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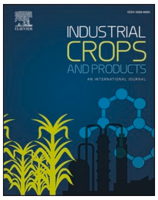

# Cellulosic fibres-based epoxy composites: From bioresources to a circular economy

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ABSTRACT

Global transition towards a bio-based economy to reduce the carbon footprints as an alternative to petroleum-based has led to the production of new cellulosic fibre-based eco-sustainable products in the construction and building industry. The present review gives an insight into how bio-based (fibre) is used as a reinforcement in polymer matrix composites. Specifics of the fibres in varied forms and their effect on the tensile characteristics of the resulting epoxy composites are reviewed. Studies reported to date have mostly focussed on improving the strength in the context of mechanical behaviour of the composites for varied structural applications. However, the scientific knowledge relating to modulus, its reinforcing capabilities are very limited in the literature. Detailed studies on the effect of dense regions with high moduli, less dense with insignificant moduli, point-to-point variation in the matrix are needed. This review proffers detailed explanations of the chemical treatments, hybridization, processing methods used from the last 22 years (2000–2022) in processing natural fibre composites (NFC) for value-added products. Future perspectives and functionalization of intelligence in bio-composites are also discussed.

## 1. Introduction and global overview

The green economy has pointed out a renewed interest towards the access of right resources in order to stimulate innovative employment policies that promote recycling, reuse of existing products over new ones, with a focus on reducing petroleum-based products via the use of the CE (circular economy) model ( Ellen Mac Arthur Foundation, 2013; Ghisellini et al., 2016; Ogunmakinde, 2019) (Ates et al., 2020; Daminabo et al., 2020; Ashvinder K. Rana et al., 2021a, 2021b; Thakur and Voicu, 2016). The real-time sustainability model has evolved with its improvised policies in nations such as China, Japan, Germany, Scotland with its implication at three levels (micro, meso, macro) globally, the concept still requires awareness for adoption in other countries (Ghisellini et al., 2016a; Ogunmakinde, 2019; Pappu et al., 2015).

The incorporation of legislative policies, social awareness of reducing the use of synthetic fibres have reconceived for novel use of bio-fibres as reinforcements in polymer matrices whilst fostering sustainability right at the production level through preventive and regenerative measures of CE (Ghisellini et al., 2016a; Ho, 2012).

Although synthetic fibres such as carbon, glass and kevlar dominated the automotive, construction and aviation industries from the 1950 s until the early 2000 s, their non-degradable nature has resulted in considerable environmental damage (Jawaid and Abdul Khalil, 2011; Sun, 2018; Vinod, 2020) (Beluns et al., 2021; Thakur et al., 2012). Moreover, traditional metal/ceramic composites employed until the 1970s also proved to be economically unviable. Despite technological advances in synthetic fibres in the twentieth century, demand for polypropylene, polyester fibres have declined since 1970, with a fall of 4.5% to 42.2 million tonnes in synthetic fibres, 10.1% to 25.2 million tonnes in animal fibres and 5.9 million tonnes only in natural fibres (Zimniewska et al., 2011a, b).

With the adoption of cradle to cradle, blue economy theories for refinement in CE, the scientific communities, composite manufacturing industries, have managed to reduce the environmental load by making significant use of plant-based raw materials in luxury cars, electric vehicles, sports cars because they possess excellent mechanical properties such as high specific strength and modulus, low density, biodegradability, renewability etc. over their synthetic counterparts (Ankit et al.,

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2021; Ghisellini et al., 2016b; Lakshmi Narayana and Bhaskara Rao, 2021; Maou et al., 2021; Pappu et al., 2019; Saxena et al., 2008; Saxena and Gowri, 2003; Sherratt, 2013; Thakur and Thakur, 2014; Uppal et al., 2019; Venkatarajan and Athijayamani, 2021; Vinod et al., 2020).

NFCs were used as early as 1896 in aircraft seats. The biggest impulse for the use of NFCs is still the construction and automotive sectors owing to their low density, production cost allowing curtailment in vehicular weight with strategies to reduce embodied energy in the construction sector (Dixit et al., 2015; Zimniewska et al., 2011b) (Ashvinder Kumar Rana et al., 2021).

European Commission under directive, /64/EC (2005) "End of Life Vehicles" program have made mandatory requirements for all the automotive manufacturers to use 95% recyclable materials by 2015 (Balla, 2019; Sun, 2018). For example, Daimler Chrysler was awarded by the European composite association for its use of Banana fibre for the first time in the exteriors of Mercedes A-class car in 2005 (Holbery and Houston, 2006; Mercedes-Benz incorporates banana fibres into A-Class, 2005). Each BMW 7 vehicle contains 24 kg of sisal and flax fibres (Saxena et al., 2011). Lotus Eco Elise, a sports car uses hemp fabric in its upholstered seats and body panels along with sisal carpets (Zimniewska et al., 2011b). Toyota manufacturers in Indonesia use kenaf and polypropylene indoor trims (Elliot, 2005; Zimniewska et al., 2011). Honda embarks usage of wood fibre in flooring of sport's vehicle-The Pilot SUV (Elliot, 2005; Holbery and Houston, 2006b). Kafus Bio-Composites/Flexform Technology, Cargill Ltd, Delphi Interior Systems in North America uses natural fibres in manufacturing auto parts (Holbery and Houston, 2006b). This strategical use of fibres has promoted green economy by consuming less energy than fibre-glass cutting the production cost by 30% (International Year of Natural Fibres, 2009; Zimniewska et al., 2011). In the construction sector, the Indian government fostered the use of jute fibre-reinforced polyester resins for building Madras House and grain elevator buildings (Sun, 2018). Decking products in North America for construction are provided by Timber Tech, Trex, AERT companies (Sun, 2018).

The market for fibre reinforced polymer composites (FRP's) in the U. S. is estimated to be US$ 2 billion in 2020 followed by the world`s second-largest economy, China which is expected to reach US$ 1.9 billion by 2027. The global market for FRP is estimated to be US$ 6.9 billion in 2020, with a compound annual growth rate (CAGR) of 6.9%, which is expected to reach US$ 11 billion by 2027 (Unmanned Systems Fiber Reinforced Polymer, 2021). The fibre reinforced polymer composites market is constantly growing and is a multibillion-dollar business (Jawaid and Abdul Khalil, 2011; Sanvezzo and Branciforti, 2021). European Commission under its new plans has adopted a strategy in January 2018 that envisages all plastic packaging will be recyclable by 2030 (EU, 2018). The government of the U.K. in its 25-year environment plans targets zero avoidable waste by 2050 (25 Year Environment Plan, 2018).

Development of lignocellulosic fibre composites relative to its synthetic part with acceptable reproducibility holds unique challenges as they show non-uniformity in morphology, crystallinity, the geometry of cell wall etc. but can be beneficially exploited combined with other unutilised waste resources to engender innovative composite materials for optimum benefits (Platnieks et al., 2021; Amar Singh Singha and Thakur, 2009, 2009; Zielińska et al., 2021).

Natural fibre reinforced composites (NFRCs) based science and engineering is an interdisciplinary area that involves characterization of raw materials, designing and fabrication of innovative materials and their performance evaluation to confirm their suitability for the targeted application(Thakur et al., 2013; Singha and Thakur, 2008). Different matrix systems can be used to fabricate and tailor the properties of composites suitably to attain improved mechanical, thermal and electrical properties(Amar Singh Singha and Thakur, 2008; A. S. Singha and Thakur, 2009a). Further, the industrial applications of composites include processing analytical techniques, materials design, and cost/-benefit trade-offs in industrial production Pappu and Thakur, 2017. Though significant achievement has been made in designing and developing such application-oriented composites, extensive innovative research is still in progress for vital applications. There is tremendous scope for setting up secondary industries for recycling and use of renewable natural fibres in making sustainable composite materials, which will generate employment and income for rural and urban populations, followed by environmental benefits and technological advancement.

Synthetic fibres such as glass, aramid, carbon fibres, are commonly used as reinforcing materials in fibre reinforced composites John and Anandjiwala, 2008; Tucker and Liang, 1999. The thermoset polymers widely utilised in polymer matrix composites are polyester, phenolics, and epoxy, whereas the thermoplastic family includes polypropylene, polyethylene, and polystyrene(Singha and Kumar Thakur, 2008; A. S. Singha and Thakur, 2009b; Thakur et al., 2011a. Cellulosic fibre-based thermoset composites have greater mechanical characteristics than thermoplastics, as evidenced by previous research, while thermoplastic composites have better design flexibility and recycling potential (A. S. Singha and Thakur, 2008; Pappu et al., 2016). In comparison to alternative matrices, epoxy resins provided improved mechanical properties (tensile and flexural strength), high thermal and chemical resistance. This review paper deals with the tensile characteristics of bast and leaf fibre reinforced composites, as well as hybrid composites in attained sustainable approach towards the circular economy.

## 2. Types of cellulosic fibres

Depending upon applications, the fibre plants are categorized as primary or secondary Summerscales et al., 2010; Trache et al., 2020. Primary plants are those that are produced for their fibre content, whereas secondary plants are those that generate fibre as a by-product Ramamoorthy et al., 2015. There are six types of cellulosic fibres namely: bast fibres (jute, ramie, hemp, flax, kenaf), leaf fibres (sisal, abaca, pineapple), seed fibres (cotton, coir, kapok), core fibres (jute, hemp, kenaf), grass and reeds (rice, corn, wheat), woods and roots (Singha et al., 2008; Thakur et al., 2011b; Pappu et al., 2016). The bast fibres are the strongest fibres among the other fibre types having an aspect ratio in the range of 1000–1200.

All cellulosic fibres are made up of filamentous units called microfibrils. These microfibrils assemble to form a fibrillar bundle which group together to form a single strand(V. K. Thakur et al., 2014). The microfibrils in the densest S2 layer (secondary cell wall) are structured in a helical pattern and the angle at which they are positioned off the filament or fibre axis is called the "microfibrillar angle" (Dittenber and GangaRao, 2012). This average angle, which changes from one fibre to another is substantially responsible for the mechanical characteristics of the fibre. Smaller microfibrillar angles indicate greater stiffness and strength, whereas larger angles signify greater ductility (Dittenber and GangaRao, 2012). Plant-based fibres are primarily composed of cellulose, hemicelluloses, and lignin (lignocelluloses) (A. S. Singha and Thakur, 2009c; Thakur and Thakur, 2015). The age of the plant, species, climatic conditions, fibre processing techniques influence the chemical content and structure of fibres, resulting in a wide range of mechanical properties of the fibre. The mechano-chemical characteristics of various natural fibres are shown in Table 1. Cotton holds maximum cellulose content whereas coir provides the highest lignin content.

### 2.1. Bast fibre

#### 2.1.1. Jute (Corchorus olitorius/ Corchorus capsularis)

In tropical regions, jute is the cheapest bast fibre after cotton. It is from the family *Malvaceae* and has currently the highest production volume among other fibres (Chand, 2008). Jute's history dates from 206 BCE-221 CE, when it was first discovered as a paper in Dunhuang, Gansu Province, China (Guo-Qing, 1989; Tsien, 1973). Some ancient records reveal the use of fibres in India during the Akbar era (1542–1605). Its

**Table 1**

Mechano-chemical properties of some natural fibres.

| Fibre | Density (g/cm$^3$) | Diameter (μm) | Tensile Strength (MPa) | Tensile Modulus (GPa) | Elongation (%) | Cellulose (wt%) | Hemi-cellulose (wt %) | Lignin (wt%) | Pectin (wt%) | Wax (wt %) | Micro-fibrillar angle (degree) | Moisture (wt%) | References |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Jute | 1.3–1.46 | 20–200 | 320–800 | 8–78 | 1.5–1.8 | 59–71.5 | 13.6–20.4 | 11.8–13 | 0.2–0.4 | 0.5 | 8 | 12.5–13.7 | (Dittenber and GangaRao, 2012; Saxena et al., 2011; Yan et al., 2016a) |
| Flax | 1.4–1.5 | 12–600 | 345–1100 | 27.6–103 | 1.2–3.3 | 62–72 | 18.6–20.6 | 2–5 | 2.3 | 1.5–1.7 | 5–10 | 8–12 | (Dittenber and GangaRao, 2012) |
| Hemp | 1.4–1.5 | 25–500 | 270–690 | 23.5–90 | 1–3.5 | 68–74.4 | 15–22.4 | 3.7–10 | 0.9 | 0.8 | 2–6.2 | 6.2–12 | (Dittenber and GangaRao, 2012; Pappu et al., 2019b; Saxena et al., 2011) |
| Kenaf | 1.4 | – | 223–930 | 14.5–53 | 1.5–2.7 | 31–72 | 20.3–21.5 | 8–19 | 3–5 | – | – | – | |
| Ramie | 1.0–1.55 | 20–80 | 400–1000 | 24.5–128 | 1.2–4.0 | 68.6–85 | 13–16.7 | 0.5–0.7 | 1.9 | 0.3 | 7.5 | 7.5–17 | (Dittenber and GangaRao, 2012) |
| Sisal | 1.33–1.5 | 8–200 | 363–700 | 9–38 | 3.0–7.0 | 60–78 | 10.0–14.2 | 8.0–14 | 10.0 | 2.0 | 16–25 | 10–22 | (Dittenber and GangaRao, 2012; Komuraiah et al., 2014; Sathishkumar et al., 2013; Saxena et al., 2011) |
| Banana | 1.35 | 12–30 | 500 | 12 | 1.5–9 | 63–67.6 | 10–19 | 5 | – | – | – | 8.7–12 | (Dittenber and GangaRao, 2012; Pappu et al., 2015) |
| Pineapple leaf fibre (PALF) | 0.8–1.6 | 20–80 | 413–1627 | 34.5–82.5 | 1.6–14.5 | 70–83 | – | 5–12.7 | – | – | 14 | 11.8 | |
| Abaca | 1.5 | – | 300–400 | 10–12 | 1.0–10 | 56–63 | 20–25 | 7–13 | 1 | 3 | – | 5–10 | (Dittenber and GangaRao, 2012) |
| Curaua | 1.4 | 7–10 | 500–1150 | 11.8–96 | 1.3–4.9 | 70.7–73.6 | 9.9 | 7.5–11.1 | – | – | – | – | (Dittenber and GangaRao, 2012) |
| Coir | 1.15–1.46 | 10–460 | 95–230 | 2.8–6 | 15–51.4 | 32–43.8 | 0.15–20 | 40–45 | 3–4 | – | 30–49 | 8.0 | (Dittenber and GangaRao, 2012) |
| Cotton | 1.5–1.6 | 10–45 | 287–800 | 5.5–12.6 | 3–10 | 82.7–90 | 5.7 | < 2 | 0–1 | 0.6 | 25 | 7.85–8.5 | (Dittenber and GangaRao, 2012; Yan et al., 2016a) |
| Oil Palm | 0.7–1.55 | 150–500 | 80–248 | 0.5–3.2 | 17–25 | 60–65 | – | 11–29 | – | – | 42–46 | – | (Dittenber and GangaRao, 2012) |
| Bagasse | 1.25 | 10–34 | 222–290 | 17–27 | 1.1 | 32–55.2 | 16.8 | 19–25.3 | – | – | – | – | |
| Bamboo | 0.6–1.1 | 25–40 | 140–230 | 11–17 | 2.5–3.7 | 26–65 | 30 | 5–31 | – | – | 2–10 | – | (Abdul Khalil et al., 2012) |

industry grew in the early 1800 s in Dundee, Scotland. Globally, around $2300 \times 10^3$ tons of jute fibre are produced. It is largely produced in India, China and Bangladesh. The jute plant has a height of around 1–4 m and the fibres are extracted by the steeping (retting) process (Yan et al., 2016a). Fibre lies in the middle of the bark and centrally located pith (Mohanty and Misra, 1995). Fig. 1(a) and 1(b) show the cultivation of Jute plants and the fibre. The tensile characteristics of jute fibres are shown in Table 1. The tensile strength and modulus of the fibre range from 320 to 800 MPa and 8–78 GPa (Dittenber and GangaRao, 2012). The micro-fibrillar angle of jute fibre is 8.1°(Yan et al., 2016a). It is the most explored fibre for reinforcements in epoxy, phenolics, PLA composites (Khondker et al., 2006; Mishra et al., 2000). Jute fibres are a typical reinforcement in the German automobile industry as they are readily available in large quantities.

### 2.1.2. Flax (Linum usitatissimum)

Flax belongs to the *Linaceae* family, which includes 13 genera and 300 species (Chand, 2008). It is the oldest cultivated plant used well before 5000 BCE in Georgia and Egypt and the first to be woven into textiles (Kvavadze et al., 2009). It is predominantly grown in France, Belgium and Canada for its fibre and linseed oil. High-grade fibres are used in textile yarns while the low grades as reinforcement in composites. The plant grows up to 90 centimetres and its structure is as follows:

At the macroscopic level- Bark, phloem, xylem, and the centre void make up the stem (Kvavadze et al., 2009).

At mesoscopic level- A bundle's cross-section consists of 10–40 fibres joined primarily by pectin (Charlet et al., 2009, 2007; Yan et al., 2014).

At microscopic level- Each fibre is composed of cell walls that vary in thickness (Yan et al., 2014).

Fig. 2 shows the plants and the fibre. At the centre of the fibre is the lumen, wh ich transports water. The primary cell wall protects the secondary wall which gives fibres their strength. Each layer comprises cellulose microfibrils that run parallel to each other and form a micro-fibrillar angle in the direction of the fibre (Charlet et al., 2007). The secondary cell wall is made up of crystalline cellulose and hemi-celluloses that are oriented at an angle of approximately 10 degrees along the fibre direction. This minimum angle gives the fibre a high tensile strength. Table 1 lists the tensile characteristics of flax fibre. The fibre has a tensile strength of 345–1100 MPa and a modulus of 27.6–103 GPa, respectively (Dittenber and GangaRao, 2012). Flax fibre has a micro-fibrillar angle of 5–10° and is feasible with both thermoplastics and thermosets (Dittenber and GangaRao, 2012).

### 2.1.3. Hemp (Cannabis sativa)

Hemp is a member of the *Cannabaceae* family and is predominantly produced for its seed and fibre in France and China. Cannabis plants were indigenous to Central Asia and were introduced to Europe during the Iron age. It is cultivated today in temperate countries such as North Korea, Chile, Japan, India, and Europe (Pappu et al., 2019b). The plant reaches a height of around 10 feet with an average fibre bundle length of 5–55 mm and a diameter of 25–500 μm. Fig. 3 shows the plants and the fibre. Hemp is utilised in a variety of products such as paper, textile fibre, seed food, oil, pulp and biofuel. It currently provides less than 0.5% of the world's natural fibre production (Shahzad, 2012). The tensile characteristics of hemp fibre are presented in Table 1. The fibre's tensile strength and modulus range from 270 to 690 MPa and 23.5–90 GPa respectively. The micro-fibrillar angle of hemp fibre ranges from 2 to 6.2° (Dittenber and GangaRao, 2012). Bayerische Motoren Werke AG (BMW), a renowned automotive firm, manufactures car parts made from hemp fibre (Kandachar and Brouwer, 2001). Hemp is a potential reinforcing material used in thermoplastic and thermosetting resins.

### 2.1.4. Kenaf (Hibiscus cannabinus)

Kenaf is a member of the *Malvaceae* family and has been cultivated since 4000 BCE (Mariod et al., 2017). It is a crop of southern Asia, central Africa and is grown primarily today in Bangladesh and India. In Bengal and India, it is termed as mesta, java jute in Indonesia, stockroot in South Africa and ambari in Taiwan. Kenaf is a 3-metre-tall herbaceous plant with a 3–5 cm woody base diameter. Fig. 4 shows the plants and the fibre.

The plant is mainly cultivated for its seed oil and fibre in tropical regions. In ambient conditions, it grows up to 10 cm per day and matures in three months (Nishino, 2004). The bark of kenaf is made up of around 40% plant material that is utilised to extract fibres, and the rest is core wood. The low-cost fibre uses 15 MJ energy for 1-kilogram kenaf, whereas glass fibres use 54 MJ. Table 1 provides the tensile properties of kenaf fibres. Kenaf has a tensile strength and modulus ranging from 223 to 930 MPa and 14.5–53 GPa, respectively and has a good potential as a thermoplastics and thermosets raw material.

**Fig. 1.** (a) Jute plants bearing yellow flowers (b) Decorticated and dried Jute fibres. (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)

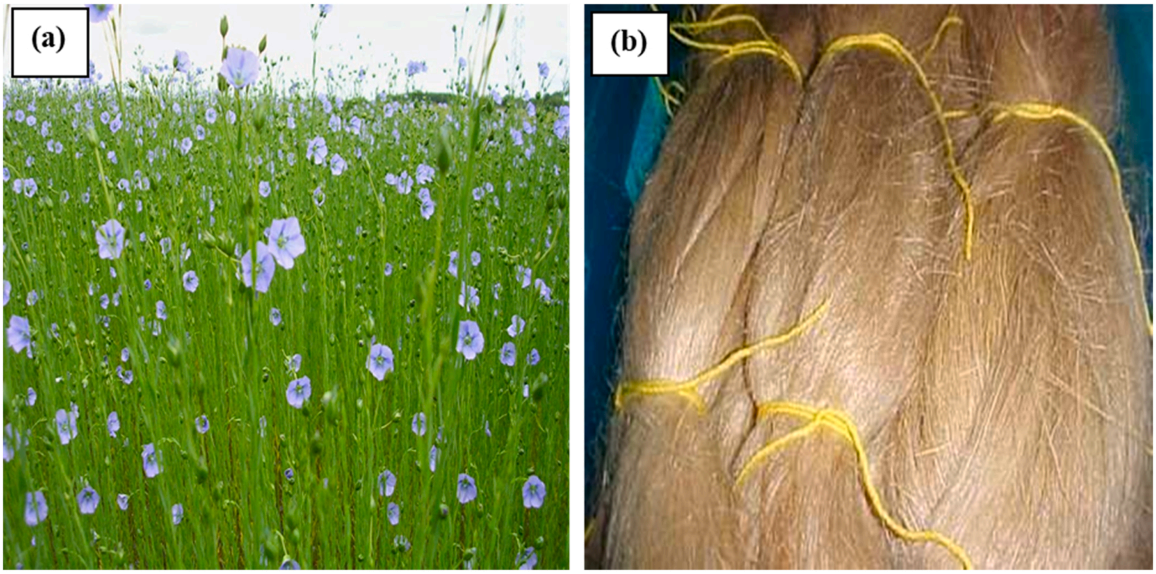


**Fig. 2.** (a) Flax plant. (b) Flax fibre.
(a) (https://www.connie.co.uk/blog/From-The-Humble-Flax-Plant-Comes-Luxury-Bed-L) (b) (http://textiletuition.blogspot.com/2015/08/history-of-flex.html).

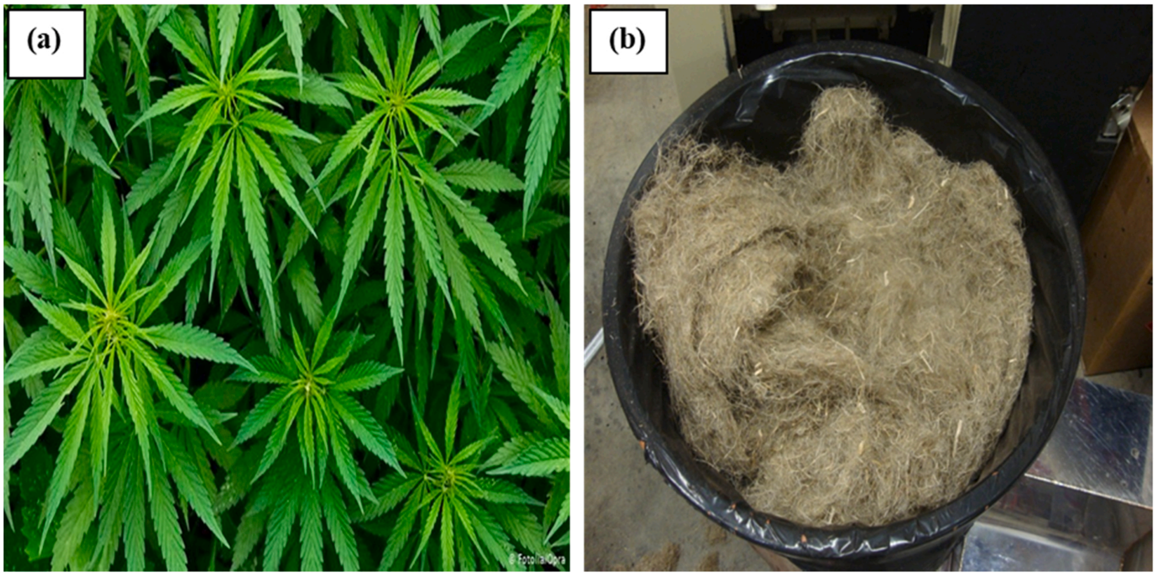


**Fig. 3.** (a) Hemp plants (b) Dried hemp fibres.
(a) (https://www.dw.com/en/cannabis-gains-acceptance-as-medicinal-product-in-germany/a-17766339).

### 2.1.5. *Ramie (Boehmeria nivea)*

Ramie comes from the nettle family *Urticaceae* and is primarily produced in Brazil and China. It has been grown in China for decades and is commonly referred to as "China Grass". It is a perennial herb that grows to a height of 1–2.5 m and survives for around 7–20 years.

Global production of these bast fibres is the lowest and is around 100 × $10^3$ tons. Fig. 5 shows the plants and the fibre. Low production and presence of impure substances like gum, pectin, makes it complicated to use as a reinforcement in composites. Due to the above-statedd problems (impurity and availability) ramie fibres are relatively less explored. The tensile characteristics of Ramie fibre are shown in Table 1. The fibre has a tensile strength of 400–1000 MPa and a modulus of 24.5–128 GPa respectively. The fibre's micro-fibrillar angle is 7.5° (Dittenber and GangaRao, 2012). Ramie fibres are used in both thermoplastic and thermosetting resins.

## 2.2. *Leaf fibre*

### 2.2.1. *Sisal (Agave sisalana)*

Sisal is a crop grown commercially in Brazil and Tanzania. It comes from the family *Agavaceae* and is extensively cultivated in Kenya and China (Medina and Dzalto, 2018). Lock explained the plant in terms of structure, production and use. Sisalana, Natale, Vergross, and Istle are the four variants of sisal found in India (Chand, 2008). The plant grows to a height of 1 m and a width of 28 mm with 200–250 leaves (Li et al., 2000; Yan et al., 2016b). It has dark green, lance-shaped leaves that grow in the form of a rosette. Fig. 6(a) and 6(b) show the sisal plants and leaves.

The lifespan of the plant is around 7–10 yrs and once mature, the fibres are ready to be extracted from the leaves through a process called decortication. A leaf has only 4% fibre of the 1000 fibre bundles (Mukherjee and Satyanarayana, 1984; Saxena et al., 2011). Based on

**Fig. 4.** (a) Kenaf plants(b) Kenaf fibres.
(a) (http://treeliving.com/kenaf-multi-use-plant/), (b) (https://www.kenafpartn ersusa.com/kenaf-fiber-industry.html).

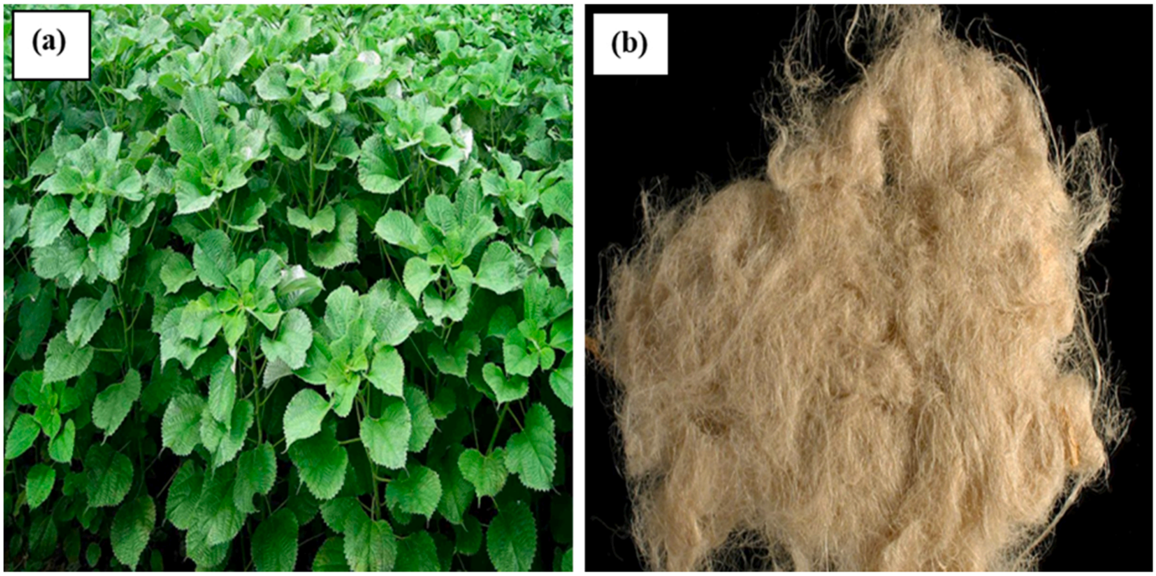


**Fig. 5.** (a) Ramie plants (b) Ramie fibres.
(a) (https://naturalfibersinfo.org/?page_id=87). (b) (http://cameo.mfa.org/wiki/Ramie).

their extraction, the fibres are classified into three types (Bai et al., 2002):

1. Mechanical fibres- They are horse-shoe shaped, arranged in a definite pattern and are extracted from the leaf's periphery. These fibres have the highest strength and rarely break during extraction.
2. Ribbon fibres - These intermediate fibres are drawn from conducting tissues along the median leaf line and have substantial mechanical strength. They are the longest and are most likely to break longitudinally during processing.
3. Xylem fibres - They are irregularly shaped with thin-walled cells, i.e., why have the lowest strength. These fibres easily get damaged during the process of extraction (Bisanda and Ansell, 1992; Li et al., 2000).

Fig. 6(c) and 6(d) show the technical fibres and SEM image of the Sisal fibres. Table 1 displays the tensile characteristics of sisal fibre. The fibre has a tensile strength of 363–700 MPa and a modulus of 9–38 GPa respectively (Dittenber and GangaRao, 2012). The fibre's micro-fibrillar angle varies from 16 to 25° (Komuraiah et al., 2014; Li et al., 2000; Sathishkumar et al., 2013). Sisal fibres are reinforced in thermoplastic and thermoset matrices like other plant fibres.

### 2.2.2. Banana (*Musa indica*)

Banana is the oldest cultivated plant that belongs to the family *Musaceae.* The word 'banan' comes from the Arabic term 'finger'. There are about 300 different species of banana, of which only 20 are consumed. The Asian, American, African, subtropical regions produce around 50 million metric tons of bananas every year. Banana plants are available from 300 N to 300 S of the equator. The plant mistakenly referred to as a tree, is a giant herb with a stem comprising overlapping leaf bases. The leaves of banana are paddle-shaped with a thick midrib. Fig. 7 shows the Banana plant and the fibres.

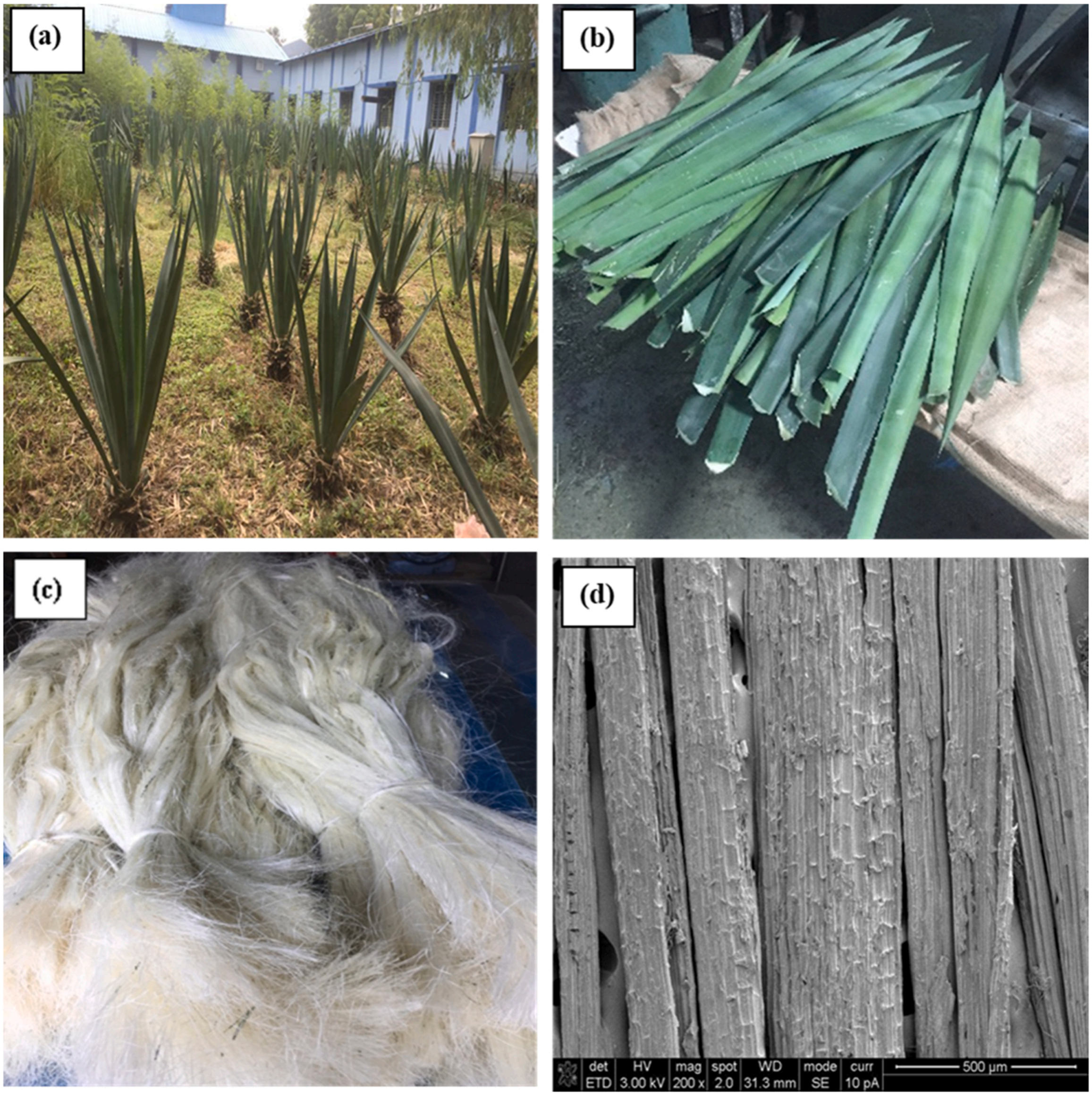


**Fig. 6.** (a) Sisal plants, (b) Sisal leaves, (c) Dried sisal fibres (d) Micrograph of Sisal fibres showing distinct parenchymatous cells.

There are varieties of Musa cultivated for the fruit, but the fibre is produced in only four varieties: Aethalpalal, Palayankottai, Rasagatali and Sentuluvan. Senthulavan is red giving reddish brown fibre while the others yield white coloured fibre. These varieties are primarily grown in Southern India in Tamil Nadu, Kerala, Kanyakumari and Tirunelveli (Chand, 2008). Banana fibres are used to make products like bags, bins, mats, etc. Table 1 reports the tensile characteristics of banana fibres. The fibre has a tensile strength of around 500 MPa and a modulus of around 12 GPa (Dittenber and GangaRao, 2012). This natural fibre is used in both thermosetting and thermoplastic matrices.

### 2.2.3. *Pineapple (Ananas comosus)*

Pineapple belongs to the family *Bromeliaceae*, is native to South America and is widely cultivated in Indonesia, Thailand, Philippines, Costa Rica, and India (Bartholomew et al., 2002; Wali, 2019). There are around 2794 species in the Bromeliaceae family, which comprises 56 genera (Wali, 2019). The architecture, cultivation of the plant was studied by Collins. Pineapple is a tropical plant that grows to a height of 0.75–1 m and produces 40–50 leaves per plant which are 0.5–1.8 m long and 0.02–0.055 m wide (Jawaid et al., 2020; Mukherjee and Satyanarayana, 1986). Fig. 8(a) and 8(b) show the plants and the leaves.

The fruit is scaly outside and contains phyllotaxies leaves on the top. Fig. 8(c) and 8(d) show the fruit and the fibre. Pineapple grows well in sandy loam soil at temperatures between 18 and 45°C (Jawaid et al., 2020). Mechanically, the principal structural component of pineapple fibre, known as "ultimate cells," is longer than most other fibres, thereby yielding long fibres (Sena Neto et al., 2013). The four major types of pineapple are Abacaxi, Smooth Cayenne, Spanish and Queen (Jawaid et al., 2020). Embrapa Cassava and Tropical Fruits have an operational bank named Pineapple germplasm with approximately 670 Ananas and other Bromeliaceous varieties. The majority of research has concentrated on commercial pineapple varieties, leaving out germplasms or other high genetic varieties, several of which are still unexplored for technical applications (Sena Neto et al., 2015). Table 1 lists the tensile properties of pineapple fibre. The fibre's tensile strength and modulus are 413–1627 MPa and 34.5–82.5 GPa, respectively. The micro-fibrillar angle of pineapple fibre is 14°. The modulus of the fibre is the highest among leaf fibres and is used in both thermosets and thermoplastics as a reinforcing material.

### 2.2.4. *Abaca (Musa textilis)*

Abaca is from the Musaceae family and is mostly cultivated in

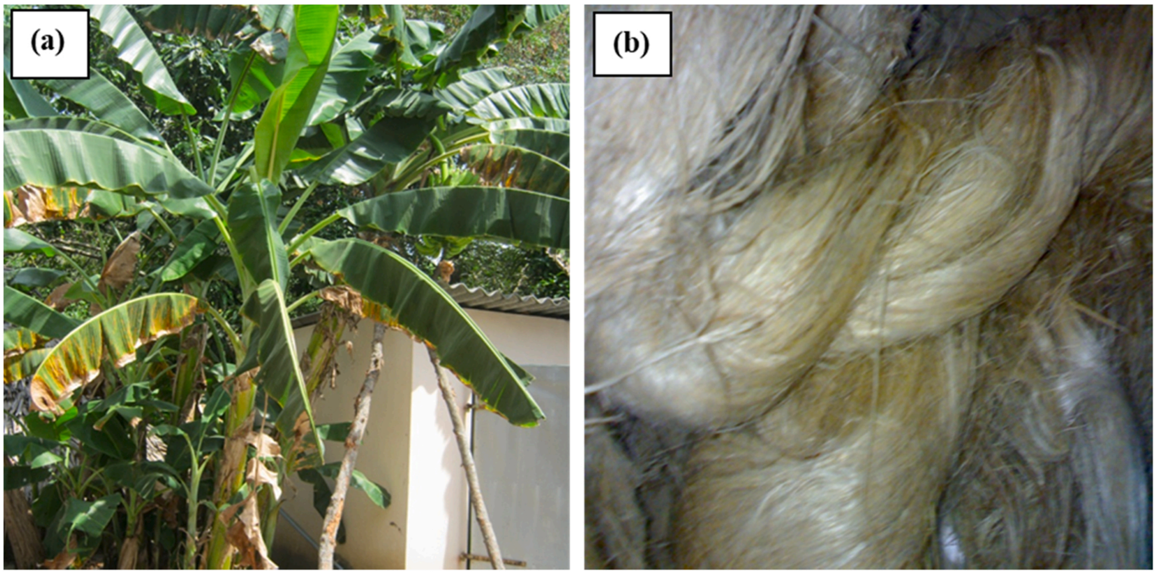


**Fig. 7.** (a) Banana plant (b) Banana fibres.

Ecuador, Costa Rica and the Philippines. Abaca was first cultivated on a larger scale in Sumatra in 1925. The lifespan of the plant is about 10 years and grows to an average height of 4 m. Fig. 9 shows the plant and the fibre.

After the initial growth, the fibres are harvested regularly (twice or thrice a year). Goltenboth and Muhlbauer described the production, extraction and preparation of abaca fibres in detail (Göltenboth and Mühlbauer, 2010). Due to its resistance to salty water, the fibre finds its use in marine ropes. Table 1 depicts the tensile characteristics of abaca fibre. The fibre's tensile strength and modulus are 400–980 MPa and 6.2–20 GPa, respectively (Dittenber and GangaRao, 2012). Abaca fibres are employed as reinforcements in both thermosetting and thermoplastic matrices because of their excellent mechanical properties.

### 2.2.5. *Curaua (Ananas erectifolius)*

Curaua, a species of the family Bromeliaceae is mainly cultivated in Venezuela and Brazil (Ladchumananandasivam and Franck, 2005). The lifespan of the plant is around 5 years, with each plant producing 7 scions a year (Ladchumananandasivam and Franck, 2005).

The leaves of the plant are hard, erect and require at least 2000 mm of annual rainfall to thrive. Fig. 10 shows the plants and the fibre. When the dry fibre content is just 5–8%, the plant produces approximately 50–60 leaves annually, which grows to 1.5 m in eight months (Leao et al., 1998). Table 1 describes the tensile properties of curaua fibres. Curaua fibre's tensile strength and modulus vary from 500 to 1150 MPa and 11.8–96 GPa respectively (Dittenber and GangaRao, 2012). The high cellulose content of the fibre accounts for its greater strength, allowing it to be used in both exterior and interior automotive applications (Silva and Aquino, 2008; Zah et al., 2007). Curaua fibres have a high potential to be used as reinforcements in both thermoplastics and thermosetting bio-composites.

## 2.3. *Seed/ fruit fibres*

### 2.3.1. *Coir (Cocos nucifera)*

Coconut belongs to the family *Arecaceae* (Roopan and Elango, 2015). Coir fibre is sourced from coconut's outer layer husk and is principally grown in India and Sri Lanka (Chand, 2008). This coconut husk (exocarp) consists of a water-resistant outer skin (epicarp) and a fibrous region (mesocarp). Mesocarp comprises vascular bundles embedded in parenchymatous connective tissue (non-fibrous) (Chand, 2008). Fibres of unripe coconut are white and turn brown on ripening (Harish et al., 2009).

It is the only relative waterproof fibre that absorbs minimal quantities of saltwater (Geethamma and Thomas, 2005; Harish et al., 2009; Viswanathan et al., 2000). Fig. 11(a) and 11(b) shows the tree and the fibres. Low water absorption in coir fibre is due to the low cellulose content and a high amount of lignin. Coir fibres have the highest percentage of lignin. Fig. 11(c) and 11(d) depict the ripe fibre. Due to a micro-fibrillar angle of 30–49°, these fibres can stretch beyond their elastic limit without fracture. The tensile characteristics of coir fibre are presen ted in Table 1. The fibre's tensile strength and modulus are around 95–230 MPa and 2.8–6 GPa respectively (Dittenber and GangaRao, 2012). They are widely used to make mattresses, doormats, cords, brushes, twines, etc. and demonstrate a great potential as reinforcement in polymeric composites.

### 2.3.2. *Cotton (Gossypium)*

Cotton comes from the family *Malvaceae* and is native to the tropics and subtropics of the world (Chand, 2008). The top three cultivators of cotton are the United States, India, and China. Cotton fibres were used since 5000 BCE and are grown in a shielded capsule called (boll). The fibre grows around the seeds and is composed of pure cellulose which imparts strength to the fibre. Fig. 12 (a), 12(b) show the cultivation of cotton plant and Fig. 12(c) shows the cotton fibre mat.

The structure of the fibre resembles the coils of a spring (Chand, 2008). During fibre development, elongations up to 1000–3000 times longer than the diameter occur. The plant produces the purest form of cellulose (Basra and Malik, 1984). The tensile characteristics of cotton fibre are summarized in Table 1. The fibre's tensile strength and modulus range from 287 to 800 MPa and 5.5–12.6 GPa (Dittenber and GangaRao, 2012). The micro-fibrillar angle of cotton fibre is 25° (Yan et al., 2016a). Cotton fibres are employed in a variety of high-value products, and cotton-based composites are used for automotive applications.

### 2.3.3. *Oil Palm (Elaeis guineensis)*

Oil Palm is the major source of palm oil and belongs to the family *Palmacea.* The African oil palm (*Elaeis guineensis*) and the American oil palm (*Elaeis oleifera*) are the two species that make up this genus. African oil palm, which is mainly found between Angola and Gambia, is native to Southwest Africa, whereas American oil palm is indigenous to South America. Most of the world supply of fibre is provided by Indonesia and Malaysia (Abdul Khalil et al., 2008). The highest yielding edible oil crop,

**Fig. 8.** (a) Pineapple plant (b) Pineapple leaves (c) Pineapple fruit with its crown (d) Dried Pineapple fibres.

Elaeis guineensis, is produced mainly outside Africa. Oil palm fibre is a secondary component derived from empty fruit bunches that being utilised in composites as reinforcement. When compared with other regions of the tree, the yield of fibres obtained from the fruit bunch is high (Rozman et al., 2000; Wirjosentono et al., 2004). The most prevalent approach for extracting the fibres is water retting (Raju et al., 2008). These fibres are hard and stiff and are similar to coir fibres in their properties (Sreekala et al., 1997). Fig. 13 shows the palm tree and the fibres. As listed in Table 1, the fibre's tensile strength and modulus range from 80 to 248 MPa and 0.5–3.2 GPa, respectively. The fibre has a micro-fibrillar angle of 42–46° (Dittenber and GangaRao, 2012). Oil palm fibre (OPF) is used in both thermosetting and thermoplastic composites.

## 2.4. *Grass*

### 2.4.1. *Bagasse (Saccharum officinarum)*

The fibrous substance obtained by crushing sugarcane stalks is known as bagasse (Xiong, n.d.).

It is from the grass family *Poaceae* (Selman-Housein et al., 2000) having a worldwide production of around (75 $\times 10^6$ tonnes). The major producers of fibre are Brazil, India, China and South Africa. Brazil is the major participant of the world sugar industry and the most competitive producer, with the lowest production cost in both the field and industrial sector (Ramajo-Escalera et al., 2006). In India, 100 tonnes of sugarcane produce around 30–34 tonnes of bagasse (Solomon, 2011). Bagasse is used in biocomposites as a raw material (Hajiha and Sain, 2015; Nikodinovic-Runic et al., 2013; Solomon, 2011), for combustion (Ramajo-Escalera et al., 2006), bioethanol production (Cardona et al., 2010; Rabelo et al., 2011) and is an important source of cellulose for dissolving pulp. Fig. 14 shows the plant and the bagasse fibres. The fibre has lower tensile properties as compared to bast and leaf fibres, with a strength of 222–290 MPa and a modulus of 17–27 GPa respectively as detailed in Table 1. It is used as reinforcement in both thermoplastics and thermoset composites.

### 2.4.2. *Bamboo (Bambusa vulgaris)*

Bamboo is an evergreen perennial belonging to the family *Poaceae* (Chand, 2008). The plant grows in monsoons to a height of 40 m and is mostly cultivated in countries like China, Indonesia and India.

It is the second most effective fibre used for pulp production in China and has more than 1250 species in the world. Fig. 15 shows the plants and the fibre. *Guadua angustifolia* Kunth, a popular bamboo species in

**Fig. 9.** (a) Abaca plant. (b) Abaca fibres.
(a) (https://matichub.com/2019/08/05/philippine-abaca/). (b) (https://naturalfibersinfo.org/?page_id=83).

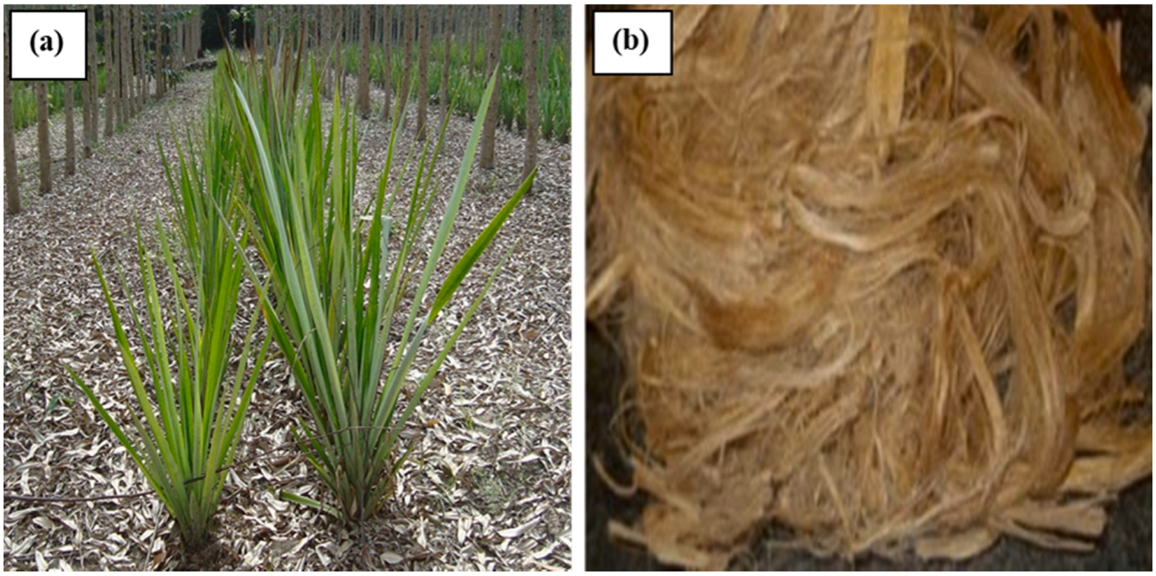


**Fig. 10.** (a) Curaua plants (b) Curaua fibres (Neves Monteiro et al., 2017).
(a) (https://commons.wikimedia.org/wiki/File:Curu%C3%A1_(cropped).JPG).

Latin America, is often used for construction in Peru, Ecuador and Colombia. In bamboo, there are two kinds of cells; leptodermous (matrix tissue cells) and sclerenchyma cells (enclosed in the matrix tissue). Vascular bundles made of the latter act as a reinforcement in bamboo (Chand, 2008). Panels and boards based on bamboo are hard and can replace hardwood items effectively. Bamboo fibre curtains absorb UV light of diverse frequencies, making them less damaging for the human body. Table 1 describes the tensile properties of Bamboo. The tensile strength and modulus of the fibre range from 140 to 230 MPa and 11–17 GPa, respectively. The fibre has a micro-fibrillar angle in the range of 2–10°(Abdul Khalil et al., 2012). Bamboo has emerged as an incredibly useful fibre in the last 15–20 years and is a superior substitute for wood. In many commercial applications, bamboo can substitute wood thereby protect and preserve the world's forests. Bamboo fibre is used in both thermoplastics and thermosets.

## 3. Comparison between fibre types and their mechanical properties

Lignocellulose (cellulose, hemicellulose, and lignin) is the most important chemical component of plant fibres (Dittenber and GangaRao, 2012). The amount of each component varies from plant to plant Savastano et al., 2009. This variation is due to several factors like plant's geographical location, age, chemical composition, species, and could also vary in different parts of the same plant. The physical properties of the fibres are partially determined by these basic components. Natural fibres are intrinsically hydrophilic due to fibre's porosity, high relative humidity, chemical composition and contain a significant amount of moisture content, which affects their mechanical properties. High cellulosic content, high pore volume, and low crystallinity are the factors that cause high moisture content. Table 1 also displays the moisture absorption content of some natural fibres. Growth, harvesting and fibre

**Fig. 11.** (a) The coconut tree (b) Coir fibres; from the outer layer (husk) of the coconut (c) Rope made from the coconut fibre (d) Technical coir fibres.

extraction at each stage are also the other multiple contributing elements that account for the variability. It is well established that the strength of the fibre is related to physical properties and microfibrillar angle. Small fibre diameter, small microfibrillar angle, high amount of cellulose and a high aspect ratio are the important factors responsible for fibre's excellent mechanical properties to be used as reinforcements in composites (Dittenber and GangaRao, 2012). In general, bast fibres appear to have superior characteristics for structural applications of which flax fibre offers the best possible combination of light weight, high cellulose content (62–72%) and thus high strength and stiffness as can be seen in Table 1. Though the tensile strength and cellulose content of jute fibres are similar to flax, the former absorbs more water thereby resulting in less strength and stiffness as compared to the latter. Kenaf has a relatively low cellulose content (31–72%) and highest lignin (8–19%) among other bast fibres. Both lignin and pectin are weak polymers and must be removed prior to using them as composite reinforcements (Dittenber and GangaRao, 2012). Hemp fibre shows a high amount of hemicellulose content (15–22.4%) which is amorphous and acts as an impurity hindering its use as reinforcement without chemical treatment. Ramie is the stiffest among all as it has the highest cellulose content (68.6–85%) and least lignin (0.5–0.7%) but the low production and high moisture content (7.5–17%) renders it difficult to be used as reinforcement for composites on a large scale.

On the other hand, leaf fibres are known to be coarser than bast fibres (Dittenber and GangaRao, 2012). Sisal and banana fibres are extensively used in composites due to their excellent mechanical properties as can be seen in Table 1. PALF has the highest tensile strength and modulus among leaf fibres due to its low microfibrillar angle (14°) and a high cellulose content (70–83%), yet its use as reinforcements for composites applications has not been fully explored as the leaves were considered as a waste. Curaua fibres have high cellulose content (70.7–73.6%) but their low production hinders its use at an industrial scale, and thus it is often substituted by other plant fibres. The high hemicellulose content (20–25%) in abaca makes it difficult to use in composites as compared to other leaf fibres. Among seed fibres, coir fibre has the lowest cellulose content (32–43.8%) and highest lignin (40–45%), thus showing low mechanical properties. On the other hand, cotton has the highest cellulose content (82.7–90%) and has the smallest diameter (10–45 μm) but the large amount of OH groups make it absorb more moisture making surface modification necessary to be used for composites or

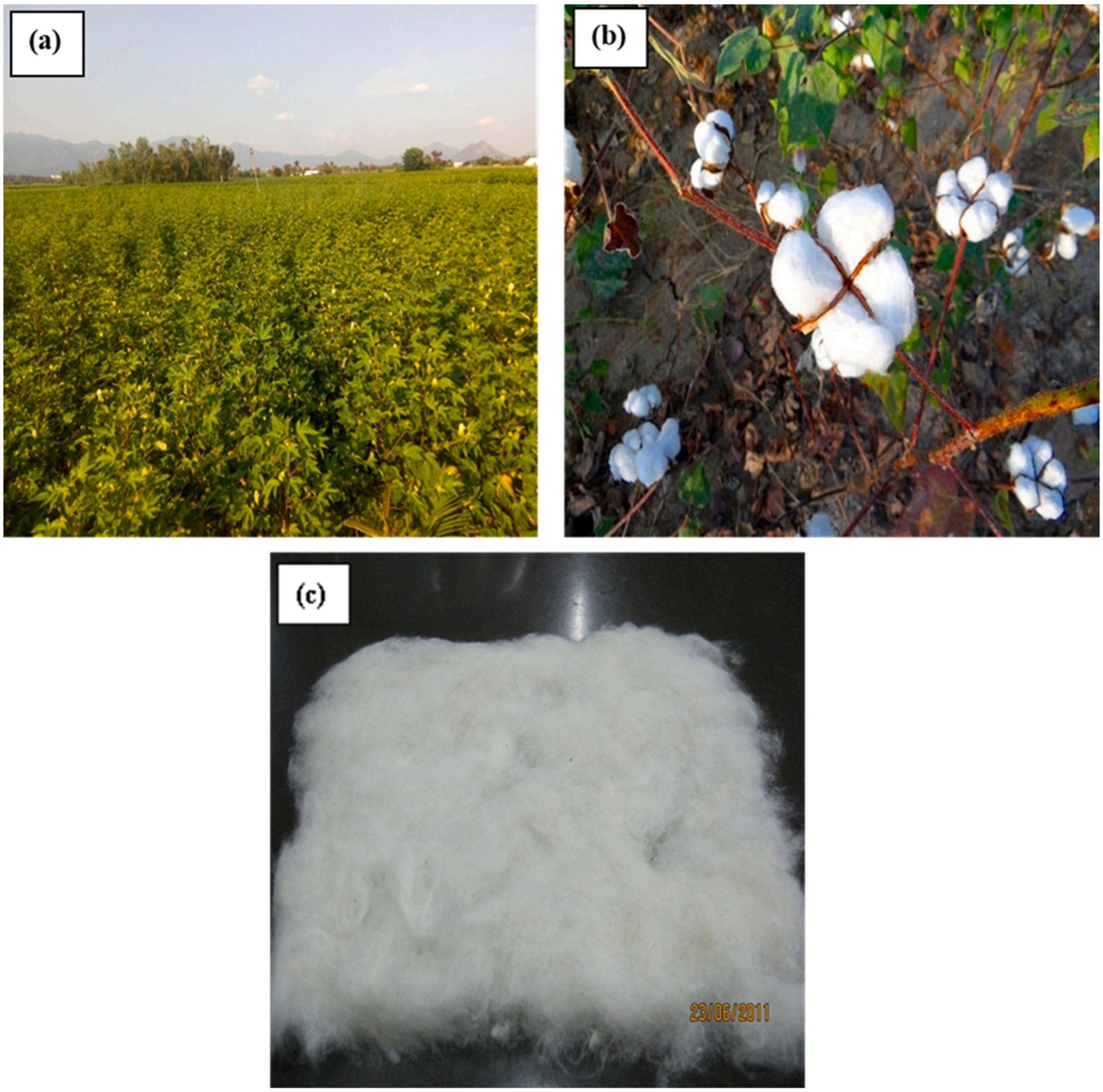


**Fig. 12.** (a) Cotton field at Singalandapuram Village, Tamil Nadu, India (b) Cotton Plant (c) Cotton fibre mat.
(a) (https://en.wikipedia.org/wiki/Cotton). (b) (https://www.agrifarming.in/cotton-farming-guide).

other technical applications. Oil palm fibre extracted from empty fruit bunch has mechanical properties similar to those of coir fibres. Its large variability in diameter (150–500 μm) and a high microfibrillar angle (42–46°) makes it less common than other seed fibres (Dittenber and GangaRao, 2012). The grass fibres have inferior mechanical properties when compared to bast and leaf fibres which have been demonstrated in Table 1.

## 4. Manufacturing processes of natural fibre reinforced polymeric composites (NFRPC)

Natural fibres (NF) can be processed to a temperature of 175–180 °C, beyond which they tend to degrade. Accordingly, the matrix systems are chosen to establish compatibility with NF's (Dittenber and GangaRao, 2012). The preliminary assessment to select the manufacturing technique is dominated by the size of the composites. For small to medium-sized components, simple and fast processing techniques like compression and injection moulding are used whereas large components are manufactured by autoclave and open moulding processes. Well-controlled temperature and pressure are two important pre-requisites to ensure the quality of NFRPC's. Optimum pressure gives desirable volume fraction and optimum temperature prevents degradation of fibres, controlling curing. However, very low temperature affects the viscosity of the resin and over-temperature gets accumulated at the localized regions thereby deteriorating the mechanical properties (Lau et al., 2018). Manufacturing processes that could be used with thermosetting matrices like polyester, phenolics, novolac and epoxies, broadly mirrors both open moulds (hand lay-up, spray lay-up) and closed mould (vacuum infusion, resin transfer moulding, compression moulding) techniques. A few of the fabrication techniques are described below:

### *4.1. Hand lay-up*

The hand lay-up method, also known as contact moulding is employed for thermosetting resins (Chanda and Roy, 2006). Both long and short fibres in varied forms; like the chopped mat, stitched or woven fabric can be used to produce NFRPC's with high fibre contents (Akay, 2015). Workability of resin accounts for low viscosity. In this method, mould is first coated with a releasing agent (silicone spray or wax) and a coating of liquid resin is brushed or rolled on top of it. Fibres are placed over and impregnated with resin. Air bubbles are worked out with the

**Fig. 13.** (a) The Palm tree (b) Palm fibres from the leaf stalk (c) Palm fibres from the leaf base.

help of rollers. Curing is done under ambient conditions (Chanda and Roy, 2006). It is a low-cost, labour-intensive process used for longer fibres and high fibre content (Akay, 2015; Gurit Guide to Composites (v5) - Gurit - PDF Catalogs | Technical Documentation | Brochure, 2000).

### 4.2. *Resin transfer moulding (RTM)*

Resin transfer moulding encompasses vacuum-assisted resin transfer moulding (VARTM), co-injection resin transfer moulding (CIRTM) techniques etc. In this manufacturing process, stationary preforms are injected with liquid resin (Ho et al., 2012b). 'Preforms' are laid in the mould cavity and the second matching mould is clamped over the first ("Gurit Guide to Composites (v5) - Gurit - PDF Catalogs | Technical Documentation | Brochure, ", 2000). Pressurized resin is injected into the heated mould using single or multiple ports. After cooling, the product is removed (Ho et al., 2012b). Any fibre form can be used with this technique. Stitched materials work well as the gaps allow rapid transport of the resin. High fibre volume composites with low void contents can be obtained through this technique (Gurit Guide to Composites (v5) - Gurit - PDF Catalogs | Technical Documentation | Brochure, 2000).

### 4.3. *Compression moulding*

A popular technique used for mass production in a shorter period is compression moulding. Pre-heating and pressurization are the requisites for moulding a composite in the desired shape. Fibres are cut, weighed and placed in a pre-heated mould. Mould is closed, pressurized before applying temperature and allowed for curing. Since no shear stresses and vigorous motion takes place, the damage to the fibre is minimal. These molten compounds take the shape of the cavity and the part is ejected when curing is complete. Both short-fibres (mat, stitched form) and long fibres can be used in this technique. The former is used to reduce the shrinkage and the latter to produce a high-volume fraction.

## 5. Mechanical properties of matrix and reinforcement

### 5.1. *Mechanical behaviour of polymeric materials*

A critical facet of designing a material that can bear the load, stresses without much deflection is governed by the mechanical behaviour of the

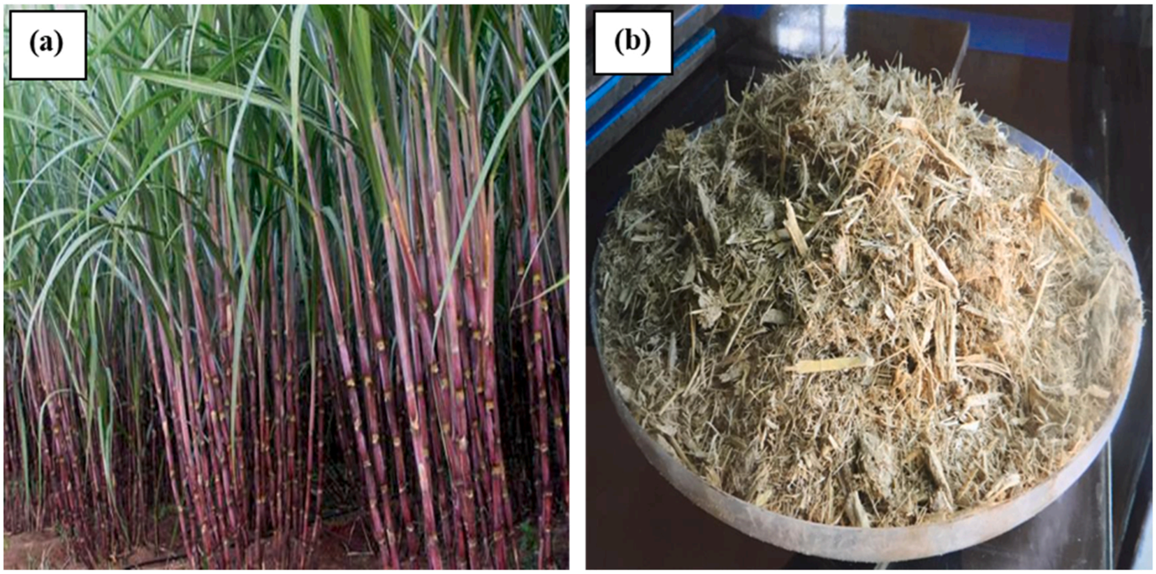


**Fig. 14.** (a) Sugarcane plant (b) Bagasse fibres.
(https://agritech.tnau.ac.in/agriculture/sugarcrops_sugarcane.html).

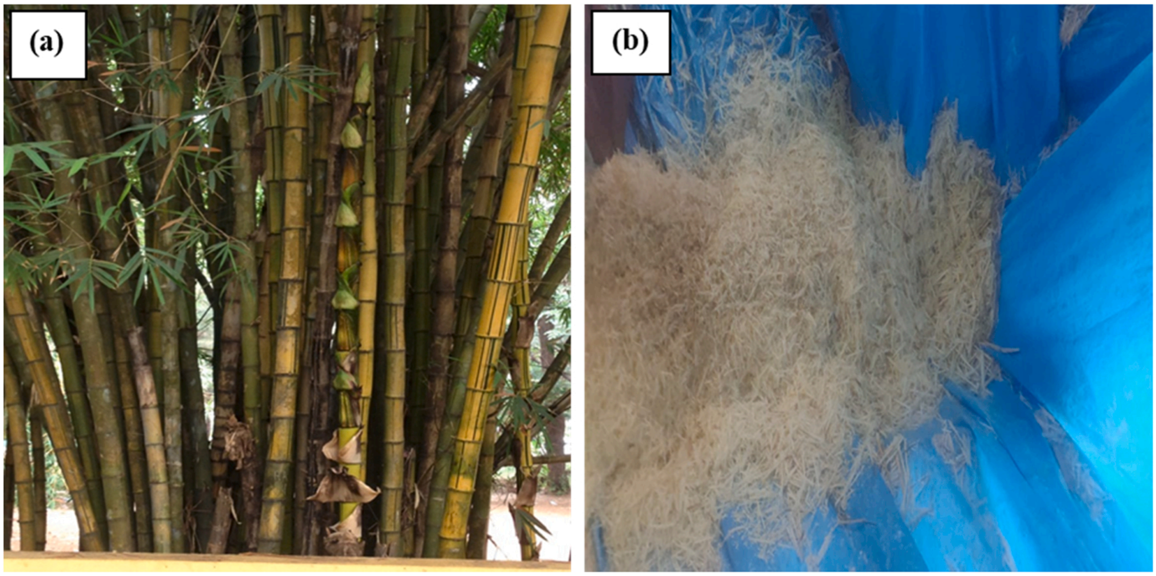


**Fig. 15.** (a) Bamboo plants (b) Bamboo fibres.

constituent materials. If a material can sustain the stresses subjected to it, the design is contemplated or if it fails, the design is iterated by choosing a new material or by changing the component design to overcome the failure (Ashby and Jones, 2006; Chanda and Roy, 2006). Unlike metal and ceramic-based composites which have constant strength and stiffness due to high melting points near ambient temperatures, the polymeric materials show a spectrum of mechanical behaviour from low to high temperatures, i.e., from brittle-elastic to plastic to rubbery to finally viscous. A polymer passes through all the mechanical states between − 20 °C to + 200 °C which can affect its strength and modulus by $10^3$ or more (Ashby and Jones, 2006).

### 5.2. *Mechanical properties of natural fibres*

Mechanical properties of cellulosic fibres are governed by various parameters like cell wall dimensions, chemical composition, defects, microfibrillar angle etc. Aspect ratio is another crucial factor that aids in estimating the strength of the composites. The length to diameter ratio (aspect ratio) should range from 100 to 200 for its optimal effectiveness. Fibres with a high aspect ratio are long and slender while lower aspect ratio fibres are short and wide in the transverse direction. Fibre architecture that enfolds fibre geometry, volume fraction, orientation and packing arrangement significantly affects the mechanical properties of the composites. Fibre geometry is a function of the morphology of tissues formed which to a certain degree is affected by the method of extraction and processing of the fibres. Orientation of fibres along the flow path during processing also causes mechanical properties to vary accordingly in different directions. The key factor that governs the mechanical properties is fibre volume fraction $V_f$. Mechanical properties increase with the increasing $V_{f.}$ to a certain extent after which the maximum value is dominated by the packing arrangement and orientation of the fibres which in turn is dictated by the adopted manufacturing processes. The strength of fibre also depends on its fibrillar angle. The tensile strength is maximum when the fibrillar angle is zero and decreases with the increasing angle.

### 5.3. *Tensile stress-strain properties of a composite material*

One of the most commonly used measurement standards for testing the mechanical parameters of composite material is the tensile test. A tensile test determines how well a material can resist tearing forces and the degree to which it stretches before fracture. As the tension steadily increases at a fixed cross-head speed, the load and cumulative elongation over gauge length is monitored and measured continuously until the specimen breaks. The test produces a stress-strain graph that varies for different materials. Tensile strength and modulus are two important strength indicators in a plastic material that illustrate a material's mechanical behaviour when subjected to tensile force (Campo, 2008).

## 6. Effect of various chemical treatments and hybridization on tensile properties of natural fibre epoxy composites

### 6.1. *Chemical modification of cellulosic fibres*

The mechanical interlocking of fibres and polymer matrix at the interface plays a crucial role to facilitate load transfer efficiency for the improved mechanical performance of the composites (Shi et al., 2017). Natural fibres are inherently polarized (hydrophilic) which renders them incompatible with the hydrophobic matrices (Faruk, 2012; Lau et al., 2018). A possible solution to improve the interaction between the two is chemical modification or pre-treatment method, which modifies the fibre surface by increasing its roughness or by introducing new surface moieties that readily interlock with the matrix (Kalia et al., 2009). Surface modification methods decrease the rate of water sorption, whereas bulk modifications additionally reduce the capacity of water sorption (Xie et al., 2010). The weak interface is the result of poor wettability, poor moisture resistance, poor adhesion etc. which can be improved by the use of compatibilizers, adhesion promoters etc (Kalia et al., 2009). A few treatment methods are briefed in the following subsections:

#### 6.1.1. *Alkaline treatment (Mercerization)*

A treatment in which lignocellulosic fibres are subjected to a strongly basic medium (usually NaOH) to attain changes in the fibre structure, mechanical properties etc. is called Mercerization. It is carried out following ASTM-D 6942–03 ("Standard Test Method for Stability of Cellulose Fibers in Alkaline Environments, 2019)). The effect of mercerization is a swelling reaction in which cellulose-I (native cellulose) is transformed to cellulose-II. $Na^+$ ions from NaOH widen the smallest pores of cellulose fibres by penetrating them forming a new sodium-cellulose-I lattice. Disruption of hydrogen bonding occurs resulting in rough surface topography (Chand, 2008). Cellulose fibres now contain -ONa groups rather than hydrophilic OH groups which gives better mechanical interlocking. The chemical reaction that takes place is as follows (Kalia et al., 2009):

$$\mathrm{Fibre-OH+NaOH \rightarrow Fibre-O-Na^{+} \quad + H_2 \quad O} \tag{1}$$

#### 6.1.2. *Silane treatment*

The chemical formula for Silane is $SiH_4$. Silanes are the coupling agent that improves cross-linking at the interface (Li et al., 2007; Yan et al., 2016b). The silicon of silane is appended with the groups in such a way that one end interacts with the matrix and the other with the hydrophilic fibre, enabling it to act as a bridge between the two. Alkoxy silanes react with moisture to generate silanols. These silanols react with the fibre's OH group, forming covalent bonds that are chemisorbed on the fibre's surface. Thus, the hydrocarbon chain as a function of silane impedes the swelling of the fibre by forming covalent bonds (Li et al., 2007; Yan et al., 2016b). In other words, these coupling agents form a tough layer by eliminating the weak one to form a strong cross-linked network. The development of this strong interphase results in an intermediate modulus between the polymer and the substrate thereby improving the wettability (Chand, 2008; Dittenber and GangaRao, 2012; Malkapuram et al., 2009).

#### 6.1.3. *Acetylation*

An esterification process that causes the plasticization of lignocellulosic fibres is called acetylation (Chand, 2008; La Mantia and Morreale, 2011). In this treatment, the hydroxyl groups of the fibres (usually hemicelluloses and lignin) are substituted with acetyl groups in the presence of acetic anhydride ($CH_3CO - O - OC - CH_3$), rendering the surface hydrophobic. The acetylation reaction of -OH group of fibre can be shown as (Chand, 2008):

$$\mathrm{Fibre-OH+CH_3CO-O-OC-CH_3 \rightarrow Fibre-O-CO-CH_3+CH_3COOH} \tag{2}$$

Acetylation thus reduces hygroscopicity thereby increasing the dimensional stability of composites (Chand, 2008).

#### 6.1.4. *Isocyanate treatment*

Isocyanate is a coupling agent having the functional group $-N{=}C{=}O$ that reacts with the –OH groups of cellulose and lignin (Chand, 2008; Li et al., 2007; Yan et al., 2016b). In this treatment, fibres are treated with NaOH, washed, dried and then soaked in $CCl_4$. A catalyst is added and the reaction is carried out with continuous stirring at a slightly higher temperature than the ambient condition. Purification is done by refluxing, subsequently drying the fibres at 100 °C. The reaction that takes place between fibres and coupling agent is as follows:

$$\mathrm{R-N=C=O+OH-Fibre \rightarrow R-NH-C=O-O-Fibre} \tag{3}$$

where R refers to different chemical groups (e.g., alkyl group). This treatment creates better compatibility between fibre and polymer thereby improving interfacial adhesion (Chand, 2008).

### 6.2. *Hybridization*

Hybrid composites are the systems in which integration of varied fibres in a single matrix or a single filler in different matrices is done to develop composites (Saba et al., 2014). Hybridization is a function of the stability of physical, chemical and mechanical properties of the fibres and the matrix which improves the overall performance of the composites. Fillers or reinforcements can be incorporated in various ways by:

1. Intermixing of two types of lignocellulosic fibres with or without modification before adding to polymers like polyesters, polyvinyl ester, polyurethane resins etc.
2. Sandwiching the fibres or using them in mat form.
3. The intermingling of natural fibres with synthetic fibre.

The properties of the hybrid composites are governed by the individual lengths of the fibre, bonding, extent of intermingling, orientation etc. and are studied by the Rule of mixtures which is a weighted sum of the individual components (Yan et al., 2016b). Hybridization can either have a positive or negative effect on the tensile properties of the natural fibre reinforced composites (Dong, 2018).

## 7. Tensile properties

### 7.1. *Effect of Jute fibres on tensile properties of composites*

Improvement in mechanical properties of laminated composites was investigated by applying two surface treatments on the jute fabric, alkali and siloxane. The efficacy of the treatments to disarray hydrogen bonding and induce compatibility was observed on two thermosets viz.

unsaturated polyester and epoxy. The 6 layered 3.5 mm laminated composites were fabricated by hand lay-up followed by compression moulding at 120 bar pressure and the properties were evaluated at 35% fibre volume fraction. Compared to mercerization, the strength of 1% siloxane treated ASJE (jute/epoxy composites) was found to be 81 MPa, which was 22% higher than AJE's (alkalized jute epoxy's) 66 MPa due to the presence of functional alkyl group in siloxanes that acted as a good dispersing agent resulting in a better interface. Table 2 highlights the mechanical properties of jute fibre composites reinforced in varied forms at varying percentages. However, a decreasing trend was observed in the case of modulus from 6.28 to 5.74 GPa with a decrement of 8.5% in ASJE than AJE (Seki, 2009).

Mechanical properties after modifying epoxy with different concentrations of oligomeric siloxane were investigated by (Sarikanat, 2010). Dimensions of the laminates, fabrication technique and fibre volume fraction were the same as aforementioned with an applied pressure of 150 bars. Results found were similar to those by (Seki, 2009) and showed that at 3% w/w of AJE (alkalized jute/siloxane modified epoxy) the tensile strength increased from 66.5 MPa to 86.73 MPa, i.e. by 30.4% over AJE composites. Tensile modulus also increased by 69.1% and was recorded as 9.28 GPa (Fig. 1). This enhancement in properties was due to the improved adhesion as a result of higher siloxane concentration which was confirmed by Scanning Electron Microscopy.

According to (Pinto et al., 2014), the vacuum-infusion technique can produce void-free composites in comparison to hand lay-up. The latter showed a good fibre volume fraction (FVF) but with a massive void content. Both alkali and silane treatments were applied on the woven unidirectional preforms to improve the fibre-matrix adhesion. Silane treatment improved the interface resulting in an increase in the tensile modulus by 12% from 11.9 to 13.4 GPa irrespective of the fibre architecture. No significant change was seen in the tensile strength as the twist in the jute yarns caused poor alignment in the loading direction thereby reducing its strength.

Hybrid composites were developed using the hand lay-up technique in which hybridization of oil palm empty fruit bunch fibres (EFB) with woven jute fibre mat was studied by (Jawaid et al., 2011) in various layering patterns. Results showed an increase in strength by hybridization. Tensile strength and modulus were found to be 32.5 MPa and 2.8 GPa respectively when woven jute fibres were used as outer layers since jute fibres withstood the stress and EFB simultaneously absorbed and distributed it evenly. However, pure jute composites showed the highest strength and modulus of 53.31 MPa and 4.6 GPa since jute fibres are stiffer and stronger than EFB. They concluded that strength can be increased by using a high strength material as the outer skin.

The effect of stacking sequence of four plies processed by hand lay-up on the mechanical properties of composites was examined by (Gujjala et al., 2014). Woven jute fibres were stacked with glass mats in different layering patterns. It was seen that the tensile strength of jute fibres was 142% more than neat epoxy i.e., 56 MPa but the laminates, when incorporated with glass fibres in 50:50 wt% ratio increased the strength to 86 MPa i.e., by 66% when compared with pure jute laminates. Modulus also increased due to the presence of stiffer and stronger glass fibres.

The tensile properties of hybrid jute-glass composites fabricated by resin infusion were reported by (Pandita et al., 2014) and the results showed that they are greatly affected by the orientation of fibres in two directions, namely 'main' and 'bias. However, jute fibres increase the stiffness in the 'main' direction but the results showed that the tensile strength of jute fibres was 47.15 MPa which was even less than pure epoxy's 80 MPa when oriented perpendicular or in the 'mains' of the loading direction. The decrease can be attributed to the fibres porous nature which acts as a hollow structure having voids. Tensile strength of the woven sandwiched composites in the 'mains' direction was found to be 74 MPa and the result aligns with the fact that properties of the fibres are direction-oriented.

Cross-plied jute-banana hybrids (mat form) were manufactured by hand lay-up technique followed by compression moulding and was evaluated for their mechanical properties by (Boopalan et al., 2013a). With 100 wt% of jute, the tensile strength and modulus were found to be 16.62 MPa and 0.664 GPa respectively and with 50:50 wt% of the hybrid, the strength increased marginally from 16.62 to 18.96 MPa. This

**Table 2**
Mechanical properties of Jute fibre reinforced epoxy composites.

| Composite | Raw material (wt%) | Processing method | Tensile strength (MPa) | Tensile modulus (GPa) | Flexural strength (MPa) | Flexural modulus (GPa) | Impact strength (KJ/m$^2$) | Water absorption (%) | Density (ρ) (g/cm$^3$) | References |
|---|---|---|---|---|---|---|---|---|---|---|
| Jute comp. | Fibres, (78:22) | Hand layup- CM | 66.36–72.96 (65.35) | 5.59 – 6.79 (6.19) | 91.08–97.08 (94.08) | 5.41–6.41 (5.91) | – | – | – | (Seki, 2009) |
| Jute/ modified | Fabric, (78:22) ($V_f$-35%) | Hand layup- CM | 82.71–90.75 (86.73) | 9.05–9.51 (9.28) | 105.8–114.46 (110.13) | 6.96 – 7.56 (7.26) | – | – | – | (Sarikanat, 2010) |
| Jute fibre LC | Unidirectional fabric (100:27) | vacuum infusion | 74.3 | 13.4 | – | – | – | 4.28 | – | (Pinto et al., 2014) |
| Oil Palm-woven jute | Fibre mat | Hand layup | 33 | 2.8 | 57 | 3.25 | – | – | – | (Jawaid et al., 2011) |
| Oil Palm fruit jute hybrid | Fibre mat | Hand layup | – | – | 49.0 | 3.07 | 57.0 J/m | – | – | (Jawaid et al., 2010) |
| Jute/Glass hybrid | Woven (10:1) | Hand layup | 86 | 4.8 | 164 | 6.5 | – | – | 1.26 | (Gujjala et al., 2014) |
| Jute/Glass hybrid sandwich | Jute plain weaves | Resin infusion | 74 | 4.8 | – | – | – | 5.38 | 1.27 | (Pandita et al., 2014) |
| Jute and banana hybrid | Jute fibres, (10:1) | Hand layup- CM | 16.62 | 0.664 | 57.22 | 8.956 | 13.44 | 0.81 | – | (Boopalan et al., 2013a) |
| Jute fibre | Jute slivers | Hydraulic press | 148.3 | 3.184 | 196.11 | 20.45 | 107.94 J/m | – | – | (Mishra et al., 2000) |
| Jute fibre | Fabric (100:32) $V_f$ – 0.25–0.27 | Hand layup- CM | | | 39 | 4.2 | 32.17 | 4.95 | | (Jabbar et al., 2016) |

**CM**-Compression moulding; **LC**-Laminate composites; () The values in the parentheses are the average of the above properties slight increase is because of weak interfacial bonding. Modulus increased by 9% from 664 to 724 MPa.

Jute slivers (unmodified and bleached) prepared by carding were used as reinforcement to strengthen epoxy. Composites with three distinct percent of fibres (40, 50, 57%) processed as laminates by the hydraulic press at 150°C and 4.35 MPa pressure for 5 min was assessed by (Mishra et al., 2000). It was found that the unmodified composites with 50 wt% fibre resulted in a higher tensile strength of 148.3 MPa and a modulus of 3.184 GPa. The increase in strength was a result of proper wetting of fibres. (Fig. 16).

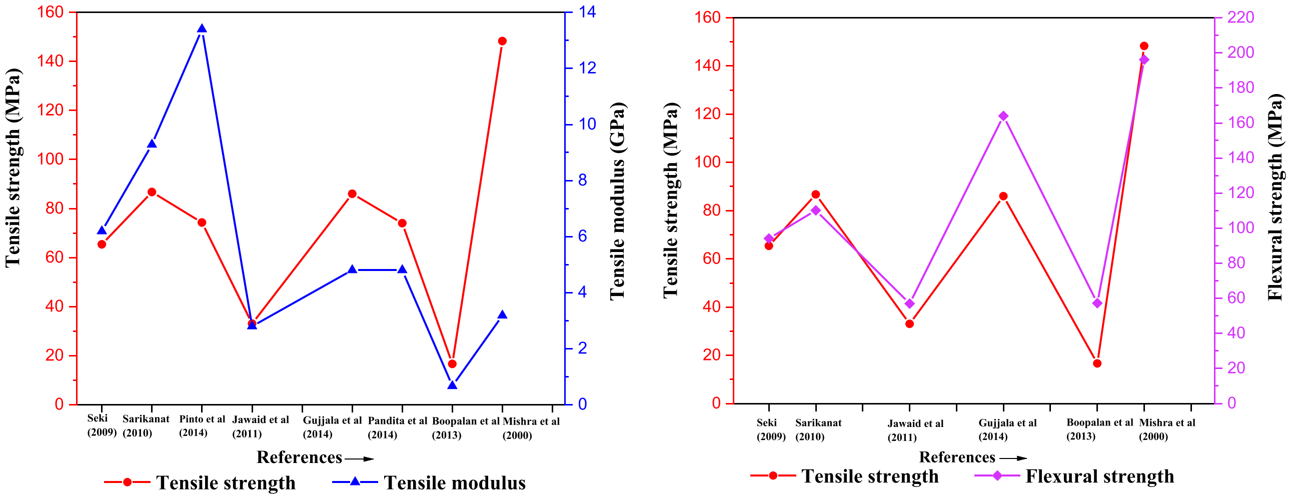
### *7.2. Effect of Sisal fibre on tensile properties of composites*

The interfacial properties of unidirectional composite laminates made by the hot-pressing technique were investigated. The processed composites with varied fibre content under diversified chemical treatments were evaluated by (Rong et al., 2001) (Table 3). It was found that alkali-treated fibre composites showed a series of improvements in the tensile strength from 190 to 380 MPa with 30–58 vol% fibre content with a maximum strength achieved at 58 vol% due to proper wetting whereas untreated fibres could reach only 320 MPa at 75% fibre content. However, improvement of 15% was seen in the strength but it was quite lower than the strength of alkali-treated sisal fibre itself. The observed nominal improvement was due to the decohesion of cells that resulted in weak intercellular adhesion and was seen as cell pull-out in the micrographs. Tensile modulus showed a decreasing trend from 8.3 to 5.6 GPa due to the lower stiffness of the treated fibres. They concluded that higher strength can be acquired by maintaining isometric conditions where shrinkage is 0% or stretching is dominant.

Chopped fibres were hybridized with banana and glass fibre to study the mechanical properties of nine laminates processed by compression moulding (Arthanarieswaran et al., 2014a). Pure sisal laminate showed a tensile strength of 23 MPa and by adding banana fibre, the strength raised by 8% to 25 MPa. However, two glass layers when added increased the strength by 126% but hybridizing banana-sisal with glass gave the highest tensile strength of 104 MPa by withstanding more tensile forces. Modulus also increased by 72% from 0.67 to 2.35 GPa.

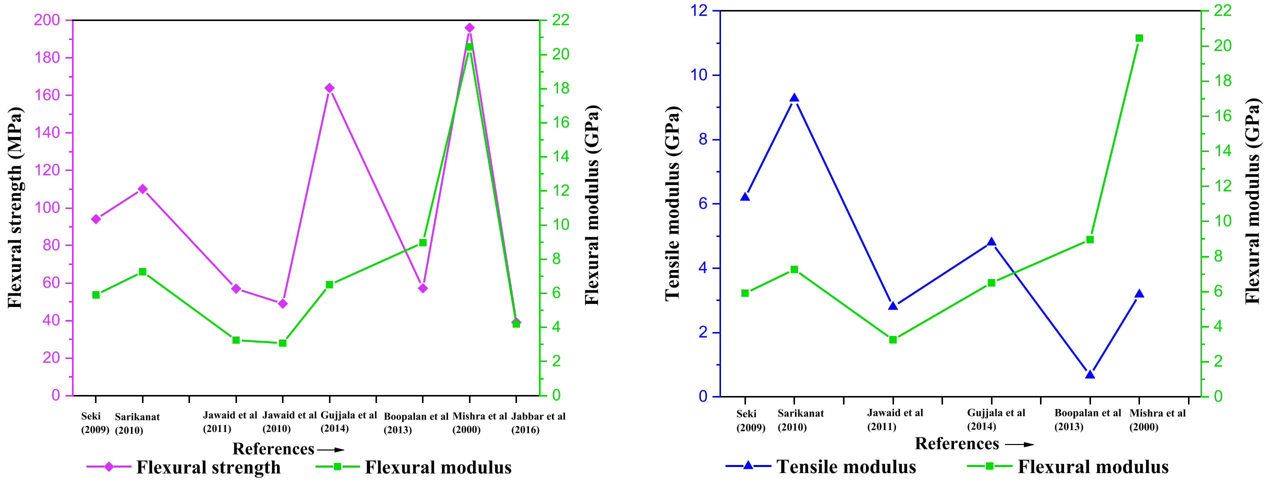
(Padmavathi et al., 2012) explored unidirectional bio-composites with modified (18% NaOH treated) sisal fibres at varied weight fractions. An improvised technique was used to manufacture the composites namely wet lay-up. The obtained linear graph showed an increase in the tensile strength with the increasing fibre fraction, i.e., the strength increased from 161.3 to 235 MPa from 0.15 $W_f - 0.40$ $W_f$. The increase in strength was due to the orientation of fibres along the axis, the chemical treatment that eliminated the impurities and decreased fibre diameter. Tensile modulus also increased linearly to a lesser extent as compared to its strength.

Mechanical properties of raw and dewaxed short-sisal fibre (20 mm)



(a) Tensile strength versus Tensile modulus

(c) Tensile strength versus Flexural strength



(b) Flexural strength versus Flexural modulus

(d) Tensile modulus versus Flexural modulus

**Fig. 16.** Tensile and Flexural Properties of Jute fibre reinforced epoxy composites.

**Table 3**
Mechanical properties of Sisal fibre reinforced epoxy composites.

| Composite | Raw material (wt%) | Processing method | Tensile strength (MPa) | Tensile modulus (GPa) | Flexural strength (MPa) | Flexural modulus (GPa) | Impact strength (KJ/m$^2$) | Water absorption (%) | Density (ρ) (g/cm$^3$) | References |
|---|---|---|---|---|---|---|---|---|---|---|
| Sisal laminates | Fibres, D-100–200 μm, (1.2/1) | Hot Pressing | 380 | 5.6 | 325 | 19 | – | – | – | (Rong et al., 2001) |
| Sisal laminates | Chopped fibres, 10:1 | CM | 23 | 0.67 | 61 | – | – | – | – | (Arthanarieswaran et al., 2014a) |
| Sisal laminates | Fibres, D-100–300 μm | Hand layup | 235 | 6.42 | – | – | – | – | – | (Padmavathi et al., 2012) |
| Sisal comp. | Fibres, D-170–300 μm 10:1 | Hand layup | 34 | | 60 | – | – | – | – | (Patra and Bisoyi, 2011) |
| Sisal comp. | Fibres 0.4 $V_f$ | CM | | | 262.1 | 17.63 | – | – | 1.24 | (Bisanda and Ansell, n.d.) |
| Sisal comp. | Fibres 10:8, $V_f$- | Hot Pressing | 43 | 1.046 | | | 20.29 | | 1.23 | (Hashmi et al., 2011) |
| Sisal comp. | Sisal fibres | CM | | | 74 | 6.4 | | | | (Gañan et al., 2005) |
| Sisal comp. | Woven Sisal fibres 10:1 | Hand layup | 25 | | 3 | | 0.4 J | | | (Sekaran et al., 2015) |
| Sisal comp. | Sisal fibre mat | RTM | 199–223 (211) | 18.2–21.2 (19.7) | | | | | | (Oksman et al., 2002) |
| Sisal bio-comp. | Sisal fibre mat | Hand layup-CM | 116.2 | 3.4 | 112 | 5.7 | 19 | | | (Sahoo et al., 2015) |

**CM**- Compression moulding; **RTM**-Resin transfer moulding; () The values in the parentheses are the average of the above properties.

epoxy composites at 15 vol%, obtained by chemical modification and fabricated by hand lay-up technique was reported by (Patra and Bisoyi, 2011). Dewaxing was done by cooking the fibres in a 1:2 mixture of ethanol and benzene to achieve a hohlraum character i.e., the free spaces. Compared to raw's 26 MPa, dewaxed fibres demonstrated a higher strength of 34 MPa, which clearly showed that the removal of cementing material increased fibres coarseness for better contact and the replacement of space by epoxy improved the adhesion forming reactive bonds. Studies regarding modulus were not reported.

Hybrid sisal-glass epoxy composites were designed and fabricated by a hot-press technique using low strength sisal paper as reinforcement rather than sisal fibre. Reinforcement used was in the range of 22.34–23.7%. Tensile strength and modulus obtained for the pure sisal paper sample were 32.54 MPa and 517 MPa in the low strain range (0–0.02). When one glass layer (3.4 vol%) was hybridized with 3 sisal layers (18.88 vol%) the strength increased up to 43 MPa and modulus reached 531 MPa. The results drawn from various sample testing were that tensile strength increased with an increase in the volume fraction of glass fibres till critical packing density beyond which it started decreasing due to non-wettability, voids etc. Modulus also showed an increasing trend. They also concluded that both these properties are highly influenced by the length of the reinforcement as short fibres do not increase the strength but do contribute to improving the modulus (Hashmi et al., 2011).

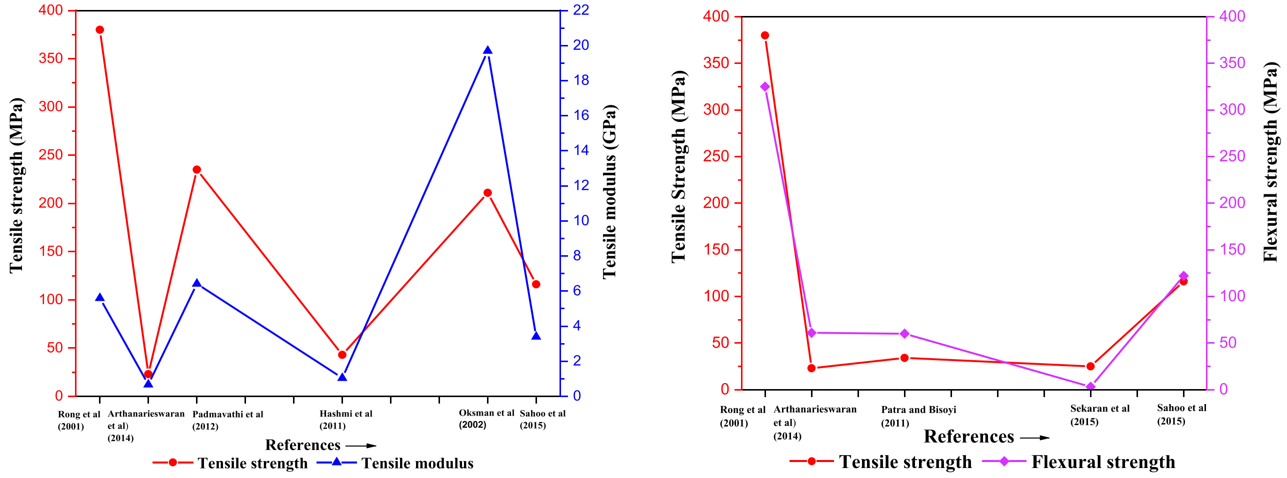
Woven Aloe vera and sisal hybrids designed by hand lay-up in the form of laminates were used to examine the mechanical properties of composites. Hybridization of the two increased the tensile strength from 25 to 27 MPa but flexural strength remains unchanged (Sekaran et al., 2015). It can be asserted from their work that proper manufacturing techniques and the selection of the correct chemical treatment for fibres can improve mechanical properties. Modulus was not reported.

The research was carried out to analyze the longitudinal tensile strength and modulus of technical fibres (mat form) reinforced in sisal-epoxy composites prepared by the Resin Transfer Molding (RTM) technique. Three different samples were prepared with different fibre content. It was found that at 46% vol the tensile strength and modulus was maximum i.e 211 MPa and 20 GPa respectively. When literature and measured data were compared, a great deal of difference was seen in the physical properties of the sisal. Mechanical testing revealed that when reinforced in the matrix, technical fibres displayed a higher modulus, greater than 20 GPa than the 7–20 GPa reported in the literature. Conversely, the strength of technical fibres was 211 MPa when compared to the measured strength of (550 ± 100) MPa. This is because of the non-uniform distribution of fibres as they are pulled out parallelly to the direction of the fibres during processing, attaining a different cross-section shape. They didn't report the data on flexural properties (Oksman et al., 2002).

The mechanical properties of sisal biocomposites were studied using two matrices, the unmodified petroleum-based and the other modified with two bio-resins namely epoxidized soybean oil (ESO) and epoxy methyl soyate (EMS) (Sahoo et al., 2015). The processing was done by hand lay-up technique followed by compression moulding. Tensile strength of EPEMSSF was found maximum (116.2 MPa) due to the addition of organic oil which increased the interfacial interaction resulting in better stress transfer. Modulus of unmodified biocomposite (EPSF) was found to be 3.2 GPa which was more than EPESOSF's 3 GPa but less than EPEMSSF's 3.4 GPa. This increase in modulus was due to the high stiffness and presence of monoglyceride esters of EMS as compared to triglycerides of ESO which reduced the modulus. (Fig. 17).

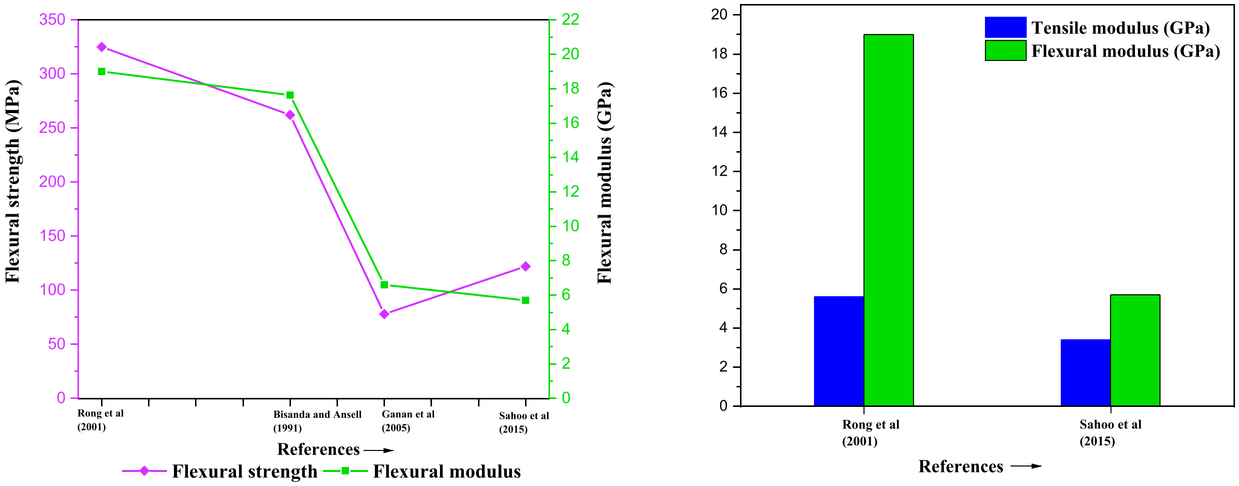
### 7.3. *Effect of banana fibres on tensile properties of the epoxy resin system*

The influence of hybridization of two natural fibres (banana and jute) and synthetic fibre (glass fibre) on the mechanical properties of composites processed by hand lay-up has been studied by (Vijaya Ramnath et al., 2015). Tensile strength of 68.42 MPa was attained with



(a) Tensile strength versus Tensile modulus

(c) Tensile strength versus Flexural strength



(b) Flexural strength versus Flexural modulus

(d) Tensile modulus versus Flexural modulus (GPa)

**Fig. 17.** Tensile and Flexural Properties of Sisal fibre reinforced epoxy composites.

banana and glass fibres. However, the strength enhanced to 85.91 MPa when three fibres were hybridized together. It is evident that hybrid fibre composites exhibited better properties than their mono parts, However, the results on the modulus were not reported.

(Arthanarieswaran et al., 2014b) investigated hybridization of glass in banana composites fabricated by compression moulding. It was recorded that pure banana laminate achieved a strength of 21 MPa. Incorporation of two and three layers of glass fibres increased the strength to 46 and 88 MPa showing an improvement of about 119, 319%. Modulus improved from 0.62 to 1.42 and 1.94 GPa (Fig. 3). Hybridization of fibres increases the tensile strength and modulus of composites.

The properties of hybrid composites of banana and jute manufactured by hand lay-up have been analyzed by (Boopalan et al., 2013b). The tensile strength and modulus of banana fibre were found to be 17.92 MPa was 0.718 GPa but by hybridizing jute and banana in 25/75, 50/50 ratio, the strength increased to 18.25 MPa, 18.96 MPa by 1.84% and 5.8% and modulus enhanced by 0.27, 0.83% to 0.72, 0.724 MPa.

The effect of fibre content and fibre length on the mechanical properties of hand lay-up processed composites were tested by (Venkateshwaran et al., 2011). Four different length's of fibre were taken (5, 10,15,20 mm) with a fibre weight of (8,12,16,20%). The tensile strength of 16.39 MPa and modulus of 0.652 GPa were reported at 5 mm fibre length with a 12 wt% ratio. It was asserted that the optimum fibre length and weight % are 15 mm and 16% below which the values decreased due to the increase in the content of fibre than the matrix and reduced interfacial adhesion.

Another experimental study carried out by (Balaji et al., 2020) concluded that the mechanical behaviour of composites was influenced by fibre length and loading percentage. The alkali-treated banana-epoxy composites were fabricated by a compression moulding technique. Strength of 30.4 MPa was achieved with 20 mm length and 15 wt% loading showing a strong adhesion between fibre and matrix. Modulus was not reported.

The experimental investigation on pseudo-stem banana woven fabric in epoxy resin prepared by hand lay-up process was reported by (Maleque et al., 2007). For banana-epoxy composites, the tensile strength was found to increase by 90% compared to unreinforced epoxy i.e., from 23.98 MPa to 47 MPa. Modulus also increased by 36% from 1.39 GPa to 1.89 GPa. The experimental data revealed that reinforced banana fibres distributed the stress uniformly from the matrix to the fibres attributing to a higher strength.

The effect of fibre length, loading, alkaline content on tensile strength of Musa acuminate (pseudo-stem) in epoxy composites was investigated by. The composites cured at room temperature showed a tensile strength of 22.86 MPa with 3.25 mm length at 29.86 wt% loading and 5.45 wt% NaOH content. Due to strong adhesion between fibres and matrix, the strength increased by 22% over neat epoxy resin.

The use of banana fibres with a 40% volume fraction in the epoxy resin system was studied by (Pappu et al., 2015). Tensile strength of 78 MPa and modulus of 0.758 GPa were reported as seen in Table 4. It was observed that banana fibres incorporation can bear a higher load as compared to pure epoxy due to the transmission of stress from epoxy to the fibres resulting in increased adhesion thereby giving a higher strength.

One of the studies conducted by (Devireddy and Biswas, 2017) on hybrid composites using varied weight percentages of banana fibres manufactured by hand lay-up, showed that the use of 30% fibre resulted in tensile strength of 49.32 MPa. Due to agglomeration, fibre entanglement and lack of epoxy-rich areas, the rise in fibre weight percent from 30 to 40 decreased the tensile properties. However, infused hybrids (banana & jute) in epoxy attained a strength of 64.75 MPa. Further details on tensile modulus have not been reported.(Fig. 18).

### 7.4. *Effect of pineapple fibre on tensile properties of the epoxy resin system*

According to (Nagarajan et al., 2016) the surface-treated composites displayed better mechanical properties as compared to the untreated composites. The effect of fibre content on the tensile properties of untreated and treated unidirectionally laid fibres, fabricated by hand lay-up technique was investigated. Fibre loading of 20% by wt gave the highest modulus of 3.44 GPa in the case of alkali-treated composites over 2.03 GPa of neat epoxy. The increase in modulus of alkali-treated fibres was due to the removal of hemicelluloses, lignin which allowed the epoxy to infuse in fibres improving the adhesion which was further verified by the results of FTIR. Increasing the fibre loading percent beyond 20, however, caused a reduction in properties.

Grafting of modified pineapple fibres using a coupling agent 3-aminopropyl-triethoxysilane on epoxy to enhance the compatibility between the two was studied by (Shih et al., 2012). The tensile strength of the modified fibre-reinforced composites was found to be 39.9 MPa which was 82% greater than the pristine epoxy (Table 5). The strength in the case of modified fibres increased more as compared to the untreated ones, due to a cross-linked network formed between the fibres and epoxy. Also, the increased fibre content of 20% by weight increased the rigidity of the composites resulting in better adhesion. Modulus of elasticity was not reported.

The effect of modified pineapple leaf fillers at varied loading percentages prepared by wet ball milling, fabricated by hand lay-up process in the epoxy composite has been investigated by (Kumar et al., 2020). For 2.5 per cent of filler loading at 2 mm/min, the highest value of tensile strength and modulus recorded was 22 MPa and 570 MPa. The increase in strength could be attributed to better stress transfer from matrix to fibres giving better mechanical interlocking. However, at high filler content, the strength decreased due to improper wetting and agglomeration of fillers.

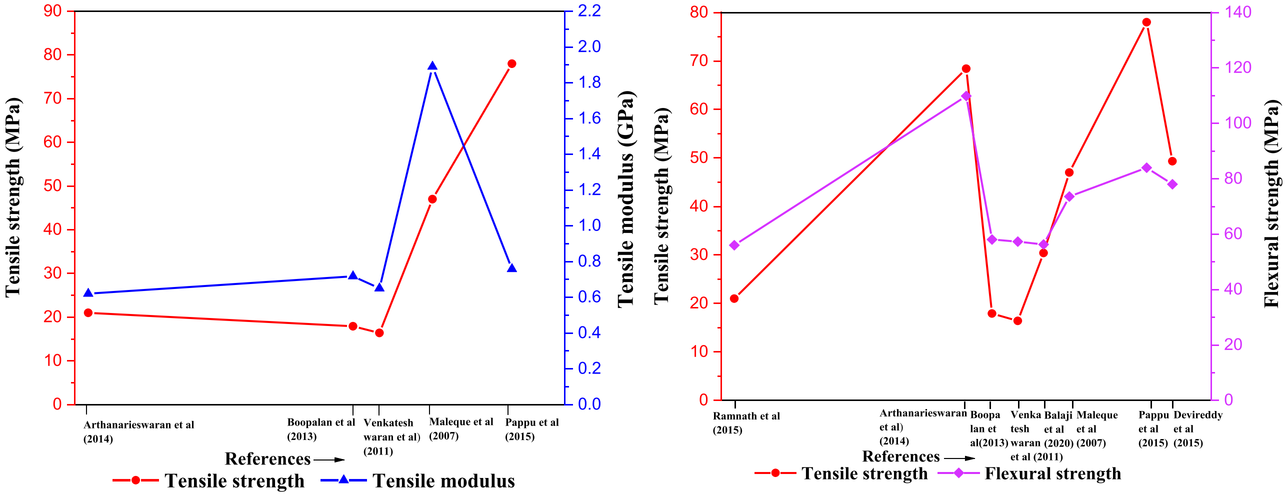
## 8. Micromechanical modelling

The design rationale to compute the structural mechanics of reinforcing fibres in polymer matrix composites embodies the approach of micromechanics. Micromechanics is the simplest mathematical estimation to determine the stiffness of the constituent materials in a composite i.e., an expression can be derived through statistics to understand what is happening at the level of matrix and fibre (Harris, 1999; Hyer and Waas, 2018; Jones, 2018). Although several mathematical models have been proposed to understand the interacting mechanism of the constituents in a heterogeneous material, they usually predict the tensile properties of continuous fibre composites only as they act as a prelude to model more realistic situations (Aruan Efendy and Pickering, 2019; Jones, 1999; Tucker III and Liang, 1999). Two important parameters that dominate the performance of composites are the strength and stiffness of the fibre. Tensile properties of continuous and uniaxially aligned fibres can be predicted accurately by the Rule of Mixtures. Perversely, in discontinuous fibre composites, the same rule is not obeyed as the properties are largely influenced by factors like elastic properties of the fibres, fibre orientation, resin-fibre interactions, load transfer efficiency from matrix to fibre via interface, etc (Aruan Efendy and Pickering, 2019). Micromechanical models are used to determine the elastic behavior of fibre composites, in which an isostress and isostrain condition is assumed with idealized geometries (Chen et al., 2014). It is presumed that fibres and matrix are perfectly bonded, and deform together (Harris, 1999). In this review, analytical modeling schemes to predict the strength and young's modulus of fibre composites are briefly elucidated.

**Table 4**
Mechanical properties of Banana fibre reinforced epoxy composites.

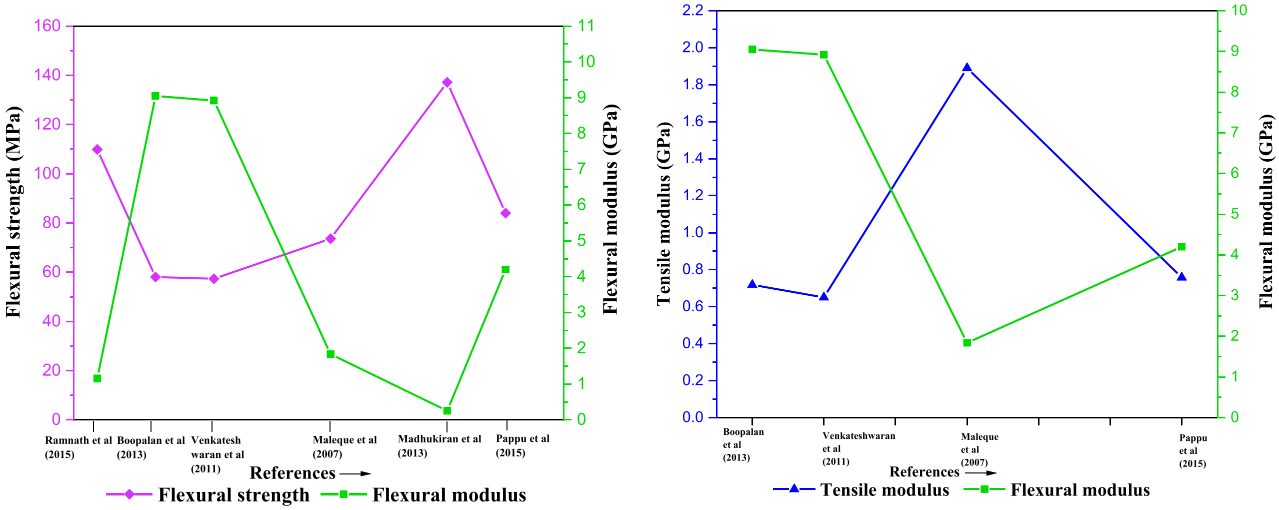
| Composite | Raw material (wt%) | Processing method | Tensile strength (MPa) | Tensile modulus (GPa) | Flexural strength (MPa) | Flexural modulus (GPa) | Impact strength (KJ/$m^2$) | Water absorption (%) | Density (ρ) (g/$cm^3$) | References |
|---|---|---|---|---|---|---|---|---|---|---|
| Banana jute hybrid | Banana fibres, Epoxy | Hand layup | 68.42 | – | 109.86 | 1.156 | 423.63(J/m) | – | – | (Vijaya Ramnath et al., 2015) |
| Banana fibres | Banana fibres, Epoxy (10:1) | CM | 21 | 0.62 | 56 | – | – | – | – | (Arthanarieswaran et al., 2014b) |
| Banana fibre hybrid | Banana fibres, Low temp Epoxy(10:1) | Hand layup-CM | 17.92 | 0.718 | 58.06 | 9.048 | 16.92 | – | – | (Boopalan et al., 2013b) |
| Banana fibre | Banana fibres, Epoxy | Hand layup | 16.39 | 0.65 | 57.33 | 8.92 | 2.3 J/m | 4.8 | – | (Venkateshwaran et al., 2011) |
| Banana fibres | Banana fibres | CM | 30.4 | – | 56.3 | – | 2.7 J/$mm^2$ | – | – | (Balaji et al., 2020) |
| Banana fibres | Banana woven fabrics, Epoxy (4:1 vol%) | Hand layup | 44–50 (47) | 1.89 | 73.58 | 1.835 | 6.95 | – | – | (Maleque et al., 2007) |
| Banana | Banana fibres, Epoxy (3:2) | Hand layup | 22.86 | – | – | – | – | – | – | |
| Banana fibres | Banana fibres, Epoxy (10:1) | Hand layup | – | – | 137.17 | 0.252 | – | – | – | ("banana 8.pdf," n.d.) |
| Banana fibres | Banana fibres, Epoxy (50:50 vol%) | Hand layup | 78 | 0.758 | 84 | 4.2 | 40 | – | – | (Pappu et al., 2015) |
| Banana fibre | Banana fibres, epoxy (10:1) | Hand layup-CM | 49.32 | – | 78 | – | – | – | 1.159 | (Devireddy and Biswas, 2017) |

**CM-** Compression moulding; () The values in the parentheses are the average of the above properties



(a) Tensile strength versus Tensile modulus

(c) Tensile strength versus Flexural strength



(b) Flexural strength versus Flexural modulus

(d) Tensile modulus versus Flexural modulus

**Fig. 18.** Tensile and Flexural Properties of Banana fibre reinforced epoxy composites.

**Table 5**
Mechanical properties of Pineapple leaf fibre reinforced epoxy composites.

| Composite | Raw material (wt%) | Processing method | Tensile strength (MPa) | Tensile modulus (GPa) | Flexural strength (MPa) | Flexural modulus (GPa) | Impact strength (KJ/m$^2$) | Water absorption (%) | Density (ρ) (g/cm$^3$) | References |
|---|---|---|---|---|---|---|---|---|---|---|
| Pineapple/Epoxy comp. | PALF, (D- 20–80 µm) (100:11) | Hand layup | – | – | 145–165 (155) | – | 33.44–50.48 (41.96) | – | – | (Lopattananon et al., 2008) |
| Pineapple/Epoxy comp. | PALF | Hand layup | – | 3.44 | – | 6.4 | 28 (J/m) | – | – | (Nagarajan et al., 2016) |
| Pineapple/Epoxy comp. | PALF, (L- 2.3–3.9 mm) | Hand layup | 39.9 | – | – | – | – | – | – | (Shih et al., 2012) |
| Pineapple/Epoxy comp. | PALF fillers | Hand layup | 22 | 0.57 | 31 | 0.89 | – | – | – | (Kumar et al., 2020) |
| Pineapple/Epoxy comp. | PALF (D- 30–50 µm) (100:11) | Hand layup | – | – | – | – | – | – | – | (Payae and Lopattananon, 2009) |

**PALF-** Pineapple leaf fibre**,** () The values in the parentheses are the average of the above properties

### 8.1. Rule of mixture (ROM Model for Young's modulus prediction)

The simplest model to predict young's modulus of a composite parallel to the principal axis using empirical equations is the Rule of Mixture (Virk, 2010). ROM renders two theoretical models: the Voigt model for upper-bound modulus (axial loading) and the Reuss model for lower-bound modulus (transverse loading) (Erkkilä et al., 2016). To understand the composite failure in an isostrain condition, Voigt in his "iso-strain model" assumed that both the constituents behave elastically and deform in a brittle fashion i.e., the fibre and matrix experience the same axial strain (Harris, 1999; Hull and Clyne, 1996). The strain generated is the application of uniform stress being applied over a uniform cross-sectional area (Facca et al., 2006; Tucker III and Liang, 1999). It is important to note that the model considers only the cross-sectional areas of the fibre and matrix, not their shapes (Hyer and Waas, 2018). Young's modulus (in the fibre direction) for a continuous unidirectional fibre composite is given by the ROM equation (Hyer and Waas, 2018; Virk, 2010):

$$E_l = E_f V_f + E_m V_m \tag{4}$$

where $E_1$ is the extension modulus in the fibre direction, $E_f$, $E_m$ is the modulus of fibre and matrix respectively and Vm and Vf are the matrices and refibre volume fractions. Since $V_f + V_m = 1$, the above Eq. (1) can be written as (Hyer and Waas, 2018):

$$E_l = E_f V_f + E_m (1 - V_f) \tag{5}$$

This equation infers that $E_1$ (extension modulus of the composite is linearly related to the moduli of the constituents and volume fraction of the fibre and is often referred to as the rule of mixtures (Hyer and Waas, 2018). The modulus of the composite is considered to be upper-bound when the Poisson ratios of the two constituents are equal (Virk, 2010) i.e.

$$V_f = V_m \tag{6}$$

where $V_f$ and $V_m$ are the fibre and matrix axial Poisson's ratio.

It should be taken into account that elastic constraints caused due to differential lateral contractions are ignored (Harris, 1999).

Reuss on the other hand estimated the transverse modulus perpendicular to the fibres. He gave an "iso-stress model" where fibre and matrix experience the same stress and deform independently when loaded in the transverse direction (Harris, 1999; Hyer and Waas, 2018; Virk, 2010). This is called the inverse rule of mixtures and is given by the equation (Facca et al., 2006; Jones, 1999):

$$E_2 = \frac{E_f E_m}{E_f V_m + E_m V_f} \tag{7}$$

where $E_2$ is the extension modulus perpendicular to the fibre's direction.

This ROM equation gives the lower-bound modulus (Jones, 1999).

Reuss approach does not account for true stress and strain distribution in the composite. Due to relatively higher stress built in the matrix, polymeric matrices tend to creep under low mechanical loads (Clyne and Hull, 2019). Experimental measurements resulting from the Voigt estimate have a higher degree of precision as compared to the Reuss estimate (Erkkilä et al., 2016). Young's modulus of most of the composites (in the fibre direction) lies between the extreme values of either ROM or IROM equations (Facca et al., 2006).

### 8.2. Halpin-Tsai model

The most accurate method is the one designed by Halpin and Tsai (Clyne and Hull, 2019). This method provides refinement by taking into account short fibres, the role of aspect ratios to develop rational interpolation methods to be used between the upper and lower bounds of the composites (Chen, 2014; Virk, 2010). To predict the tensile modulus of short fibre reinforced composites, Halpin-Tsai gave the following equation (Clyne and Hull, 2019; Facca et al., 2006):

$$E_l = E_M \left( \frac{1 + \xi \eta V_F}{1 - \eta V_F} \right) \tag{8}$$

Parameter η in (Eq.5) can be written as:

$$\eta = \frac{(E_F/E_M) - 1}{(E_F/E_M) + \xi} \tag{9}$$

where ξ is the shape fitting parameter used to fit the experimental data. The value of ξ varies from 0 to ∞ (Tucker III and Liang, 1999). The noteworthiness of ξ is its quantification of geometry and packing arrangement of the reinforcing fibres. The way how the load is introduced in the composite can also be detailed by this method (Facca et al., 2006; Hyer and Waas, 2018; Jones, 1999; Tucker III and Liang, 1999). As found in the literature, various empirical equations for ξ are available which can be correlated to particle shape and predicted modulus of the composite. If the modulus is to be found in the fibre direction and the shape of the fibre is circular or rectangular, then ξ is given by the expression (Facca et al., 2006):

$$\xi = 2\left(\frac{L}{T}\right) \text{or} \xi = 2\left(\frac{L}{D}\right) \tag{10}$$

where L represents fibre's length in one direction, T or D gives fibre's thickness and diameter in three directions. According to Eq. (7), when $L \longrightarrow \infty, \xi \longrightarrow \infty$, then Halpin-Tsai follows ROM equation and when $L \longrightarrow 0$, $\xi \longrightarrow 0$ then it follows IROM equation.

### 8.3. Cox model

Cox (1952) was first to provide refinement by delineating the orientation parameter, reinforcing the effect of short fibres when predicting Young's modulus of discontinuous fibre composites. His work is commonly referred to as the shear-lag theory (Chen et al., 2014; Facca et al., 2006). The model is centred on the equilibrium of tensile and shear forces with an assumption of a single fibre embedded in a concentric cylinder of the matrix, also referred to as the "Concentric cylindrical model" (Harris, 1999). Cox's theoretical work assumes three stiffness measurement regimes: First, a perfect elastic response exists between matrix and fibre and stress is transferred without slip or yielding. Second, fibres are aligned and packed in an array. Third, interfacial shear stresses are responsible for load transfer from the polymer matrix to fibre with zero tensile stress at the extremities or ends of the fibres. Consequently, the ends of the fibre exhibit maximum shear stress whereas the central portion of the fibre bears the maximum tensile stress (Facca et al., 2006; Harris, 1999). As the ends constitute ineffective length, the reinforcement efficiency of the fibre is reduced which allows modification in the rule of mixture taking into consideration the fibre-end effect. The ROM is now modified by another efficiency factor, $\eta_l$ and the equation can be written as(Harris, 1999; Virk, 2010):

$$E = \eta_l E_f V_f + E_m V_m \tag{11}$$

where

$$\eta_l = 1 - \frac{\tanh(\beta l/2)}{\beta l/2} \tag{12}$$

β refers to the rate at which stress is being built at the fibre ends.

$$\beta = \sqrt{\frac{2\pi G_m}{E_f A_f \ln(R/r_0)}} \tag{13}$$

l is the length of the fibre, $G_m$ is matrix shear modulus, $A_f$ is the cross-sectional area of the fibre, R is the radius of matrix cylinder and $r_0$ is the fibre radius.

For fibres with circular cross-section and different packing arrangements the volume fraction of fibre is given by the following equations (Virk, 2010):

For Square packed array:

$$V_f = \frac{\pi r_0^2}{4R^2} \tag{14}$$

Hexagonal packed array:

$$V_f = \frac{2\pi r_0^2}{\sqrt{3}R^2} \tag{15}$$

### 8.4. Model for strength prediction

Prediction of strength is comparatively difficult than elastic properties due to the complex mechanism of damage accumulation. It is difficult to ascertain how damage reaches a critical level before final failure (Harris, 1999).

#### 8.4.1. Tensile strength of unidirectional composites (continuous fibre)

Based on the foregoing discussion, to determine the strength of a simple unidirectional composite it is assumed that all the reinforced fibres have the same strength and both the matrix and the fibre experience the same strain when subjected to loading. The longitudinal tensile strength depends on the fibre's failure strain (Harris, 1999; Virk, 2010). If the failure strain of the fibres is less than the matrix then the strength is determined by the Kelly-Tyson equation (Virk, 2010):

$$\sigma_c = \sigma_f V_f + \quad (\sigma_m)\varepsilon_f \left(1 - V_f\right) \tag{16}$$

where $\sigma_c$, $\sigma_f$ are the tensile strength of the composite and fibre respectively. $(\sigma_m)\varepsilon_f$ is the matrix stress at a strain equal to fibres failure strain. However, for low fibre volume fractions, the above equation is not true. The strength at low fibre volume fraction is given by:

$$\sigma_c < \sigma_m \left(1 - V_f\right) \tag{17}$$

Where $\sigma_m$ is the maximum tensile strength of the matrix.

The strength of the composite is given by the higher value calculated from Eqs. 16 and 17.

## 9. Conclusions

Bio-mechanics of the cell wall is still emerging research. Anisotropy of individual cell walls, fibrillar angle has been modelled and discussed by several researchers without much investigation of the role of young's modulus. An understanding of the interplay of variable young's modulus between the hydrogen-bonded microfibrils and load-bearing cross-links building shear forces is required. When subjected to large strains cell wall responds elastically. Tethering network associated with the breakage of hydrogen and covalent bonds whilst forming new microfibrils and newer cross-links needs to be understood well in the cell-wall growth mechanism. More experimental data on elastic response can help in better estimation of hyperelasticity and determining cell-wall geometry. Another aspect although undesirable that lowers the modulus is the fibre flaws mainly dislocations, crimps, micro compressions etc. either on the surface or in the internal structure which renders for more profound research in genetic modification of fibres for an improved fiber yield, reducing imperfections, thereby promoting agronomy.

From a chemical standpoint, further research in recycling heterogeneous plastics is required. Automated sensors and resin detectors have proved to be an effective means in the segregation of commingled plastics by employing single wavelength NIR (near infra-red spectrum) yet thermoset disposal faces recycling issues owing to its network structure. At present, recycling of cured epoxies with nitric acid has proved a promising method in this regard, by converting the wastes back to their original constituents, but more new chemical routes in recycling needs to be explored for better calculations in terms of economic value recovery. With the global target of an emerging need for sustainable development in improving composites performance, smart composites made from renewable resources are the future of the construction industry. The concept of stimulating rural economies in conjunction with advanced technology has led to the fabrication of smart thermochromic composites made of natural fibre. The intelligent composites are developed by the incorporation of colour-sensitive microcapsules with varied concentrations which change their colour when subjected to external stimuli. These innovative materials would help in building modern homes while using traditional ones for future structural designs. Empirical findings on durability studies are also needed for long-term relevancy in civil structures which could help in the commercialization of the end-products.

## CRediT authorship contribution statement

**Neha Uppal:** Conceptualization, Data Curation, Formal analysis, Methodology, Visualization, Writing – original draft preparation, **Asokan Pappu:** Methodology, Resources, Visualization, Supervision, Writing – review & editing, **Vijaykumar Sorna Gowri:** Resources, Visualization, **Vijay Kumar Thakur:** Supervision, Writing – review & editing.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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