 50% humidity, has shown the composites to be more stable than PP without natural fibre for natural and accelerated weathering conditions, although MAPP was found to reduce stability [142]. It was suggested that lignin acting as a natural antioxidant and it darkening on exposure, increases protection from UV, as well as enhanced adhesion due to oxidation of the matrix provided stability in the composites.

6. Applications

Over the last couple of decades, increasing numbers of car models, first in Europe encouraged by government legislation and then in North America, have featured natural fibre-reinforced polymers in door panels, package trays, hat racks, instruments panels, internal engine covers, sun visors, boot liners, oil/air filters progressing to more structurally demanding components such as seat backs and exterior underfloor panelling. Now, all of the main international automotive manufactures use these materials and their use is expected to increase in this area [47]. In India composite board has been developed as an alternative to medium density fibreboard which has been assessed for use in railcars [17]. The aircraft industry has also been adopting NFCs for interior panelling [96]. They have been used in applications as diverse as toys, funeral articles, packaging, marine railings and cases for electronic devices such as laptops and mobile phones as a replacement for synthetic fibre [96]. In sports, surfing appears to be of particular note with respect to embracing environmentally friendly materials. A number of companies are now advertising surfboards incorporating NFCs. One of the earliest was the "Ecoboard" produced by Laminations Ltd, using bio-based resin and hemp fibre [17]. A recent study supported the production of natural fibre surfboard fins through RTM to be possible regarding provision of mechanical performance and economic viability [143]. Fishing rods are also being produced using material developed by CelluComp Ltd who extract nanocellulose from root vegetables [17,144]. Recent research showing RTM flax reinforced polyester turbine blades to be a potential replacement for those reinforced with glass fibre obtained recognition by way of the Asia 2013 Innovation Award from the JEC composites group for the world's first functional flax composite wind turbine blade [145]. Research has also shown the potential of NFCs for top-plates of string musical instruments [146].

In the construction industry, wood fibre/PP or fibre/PE has been used extensively in decking, particularly in the USA. Natural fibre reinforced composites have also been gaining popularity in non-structural construction applications and used for door and window frames, 21 wall insulation and floor lamination [147–150]. Assessment for replacement of wooden laminates in insulating structural panels have found NFCs to have better mechanical properties [151]. The possibility of using natural fibre composite sheet piles by evaluating the flexural behaviour of extruded hollow cross-section wood-plastic composites (WPCs) with 50 m% wood flour has been investigated [152]. Results highlighted significant promise for natural fibre light duty sheet piling structures to replace conventional sheet piles made from concrete and steel. There has also been suggestion of similar materials to be used for beams and slabs [153]. Reinforcement of cement by natural fibres for building materials is also being assessed [47].

Overall, the global NFCs market was estimated at US$2.1 billion in 2010 and projected to rise 10% annually until 2016 [96] reflecting further potential seen across a range of industries including automotive, aerospace, construction, civil and the sports and leisure industries.

7. Conclusions

Much research and progress has occurred in recent decades in the mechanical performance of NFCs. Improvement has occurred due to improved fibre selection, extraction, treatment and interfacial engineering as well as composite processing. This paper has reviewed the research that has focussed on improving strength, stiffness and impact strength including the effect of moisture and weathering on these properties; long and short term performance was addressed. NFCs now compare favourably with GFRPs in terms of stiffness and cost; values of tensile and impact strength are approaching those for GFRFs. The lower densities for NFCs lead to better comparison for specific properties. Applications of NFCs have extended dramatically including load bearing and outdoor applications such as automotive exterior underfloor panelling, sports equipment and marine structures. Further research is still required to extend their application range including improvement of moisture resistance and fire retardance. Overall, growth of NFC uptake continues at a rapid rate and there would appear to be a very positive future ahead for their application.

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