Selection and Processing of Natural Fibers and Nanocellulose for Biocomposite Applications: A Brief Review

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In this study the recent developments in raw materials, manufacturing processes, and applications of natural fiber composites (NFCs) were reviewed. Natural fibers can represent a substitute for man-made fibers (including glass, aramid, and carbon) in a variety of biocomposite applications. Physical and chemical properties of the natural fibers are given and compared with the synthetic fibers. Advantages and disadvantages of NFCs in comparison with synthetic fibers such as glass and carbon fibers have been proposed. Criteria are described for the selection and processing of natural fibers for polymer composites used in different sectors such as automotive and building industries. The nanocellulose production methods, unique properties, and its recent industrial application in various sectors are given. This short review on NFCs considers their chemical, physical, and mechanical characteristics, as well as their various applications.

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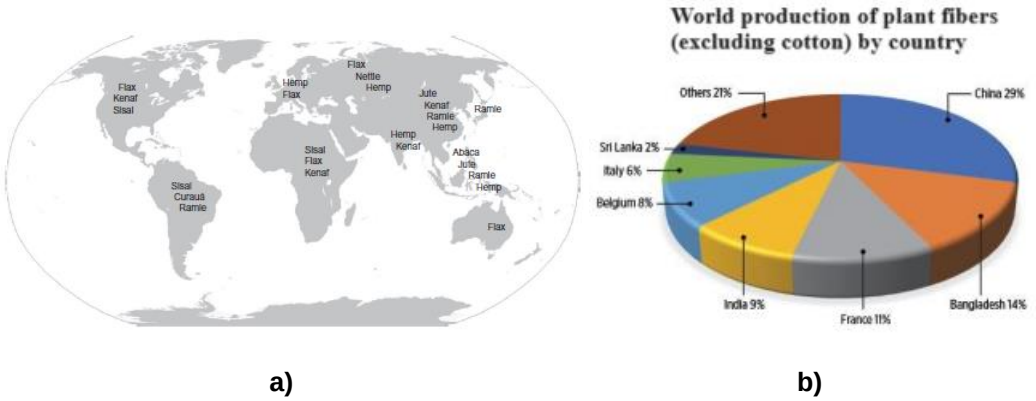
INTRODUCTION

In the last decades, natural fibers have gradually been proposed as a potential alternative to man-made fibers in polymer composites in a wide range of engineering applications. These initiatives are motivated by their low costs and acceptable mechanical properties, sustainable and abundant natures, and lessening of the carbon footprint (Wambua *et al.* 2003; Alhijazi *et al.* 2020). The goal of making natural fiber composites (NFCs) with limited environmental impact and tending to carbon dioxide neutrality has been gaining attention, as regards to both the use of thermosets and thermoplastics (Mohanty *et al.* 2002). From these early studies, the growth in the market of natural fibers aimed at the biocomposite sector has been steadily increasing throughout the world. It can

be easily suggested that in any region in the world, there will be one or more natural fibers having potential to be exploited in composites. According to some recent studies, the output of natural fiber composites manufacturers was expected to increase by 10% to 400% globally within the span of several years (Uddin 2013; Akter *et al.* 2022). Further developments specific to their application in composites depend on the tailoring of hybridization processes between carbon, glass, aramid, and natural fibers, and the use of different, more focused treatments, even moving away from the technologies that have become established in textile products (Khalid *et al.* 2021). Geographic distribution of the commercially important fibers for biocomposite manufacture is presented in Fig. 1. China is the leading country in the production of plant fibers followed by Bangladesh, France, India, and others. Flax, hemp, kenaf, sisal, jute, ramie, and abaca fibers are the most abundant fibers all over the world.



a) **b)**

Fig. 1. Geographic distribution of the commercial fibers for biocomposite manufacture. b) World production of plant fibers (excluding cotton) by country. Reproduced with permission from Gardner Business Media, Inc. Source: https://www.compositesworld.com/articles/natural-fiber-composites-whats-holding-them-back

Integrating, in a sound and durable way, tough and lightweight natural fibers into polymeric materials can produce composites having high specific strength, both in the case of thermoplastics and thermosets. Table 1 lists some natural fibers that are both widely used and commercially viable worldwide, and the total amount of fiber produced globally. The use of eco-friendly composites in the production of automobile parts is also becoming increasingly widespread (Dahlke *et al.* 1998).

Table 1. The World's Natural Fibers and Their Global Production

Source of Fiber	Production in the World (10^3 tons)
Cotton	18450
Oil palm	7227
Rice-stalk	7150
Bamboo	10000
Wheat barn	1500
Rice Husk	1200
Flax	830
Jute	2300
Kenaf	970
Okra	7896
Date palm	8384

***** Data from: (Faruk *et al.* 2012; Sathishkumar *et al.* 2022)

This review concentrates on the processing techniques for NFCs and on their mechanical properties and prospective applications. The materials considered are obtained with different fibers and using industrial (not purposely synthesized) matrices, both oil-based and bio-based. This brief review generally does not consider the question of natural fiber selection, which would involve their full characterization and considerations on their market/availability. However, some indications on the chemical composition of the relevant natural fibers (lignin, cellulose, hemicellulose content) are offered as a means to give some suggestions on their more or less hydrophilic/hydrophobic character. The classification of natural fibers, their physical and mechanical properties, NFC manufacturing methods, and recent developments are given. Moreover, the significance of nanocellulose, its classification, its unique properties, and some recent industrial applications of nanocellulose in biocomposites are reviewed.

Natural Fibers

Natural fibers can originate from plants, animals, or minerals (Célino *et al.* 2014). Notable among animal fibers are protein fibers, such as wool, silk, and even human hair (Suman *et al.* 2016). Figure 2 shows some examples of fiber classification. It is possible to classify some plant-based fibers according to their origin: stalk, grass, wood, leaf, fruit, bast, and seed. In plant fibers, the prevalently crystalline cellulose is held together in cell walls exploiting the random orientation of hemicellulose and lignin (Fig. 2).

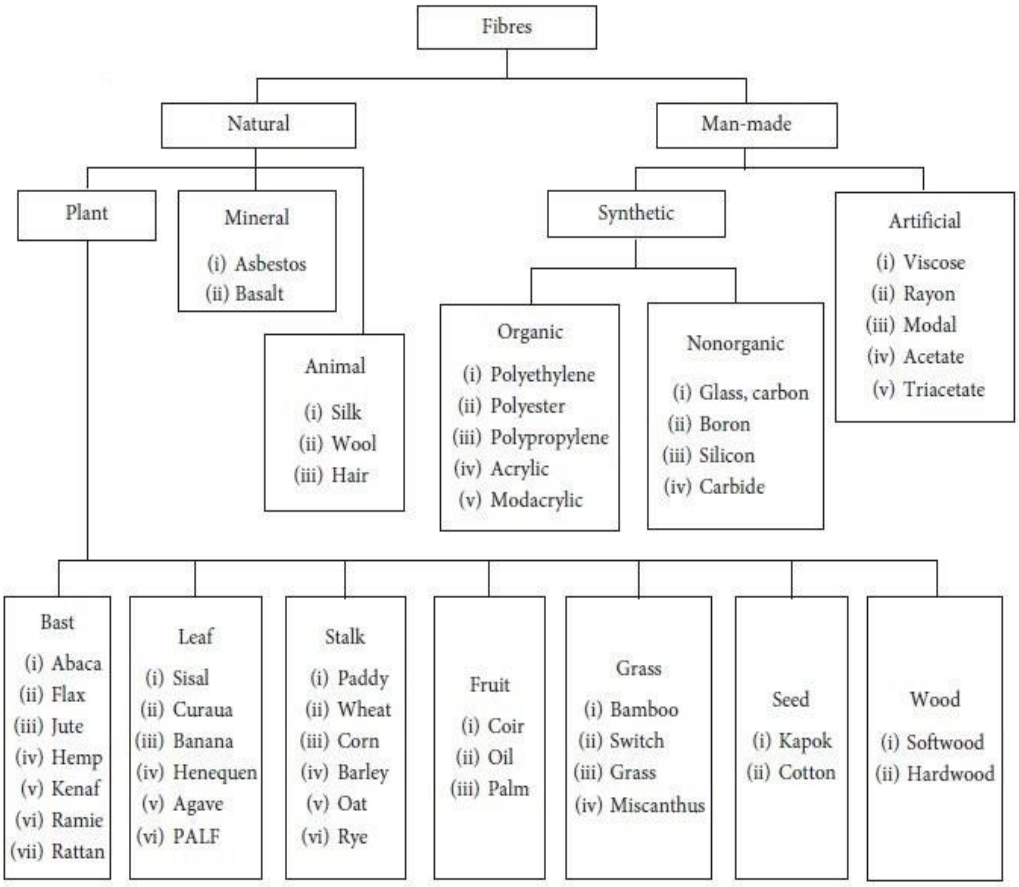


Fig. 2. Fiber Classification. (Source: Ahmad *et al*. 2019. *Advances in Civil Engineering* article ID: 5185806, Creative Commons BY 4.0)

In natural fibers, the structural assembly formed by cellulose often contains waxy substances. The natural matrix material can include hemicellulose or pectin, or mixtures of organic polymers, naturally synthesized from phenol-propane units, giving rise to lignin (Chen 2014). The coverage provided by amorphous lignin adds strength to the network of hemicellulose and cellulose, and is likely to create a protective layer within the fibrous structure (Mohanty *et al.* 2005). Hemicellulose is a cementing element in the cell wall, which forms a matrix embedding the microfibrils of crystalline cellulose (Fig. 3). The secondary walls of the crystalline cellulose microfibrils, constituted by three layers, of which the middle one is the thickest, as effectively described *e.g.*, by Rahman *et al*. (2015), determine the mechanical performance of the fiber. In the case the crystalline fraction of cellulose is sufficiently high, the fiber may offer excellent tensile strength, in a micrometric or even nanometric form (Ibrahim *et al.* 2010). This arrangement has been found to have a positive impact, *e.g.*, on the mechanical characteristics of polypropylene after surface modification (Joonobi *et al.* 2010), which appears to be accountable for the effective bonding and reduced proneness to degradation of natural fibers in composites than other layers, which is the approximately 80% of total thickness and the main load bearing layer (Kalia *et al.* 2014).

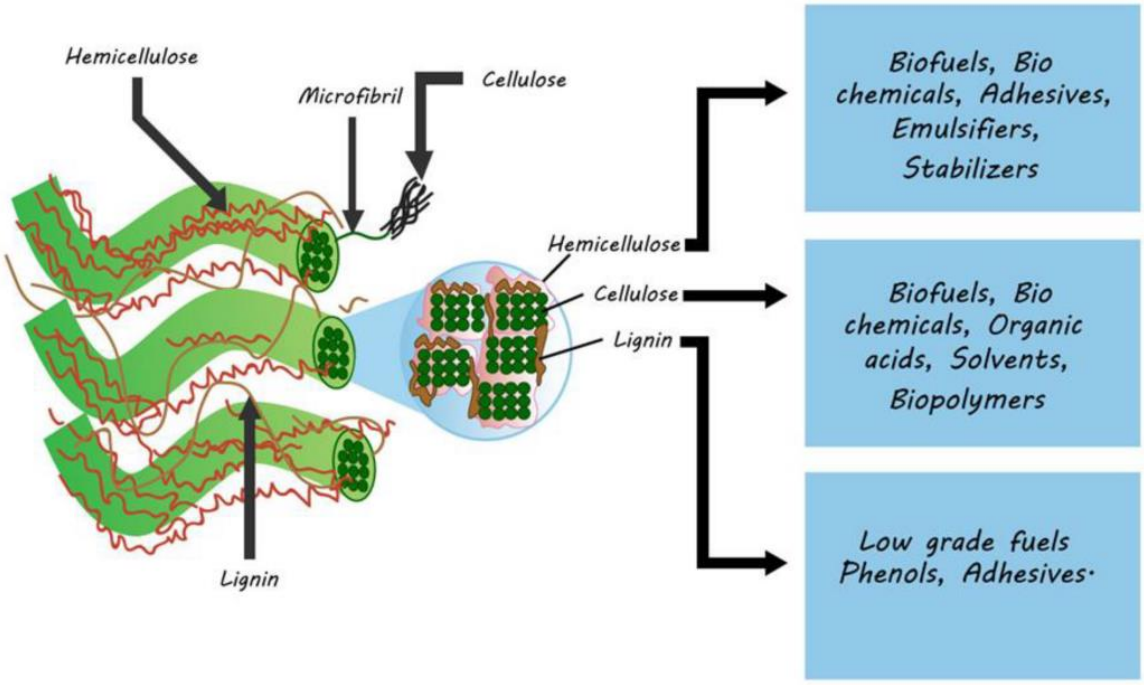


Fig. 3. Representation of structures of cellulose, lignin, and hemicellulose. (Source: Haq *et al*. 2021, *Catalyst* 11, article 309, Creative Commons BY 4.0)

The strength of wood fibre is determined by the amount of and orientation degree of microfibrils in these layers, as well as their polymerization degree and crystallization. Thus, the orientation degree of microfibrils at S2 layer determines the physical and mechanical properties of the wood fibre (Fig. 4). The modulus of the fibre improves with decreasing microfibril angle. In general, fibers with a high cellulose content and lower microfibril angle possess a good strength with a lower elongation at break.

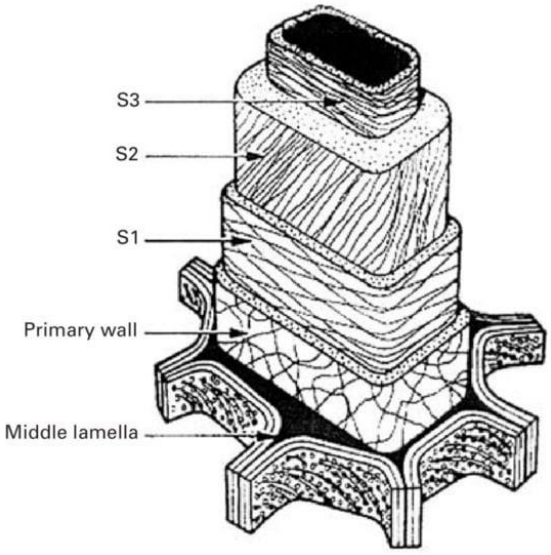


Fig. 4. Structure of wood cell of lignocellulosic plant fiber. (Source: Zhang *et al*. 2022, *Forests* 13, 439. Creative Commons BY 4.0)

Among the natural fibers, flax, jute, hemp, sisal, kenaf, oil and date palm, coir, banana, abaca, areca nut, and cotton are the most important plant fibers from a commercial and technological standpoint. In particular, three natural fibers used in composites from the early period of experimentation are hemp, jute, and flax, though over time the field of exploration has extended to a very large number of species. Jute is a fiber with important mechanical qualities for use in ropes and sacks, grown in Bangladesh, India, Thailand, and some Latin American countries (Mwaikambo 2006). Flax is Europe's most essential bast fiber, though some cultivars of it are also cropped in China (Wang *et al*. 2018). Some of the commercial natural fibers are presented in Figure 5.

Fig. 5. Some industrially used plant fibers. (Source: Nurazzi *et al*. 2021, *Polymers* 13, 646. Creative Commons BY 4.0)

A comparison of the natural fibers and man-made fibers in terms of cost and per weight ($/kg) is displayed Fig. 6. The man-made fibers, especially carbon fibers, are quite expensive in comparison to natural fibers. Furthermore, the carbon footprint of the natural fibers (4 GJ/ton) is greatly lower than synthetic fibers such as glass (30 GJ/ton) and carbon fibers (130 GJ/ton) due to the fact that lower energy requirement and lower CO_2 emission energy as compared to the synthetic fibers (Maiti *et al*. 2022).

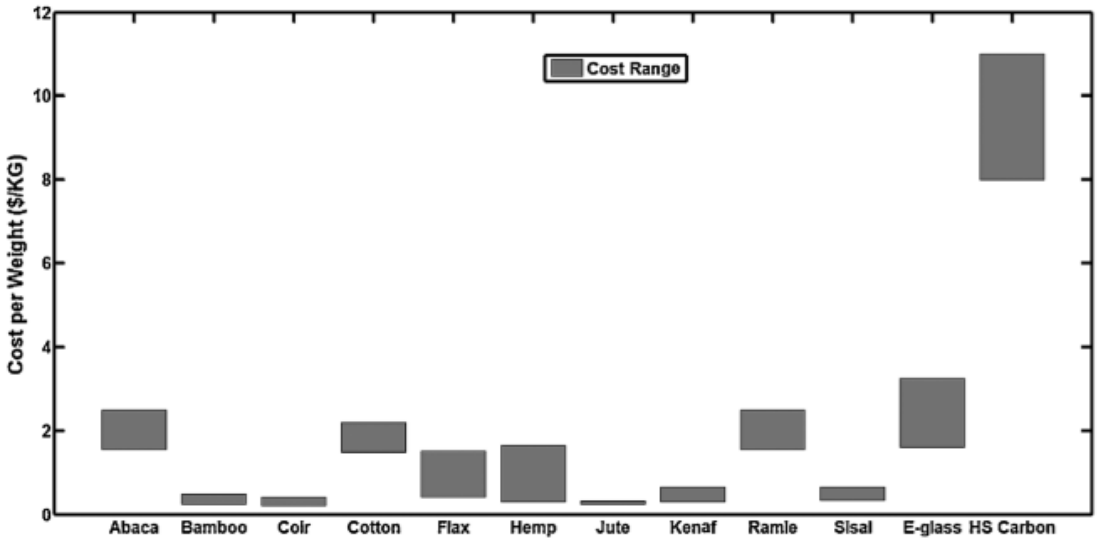


Fig. 6. Comparison of natural fibers and man-made fibers in terms of cost/weight. (Source: Ahmad *et al.* 2015, *Macromolecular Materials and Engineering* 300, 10-24. Creative Commons BY 4.0)

A complex mix of agronomical factors, resulting in chemical, physical, and mechanical characteristics of the fibers, is encountered when dealing with the performance of plant fibers. The species and cultivars from which fibers are extracted, their harvesting time, and their chemical composition and cellulose crystallinity are obviously determinant for the characteristics of the composite. Other inherent characteristics of the fibers are their microfibrillar angle, *i.e*., the angle within the dominant S2 sublayer by which the fibrils are wound together to form the technical (extractable) fiber, the dimension of lumens and porosity, and the cell length and diameter, which influence the relevant fiber's aspect ratio (Ku *et al.* 2011; Thakur and Thakur 2014). All the aforementioned characteristics are able to influence the fibers' mechanical characteristics. To offer some elements for comparison, the mechanical properties of a number of natural fibers are compared with E-glass fibers in Table 2. When passing to the composite, fiber extraction, matrix selection, interfacial strength, fiber dispersion, composite production process, orientation of fiber, and porosity are the primary elements impacting mechanical performance (Pickering *et al.* 2016). Also, the modulus of the natural fiber has been reported to diminish as its diameter increases (Van den Oever *et al.* 2000). This can be attributed to the possible separation of fibrils, normally referred to as "fibrillation" and to the subsequent higher percent of porosity within the fibers, which is typical of cellulose, especially when stiffened by applying chemical treatments (Sharma *et al.* 2015). Due to their chemical composition, natural fibers have however a most unfavorable property for their use, which is hygroscopicity. Absorbing moisture has negative consequences on their characteristics and, as a result, their performance over a long service term (Wang *et al.* 2006). According to Table 2,

among the commercial natural fibers, the flax fiber has the highest tensile strength, which makes it attractive for polymer composites used in automotive industry, followed by hemp, ramie, sisal, and jute fibers.

Table 2. Mechanical Characteristics of Some Fibers*

Fibers	Density (g/cm^3)	Tensile Strength (MPa)	Specific Tensile Strength $(MPa)/(g/cm^3)$	Stiffness (GPa)	Specific Stiffness $(GPa)/(g/cm^{-3})$
E-glass	2.5	2000 to 3000	800 to 1400	70	29
Wool	1.3	50 to 315	38 to 242	2.3 to 5	1.8 to 3.8
Chicken feather	0.9	100 to 203	112 to 226	3 to 10	3.3 to 11
Ramie	1.5	400 to 938	270 to 620	44 to 128	29 to 85
Flax	1.5	345 to 1830	230 to 1220	27 to 80	18 to 53
Jute	1.3 to 1.5	393 to 800	300 to 610	10 to 55	7.1 to 39
Hemp	1.5	550 to 1110	370 to 740	58 to 70	39 to 47
Cotton	1.5 to 1.6	287 to 800	190 to 530	5.5 to 13	3.7 to 8.4
Sisal	1.3 to 1.5	507 to 855	362 to 610	9.4 to 28	6.7 to 20
Silk	1.3	100 to 1500	100 to 1500	5 to 25	4 to 20

***** Adapted from Pickering *et al.* (2016).

Manufacturing Techniques of NFCs

Depending on the geometry of the component to be manufactured, fiber-reinforced plastics can be obtained in various ways, all of which are based on the polymerization concept. Table 3 provides a list of works on natural fiber composites obtained with different manufacturing methods and processes.

Table 3. Different Types of Manufacturing Methods and Processes of Composites

Method	Composite Produced	References
Hand layup	Polyester with sisal, jute, and glass fibers	(Ramesh *et al.* 2013)
	Epoxy with bi-directional jute fiber	(Mishra and Biswas 2013)
	Epoxy with banana fiber	(Ramesh *et al.* 2014)
	Epoxy with glass and sisal/jute fibers	(Ramesh *et al.* 2013)
	Polyester with *Calotropis Gigantea* fruit fiber	(Babu *et al.* 2014)
Spray layup	Rice straw waste in Kraft paper box with natural rubber binder	(Klinklow *et al.* 2013)
	Composites with nanosilica particles and various flax structures	(Ashworth *et al.* 2016; Siengchin and Dangtungee 2014)
	Epoxy with coconut sheath fiber	(Kumar *et al.* 2014)
	Epoxy with sisal and jute fibers	(Costa and d'Almeida 1999)
Filament winding	Cellulose and kraft paper into natural rubber (NR) or cellulose aceto-butyrate (CAB)	(Ly *et al.* 2008)
	Composites with ramie fiber yarn reinforcement	(Ma *et al.* 2016)

	Composites made from jute yarn and biopol	(Mohanty *et al.* 2000)
	Sisal fiber high density polyethylene pre-preg	(Dun *et al.* 2019)
	Filament-wound epoxy composites with natural fiber reinforcements	(Lehtiniemi *et al.* 2011)
Compression moulding	Reinforced polyethylene short natural fiber with cellulose/natural rubber composites	(Abdelmouleh *et al.* 2007)
	Jute fibers in polyester/epoxy matrices	(Gopinath *et al.* 2014)
	Composites reinforced with banana and sisal	(Ramesh *et al.* 2013)
	Natural fibers used to reinforce PLA composites	(Oksman *et al.* 2003)
	Polypropylene/sugarcane bagasse fibers composite	(Luz *et al.* 2007)
Injection winding	Woven sisal fibers with natural rubber modified epoxy matrix	(Rashnal *et al.* 2013; Srisuwan *et al.* 2014)
	Vetiver/polypropylene	(Ruksakulpiwat *et al.* 2007)
	Bamboo-glass fibers/polypropylene hybrids	(Thwe and Liao 2002)
	Polypropylene composites reinforced with sugarcane bagasse fibers	(Luz *et al.* 2007)
	Palm fibers reinforced polypropylene	(Goulart *et al.* 2011; Mishra and Biswas 2013)

Hand Layup

This method of moulding involves placing fiber reinforcements by hand and pouring polymer resin on top of them. A secondary layer of fiber reinforcements is placed on the top of the polymer matrix surface, where a slightly pressurized roller is moved on the reinforcement fibers to avoid air penetrating among the layers and remaining among them (Summerscales *et al.* 2013; Fiore *et al.* 2015). After that, the process is repeated for each polymeric matrix and fiber layer until all necessary layers are stacked, and cure can be allowed for the time necessary. This method is best suited for smaller batches of composites. The viscosity of a resin must be low enough for it to be worked manually due to high diluent/styrene levels; this necessary step decreases somewhat the mechanical characteristics though. When hand layup is used on one side only, this makes a smooth, high-quality finish. In addition, it has a greater degree of material design flexibility, though requiring a longer cycle for curing, up to 24 to 48 h (Mishra and Biswas 2013; Ramesh *et al.* 2014).

Spray Layup

Similar to hand layup, spray layup is a hand moulding technique that extends the hand layup method. This method has gained considerable attention also on natural fiber composites (Srinivas *et al.* 2017). In this procedure, spraying pressurized resin and reinforcement into a chopped fibers' geometry is accomplished by using a spray gun. Matrix material, as well as reinforcement, could be sprayed at the same time or at different moments in time, in a sequential manner. A roller is passed over the sprayed surface with a small amount of strain to remove any air that may have been trapped in the layups. Immediately after spraying up to the desired thickness, the material is allowed to cure at room temperature for some time before being removed from the mould (Ku *et al.* 2011). Low viscosity matrices are preferably used with this method, which might affect their mechanical properties, so that this manufacturing process is best suited for low volume production to offer a low cost composite with good surface finish on only one side (Omrani *et al.* 2016).

Filament Winding

Filament winding is primarily used to generate closed and open-end structures, depending on the application. In this system, the composite is obtained by winding tended filaments on a mandrel rotating around a spindle, while a shipping eye is carried in axis with the rotating mandrel, laying down fibers as per request. Convex shapes can be created using this method. Low viscosity resins are typically preferred (Belaadi *et al.* 2014). In natural fiber composites, tailored types of epoxies have been used (Lehtiniemi *et al.* 2011).

Compression Molding

When it comes to thermoplastic matrices, compression molding is most commonly used with unfastened chopped fiber, short or long fiber mats, which are either highly irregular or aligned, though it might be also applied in the case of thermosetting matrices. Before applying stress and heat, the fibers are typically stacked variously with thermoplastic resin sheets to create a more substantial structure. The molding material is usually preheated, then placed in an open hollow space, which is more substantially heated (Rong *et al.* 2001; de Andrade Silva *et al.* 2008) before being pressed into the mold. An example of hot-press molding process for automotive doors is presented in Fig. 7.

Fig. 7. Hot press molding of non-woven hemp-pp composites used in manufacutre of the door of the car. (Source: https://www.globalhemp.com/blog/automotive-composites/). Used with permission from Global Hemp Company

Injection Molding

In injection molding, warming bands, and the frictional motion of an oscillating screw barrel combine to melt the material in a heated barrel. Plastic is then introduced into the mold cavity through an injection nozzle, where it hardens during cooling down phase to conform to the geometry of the cavity's internal structure. When the parts are solidified, the plate is opened, and the component is thrown out using executor pins. The mildew tool is installed on a movable container. This procedure results in a smooth surface finish and is also appropriate for higher volume applications. The tensile strength of this process is slower than that of most thermoset systems (Zou *et al.* 2011).

Fig. 8. Injection molded biocomposite. (Source: https://www.arburg.com/en/solutions/by-technology/special-injection-moulding-processes/). Used with permission from Arburg Company Press

Density

The maximum theoretical density for natural lignocellulosic fiber is related to that of pure microcrystalline cellulose, which is just below 1.6 g/cm^3 (Sun 2005). Of course, natural fibers always have a lower density than that because they contain not only cellulose, but they have significant levels of porosity. Even lower is the density of natural fiber composites, where the matrices are normally lighter than the fibers. To provide examples, Table 4 shows the density of natural fiber enhanced polymer composites (NFRPs).

Table 4. Different Natural Fibers and their Densities

Fiber	Density (g/cm^3)	Reference
Abaca	1.5	(Dittenber and GangaRao 2012)
Alfa	0.89	(Dittenber and GangaRao 2012)
Bamboo	0.6 to 1.1	(Jayamani *et al.* 2014)
Banana	1 to 1.5	(Naidu *et al.* 2013)
Coir	1.25	(Wambua *et al.* 2003)
Cotton	1.5 to 1.6	(Naidu *et al.* 2013)
Flax	1.4	(Wambua *et al.* 2003)
Hemp	1.48	(Wambua *et al.* 2003)
Henequen	1.2	(Dittenber and GangaRao 2012)
Jute	1.3 to 1.49	(Jayamani *et al.* 2014)
Kenaf	1.45	(Omrani *et al.* 2016)
PALF	1.53	(Devi *et al.* 1997)
Palm	1.03	(Suman *et al.* 2016)
Ramie	1.5	(Wambua *et al.* 2003)
Sisal	1.33	(Wambua *et al.* 2003)
Wool	1.3	(Naidu *et al.* 2013)

Chemical Characteristics of Natural Fibers

The chemical characteristics of natural fibers depend on the respective amounts of cellulose, hemicellulose, and lignin. Beyond that, the amount of crystallinity also affects the structural potential of the fibers. Pectin and waxes generally are present often in negligible quantities in ordinary fiber, whereas remaining ash does not normally exceed a few percent. Fibers defined as lignocellulosic and used as such in natural fiber composites contain less than 50% lignin, in the latter case being defined as "wood". A selected number of studies that report the values of cellulose, hemicellulose, and lignin are reported in Tables 5, 6 and 7, respectively.

Table 5. Cellulose Content of Some Natural Fibers

Fiber Name	Cellulose (%wt)	Reference
Abaca	56 to 66	(Dittenber and GangaRao 2012)
Alfa	45	(Dittenber and GangaRao 2012)
Bamboo	30 to 65	(Jayamani *et al.* 2014)
Banana	62 to 64	(Badrinath and Senthilvelan 2014)
Cotton	82 to 90	(Dittenber and GangaRao 2012)
Flax	71	(Li and Tabil 2007)
Hemp	57 to 75	(Li and Tabil 2007)
Henequen	60 to 77.6	(Dittenber and GangaRao 2012)
Jute	59 to 71.5	(Jayamani *et al.* 2014)
Kenaf	45 to 57	(Omrani *et al.* 2016)
Ramie	68 to 91	(Li and Tabil 2007)
Sisal	78	(Nordin *et al.* 2013)

Table 6. Hemicellulose Content of Some Natural Fibers

Fiber Name	Hemicellulose (%wt)	Reference
Abaca	20 to 25	(Dittenber and GangaRao 2012)
Alfa	38.5	(Dittenber and GangaRao 2012)
Bamboo	30	(Jayamani *et al.* 2014)
Banana	19	(Badrinath and Senthilvelan 2014)
Cotton	5.7	(Dittenber and GangaRao 2012)
Flax	18.6 to 21.6	(Li and Tabil 2007)
Hemp	14 to 22.4	(Li and Tabil 2007)
Henequen	4 to 28	(Dittenber and GangaRao 2012)
Jute	13.6 to 20.4	(Jayamani *et al.* 2014)
Kenaf	8 to 13	(Omrani *et al.* 2016)
Ramie	5 to 16.7	(Li and Tabil 2007)
Sisal	25.7	(Nordin *et al.* 2013)

Table 7. Lignin Content of Some Natural Fibers

Fiber	Lignin (%wt)	Reference
Abaca	7 to 13	(Dittenber and GangaRao 2012)
Alfa	14.9	(Dittenber and GangaRao 2012)
Bamboo	5 to 31	(Jayamani *et al.* 2014)
Banana	5	(Badrinath and Senthilvelan 2014)
Cotton	<2	(Dittenber and GangaRao 2012)
Flax	2.2	(Li and Tabil 2007)
Hemp	3.7 to 13	(Li and Tabil 2007)
Henequen	8 to 13.1	(Dittenber and GangaRao 2012)
Jute	11.8 to 13	(Jayamani *et al.* 2014)
Kenaf	21.5	(Omrani *et al.* 2016)
Ramie	0.6 to 0.7	(Li and Tabil 2007)
Sisal	12.1	(Nordin *et al.* 2013)

Mechanical Characteristics of NFCs

The mechanical properties of a substance include a response to a connected load, such as elastic deformation. The mechanical characteristics determine its range of usefulness and contribute to the length of time it can be used in day-to-day administration. Mechanical characteristics are also used to characterize and distinguish between different types of materials and alloys. A summary of the mechanical characteristics of fiber-reinforced composites is shown in Table 8.

Table 8. Mechanical Characteristics of Different NFCs

Fiber	Matrix	Fiber wt%	Tensile Strength (MPa)	Flexural Strength (MPa)	Impact Strength	Reference
Abaca	PLA	10	74	124	5.3 kJ/m2	(Bledzki *et al.* 2009)
Bamboo	Epoxy	40	23	58	-	(Sathish *et al.* 2017)
Bidirectional jute	Epoxy	48	110	55	4.9J	(Mishra and Biswas 2013)
Harakeke	Epoxy	45	136	155	-	(Le 2016)
Abaca/glass fibre	Epoxy		44.5	12.5	16J	(Ramnath *et al.* 2013)
Kenaf	PLA	80	223	254	-	(Ochi 2008)
Coconut spathe	Epoxy	10	20	30	4J	(Vijayakumar *et al.* 2014)
Sisal unidirectional at 90°	Epoxy	12	56.5	371.33	1.3kJ/m2	(Badrinath and Senthilvelan 2014)
Sisal	Epoxy	15	66.74	204.3	-	(Gupta and Singh 2018)
Sisal mat	Epoxy	30	89.3	152.12	-	(Gupta and Singh 2018)

Industrial Applications of Natural Fibers in NFCs

The use of natural fibers has become increasingly common in a large spectrum of engineering applications, where they can constitute an alternative to fiberglass, though less so to carbon fiber composites. Assets include limited specific weight, low cost, end-of-life biodegradability, and renewability. NFCs have increasing success in the automotive industry, particularly for interior applications such as seatbacks, door panels, sun visors, exterior underfloor paneling, and boot linens (Botelho *et al.* 2006; Pickering *et al.* 2016). Natural fibers were used by the BMW Group in the amount of approximately 10,000 tons already in 2004. NFCs also are being used by the aerospace industry in the paneling of the aircraft's interior (Faruk *et al.* 2012). An increasing number of studies have indicated that NFCs can be transformed into load-bearing structural components for infrastructure and structural applications (Ticoalu *et al.* 2010). Ongoing research aims to advance the use of bamboo fiber-reinforced structural pavements, and ongoing research seeks to promote coir and sisal fiber composites to replace asbestos in roof components (Mohammed *et al.* 2015). Sports, even water sports, are another industry widely involved in the application of natural fibers. This is justified by the higher performance achievable with their use, is generally sport, and even water sports. Some of the advanced manufacturing utilizations of natural fibers in NFCs are listed in Table 9.

Table 9. Current Applications of Some Natural Fibers in NFCs*

Fiber	Application
Coir	Mirror casings, brushes and brooms, flush door shutters, seat cushions, seat cushions, filling material for upholstery, ropes and yarns for nets, roofing sheets
Cotton	Textile and yarn industries, furniture industries
Flax	Snowboarding, tennis rackets, window frames, decking, fencing, bicycle frames
Hemp	Furniture, electrical, cordage, paper, and textile industry
Jute	Packing materials, door frames and shutters, geotextiles, building panels, and chipboards
Kenaf	Cases for mobile devices, insulations, bags, packing materials, and animal beddings
Ramie	Industrial sewing threads, household furnishings, packaging material, fishing net, clothing, and paper manufacturing
Sisal	Door, panels, roofing sheets, and other items in construction-related industries

***** (Mohammed *et al.* 2015)

Nanocellulose and its Recent Applications

Nanocellulose has gained significant attention in the composite industry due to its unique properties such as high specific surface area, thermal stability, and good strength properties. According to the morphological structure, cellulose can be classified as (a) cellulose nanocrystals, (b) cellulose nanofibrils (or nanofibrillated cellulose), and (c), bacterial cellulose. The nanocrystals can be rod-like or spherical. Nanofibrils are similar to spaghetti because they are longer (a micrometer or more), flexible, and can tangle easily. The tensile strength and modulus of the nanocrystals are significantly higher than nanofibrils because the nanocrystals do not contain the amorphous regions of the cellulose while nanofibrils contain both amorphous and crystalline regions. As shown in Fig. 9, the cellulose microfibril can be separated from wood cell walls by breaking down multi-layered structure and hydrogen bonding by strong shearing force during mechanical fibrillation process using disk mill, cryocrushing, or using a high pressure homogenizer (Bhatnagar and Sain 2005; Kalia *et al.* 2014; Crotogino 2012).

Table 10. The Family of Nanocellulose Materials (Klemm *et al.* 2011)

Type of Nano Cellulose	Synonyms	Average Size
Nanofibrillated Cellulose (NFC)	Cellulose nanofibril (CNF),	diameter: 5 to 60 nm length: several micrometers
Nanocrystalline Cellulose (NCC)	Cellulose nanocrystals, crystallites, whiskers, rodlike cellulose microcrystals	diameter: 5 to 70 nm; length: 100 to 250 nm (from plant celluloses)
Bacterial nanocellulose (BNC)	Bacterial cellulose, microbial cellulose, biocellulose	diameter: 20 to 100 nm; different types of nanofiber networks

Fig. 9. Masuko Sangyo Super Masscolloider grinder for nanocellulose production. The photos were taken by N. Ayrilmis.

The diameter of NFC has a range of 10 to 40 nm, depending on different fibrillation equipment and number of repeated cycles; the aspect ratios range from 100 to 150 (Siro and Plackett 2010). Cellulose nanocrystals (CNCs) mainly are isolated using acid hydrolysis (Fig. 10) (Siqueira *et al.* 2010). The amorphous region of cellulose is degraded preferentially because of high resistance of crystalline domain (Kalia *et al.* 2014). A typical CNC particle has a width of 4 to 6 nm and a length of 100 to 250 nm with high degree of crystallinity (Habibi *et al.* 2010).

Nanocellulose particles, in general, have great potential as reinforcing filler of nanocomposites because of high tensile strength properties and high specific surface area. Bhatnagar and Sain (2005) reported that the tensile strength of polyvinyl alcohol-based nanocomposite were increased from 60 to 118 MPa by adding nanocellulose to the neat polymer.

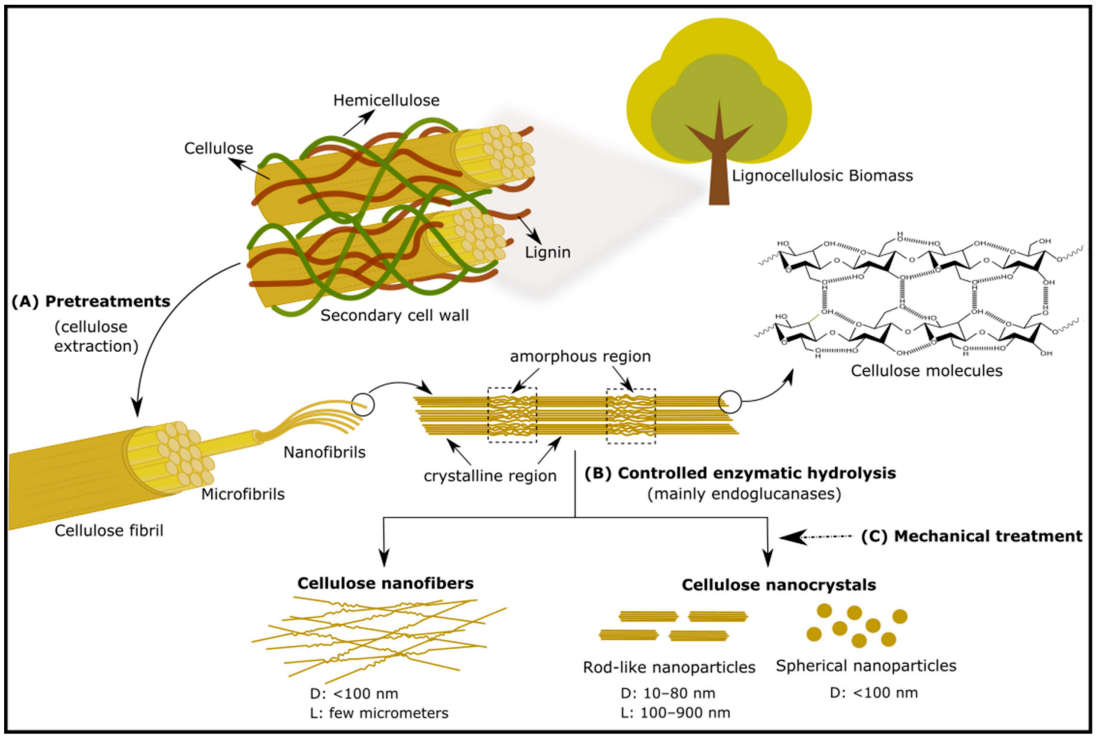


Fig. 10. Schematic view from wood to cellulose. (Source: Michelin *et al*. 2020, *Molecules* 25, 3411. Creative Commons BY 4.0)

The crystal structure of nanocellulose consists of packed needle-like crystal arrays. These crystal structures are incredibly strong, with strength values almost eight times higher than stainless steel (Anonymous 2017).

Table 11. Comparison of Nanocrystalline Cellulose (NCC) and Man-Made Fibers in Terms of Tensile Properties and Production Cost (Nelson *et al.* 2016)

Material	Density g/cm^3)	Tensile Strength (MPa)	Tensile Modulus (GPa)	Cost ($/kg)
High Strength steel	7.9	600	210	~1
Aluminum 6061-TL	2.7	275	70	~2
E-glass	2.5	3500	80	~2
Carbon fiber	1.8	4000	230	~20
Nanocrystalline cellulose	1.5	7500	135	4 to 10

When properly aligned, nanocellulose offers an alternative to stronger applications, such as replacing DuPont's Kevlar®. Its use in bulletproof vests and ballistic glass is being investigated by the Ministry of Defense (Anonymous 2014). With these optical and other properties, it is a unique additive for inks, paints or varnishes that increases color brightness while providing durability. The crystal structure of nanocellulose consists of packed needle-like crystal arrays. These crystal structures are incredibly strong, with strength values almost eight times higher than stainless steel (Fig. 8). It follows that the strength of a composite including such materials can be highly dependent on the bonding between the cellulose and the matrix.

Generally, the separator parts inside batteries are made of thick and hard material that cannot be used in bendable applications. By combining flexible and thin nanocellulose

with graphene material, a flexible battery, one of the biggest dreams of the electricity industry, can be produced (Anonymous 2017) (Fig. 10). NFC can be used in a wide variety of applications such as medical, food products, resins, paints, and separator membranes. For example, Sehaqui *et al.* (2010) produced a rapid preparation method for optically transparent cellulose nanopapers (CNP) using a semiautomatic sheet former. These environmentally friendly and low-cost cellulose CNP separators were mainly developed for the utilization in lithium-ion batteries, which considerable enhanced the ionic conductivity, electrolyte wettability, and thermal shrinkage (Chun *et al.* 2012; Willgert *et al.* 2014; Martinez *et al.* 2023). Some technology companies such as Pioneer Electronics changed the transparent nanocelluose films with the glass in flexible displays due to good durability and optical properties of the nanocellulose films. In another study, Pan *et al.* (2019) produced a thin and flexible (2.5 µm) cellulose nanofiber layer having the excellent properties such as thermally stable and hydrophilic properties using paper production method. Both sides of a plasma-treated polyethylene separator were faced with the nanocellulose film. Some of the applications of nanocellulose in different products are displayed in Fig. 10.

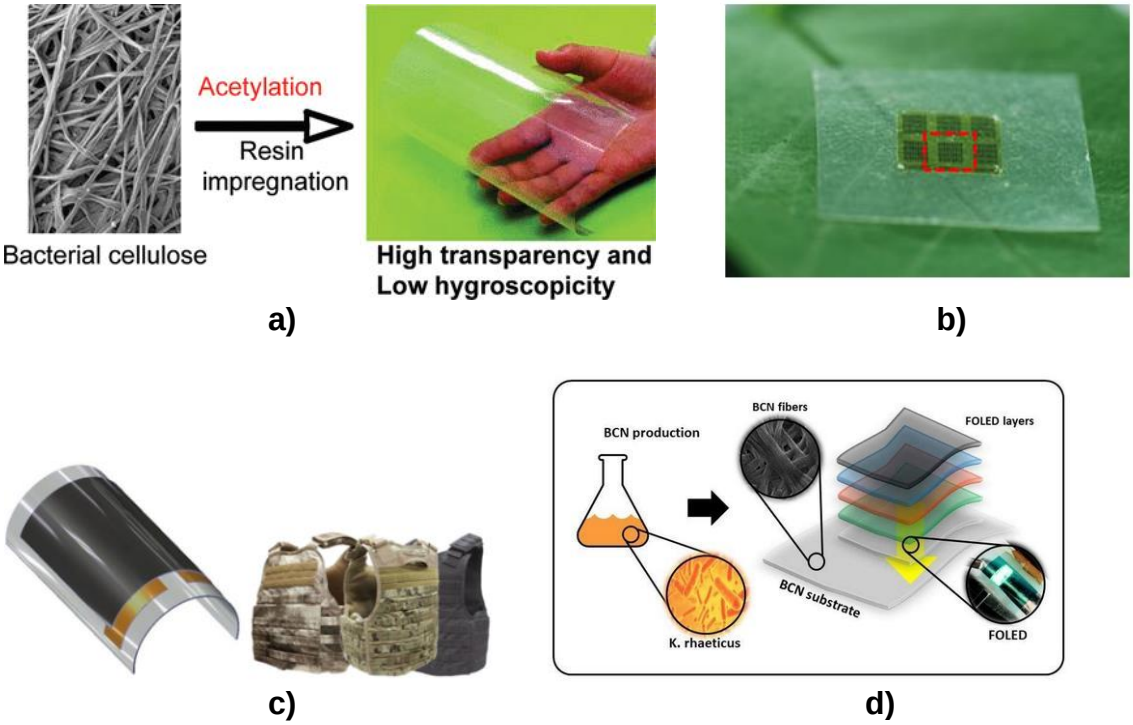


Fig. 11. a) Transparent nanocellulose film. Reproduced with permission from Ifuku *et al*. (2007), [*Biomacromolecules*]; published by ACS publisher. b) Photograph of an array of heterojunction bipolar transistors (HBTs) on a CNF substrate put on a tree leaf. Reproduced with permission from Jung *et al*. (2015), *Nature Communications* 6, 7170. Creative Commons BY 4.0) c) Separator membranes derived from nanocellulose paper (CNP separators) for use in lithium-ion batteries and body armor from the NC; (Source: https://medium.com/@nanografi/incredible-uses-of-nanocellulose-a0faa08bb9cf. Used with permission from Medium internet blog); d) Bacterial nanocellulose as substrate for conformable and flexible organic light-emitting diodes (OLED). (Source: Faraco *et al*. 2023, *Polymers* 15, 479. Creative Commons BY 4.0)

A research team from Purdue developed a new composite film based on cellulose nanocrystals as advanced barrier coatings for food packaging (Youngblood 2018). This barrier film was described as low-cost, sustainable, nontoxic, and having high specific strength and transparent materials likely suitable for food packaging applications (Fig. 12).

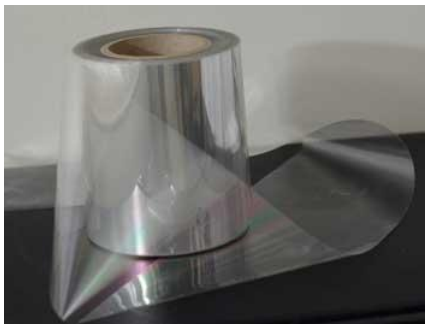


Fig. 12. Nanocellulose-Coated PET Film. (Source: https://www.purdue.edu/newsroom/releases/2018/Q2/changing-the-grocery-game-manufacturing-process-provides-low-cost,-sustainable-option-for-food-packaging.html). Used with permission from Purdue University Press.

VTT Technical Research Centre of Finland developed dental crowns produced from nanocrystals and protein for dental implants, which resembled human teeth (Mohammadi 2021). The new dental crowns is stronger and lighter than the traditional ceramics used dental-crowns, as shown in Fig. 13.

Fig. 13. Nanocellulose for dental applications. (Creator: Mohammadi, 2021. Source: https://forest.fi/products-services/nanocellulose-for-dental-implants/#3642812e). Used with permission from Finnish Forest Association

CONCLUSIONS

Natural plant-based fibers, which are also called cellulosic fibers, can be used in place of the reinforcing materials in established composite materials in many fields. When natural fiber-based composites and other reinforced composites were compared, it was found that natural fibers were better for industrial use in several respects. Thus, material scientists and engineers have recently researched ways to combine the natural fiber reinforced polymer composites with the technologies for personal protective armor. All natural fibers have chemical properties that are different from one another. Hemicellulose

is more abundant in bamboo and alfa, while cellulose is more common in cotton. Lignin is more often found in kenaf and bamboo. Density generally is lower in natural fibers than synthetic ones. Because of this, natural fibers are used or can be considered in many fields and are often seen as an alternative to synthetic fibers. Also, natural fibers can be used in different engineering applications and in the construction industries. Utilization of such resources can help rural areas to grow economically. Due to the unique properties of nanocellulose, its industrial utilization considerably increased in polymer composites in the last two decades. Nanocellulose improves physical, mechanical, and thermal properties of polymer composites, such as dental crowns, high performance films, transparent films, separators for lithium-ion batteries, and body armor for military purposes. It is believed that nanocellulose will be one of the strategic materials for high value-added products in the near future. The study results show that the properties of composites made with natural fibers often can be improved.

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Data Availability Statement

Data available on request from the authors.

Declaration of Conflicting Interests

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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