Cellulose (2021) 28:4511–4543
https://doi.org/10.1007/s10570-021-03771-4

REVIEW PAPER

Towards a cellulose-based society: opportunities and challenges

Jinwu Wang · Lu Wang · Douglas J. Gardner · Stephen M. Shaler · Zhiyong Cai

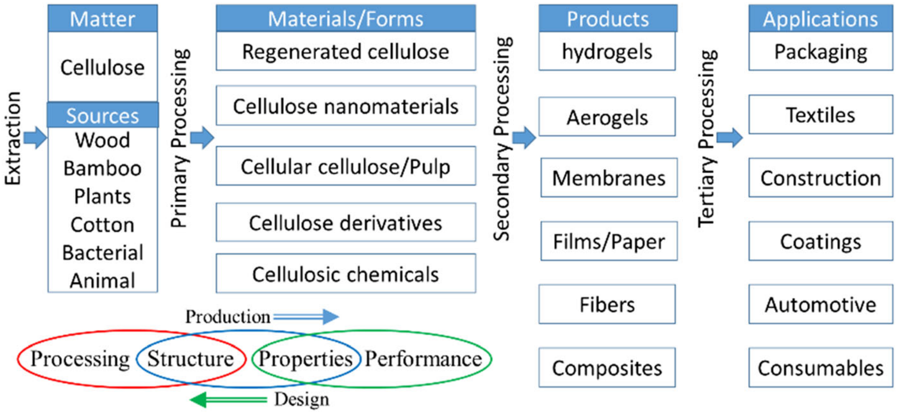
Received: 29 August 2020 / Accepted: 11 February 2021 / Published online: 27 March 2021

Abstract The current materials predominately come from fossil feedstocks and minerals. The pressures from climate change and plastic pollution challenge us to develop a bioeconomy, replacing petroleum-based products with bio-based and biodegradable products. Cellulose emerges as a versatile biopolymer to make hydrogels for absorbents, aerogels for insulation, membranes for filters, films for packaging, and fibers for textiles and reinforcements. Wood-based cellulose is increasingly perceived by relevant stakeholders to be renewable, biodegradable, and sustainable. Can the properties of cellulose-based materials compete with conventional synthetic materials? Knowledge and discoveries concerning cellulose properties and applications are scattered throughout the scientific literature base. This paper surveys the mechanical properties of cellulose-based materials in the literature using tensile properties as indicators and visualizes the data compared with other competitive materials. The goal is to provide insights into the potential and challenges of using cellulose-based products to replace synthetic materials for a sustainable society.

J. Wang · Z. Cai (✉)
Forest Products Laboratory, Madison,
WI 53726, USA
e-mail: zhiyong.cai@usda.gov

J. Wang · L. Wang · D. J. Gardner · S. M. Shaler
Advanced Structures and Composites Center, School of
Forest Resources, University of Maine, Orono,
ME 04469, USA

Graphic abstract



Keywords Materials use · Cellulose nanomaterials · Regenerated cellulose · Fiber · Composites

Current patterns of materials use relevant to cellulose

Our needs for clothing, food, shelter, and mobility are provided by natural resources including biomass, fossil feedstocks, metallic and non-metallic minerals. Humanity consumes enormous quantities of extracted materials from the surface and subsurface of the earth. About 89,000 million tonnes (Mt) of raw materials were used in 2017 (Fig. 1a) (OCDE 2019). The majority of the raw materials are non-metallic minerals used for construction, notably sand, gravel, and limestone. Non-metallic minerals are often recovered only in degraded form, e.g. usable only for downcycling. For example, concrete waste is used as a road filler, and glass into glass wool for insulation. Global oil and gas production reached 4016 Mt and 2272 Mt respectively (Fig. 1b). Most fossil feedstocks were expended as fuels through combustion for transportation and heating. A small portion of them was used to make plastics (348 Mt in 2017). Packaging represents the largest end-use market of plastics. About 39.9 percent of the plastics demand in Europe was packaging in 2018 (Plastics Europe 2019); plastic packaging took up 41 percent of total plastics in the 2017 U.S. municipal solid waste (EPA 2019). Globally, plastic packaging represents about 26 percent of the total volume of plastics used (Neufeld et al. 2016), i.e. that was about 90 Mt in 2017. Most other plastics were used for building, construction, and transport. Besides, the world produced 64 Mt synthetic fibers for textiles in 2017 (Textile Exchange 2020). The combined global production of plastic materials (plastics and synthetic plastic fibers) was around 412 Mt, which is in agreement with the trend from (Geyer et al. 2017). Plastics are valuable resources that bring numerous benefits to society. However, only 14 percent of the plastic packaging was collected for recycling and 32 percent of it was estimated to leak into environments. It was estimated at least 8 Mt of plastics leak into the ocean each year (Neufeld et al. 2016). Washing clothes made from plastic fibers has also been identified as a potentially important source of microplastics in water (Napper and Thompson 2016). The world harvested 20,000 Mt biomass in 2017, representing 25 percent of the total raw materials. Most biomass was consumed as food and feed, then being discharged into environments. Among the biomass production, 2,492 Mt were wood taking up 11 percent of the biomass (Fig. 1c). Most of the wood was consumed as fuel including 52 Mt charcoal and 37 Mt wood pellets. The world produced 185 Mt virgin wood pulp in 2017 (Fig. 1d), including 8.3 Mt dissolving pulp which was used to make manmade cellulosic films and fibers (regenerated cellulose and cellulosic derivatives). The world produced 422 Mt cellulosic pulp (wood pulp, plant pulp, and recovered paper) for making 417 Mt paper and paperboard, which was

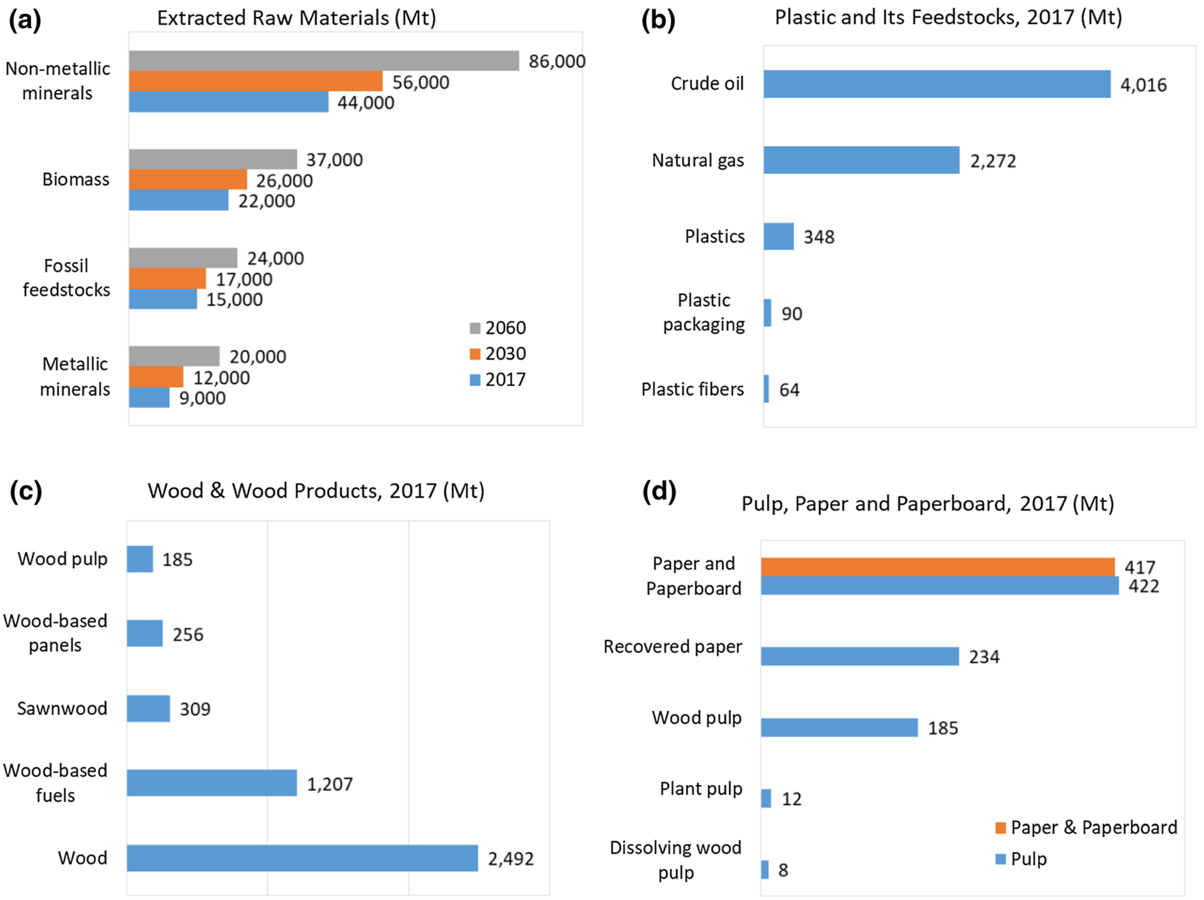


Fig. 1 Actual and projected global material production: **a** extracted raw materials (OCDE 2019), **b** oil and gas feedstocks and plastics (Neufeld et al. 2016; OCDE 2019; Plastics Europe. 2019; Textile Exchange 2020), **c** Wood and wood products (FAO 2017). The weights were estimated by assuming wood specific gravity at 0.62, and **d** cellulosic pulp, paper, and paperboard (FAO 2017)

slightly more than the production of the plastics and plastic fibers.

Textiles are everyday necessities and an important aspect of self-expression for many people. The global fiber production for textiles has increased by 250 percent from 32 Mt in 1975 to 111 Mt in 2019. In the same period, however, plastic textile fibers have increased 726 percent, far higher than the 105 percent increase of natural textile fibers including cotton, plant fibers, wool, down, and silk as well as semi-synthetic manmade cellulosic fibers (Textile Exchange 2020). Figure 2a shows the change of market shares by fiber material since 1975. The plastic textile fiber share increased from 26 percent in 1975 to 63 percent in 2019 and only less than 1 percent of which was bio-based. The cotton share decreased from 35 to 23 percent. The average share of cellulosic fibers was around 6.3 percent in the period with a trend of decreasing to the year 2005 and then slightly increased in recent years. But, the volume of global cellulosic fibers had more than doubled from around 3 Mt in 1990 to approximately 6.5 Mt in 2017 and is expected to further grow in the coming years (Textile Exchange 2020).

The volume of municipal solid waste (MSW) reflects the consumption of consumer goods. The USA had 171 Mt of total materials in products in MSW in 2017, among which 35 Mt was plastics waste, 73 Mt packaging waste, 13 Mt plastic packaging waste, and 17 Mt textile waste (EPA 2020). Figure 2b shows the weight percentage of material type in total materials in products. Cellulose-based materials including wood, paper and paperboard, and cellulosic fibers took up near the half volume. The material

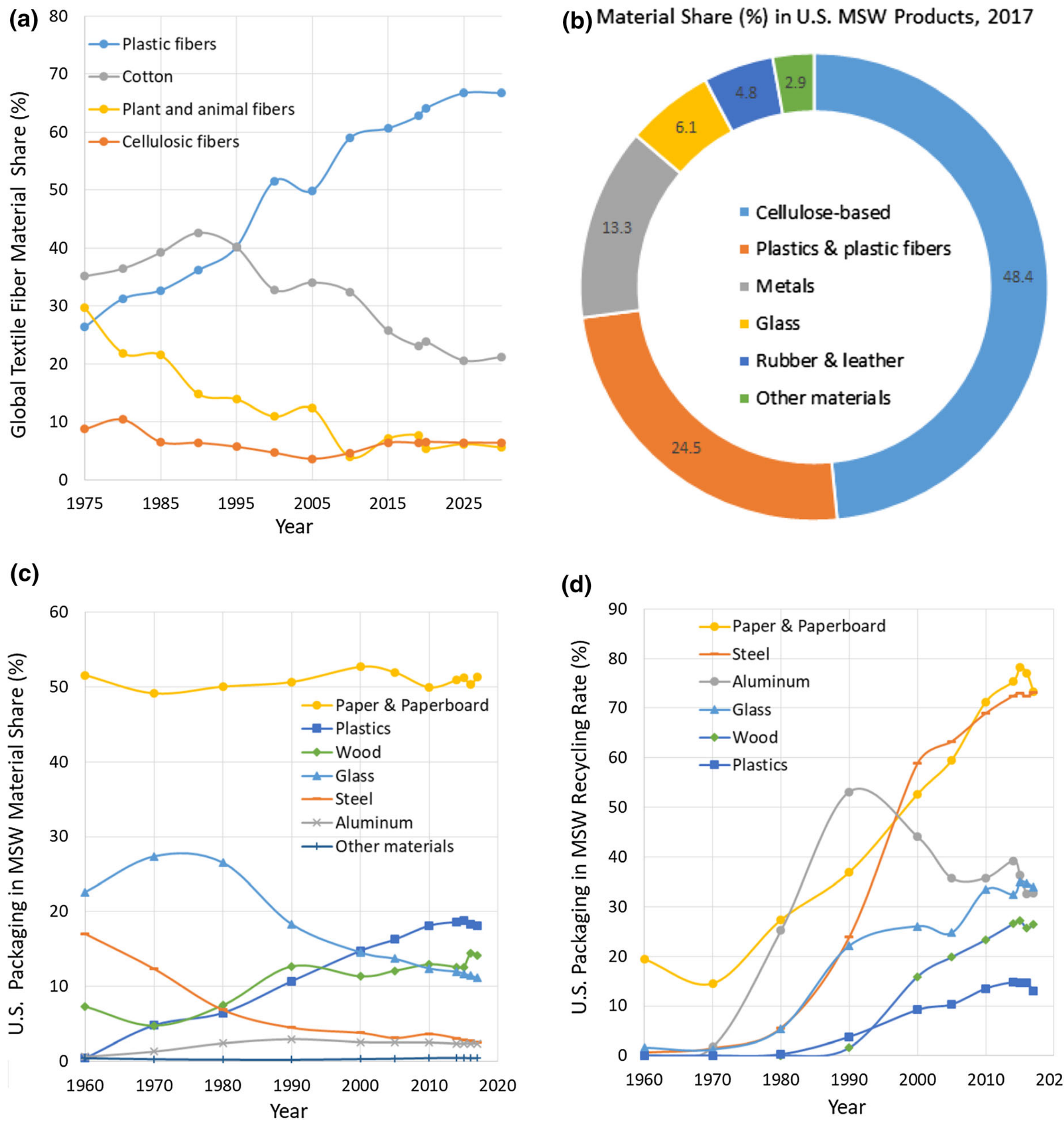


Fig. 2 Materials distributions by weight: **a** in the global production of textile fibers (actual to 2019 and projected to 2030)(Textile Exchange 2020), **b** in the U.S. packaging in municipal solid waste (MSW), and **c** in the 2017 U.S. total materials in products in MSW (EPA 2020). Cellulose-based materials include wood, paper and paperboard, plant fibers, and cellulosic fibers. The textile waste was attributed to the plastic fibers, cellulose-based, and other materials according to their estimated shares in textiles (Textile Exchange 2020), **d** the recycling rates by materials of packaging in the U.S. MSW(EPA 2020)

distributions of the packaging waste are shown in Fig. 2c. Cellulose-based packaging (wood, paper, and paperboard) took up 65.4 percent, which might be mostly used as secondary or tertiary packaging for transport. The share of plastic packaging waste has steadily increased in the past sixty years up to 18 percent in 2017, which is in the range of the global plastic packaging share at 17–25 percent of total global packaging volume (Neufeld et al. 2016). Plastic packaging might be predominantly used as primary packaging directly contacting with food or other contents. It is generally believed that plastic packaging can reduce food waste by extending shelf life and can reduce fuel consumption for transportation by bringing packaging weight down. However, the recycling rate of the collected plastic packaging was only around 13 percent and far lower than that of paper and paperboard at 73 percent in 2017 (Fig. 2d). After China's ban on importing recyclables in July 2017, it was a 4 percent decrease for paper and paperboard from 2016 whereas the overall recycling rate only decreased 2 percent. Cellophane, a regenerated cellulose film, was invented in the 1920s and had been the primary packaging for food since then, but it had been gradually substituted by plastics, especially polypropylene. Biopolymer-based films of cellulose, starches, chitin, and proteins are moisture sensitive and are needed to be processed to increase moisture resistance such as coated with resins. Currently, a very small amount of biopolymer-based primary packaging is in use and active research in this area is ongoing (Peelman et al. 2016).

Fiber-reinforced plastics are a composite material made of a polymer matrix reinforced with fibers. The global market for fiberglass used in composites is approximately 5 Mt per annum; the market for carbon fibers used for composites is less than 2% of the market size for glass fibers (Brosius 2016). A total of 1.1 Mt of tire textile cords was consumed per annum globally, among which was 51,350 tonnes or around 6% from rayon (regenerated cellulose) whereas 93 percent of the tire cords were polymer-based (NOKIAN TYRES PLC 2015). Cellulose-based fibers such as wood flour, wood pulp, regenerated cellulose fiber, and cellulose nanofibers have also been used to reinforce thermoplastic polymers. Wood flour reinforced wood plastic composite decking is a common product in backyards. Substantial efforts are ongoing to make wood fiber-reinforced composites for automotive manufacturing.

With the current trends of increasing populations and living standards in developing countries, it is projected that the world will extract 167,000 Mt raw materials in 2060. While the total amount of biomass often continues to grow, its share decreases in industrialized countries since the 1970s, especially the share of wood as fuel and building material for which nonbiomass alternatives are most easily substituted (OCDE 2019). If no action is taken, it is estimated that the global plastic production will reach 1124 Mt with plastic packaging of 318 Mt in 2050 (Neufeld et al. 2016). It is expected that the global textile fiber production will increase to 146 Mt with plastic fibers of 98 Mt in 2030 if business as usual continues. Against this background, urgent action of policy and technical interventions is needed to change the current production and consumption patterns of materials and promote the growth in sustainable materials. Some materials are usually designed with specific characteristics that make them ideal for their intended applications with which other materials are hard to compete in cost and performance; metals and some minerals are often such materials. However, it seems that there are spaces for bio-based materials to grow in the markets of textiles, primary packaging, and composites.

Bioeconomy refers to the economic activity of producing goods, services, and energy from biomass, mainly non-food biomass, whose major component is cellulose, the most abundant and renewable biopolymer on the planet. Cellulose is eminently suitable both as a chemical base and as a fiber filler to reinforce plastics composites. Cellulose-based materials here refer to any materials containing cellulose or derived from cellulose. Emerging technologies are cellulosic biofuels such as ethanol, biogas, and pyrolysis oil and cellulosic chemicals for bio-based products and polymers. Traditionally, cellulose is isolated and purified either at the cellular level or the molecular level and then reassembled into paper, paperboard, and regenerated cellulose such as cellophane, rayon, and lyocell, or is derivatized into cellulose derivatives such as cellulose ethers and cellulose esters, or used as reinforcements for wood-plastic composites. With the awareness of their renewability and biodegradability, the global production of dissolving pulp for manmade cellulosic fibers is projected to double in 2025 (Textile Exchange 2020). In recent years, another form of cellulose called nanocellulose has been intensively researched. Nanocellulose is a term referring to nano-sized cellulose isolated from plant cell walls, cotton linters, or bacteria cellulose by either chemical, mechanical, and/or enzymatic means. This may be either cellulose nanocrystals (CNCs) or cellulose nanofibrils (CNFs). They can be used as functional additives (rheological modifiers or reinforcements) in a wide range of matrices such as polymers, cement, or cosmetics, leading to enhanced process or product performance. They can also be assembled into cellulosic gels, foams, membranes, films, and fibers for a wide range of applications from absorbents, filters, and packaging to textiles, without dissolution and regeneration of cellulose.

The sources of cellulosic materials are wood and plant cell walls as well as plant seeds such as cotton. Like any other materials, the consumption of cellulose-based materials for building, construction, textiles, and packaging drives the extraction activity for the agricultural and forest sectors. Potential environmental impacts are an intensification of land use, soil erosion, loss of biodiversity, forest degradation, deforestation, habitat alteration, carbon sink depletion, desertification, alteration of watersheds. Generally, trees can grow on marginal lands without intensive use of water and chemicals. Forests have been advocated to mitigate global warming and provide feedstocks for materials with less environmental footprints. Fibers from sustainably managed forests have significantly gained in dynamics recently. Cellulose-based products are increasingly perceived by relevant stakeholders to be renewable, biodegradable, and sustainable. It is the mixture of durability, low cost, and massive scale that has made plastic so successful globally and difficult to match by any competing ecological solutions. In this sense, the economy of scale and stability of wood fiber supply from sustainably managed forests are huge advantages for wood cellulose in comparison to other sourced cellulose in the competition with plastics. Knowledge and discoveries concerning cellulose are scattered throughout the scientific literature. This lack of connection is an inherent challenge to the advancement of cellulose technology. This review surveys the mechanical properties of cellulose-based materials in the literature using tensile properties as an indicator. If data are presented in graphs and charts in original publications, they were then extracted by a software tool WebPlotDigitizer (Rohatgi 2018). The paper presents the data compared with other competitive materials, provides insights into future research, and discusses the potential and challenges of cellulose-based products to replace polymers and other materials. In this review, the word fiber may refer to thick individual plant cells, an aggregate of plant cells, staple fibers, or filaments.

Crystalline cellulose

In plant cell walls, linear cellulose chains are stabilized laterally by hydrogen bonds to form microfibrils with alternate crystalline and amorphous phases. Crystalline phases are a few nanometers laterally and hundreds of nanometers along the chains. The fact that the yield loss upon controlled acid hydrolysis is small and that there exists a level-off degree of polymerization implies that amorphous regions are shorter than crystalline regions. The amorphous region may be merely 4–5 anhydroglucose units in length (Nishiyama et al. 2003). Amorphous phases deform under stress to provide flexibility to microfibrils. Moreover, crystalline phases plasticize themselves by localized shear through a consequent dissipative molecular stick–slip deformation (Molnár et al. 2018). Without these means, cellulose microfibrils would be very brittle. In nature, microfibrils are embedded in the amorphous matrix of a mixture of hemicellulose and lignin to form hollow cells, which are glued together by a middle lamella to form plant support structures. In this hierarchical structure, at each higher level, flaws and disorder are introduced to dissipate energy under stress and increase the toughness of materials, but decrease the strength and stiffness. The plant cell wall structural properties are dictated by defects: the larger the size, the more flaws the structure contains, the less structural integrity it becomes.

There exist several cellulose crystalline lattices. The naturally occurring cellulose crystalline lattice is cellulose I; regenerated cellulose technologies break down cellulose I structure into molecules and reassemble them into cellulose II structure. Never does a piece of cellulose crystal on a human scale exist for mechanical testing. Isolated cellulose nanocrystals have small diameters and direct measurement of their tensile strength has not been reported. The modulus of elasticity of perfect crystalline cellulose is estimated by various analytical techniques such as x-ray diffraction, Raman spectroscopy, and inelastic neutron scattering or calculated through multiscale modeling. A broad range of crystalline cellulose moduli of elasticity values between 56–196 GPa are observed and the average experimental and predicted tensile modulus of cellulose I crystallites is 124 GPa and 134 GPa, respectively (Dufresne 2017). It is reasonable to consider that the tensile modulus of crystalline cellulose I is around 130 GPa (Dufresne 2017). Crystalline cellulose I is estimated to be 7.5–7.7 GPa in tensile strength. Molecular modeling of pure crystalline forms indicates cellulose II is slightly weaker and less stiff than cellulose I and is in the region of 77–167 GPa in modulus and 4.9–5.4 GPa in strength (Eichhorn et al. 2005; Eichhorn and Davies 2006).

Seldom do materials occur in a perfect crystalline structure macroscopically. Most highly ordered materials are aggregates of macroscopic fine fibers. The tensile properties of the state-of-the-art highly ordered fibers through post-drawing are also listed in Table 1. It can be seen that cellulose possesses remarkable properties. In its perfect packing phase, cellulose is superior to PET in intrinsic modulus but inferior to carbon, carbon nanotube, polyester, nylon, and aramid fibers. Cellulose fibers have achieved in a lab the same order of stiffness as glass fibers but inferior in strength.

Neat bulk cellulose materials

Cellulose microfibrils in plant and cotton fibers have been isolated using various methods into fibrils in submicron dimensions. The smallest isolated element is supposed to be from crystalline phases, i.e. cellulose crystallites, technically named cellulose nanocrystals (CNCs). Theoretically, CNCs should exhibit all the attributes of crystalline cellulose, i.e. with very high mechanical properties, but it is very difficult to directly measure the properties of individual CNCs. It seems that there are not any mechanical data for individual CNCs. The intermediate technical products of isolating microfibrils are cellulose nanofibrils via biologically, chemically, or thermally assisted mechanical disintegration, widely believed to possess alternate crystalline and amorphous phases. Coarser separated fibrils are loosely named microfibrilated cellulose or cellulose microfibrils. This latter term should not be confused with the concept of ontological cellulose microfibrils existing in plant cell walls. These isolated fibrils can be reassembled into neat cellulose hydrogels, aerogels (foams), membranes, films, and fibers (Fig. 3) by various processes including crosslinking, drying, vacuum filtration, solution casting, layer-by-layer assembly, coating, forming, extruding, spinning and drawing. Except for breaking down cellulose into fibrils, cellulose has been dissolved directly in several solvent systems such as ionic liquids, mixtures of NaOH/Urea/Water, N-methylmorpholine oxide (NMMO) (lyocell), or organic solvents, or can be derivatized first and then dissolved in solvents (viscose). Dissolved cellulose has been

Table 1 The crystalline lattice modulus in the direction of the polymer chain axis measured or simulated through molecular dynamics and technically achieved tensile properties of macroscopic materials

Material	Intrinsic modulus of elasticity (GPa)	Intrinsic Strength (GPa)	Actual modulus of elasticity (GPa)	Actual strength (GPa)
Cellulose I (Dufresne 2017; Mittal et al. 2018)	130	7.5–7.7	86	1.57
Cellulose II (Northolt et al. 2001; Dufresne 2017)	77–163	4.9–5.4	45	1.3
Carbon Fiber (Lee et al. 2008) (Okuda et al. 2016)	1000	130	294	13
CNT (Barber et al. 2006; Takakura et al. 2019)	1000	100		133
Polyester (PEN) (Nakamae et al. 1995; Suzuki and Koide 2000)	145		33	1.1
Polyester(PET) (Thistlethwaite et al. 1988; Zhang et al. 2017)	110		20	0.93
Aramid p-phenylene terephthalamides (Mercer 2016)	281–336		124	3.6
Nylon 6.6 (Dasgupta et al. 1996)	261		7.5	0.95
E-Glass (Unterweger et al. 2014)			72	3.5

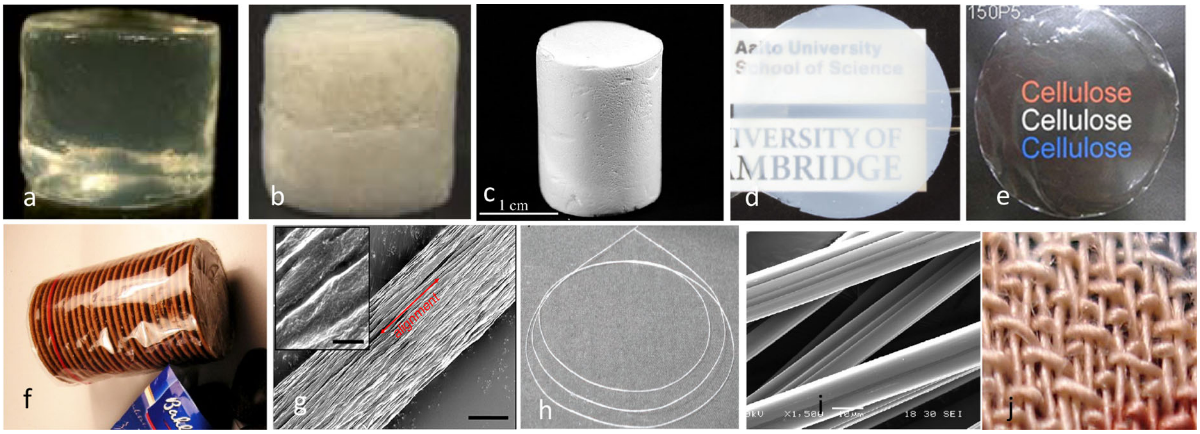


Fig. 3 Bulk cellulose materials: **a** 0.4% w/v TEMPO-CNFs hydrogel; **b** aerogel (Saito et al. 2011); **c** regenerated cellulose foam (Gavillon and Budtova 2008); **d** porous CNF membrane (Toivonen et al. 2018); **e** TEMPO-CNF film (Wakabayashi et al. 2020), **f** regenerated cellulose film (cellophane); **g** TEMPO-CNF fiber (Mittal et al. 2018) and **h** the image of a macroscale fiber spun from cellulose nanofibrils (Walther et al. 2011); **i** regenerated cellulose fibers (Shamsuddin et al. 2016) and **j** regenerated cellulose fabric from a skirt photographed with a macro lens. Reproduced from the reference with permission from The Royal Society of Chemistry for (a) and (b); from Wiley–VCH, copyright 2018, CC-BY-4.0 for (d); from Frontiers in Chemistry, copyright 2020, CC-BY-4.0 for (e); from American Chemical Society for (c) and CC-BY-4.0 for (g); from Wiley–VCH, copyright 2011 for (h); from Elsevier, CC-BY-4.0 for (i). Both (f) and (j) are in the public domain (Wikipedia Commons)

reassembled and regenerated into cellulose hydrogels, aerogels, membranes, films (e.g. cellophane), and fibers (rayon, lyocell, modal, cellulose acetate) through inversion precipitation and wet spinning, etc. In summary, neat bulk cellulose materials can be fabricated through either nanocellulose or regenerated cellulose.

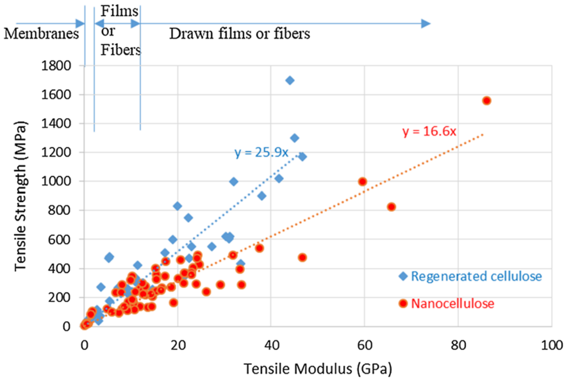


Fig. 4 Tensile properties of neat cellulose membranes, films, and fibers via nanocellulose and regenerated cellulose routes (Bhat and Makwana 1988; Northolt et al. 2001; Oliva et al. 2005; Gindl and Keckes 2006; Ganster and Fink 2006; Gindl and Keckes 2007; Gindl et al. 2008; Henriksson et al. 2008; Sun et al. 2010; Sehaqui et al. 2011; Panthapulakkal and Sain 2012; Sehaqui et al. 2012; Koga et al. 2013; Baez et al. 2014; González et al. 2014; Hamedi et al. 2014; Torres-Rendon et al. 2014; Sixta et al. 2015; Toivonen et al. 2015; Mertaniemi et al. 2016; Wanasekara et al. 2016; Mohammadi et al. 2017; Wang et al. 2017; Yao et al. 2017; Cazón et al. 2018; Lundahl et al. 2018; Mittal et al. 2018; Kim et al. 2019a; Lossada et al. 2019; Nechyporchuk et al. 2019)

Figure 4 compares tensile modulus and strength of regenerated cellulose and cellulose nanostructured materials. Rarely were hydrogels and aerogels characterized with tensile properties, so no data are presented here for these materials. The tensile properties are dominantly determined by pores and fibril orientations, which can be finely tuned by processing methods. The terms of membrane and film both refer to a flat artifact that is thin relative to its length and width; the difference is that membranes utilize their pores whereas films function with other properties. Cellulose membranes are typically formed by immersion precipitation and thus have a controlled percentage of porosity at around 40–86%, which is intermediate between water-dried films and freeze-dried aerogels. Their tensile properties decrease with increasing porosity (Sehaqui et al. 2011) and are lower than films, which are often prepared by solution casting, vacuum filtering, dried, or consolidated by pressure to yield leading to being denser, stiffer, and stronger. For example, aqueous solution cast nematic ordered CNF films through control of the drying conditions are 15.4 GPa in modulus and 359 MPa in tensile strength (Zhao et al. 2018). Random-oriented dense films can be up to 12 GPa and 259 MPa (Zhao et al. 2018), and 11.2 GPa, 230 MPa (Österberg et al. 2013). Most nanocellulose films are around 60–120 MPa. The tensile strength and Young's modulus of regenerated films without orientation can be up to 263 MPa and 7.3 GPa, respectively (Yang et al. 2012). Fibers are wet spun through a small capillary hole resulting in orientation to a certain extent during the spinning process, and post-spinning drawing further orients the fibrils or cellulose chains along the fiber axis leading to enhanced mechanical properties in the drawing direction. Drawing can also orient films and thus improve the tensile properties along the draw direction. The mechanical properties of the resulting fibers appeared to be highly dependent on the extrusion and reeling speed. Oriented CNF filaments achieved 1570 MPa in tensile strength and 86 GPa in Young's modulus (Mittal et al. 2018). A tensile strength of 1300 MPa and an elastic modulus of 45 GPa were achieved through regenerated cellulose by increasing the degree of orientation of cellulose chains along the fiber axis (Northolt et al. 2001). The maximum tensile properties achieved by the current technology are still well below the theoretical prediction for perfect cellulose crystalline lattices (Table 1). These low strengths have been ascribed mainly to the presence of amorphous domains, pores, and flaws in fibers. Crystallites may also contain an array of defects, including imperfect chain alignment, impurities, and sheet-stacking faults. The extent of perfection that can be achieved also depends on the process used to obtain collectible fiber forms. Lyocell fibers have a higher degree of polymerization, crystallinity, and molecular orientation than viscose fibers (Kreze and Malej 2003). Generally, textile fibers do not possess as high mechanical strength as reinforcement fibers since the latter is usually subject to post spinning drawing. interfibrillar hydrogen bonds resulting in very strong films.

It is very difficult to organize fundamental building blocks below the nanoscale into precisely ordered hierarchical structures on the macroscale without introducing flaws and disorder at a larger length scale, especially in a facile, low-cost, and environmentally friendly manner. New processing methods like the one assembling CNFs into the strongest filament (Mittal et al. 2018) are not economically viable, so do other membrane and film formation processes. Moreover, a three-dimensional ordered bulk material would be very brittle and difficult to be used. The current technology is to fabricate 1D fine filaments with high strength in the direction of the highly orientated molecular chains, with the mechanical properties significantly lower in the lateral directions. These filaments are then used as thread constituents in two-or three-dimensional woven fabric structures or as reinforcements in high-performance (typically) polymer-matrix composites, in the same way as carbon and glass fibers.

It can be seen from Fig. 4 that regenerated cellulose-based materials are comparable or even stronger and more flexible than those fabricated from nanocellulose. Regenerated cellulose such as rayon and cellophane have been used for textiles and food packaging since the 1920s and waned since the 1960s attributable to the replacement by plastic films. Since these processes involve solvents, they are not very green to the environment and the workers, thus they are designated as semisynthetic fibers, not natural fibers. Clothing is one of the four necessities (food, clothing, shelter, and mobility). The low-cost production of textiles from wood fibers could be a hot research and development topic for generations to come. Using nanocellulose prepared from a mechanical process instead of chemicals could produce textiles. Both regenerated cellulose and nanocellulose technologies are currently under development driven by lowering plastic pollution. For nanocellulose, it is to develop processes of assembling nanocellulose into macroscopic structures useful for implementation; for regenerated cellulose, it is to decrease the cost and toxicity of production.

Composite materials from cellulose with hemicellulose and lignin

Fibers for textiles

Hemicellulose and lignin are generally weak materials, but they can be combined with cellulose to form various materials on the human scale. In nature, cellulose is combined with hemicellulose and lignin to form various elongated cells with a lumen. Plants contain a variety of cells that perform specific functions. Elongated thick-walled cells are strong, providing most of the strength and support in plants.

Technically, individual fibers refer to sclerenchymatous fibers in vascular bundles (such as those in bamboo and bast fiber crops), fibers in hardwood, and tracheids in softwood, etc.. The cell wall consists of oriented cellulose microfibrils embedded in a matrix of hemicellulose and lignin. The dimensions of commercially important fibers are summarized in Table 2. Figure 5 shows the tensile modulus and strength of individual fibers. There are two populations of fibers as designated by the two dotted trend lines. The black dashed line corresponds to bast fibers and other plant fibers with high stiffness. The red dashed line represents a group of fibers with lower stiffness, therefore, may be better suited for textile manufacture.

The variations of the tensile properties are ascribable to fiber morphology (shape, size, cell wall thickness, the fraction of the secondary wall), chemical composition (fractions of cellulose, hemicellulose, and lignin), the structure of cellulose (crystallinity and microfibril angle to the fiber axis), and the presence of flaws (type, size, distribution) as well as measurement methods (Everitt et al. 2013). The microfibrillar angle has a major influence on the tensile properties of plant fibers. The fiber strength generally decreases with an increase in microfibril angle. Since cotton fibers have a large average microfibril angle, their strength and stiffness are also low. Besides, increasing the fiber diameter results in decreasing the tensile strength of the fiber since coarser fibers may have a high probability to contain more flaws (Tomczak et al. 2007). It may be worth noting that the measurement of the technical properties of bast fibers is sometimes not on individual fibers but the bundle of individual fibers (also known as technical fibers). Individual fibers should have much higher strength than fiber bundles because the break might occur at the connection between individual fibers in a bundle. When tested as a filament of fiber bundles, these technical fibers typically decrease in strength with increasing gauge length since the long gauge length may cross different individual fibers. The connection between individual fibers comes into play in degrading the tensile performance by slipping over each other (Tomczak et al. 2007; Zhu et al. 2013). This type of behavior is described by Weibull statistics, in which flaws are assumed to be randomly distributed throughout the tested length. Individual fibers may deform considerably under stresses incurring kink bands, nodes, dislocations, and slip planes. The tensile strength of fibers is controlled by the strength of the "weakest link," i.e., fracture occurs at the largest flaw present in the sample. With increasing gauge length, the chance of finding a severe flaw increases, so fiber

Table 2 Physical properties of plant fibers and other natural textile fibers (Han 1998; Ansell and Mwaikambo 2009)

Fiber	Length (mm)	Diameter (μm)	Wall thickness (μm)	Microfibril Angle (°)
Cotton(USDA 2001)	20–38	11–17	1–4	20–30
Kapok	8–32	15–35	1–3	0
Flax	20–50	15–20	4–12	5
Hemp	20–50	17–23	4–12	6
Ramie	60–250	28–35	4–12	8
Mulberry	12–35	13–19		
Kenaf	2–6	17–22	4–12	
Jute	2–3	16–21	4–12	8
Bamboo (Osorio et al. 2018; Rusch et al. 2019)	1–3	12–20	5–7.5	2–10
Alfalfa	2–5	5–10	–	
Wool	90–180	21–26	Solid	
Human hair		50–100	Solid	
Silk	Filament	10–20	Porous	
Spider silk	Filament	2–5	Solid	
Softwood (tracheid)(Mander and Liu 2010)	1.18–7.39	20–40	2–5	5–20
Hardwood (fiber)	0.77–2.30	15–30	3–6	5–20

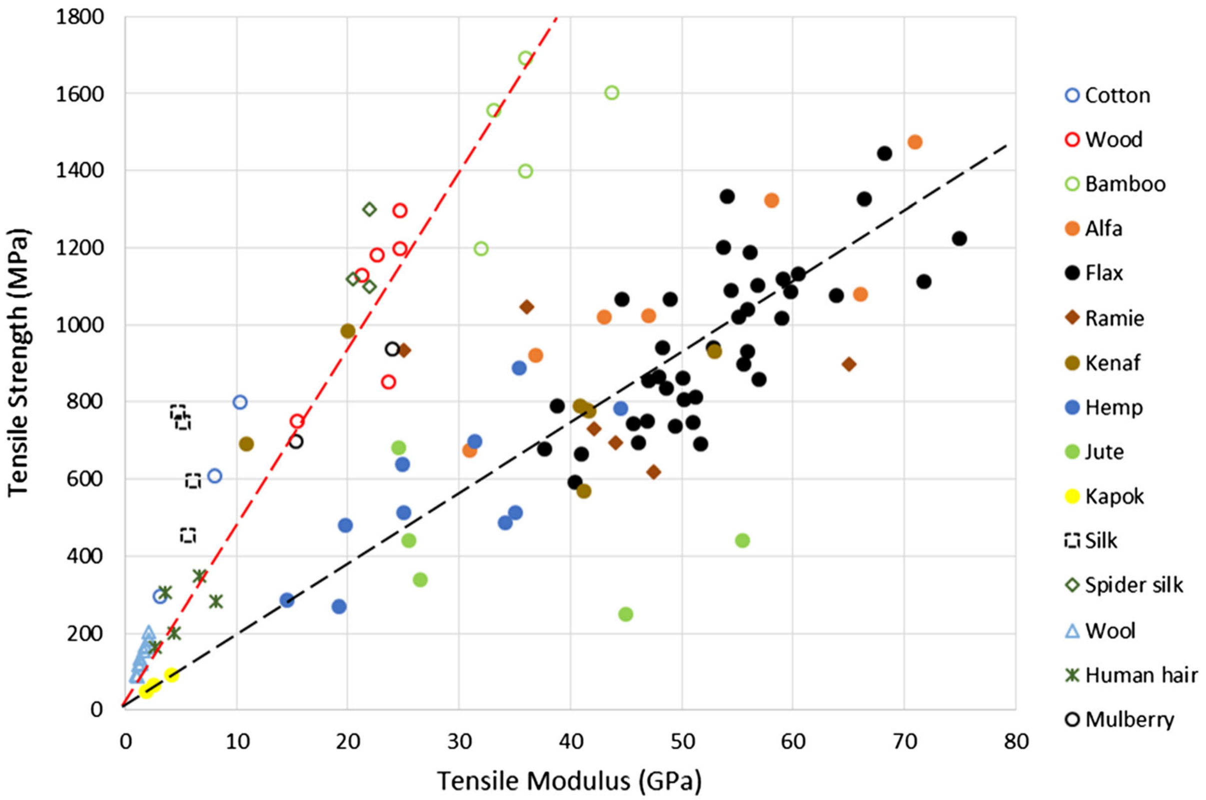


Fig. 5 Tensile modulus and strength of individual cells of various sources. It can be classed into two populations as strong (red) vs stiff (black) fibers designated by the dash lines (Cunniff et al. 1994; Gosline et al. 1999; Bunsell 2009; Cicala et al. 2009; Virk et al. 2012; Mahjoub et al. 2014; Unterweger et al. 2014; Du et al. 2015; Ashori et al. 2016; Dong et al. 2017; Bourmaud et al. 2018; Kunchi et al. 2018)

strength decreases. Because each of the measurements was based on different fiber diameter, *d*, and gauge length L for different fibers under different investigations and has not been normalized to a standardized diameter and gauge length, the data are only good for interpreting the trends (Yang et al. 2017).

Table 2 shows typical fiber lengths and diameters. These two parameters have important implications for applications, in contrast, mechanical properties are less important since most widely used fibers for textiles such as wool, silk, and cotton do not have the best tensile properties as shown in Fig. 5. Textile quality relates to fiber geometry. Finer fibers render fabrics softer; longer and coarser fibers pill less in yarns and fabrics. Fiber length affects yarn strength, yarn evenness, and the efficiency of the spinning process. Only those fibers long enough can be dry spun into yarns for textiles, including cotton, flax (linen), hemp, ramie, and kapok fibers. A high content of less than 12.7 mm-long fibers leads to high waste in the spinning process and weak, uneven, and hairy yarns (Jitjaicham and Kusuktham 2016). Premium fiber geometry depends on species. Premium cotton fibers are around 13–16 µm (USDA 2001). The diameters of most synthetic polymer textile fibers also fall in this range. In this sense, spider silk may be too fine, and human hair may be too coarse for producing good textile materials. Kapok fibers are generally fairly short, thin-walled, and smooth, making them less efficient to be woven into yarns, but their textiles are extremely lightweight. Ramie fibers have large and distributive diameters and dry spinning may result in coarse yarns and fabrics. It is reported that mulberry inner bark fibers can be spun into yarns with a higher tenacity and elongation but a lower unevenness than flax yarns (Dong et al. 2017). Alfa, bamboo, jute, and kenaf individual fibers are too short to be dry spun into yarns. But those fibers exist in nature as fiber bundles and their technical fibers may be long enough to be spun into coarse yarns and fabrics, usually not good enough for fine clothing in modern society.

Cotton and flax fibers are distinguishable by their superiority to other fibers such as non or very low lignin content. Since flax fibers are longer and coarser than cotton fibers, linen fabrics feel slightly thicker and stiffer than cotton fabrics. Flax fibers appear line patterned in the weave of a linen garment, whereas cotton can appear smoother and more uniform. A linen item may also have more natural creases than a cotton piece. Cotton fibers are less stiff and weaker and smaller in diameter, lending them softness and fluffiness. Excluding plastic fibers, cotton constitutes the largest portion of plant fibers for textiles. Today, linen is usually an expensive textile produced in relatively small quantities. Other plant fibers such as ramie and kapok fibers just have niche markets in special cultural and geological contexts. Hemp fibers should be comparable with flax fibers. Because of the concern of the psychoactive ingredient ‘tetrahydrocannabinol, hemp fibers are not widely grown in the US. In recognizing the distinction between industrial hemp crop and recreational ‘marijuana’, hemp production has been resurgent in Europe and North America.

Since wood or bamboo fibers are stiffer and stronger than cotton fibers and not as long as bast fibers, directly dry spinning wood or bamboo pulp fibers into yarns has not been realized even after the removal of lignin and hemicellulose. Instead, they need to be dissolved in a solvent and then spun into filaments or yarns using a wet-spinning process such as lyocell and viscose processes. These fibers are solid and continuous filaments similar to plastic fibers in contrast with short natural plant fibers with lumens, which make textiles insulative and light. There are trade-offs between crop fibers and wood fibers. Wood fibers are abundant but require a costly wet spinning process to produce yarns. Dry spinning processes for crop fibers are economical but its plantation competes with lands for food and uses pesticides and water. With a growing population, we may need to extract more fibers for textiles from wood, spare arable lands for food, and reduce plastic fibers to eliminate plastic pollution at the same time. Innovative ways of using wood fibers for textiles should be intensively investigated in the future, starting from fiber crop cultivation, machine-aided harvesting to pulp manufacturing, and yarn spinning.

Fibers for paper and paperboard

Paper is the assembly of partially or fully delignified and individualized plant fibers. Historically, it has been made from mulberry bast fibers, and then from rags that were waste hemp, linen, and cotton textiles. Although the word paper originates from the word papyrus, Egyptian papyrus paper is made from flattened piths of papyrus plants glued by its sap or woven together in an orderly manner Such a product is not real paper but a sheet of biocomposite for writing. Today, the paper is mostly made from wood pulp with a range of tensile properties as shown in Fig. 6. Paper mechanical properties are just needed to be sufficient to support its functionalities. The variations of the tensile properties are attributable to pulp grades (mechanical pulps or chemical pulps, bleaching, refining), processing methods (paper chemicals, forming, calendering), and species (hardwood or softwood). Cell wall thickness, fiber length, and fiber strength all influence paper strength. Cotton and bast fibers are often used for currency notes to last longer than wood pulp paper. Wood pulp paper is used for writing, printing, and packaging. Secondary and tertiary packaging paper, paperboard, and containerboard are mainly made from mechanical pulp. The primary food packaging (contact with food) is dominated by plastic and aluminum films. The challenge of substituting plastics with paper is that paper does not resist water moisture or grease, plus its opacity. Food serving paper usually contains chemicals such as per- and polyfluoroalkyl substances (PFASs) to enhance its moisture and grease resistance. These chemicals might have adverse effects on health and the environment

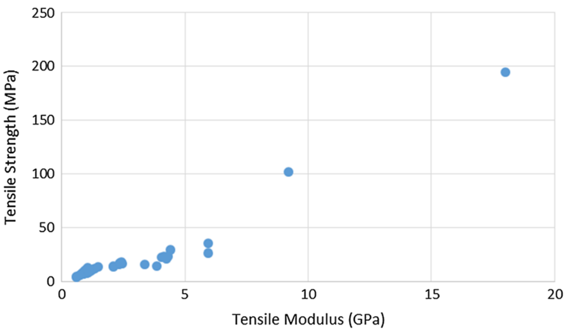


Fig. 6 tensile modulus and strength of various paper materials (Osong et al. 2014; Vallejos et al. 2016; Larsson et al. 2018; Motamedian et al. 2019)

(Schaider et al. 2017), therefore, will be a major hindrance to their being included in primary packaging. PFASs-free paper for food contact packaging would be a severe challenge to be overcome.

Wood—a natural composite of cellulose with hemicellulose and lignin

An array of cells are bound together by a middle lamella to form wood. Nature produces several different cells and arranges them in many ways to form many wood species. Softwoods consist of more than 90% tracheid cells and the remaining parenchyma or thin-walled fibers. Hardwoods contain 50% wood fibers, 30% vessels, and 20% parenchyma. Bamboo contains 50% parenchyma, 41% fibers, and 9% vessels (Qi et al. 2014). Figure 7 shows tensile strength plotted against tensile modulus for selected plant materials. Note the large range in properties produced by varying the arrangement of the three building blocks (cellulose, hemicellulose, and lignin) in the cell wall as well as the cellular structure (Gibson 2012). From crystalline cellulose (the upper right in Fig. 7) to thin parenchyma tissues (the bottom left in Fig. 7), increasing dimensions that increase the probability of the occurrence of structural defects, decreasing microfibril angles to the tension direction, and thining cell walls lead to decreasing tensile properties. Since major thick-walled fibers orient along tree height to form the wood grain for supporting the whole tree, the tensile properties parallel to grain are higher than those perpendicular to the grain.

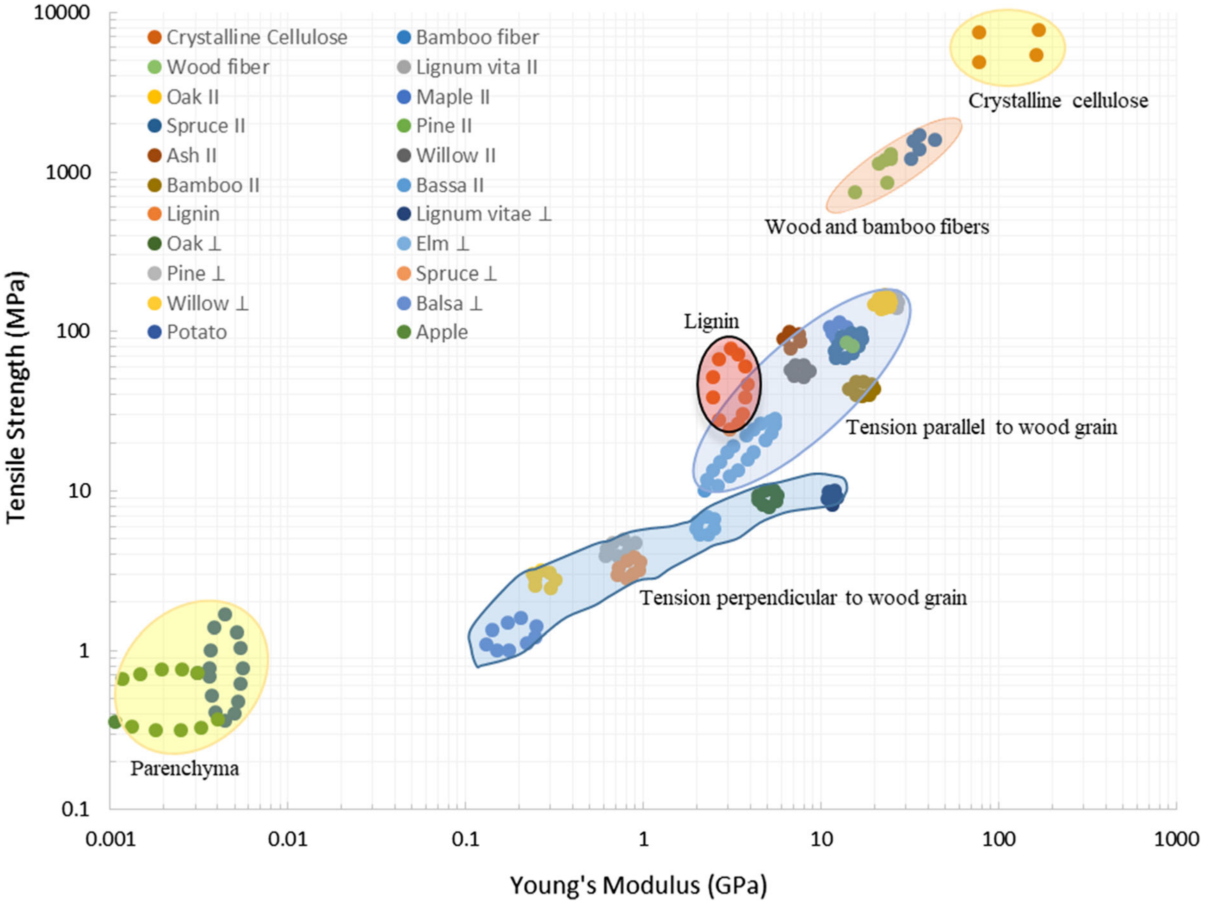


Fig. 7 Tensile properties of wood and wood fibers. From crystalline cellulose, the wood of mixing thin-, and thick-walled cells to thin-walled tissues, tensile properties decrease six orders of magnitude for modulus and five orders for tensile strength. Well-organized and defect-free tiny crystalline cellulose has very high tensile properties. With increasing the size, the material has a higher probability of containing flaws, which decreases tensile properties (Moon et al. 2011; Gibson 2012; Dufresne 2017; Bourmaud et al. 2018)

Apple and potato tissues are made of parenchyma and have the lowest tensile properties. The lightest density wood species balsa is only 160 kg/m^3 whereas *lignum vitae* is the world's densest wood at 1260 kg/m^3. Figure 8 shows their microstructure. It can be seen that the lumens of *lignum vitae* wood are very small and balsa wood cells have large lumens. These account for their lowest and highest tensile properties as shown in Fig. 7. All other wood-species properties are located between these two species. Nature has not perfectly arranged cellulose to achieve its best potential, while humans have invented many ways to improve wood properties such as condensed wood, plywood, cross-laminated timber, etc. Wood or logs can be broken down into many building blocks such as lumber, veneer, strands, and particles, which are then reassembled into engineered wood and wood products. These ways of utilizing wood will continue to be predominated.

Nature fiber reinforced polymer composites

Flax fiber-polypropylene composites

Bast fibers originate from the phloem of the plants flax, hemp, jute, and kenaf (Fig. 10a). Individual cells of flax bast fibers, also called elementary or individual fibers, are relatively long with thick cell walls. They are 15–20 µm in diameter and 20–50 mm in length (Van den Oever et al. 2000). Individual fibers orient themselves vertically and are glued together by pectin into a bundle (technical fibers), appearing in the cross-section at 50–100 µm in diameter (about 10–40 cells). The length of the technical fibers is dependent on the length of the plant (Mokshina et al. 2018). Through retting, scotching, and hackling, bundles of fibers are separated from stems into technical bast fibers (Fig. 9b). The chemical composition of the bast fibers is similar to the exception of jute, which contains much more lignin than the other bast fibers. The fibers may differ in coarseness, length, strength, and stiffness (Fig. 5 and Table 2). The natural origin of fibers accounts for the variations in the composites.

Bast fiber bundles (Fig. 9b) can be cut into short fibers of several inches, called staple fibers (Fig. 9c), then overlaid into mats (Fig. 9d), spun into yarns (Fig. 9e), or woven into fabrics (Fig. 9f). Bast fibers can also be processed through traditional pulping such as Kraft pulping into individual cells or bast fiber pulps (Fig. 9g). In the past decades, staple bast fibers, bast fiber mats, bast fiber yarns have been widely investigated to be reinforcements for polymer composites in the hope of substituting for glass fibers as light renewable materials for automotive structural components. Figure 10 shows the stiffness and strength of flax fiber reinforced polypropylene composites from the literature. The variations are ascribed to the cleanness and forms of flax fibers, polypropylene grades (co-polymer, or homo-polypropylene, molecular weight), formulations such as additives (coupling agents such as maleic anhydride grafted polypropylene), fiber fractions, processing methods, and degree of orientation. Generally, flax pulp does not reinforce

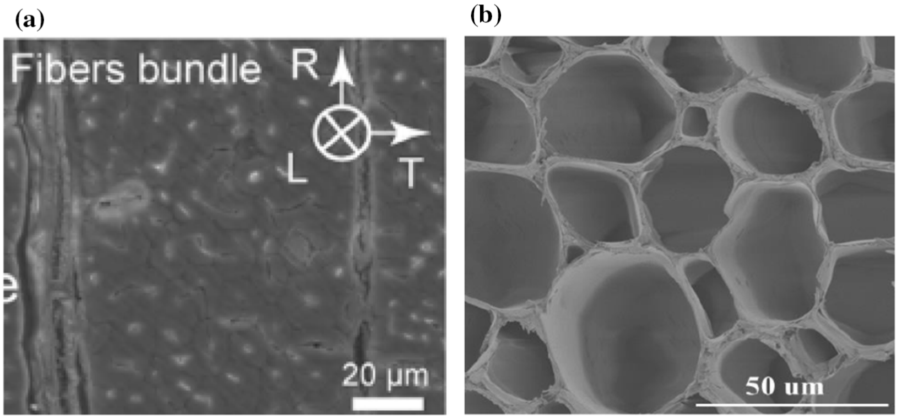


Fig. 8 The microstructure of cross-section of the densest wood: **a** *Lignum vitae* (Yin et al. 2016), and **b** the lightest balsa wood (Fu et al. 2017), Reproduced from the reference with permission from Springer Nature, CC-BY-4.0 for **a**, and from American Chemical Society for **b**

Fig. 9 **a** Flax stem cross-section showing locations of underlying tissues: Ep = epidermis; C = cortex; BF = bast fibers; P = phloem; X = xylem; Pi = pith. The stem of flax is around 2–3 mm in diameter; **b** the technical fiber is around 50–100 µm and consists of about 10–40 individual fibers in the cross-section. Individual fibers generally with diameters around 15–20 µm and lengths between 20–50 mm. The multiple bundles after scutching are up to 200 µm in diameter (Van den Oever et al. 2000; Oksman et al. 2003). **c** Chopped staple fibers (Bachmann et al. 2018), **d** flax yarns (Yan et al. 2014), **e** flax fiber mat (Claramunt et al. 2017), **f** flax fabric (Radkar et al. 2019), **g** flax bast pulp. Reproduced from the reference with permission from Elsevier for **b** and **d**; from MDPI.COM, CC-BY-4.0 for **c**, **e** and **f**; **a** and **g** Wikipedia Commons

polypropylene because pulp fibers are likely shorter than critical length below which the matrix cannot effectively transfer stress to the fibers and thus cannot take advantage of individual elementary fiber strength and stiffness. Staple fibers provide better strength while mats enhance the stiffness. The orientation of staple fibers in the mat or polymer matrix creates the anisotropy with enhanced or reduced properties in the opposite directions. Oriented yarns provide high mechanical properties in the oriented direction but decrease them along the transverse direction.

Figure 11 compares the mechanical properties of individual flax fibers, glass fibers, and polypropylene composites. It indicates that both individual flax fibers and their staple fiber reinforced PP composites are inferior to glass fibers and glass fiber reinforced PP composites. The mechanical properties in the oriented direction can surpass glass fiber reinforced PP composites. It implies that bidirectional woven fabrics of flax yarns may be a good reinforcement to achieve similar mechanical properties of the glass fiber composites biaxially as shown in Fig. 10. However, very little work has focused on using flax fabrics, which are expected to give higher strength performance attributable to its structure but the processing needs to be optimized to achieve efficiency as those applied as staple fibers.

Cellulose-polyvinyl alcohol composites

Nanocellulose is typically prepared and stored in aqueous suspensions. Polyvinyl alcohol (PVOH) is a water-soluble polymer with many hydroxyl groups. There have been considerable investigations to take advantage of the hydrophilicity of nanocellulose and PVOH over the past several decades. They have been

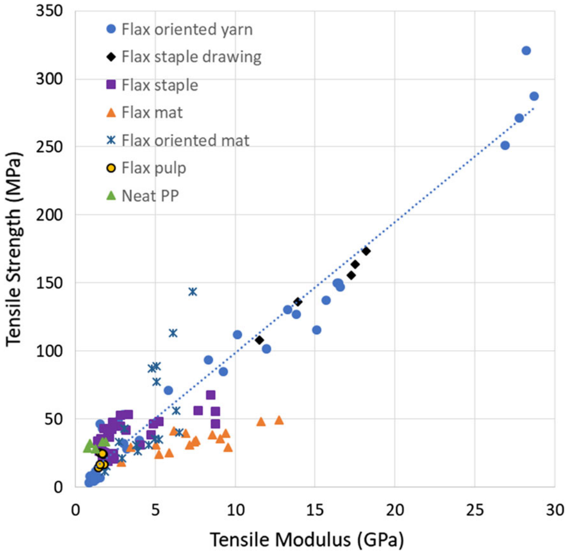


Fig. 10 Tensile strength versus tensile modulus of flax fiber reinforced polypropylene composites with flax fibers in the forms of pulp, staple fibers, mats, and yarns, random or oriented (Heijenrath and Peijs 1996; Oksman 2000; Van den Oever et al. 2000; Cantero et al. 2003; Madsen and Lilholt 2003; Arbelaiz et al. 2005a, b; Miao and Shan 2011; Kannan et al. 2013; Derbali et al. 2016; Wu et al. 2016; Bar et al. 2018, 2019)

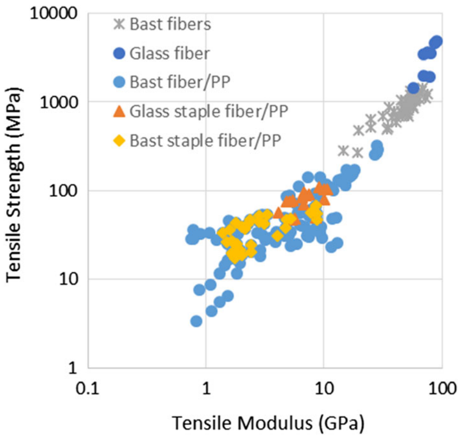


Fig. 11 Comparison of flax fibers and staple fiber polypropylene (PP) composites with glass fiber and glass staple fiber polypropylene composites (Ganster et al. 2006; Unterweger et al. 2014; Franciszczak et al. 2017; Bourmaud et al. 2018)

mixed in water and then processed into hydrogels, electrospun mats, films, and fibers through electrospinning, solution casting, and conventional wet spinning. Figure 12 shows the tensile elastic modulus and strength of these fabricated materials. Generally, the form of the composite is a deciding factor on its tensile properties, i.e. fibers have better properties than films, films better than electrospun mats, and mats or membranes better than hydrogels. Within each categorical form of material, the addition of nanocellulose skews the property slightly towards the elastic modulus axis, i.e. increasing the elastic modulus of the composites. Most often, it also slightly enhances the strength. Nanocellulose reinforcement seldom overcomes processing disadvantages to improve the properties to an upper category. In other words, fabricating methods determine the product performance window. The variations are ascribed to nanocellulose type, morphology, its volume fractions in the composites and PVOH grades (fully or partially hydrolyzed PVOH), etc. It was observed that partially hydrolyzed PVOH does not fully interact with nanocellulose (Peresin et al. 2010). There is also a trend towards lower tensile properties as the diameter of fibers increase.

It is worth noting that electrospun mats have low tensile properties. However, individual nanofibers should have similar mechanical properties with those fibers spun by conventional spinning processes since the individual nanofibers are subject to high shear stresses during electrospinning. However, there are few investigations reported to characterize individual electrospun fibers in the literature. Drawing is commonly used to improve the strength and elastic modulus of conventional fibers. Similarly, drawing on electrospun fibers can be done through the use of a rotating drum collector for collecting the fibers during electrospinning. Inai et al. (2005) tested the effect of the take-up velocity of a rotating disc collector on electrospun poly-L-lactide (PLLA). With a higher take-up velocity of 630 m/min, the tensile strength of single-strand PLLA nanofibers were increased from 89 to 183 MPa. The increment in tensile strength and modulus was attributed to higher molecular orientation as a result of a greater take-up velocity. However, the tensile elastic modulus and strength of electrospun fibers were lower than melt-spun fibers with similar take-up velocity.

Figure 13a compares the tensile strength and elastic modulus of neat nanocellulose, neat polyvinyl alcohol, neat carbon nanotubes, and their composites in films and fibers. Figure 13b is the amplified portion of Fig. 13a near the origin, mainly for films with different levels of porosity. The data points are quite scattered attributable to different additives, surface

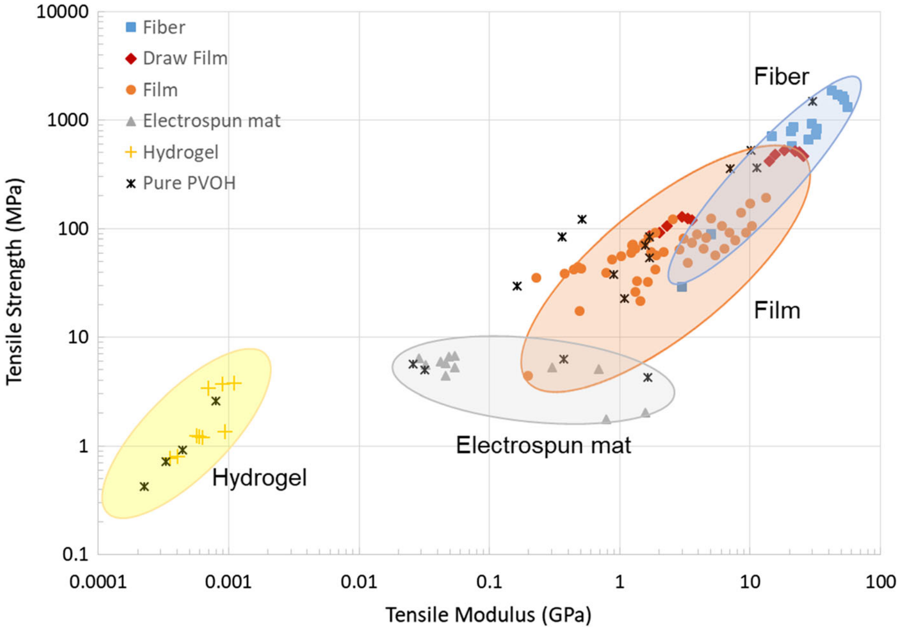
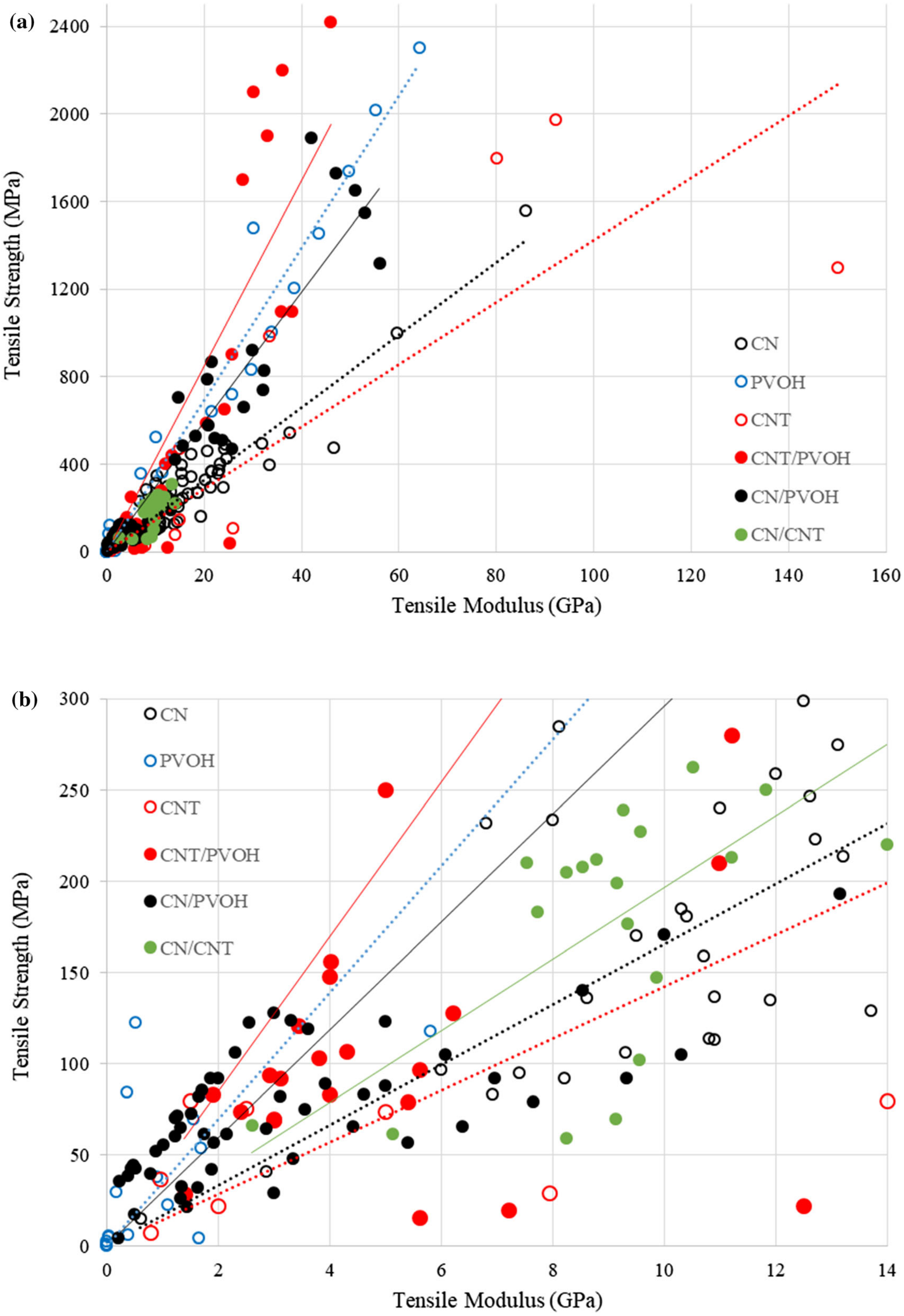


Fig. 12 The composites of nanocellulose and polyvinyl alcohol in various forms (Jalal Uddin et al. 2011; Liu et al. 2013; Gonzalez et al. 2014; Peng et al. 2014, 2015; Virtanen et al. 2014; Clemons 2016; Huan et al. 2016; Lee et al. 2016; Fahma et al. 2017; Tummala et al. 2017; Song et al. 2018; Park et al. 2019; Sanders et al. 2019)

treatments, and processing conditions. The linear trend lines are regressions of the scattered points for easier comparison. CNTs show a much larger scatter than nanocellulose and PVOH. Neat CNTs and neat nanocellulose are relatively shifted towards the elastic modulus axis while neat PVOH relatively leans towards the strength axis. These neat materials can be shaped into fine fibers by spinning and then oriented by drawing leading to very high tensile properties. The common-sense notion that polymers are soft is based on our daily perception. Each material can be made stronger and stiffer with proper techniques. Yet the process used to achieve the desired results may have different economic implications. When two materials are combined, the resultant composites possess the tensile properties that fall between them for CN/PVOH, but beyond them towards the strength axis for CNT/PVOH and CNT/CN composites. Elastic modulus is a measure of resisting the deformation of materials. Strength is a measure of resisting failure. Initial elastic modulus often reflects the intramolecular hydrogen bonding and strength reflects inter-chain or inter-fibril hydrogen bonds or covalent bonds (Du et al. 2011). It can be seen that these materials can be formulated to optimize the properties of resulting composites. It is also worth noting that reinforcement is usually discussed concerning a specific property. Nanocellulose and CNTreinforce each other in strength. But their neat materials are stiffer as compared with their composites. The same is true on CNT/PVOH. Nanocellulose stiffens PVOH towards the elastic modulus axis but may decrease its strength. Either using a single material or combined composites depends on both targeted properties and process economics, as well as sustainability and recyclability.

Figure 13a shows the trend that the nanocellulose/PVOH composites are less stiff but stronger than neat cellulose materials, which provides insight that the composites of nanocellulose and PVOH are superior for textiles to neat nanocellulose. Surprisingly, only a few investigations were conducted on the composites of regenerated cellulose and PVOH for films and fibers. It is expected that the composites of regenerated cellulose and PVOH composites can further skew the trend line towards the tensile strength axis leading to more favorable materials than neat regenerated cellulose. This observation also implies that PVOH does have some interactions with cellulose and enhances strength.


(a)
Tensile Strength (MPa)
2400
2000
1600
1200
800
400
0
Tensile Modulus (GPa)
0
20
40
60
80
100
120
140
160
CN
PVOH
CNT
CNT/PVOH
CN/PVOH
CN/CNT
(b)
Tensile Strength (MPa)
300
250
200
150
100
50
0
Tensile Modulus (GPa)
0
2
4
6
8
10
12
14
CN
PVOH
CNT
CNT/PVOH
CN/PVOH
CN/CNT

◀**Fig. 13** **a** Comparing neat materials with cellulose nanomaterials (CN), CNT and PVOH composites, **b** The plot near the origin (Kunugi et al. 1990; Vigolo et al. 2000; Dalton et al. 2003; Zhang et al. 2003, 2004; Paiva et al. 2004; Chen et al. 2005, 2018; Liu et al. 2005, 2018; Bhattacharyya et al. 2006; Xu et al. 2010; Di et al. 2012; Mercader et al. 2012; Uddin et al. 2012; Behabtu et al. 2013; Koga et al. 2013; Salajkova et al. 2013; Hamedi et al. 2014; Lu et al. 2014; Wei et al. 2015; Yee et al. 2018)

Cellulose-polypropylene composites

Polypropylene is a commodity polymer and has been reinforced with glass fibers for structural applications. Whether cellulose fibers can achieve similar performance to glass fibers has been a pressing question for decades. Figure 14 compares the tensile properties of neat polypropylene and its composites with cellulosic fibers. The variations of commercial neat PP properties are ascribed to different PP grades, such as PP block copolymers, random PP copolymers, or PP homopolymers. PP homopolymers generally have high mechanical properties than PP copolymers (Ganster et al. 2006). Cellulose pulp stiffens PP but usually weakens its strength. Wood flour also stiffens PP but weakens PP strength even more than cellulose pulp. Nanocellulose usually stiffens PP but does not strengthen PP. Sometimes, nanocellulose reinforces PP in both elastic modulus and strength at high volume fractions as fabricated by the pulp/polymer in-situ kneading fibrillation method (Suzuki et al. 2014). CNTs generally strengthen PP but do not stiffen PP.

As shown in Fig. 14, regenerated cellulose and glass fibers are both chopped short fibers from spun filaments, which can both reinforce (stiffen and strengthen) PP. Glass-fiber reinforcing effects are the best. The high-performance zone of short regenerated cellulose fibers and nanocellulose is located near the low-performance zone of glass fibers, i.e. precision manufactured cellulose reinforced PP can be only comparable with loosely fabricated glass fiber reinforced PP. This implies there is a long way to go with cellulose fibers to replace glass fibers. However, cellulose can be processed to reinforce PP for some

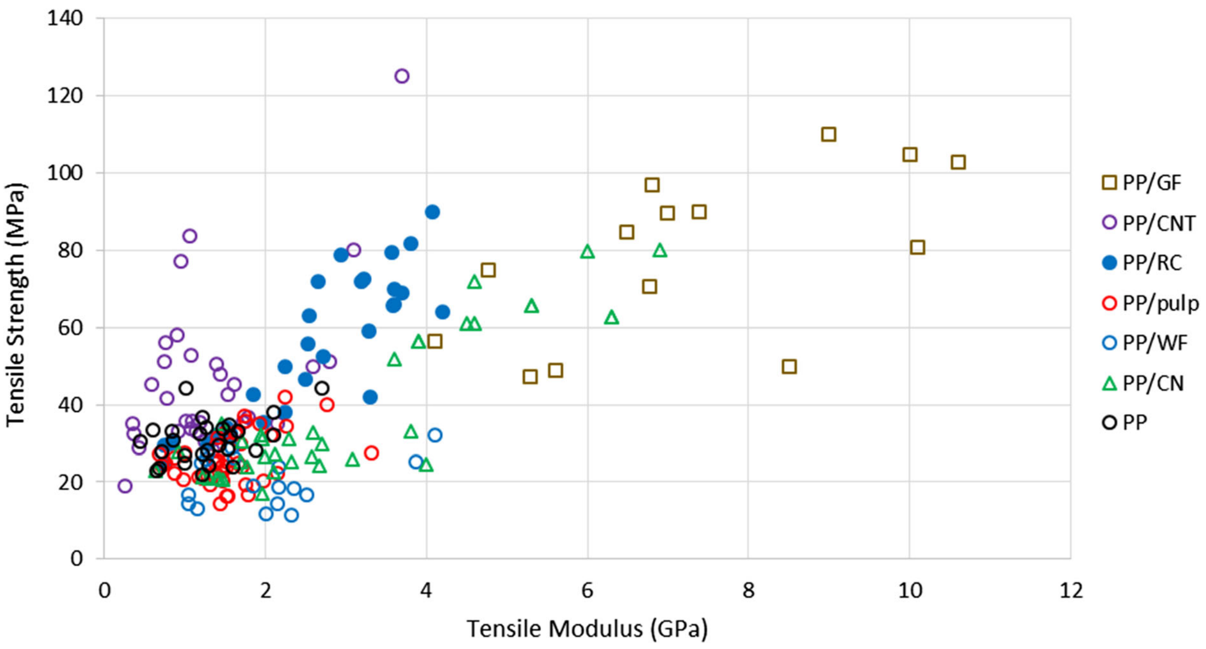


Fig. 14 Fiber-reinforced polypropylene (PP) composites. GF-glass fiber, CNT-carbon nanotubes, RC-regenerated cellulose fiber, WF-wood flour, CN-cellulose nanomaterials (Belgacem et al. 1994; Cantero et al. 2003; Nunez et al. 2003; Qiu et al. 2003; Xia et al. 2004; Manchado et al. 2005; Borja et al. 2006; Ganster et al. 2006; McIntosh et al. 2006, 2007; Ganster and Fink 2006; Cheng et al. 2007; Zhao et al. 2007; Johnson et al. 2008; Masuda and Torkelson 2008; Prashantha et al. 2009; Yang and Gardner 2011; Huque et al. 2012; Suzuki et al. 2013, 2014; Cao et al. 2014; Iwamoto et al. 2014b; Khoshkava and Kamal 2014; Unterweger et al. 2014; Matuana and Stark 2015; Peng et al. 2016; Franciszczak et al. 2017; Risnasari et al. 2018; Wang et al. 2018; Kim et al. 2019b; Mihalic et al. 2019)

applications, which do not require the high performance achieved with glass fibers, rather only higher than neat PP.

Cellulose-polylactic acid (PLA) composites

Figure 15 shows scattered points of tensile elastic modulus against the tensile strength of cellulose reinforced PLA composites. A one-way analysis of variance at the significance level of 0.01 indicates there are no significant differences in both elastic modulus and strength among different cellulose forms (Fig. 16). Such results indicate that cellulose is less effective in reinforcing PLA for higher tensile performance.

Cellulose-epoxy composites

As shown in Fig. 17, nanoparticles generally increase the tensile modulus of epoxy. If dispersed well, they also increase tensile strength. Agglomerates in the matrix often decrease the strength to below neat materials. Carbon nanotubes enhance mechanical properties at lower fiber volume fractions than cellulose nanoparticles.

Cellulose-high density polyethylene (HDPE) composites

The tensile modulus and strength of cellulosic particle reinforced HDPE composites are summarized in Fig. 18. Generally, fibrous cellulosic particles have better reinforcing effects than flour. Nanocellulose does not offer better tensile properties than other fibrous cellulosic particles (Hubbe and Grigsby 2020).

Discussion

Many factors affect the performance of fiber-reinforced polymer composites. Either a coupling agent or specific treatment of cellulose surface makes fibers more compatible with polymers and generally results in higher strength (Hubbe and Grigsby 2020).

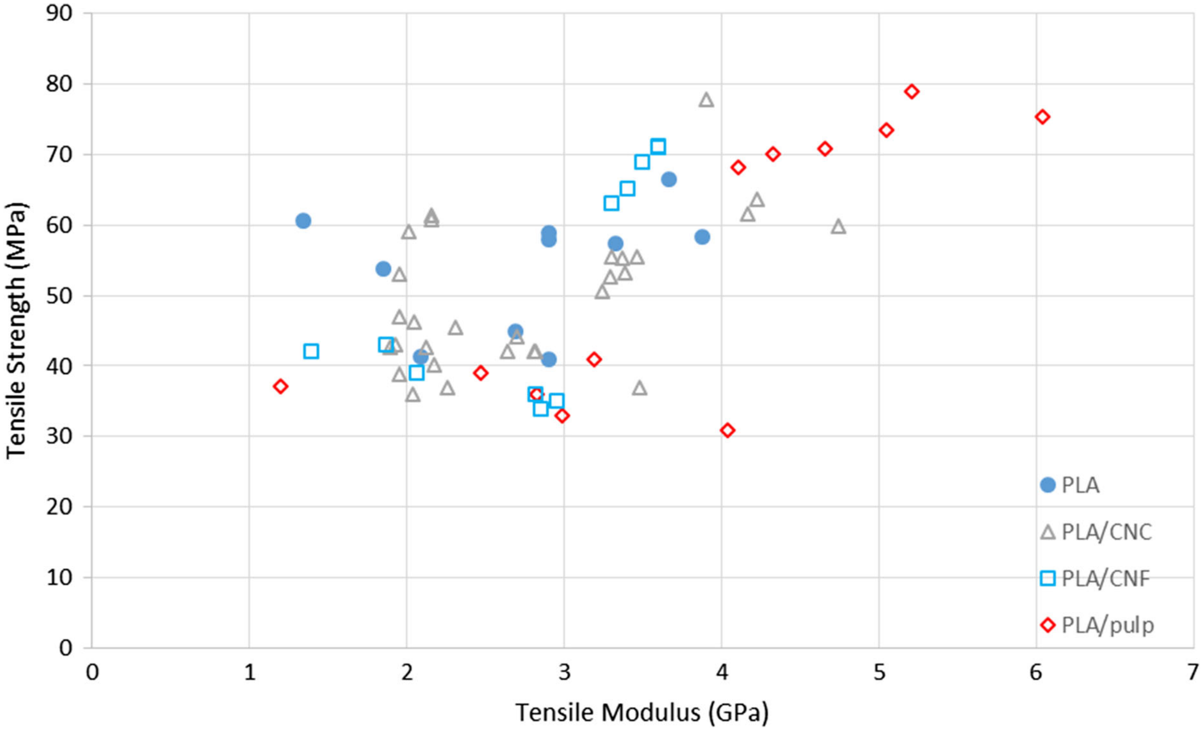


Fig. 15 Cellulose fiber reinforced polylactic acid (PLA) composites. CNC-cellulose nanocrystals, CNF-cellulose nanofibrils (Lee et al. 2009; Jonoobi et al. 2010, 2012; Tomé et al. 2011; Martínez-Sanz et al. 2012; Ambrosio-Martín et al. 2015; Robles et al. 2015; Muiruri et al. 2017; Shojaeiarani et al. 2018)

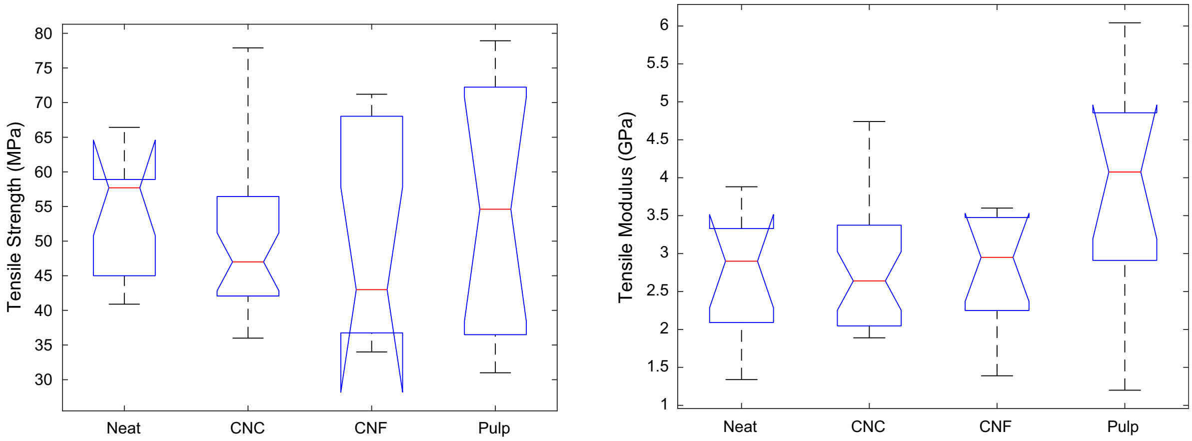


Fig. 16 Boxplot of the distribution of tensile strength and modulus of PLA composites reinforced by three cellulose forms: CNC, CNF, and pulp. Each box indicates min, lower quartile, median, upper quartile, and max as well as a notch for 95% confidence interval of the median

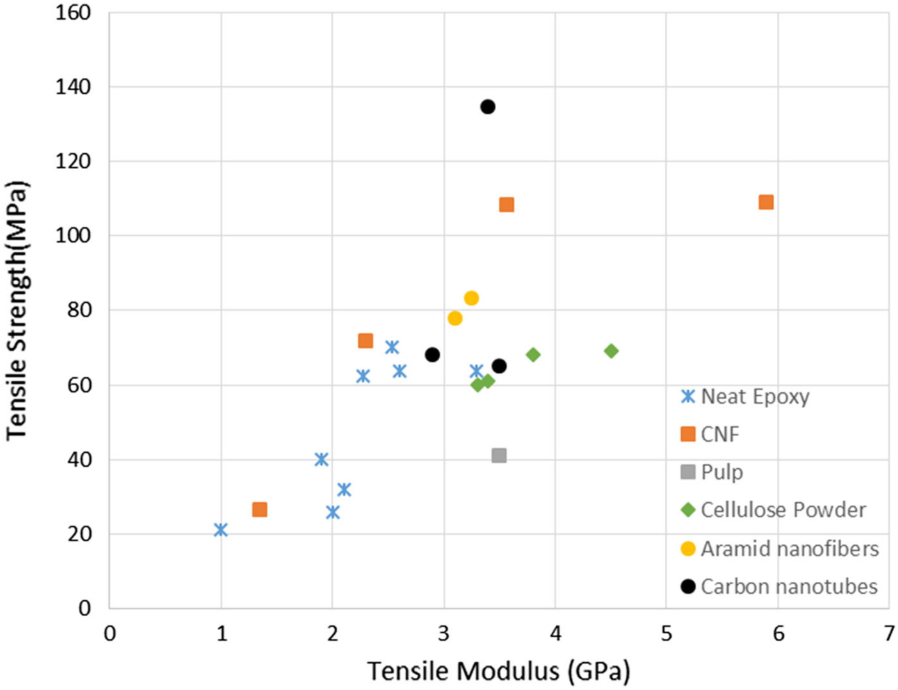


Fig. 17 Tensile modulus and strength of nanoparticle-reinforced epoxy composites (Gojny et al. 2005; Nadler et al. 2009; Ansari et al. 2014, 2015; Wang et al. 2014; Wernik and Meguid 2014; Kiziltas et al. 2016; Lin et al. 2017; Saba et al. 2017; Nair et al. 2019)

Growing and processing may introduce a lot of defects into fibers such as dislocations, slip planes, and kinks (Usov et al. 2015). Figure 19a shows that cellulosic fibers may be twisted, folded, coiled, and aggregated to further reduce the aspect ratio. The curved and kinked fibers may be difficult to orient themselves in parallel and increase out-of-plane orientation, leading to a negative influence on stiffness (Matveeva et al. 2014). In contrast, short synthetic staple fibers such as glass fibers (Fig. 19b) and rayon fibers (Fig. 19c) preserve individualized and aligned fibers, which are pulled out at tension failure and increases energy dissipation, i.e. increased toughness. Fiber

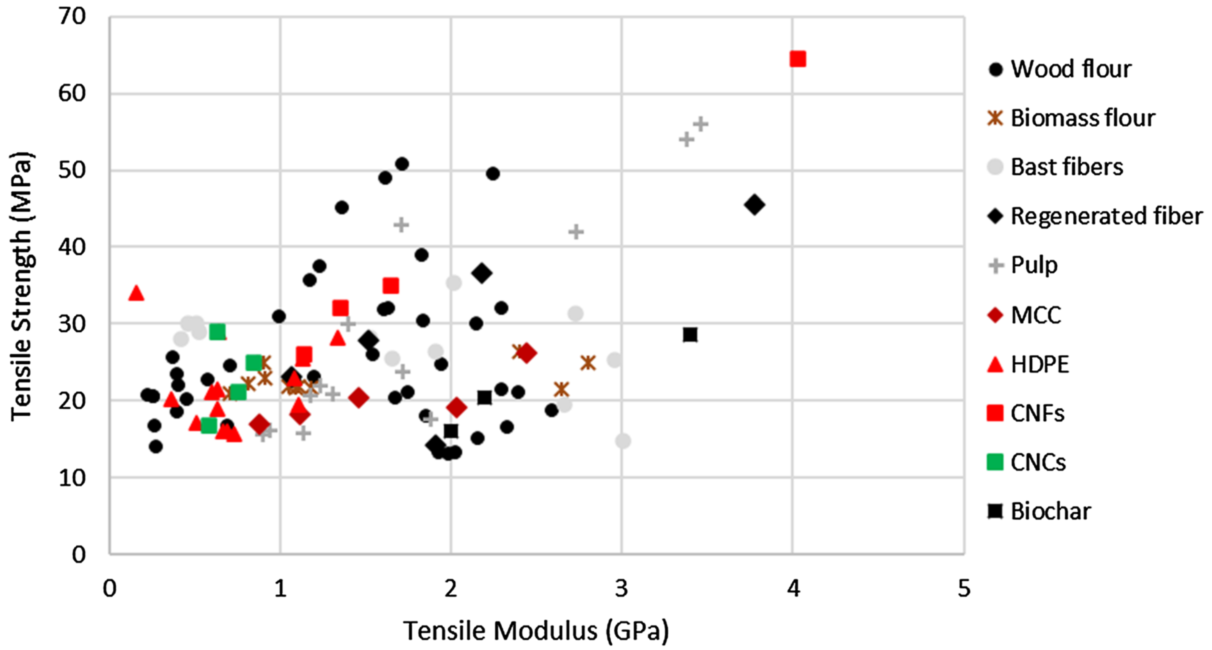


Fig. 18 Cellulose fiber-reinforced polylactic acid (PLA) composites. MCC: microcrystalline cellulose (Hubbe and Grigsby 2020)

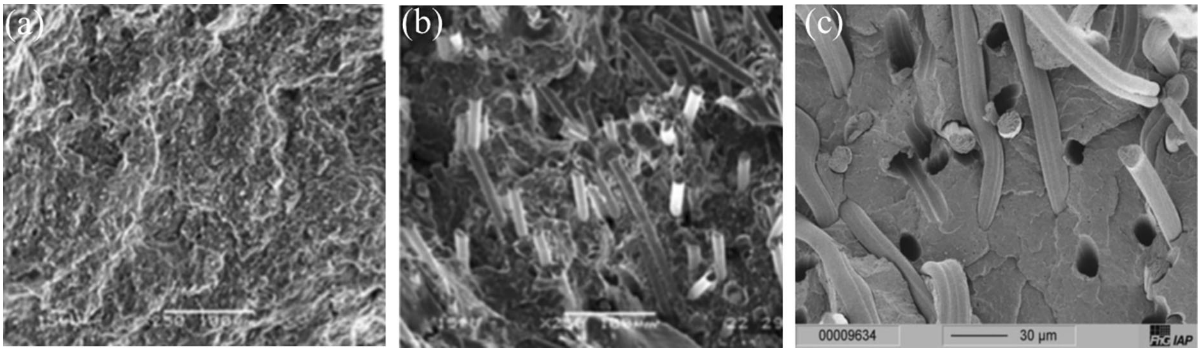


Fig. 19 SEM micrographs of fractured cross-sections: **a** cellulose filament reinforced polyethylene (Diallo et al. 2019), **b** short glass fiber reinforced polyethylene(Diallo et al. 2019), and **c** Rayon reinforced PP (Ganster et al. 2006). Reproduced from the reference with permission from John Wiley and Sons for **a** and **b**, and Elsevier for **c**

morphology and fiber/matrix affinity are the most important factors and will be discussed as follows.

Critical aspect ratio

Assuming (1) both fiber and matrix undergo elastic deformation, (2) fiber ends bear no axial loads, and (3) the presence of a fully bonded fiber-matrix interface, and (4) idealized geometry of oriented in the plane or parallel without coiling. Iso-strain assumes stress is applied parallel to the aligned continuous fillers and the modulus of fiber-reinforced polymers (E_c) correlates with fiber and matrix volume fractions (V_f, V_m) and fiber and matrix moduli (E_f, E_m) by:

$$E_c = V_f E_f + V_m E_m \tag{1}$$

For short fiber-reinforced composites, the Cox-Krenchel model predicts elastic modulus based on a classical shear-lag model, accounting for both the aspect ratio and orientation of fibers.

$$E_c = \eta_o \eta_{l/d} V_f E_f + E_m (1 - V_f) \tag{2}$$

where η_o is the orientation factor, which is directly proportional to the Hermans order parameter. The latter can be characterized by x-ray diffraction patterns, or $\eta_0 = 3/8$ by assuming an in-plane isotropic random orientation of fibers in nanocomposites. $\eta_{l/d}$ the fiber efficiency factor, a short fiber's load-bearing capability relative to the analogous continuous fiber and is mainly a function of the aspect ratio factor and can be estimated from the shear-lag model (Coleman et al. 2004):

$$\eta_{l/d} = 1 - \frac{\tanh \frac{\beta L}{2}}{\frac{\beta L}{2}} \tag{3}$$

$$where,\ \frac{\beta L}{2} = \frac{2L}{D}\sqrt{\frac{G_m}{E_f \ln \frac{\pi}{X_i V_f}}} \tag{4}$$

where L is the length and D is the diameter of fibers. G_m is the shear stiffness of the matrix, $E_m = 2G_m(1+\nu)$, ν is the Poisson's ratio; X_i is a constant controlled by the geometrical packing pattern of the fiber, which is assumed to be hexagonal packing in this case with a value of $2\sqrt{3}$ (Landel and Nielsen 1974).

Although the Cox-Krenchel model has been shown by comparison to numerical calculations to be inaccurate for some materials (Nairn 2011), it provides a capability to predict the trending effects of fiber aspect ratio and fiber volume fraction on the composite modulus. For practical purposes, 90% efficiency can be arbitrarily selected as a benchmark critical aspect ratio for an excellent short-fiber composite. Figure 20 shows that the critical aspect ratio of fiber contribution to the stiffness of composites is volume fraction dependent and decreases significantly with increasing the fiber volume fraction. High aspect ratio at high fiber volume fractions can cause a dispersion problem, leading to the formation of flaws and a decrease of mechanical properties, especially strength. Literature data have supported this prediction as discussed as follows.

At a lower volume fraction of CNTs, the modulus increases with the fiber aspect ratio for epoxy/CNTs composites (Martone et al. 2011) and polylactide/CNTs composites (Wu et al. 2010). Fiber-reinforced-rubbers gained an increase in modulus, hardness, and torque with the aspect ratio surging up to about 300, but decrease over 400. However, the fiber decreased the tensile strength at all loads. Such decreases are aggravated with high aspect ratios at high volume fractions (Lee and Ryu 1999). It was also found that 10-mm coir fiber gave better mechanical properties than 6-mm and 14-mm coir fibers (Geethamma et al. 1995). Injection molding and extrusion often shorten plant fiber length and aspect ratio, without necessarily leading to lower mechanical properties which also depends on fiber volume fraction levels (Ausias et al. 2013). An optimal range of the aspect ratio of nanofibrils larger than 100 is required to achieve higher fracture toughness of cellulose nanopaper (Meng et al. 2017).

Table 3 shows the aspect ratios of typical cellulose fibers and CNTs. CNC aspect ratio is around 15–40 for plant fibers and up to 140 from Tunicate and spinifex (Amiralian et al. 2017). Typically, stronger acid conditions, longer reaction times, and higher temperatures tend to yield shorter CNCs. TEMPO-mediated oxidation of wood pulp yields extremely thin nanofibrils with an aspect ratio ranging from 20 to 380. Mechanically refined nanofibrils without biological or chemical assistance typically appear like a network. Short staple fibers such as glass, carbon, and rayon fibers are chopped fibers from filaments, which aspect ratio can be purposely controlled. These aspect ratios are in good agreement with the literature data for the strength of fiber-reinforced PP composites. As shown in Fig. 14, the lowest aspect ratio of wood flour corresponds to its lowest tensile strength for its PP composites, followed by CNCs and pulp. As compared with pure polypropylene, these fibers with a small aspect ratio still can increase the modulus but generally decrease the strength. CNCs may have some nanosized effect offsetting their smaller aspect ratio as compared with pulp and wood flour. CNFs at high volume fractions increase both elastic modulus and strength, especially when uniformly distributed in the matrix, such as those by in-situ kneading with powdered PP (Suzuki et al. 2014). For this kind of CNFs, the aspect ratio may not be an effective parameter since they are a network at high volume fractions. Short staple fibers such as chopped glass, rayon, and flax short fibers substantially increase the strength and modulus, and so do flax mats and yarns. But flax pulp does not reinforce the composites. CNTs have a relatively high aspect ratio and increase the strength of composites but not modulus. This may be related to their small diameter. Nairn (Nairn 2011) applied shear-lag approaches to evaluate the aspect ratio requirements for CNTs in a composite and

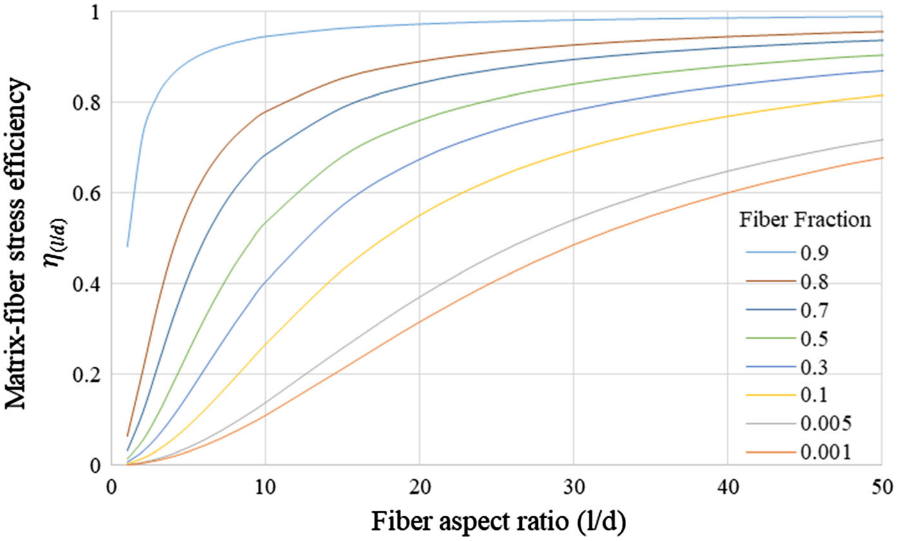


Fig. 20 The Cox-Krenchel model gives the fiber efficiency factor as a function of the volume fraction (as shown by the legend) and aspect ratio (x-axis) of the fiber. As the aspect ratio increases, the factor approaches 1. The graphs were calculated by assuming the axial modulus of cellulose as 86 GPa and the matrix (polypropylene) as 1.6 GPa in modulus and 0.43 in Poisson's ratio

Table 3 Cellulosic fiber dimensional characteristics

Fiber	Diameter (μm)	Length (μm)	Aspect ratio
CNCs	0.005–0.01	0.3–0.5	15–40
TEMPO-CNFs(Isogai 2017)	0.003–0.014	0.138–1.1	20–380
CNFs(Iwamoto et al. 2014a)	0.004–0.02	0.4–2	140–500
Short staple fibers(Martone et al. 2011)	8–18	10,000–40,000	1000–2000
CNTs	0.005–0.07	0.1–10	10–1000
Wood pulp(Migneault et al. 2014)	15–30	1000–5000	50–200
Wood flour(Khonsari et al. 2015)	250–800	1000–3000	2–6

contrasted it to micro-sized fibers. He pointed out that if the interface among matrix and the nanofibers is not perfect, those fibers will not provide actual reinforcement. In other words, small-diameter fibers put more burden on the interface than large-diameter fibers, that is to say, conventional micro-sized fibers may have robust reinforcing effects on most polymers.

Fiber/matrix interfacial shear strength and critical length

Interfacial shear strength and critical length are measured with a single fiber pullout test or fragmentation test. Raman micro-spectroscopy can examine stress in the fiber under tension and shear stress distribution along with the interface of fiber/polymer (Tze et al. 2007). Under the single-fiber pullout test, the embedded length for which the fiber fails is known as critical length (Lc). Lc correlates with the average interfacial shear strength (τ_c) of fiber/matrix and fiber tensile strength σ_f by the Eq. (5) (Barber et al. 2006)

$$\frac{L_c}{D} = \frac{\sigma_f}{4\tau_c} \tag{5}$$

Equation (5) shows the critical length or aspect ratio is inverse proportional to interfacial shear strength. Increasing interfacial shear strength would decrease the requirements on aspect ratio. The interfacial shear strength with polypropylene is 5.7 MPa for glass fibers, 4.8 MPa for ramie fibers, and 4.1 MPa for lyocell fiber (Adusumali et al. 2006). Chemically modified fibers increase shear strength and reduce critical length. Acetylation and heat treatment of cellulosic fibers were found to increase their interfacial shear strength with polystyrene from 3.06 to 10.0 MPa (Liu et al. 1994). CNTs typically have a larger shear strength than glass and cellulosic fibers and thus require a lower critical length (Table 4). Fine fibers also require a higher aspect ratio but lower critical length (Table 4). Coupling agents also decrease critical length and aspect ratio (Table 4). Short plant CNCs were found less efficient in stress transfer than long tunicate CNCs (Rusli and Eichhorn 2008). Most CNCs aspect ratios range from 10 to 50 and may be too small to be sufficient reinforcement (Shrestha et al. 2018). Cellulose nanofibrils are around 100 aspect ratio and close to the requirements (Lee et al. 2014). Improving the adhesion between the fibers and matrix will decrease aspect ratio requirements. The rough surface may increase interfacial shear strength. Low critical aspect ratio means fibers of large diameters can be used, which may ease the strict processing conditions for achieving smaller fibers. The aspect ratio affects strength more than the modulus. The initial modulus of a composite is determined primarily by the elastic properties of the constituents and is less affected by fiber-matrix adhesion. This is why natural fibers often stiffen but weaken polymers.

Future of cellulose

Cellulose can be extracted from various resources and then reassembled into diverse neat bulk materials such as hydrogels, aerogels, membranes, films, and fibers as well as combined with other materials as composites. Naturally occurring wood is a composite of cellulose with lignin and hemicellulose. Cellulose will be used largely as a constituent of wood for shelters, furniture, paper, and containerboard. Emerging applications of cellulose to compete with plastics for primary packaging, textiles and other value-added materials face the challenges of the variability of properties, long-term durability, and high production costs. Cellulose microfibrils in plant cell walls have invoked great interest as excellent building blocks to produce new materials. Translating spectacular properties of individual cellulose nanomaterials into high-performance macroscale materials for various engineering applications are currently under intensive investigation. The results thus far do not point to competitive advantages of cellulose nanomaterials as real structural reinforcements at low fiber fractions when compared to bast or rayon staple cellulose fibers, in a similar way as CNTs inferior to carbon fibers as reinforcements (Kinloch et al. 2018). As the Cox-Krenchel model predicts that such short nanoscale fibers would have been advantageous at high fraction volumes under which though dispersion issues emerge. It might be an advantage to secure a niche market for cellulose nanomaterials whose modifying capabilities are in viscoelasticity, mass permeability, and interfacial compatibility to other materials used in cosmetics, food, pharmaceutics, concretes, and coatings applications which this review has not included. The answers would have been from trial and error–based attempts to control dispersion, fiber volume fraction, and interfacial engineering of fiber/matrix. Regenerated cellulose or cellulose

Table 4 Interfacial shear strength, critical length, and aspect ratio of fibers in polymers

Interface	Shear strength (MPa)	Critical Length (μm)	Critical aspect ratio
CNT/Epoxy(Chen et al. 2015)	130	0.21	100
CNT/PMMA(Chen et al. 2015)	45	0.35	170
Glass fiber/PP(Etcheverry and Barbosa 2012)	3.5–7.4	4100–8430	150–300
Coir fiber/epoxy(Luz et al. 2018)	0.71	12,400	40
Pineapple leave fiber/epoxy(Luz et al. 2018)	2.46	7300	30
Flax individual fiber /PP(Van den Oever and Bos 1998)		980	59
Flax individual fiber/MAPP/PP(Van den Oever and Bos 1998)		520	31
Flax fiber bundle/PP(Van den Oever and Bos 1998)		3800	36
Flax fiber bundle /MAPP/PP(Van den Oever and Bos 1998)		2800	26

nanomaterials along with their composites can be used to produce continuous fibers for textiles, dense films for packaging, porous membranes for filters, hydrogels, or aerogels for absorption or insulation applications. There is a concerted effort needed for these value-added cellulose-based products to substitute their competitive synthetic products. Comparable quality, the widespread availability of these materials in large volumes and at relatively low cost will dictate their ultimate success of applications. A few successes of commercial applications are needed to drive optimization; costs will come down as processes are perfected. With an increase in the world's population, energy consumption, and climate change, continuing research on cellulose-based products will be imperative.

Funding The research was supported by the USDA NIFA Agriculture and Food Research Initiative program under grant number of 2018-67010-27906.

References

Adusumali R, Reifferscheid M, Weber H et al (2006) Mechanical properties of regenerated cellulose fibres for composites. Wiley, Hoboken, pp 119–125

Ambrosio-Martín J, Lopez-Rubio A, Fabra MJ et al (2015) Assessment of ball milling methodology to develop polylactide-bacterial cellulose nanocrystals nanocomposites. J Appl Polym Sci 132:1–8

Amiralian N, Annamalai PK, Garvey CJ et al (2017) High aspect ratio nanocellulose from an extremophile spinifex grass by controlled acid hydrolysis. Cellulose 24:3753–3766

Ansari F, Galland S, Johansson M et al (2014) Cellulose nanofiber network for moisture stable, strong and ductile biocomposites and increased epoxy curing rate. Compos A Appl Sci Manuf 63:35–44

Ansari F, Sjöstedt A, Larsson PT et al (2015) Hierarchical wood cellulose fiber/epoxy biocomposites–materials design of fiber porosity and nanostructure. Compos A Appl Sci Manuf 74:60–68

Ansell M, Mwaikambo L (2009) The structure of cotton and other plant fibres. In: handbook of textile fibre structure. Elsevier pp 62–94

Arbelaiz A, Fernandez B, Cantero G et al (2005a) Mechanical properties of flax fibre/polypropylene composites. Influence of fibre/matrix modification and glass fibre hybridization. Compos A Appl Sci Manuf 36:1637–1644

Arbelaiz A, Fernandez B, Ramos J et al (2005b) Mechanical properties of short flax fibre bundle/polypropylene composites: influence of matrix/fibre modification, fibre content, water uptake and recycling. Compos Sci Technol 65:1582–1592

Ashori A, Menbari S, Bahrami R (2016) Mechanical and thermo-mechanical properties of short carbon fiber reinforced polypropylene composites using exfoliated graphene nanoplatelets coating. J Ind Eng Chem 38:37–42

Ausias G, Bourmaud A, Coroller G, Baley C (2013) Study of the fibre morphology stability in polypropylene-flax composites. Polym Degrad Stab 98:1216–1224

Bachmann J, Wiedemann M, Wierach P (2018) Flexural mechanical properties of hybrid epoxy composites reinforced with nonwoven made of flax fibres and recycled carbon fibres. Aerospace 5:107

Baez C, Considine J, Rowlands R (2014) Influence of drying restraint on physical and mechanical properties of nanofibrillated cellulose films. Cellulose 21:347–356

Bar M, Alagirusamy R, Das A (2018) Properties of flax-polypropylene composites made through hybrid yarn and film stacking methods. Compos Struct 197:63–71

Bar M, Das A, Alagirusamy R (2019) Influence of flax/polypropylene distribution in twistless thermally bonded rovings on their composite properties. Polym Compos 40:4300–4310

Barber AH, Cohen SR, Eitan A et al (2006) Fracture transitions at a carbon-nanotube/polymer interface. Adv Mater 18:83–87

Behabtu N, Young CC, Tsentalovich DE et al (2013) Strong, light, multifunctional fibers of carbon nanotubes with ultrahigh conductivity. Science 339:182–186

Belgacem M, Bataille P, Sapieha S (1994) Effect of corona modification on the mechanical properties of polypropylene/cellulose composites. J Appl Polym Sci 53:379–385

Bhat N, Makwana D (1988) Effects of swelling treatments on fine structure and mechanical properties of cellophane film. Text Res J 58:233–238

Bhattacharyya S, Salvetat J-P, Saboungi M-L (2006) Reinforcement of semicrystalline polymers with collagen-modified single walled carbon nanotubes. Appl Phys Lett 88:233119

Borja Y, Rieß G, Lederer K (2006) Synthesis and characterization of polypropylene reinforced with cellulose I and II fibers. J Appl Polym Sci 101:364–369

Bourmaud A, Beaugrand J, Shah DU et al (2018) Towards the design of high-performance plant fibre composites. Prog Mater Sci 97:347–408

Brosius D (2016) In defense of glass fiber. https://www.compositesworld.com/articles/in-defense-of-glass-fiber

Bunsell AR (2009) Handbook of tensile properties of textile and technical fibres. Elsevier

Cantero G, Arbelaiz A, Llano-Ponte R, Mondragon I (2003) Effects of fibre treatment on wettability and mechanical behaviour of flax/polypropylene composites. Compos Sci Technol 63:1247–1254

Cao L, Su D, Su Z, Chen X (2014) Fabrication of multiwalled carbon nanotube/polypropylene conductive fibrous membranes by melt electrospinning. Ind Eng Chem Res 53:2308–2317

Cazón P, Vázquez M, Velazquez G (2018) Novel composite films based on cellulose reinforced with chitosan and polyvinyl alcohol: Effect on mechanical properties and water vapour permeability. Polym Test 69:536–544

Chen C, Bu X, Feng Q, Li D (2018) Cellulose nanofiber/carbon nanotube conductive nano-network as a reinforcement template for polydimethylsiloxane nanocomposite. Polymers 10:1000

Chen W, Tao X, Xue P, Cheng X (2005) Enhanced mechanical properties and morphological characterizations of poly (vinyl alcohol)–carbon nanotube composite films. Appl Surf Sci 252:1404–1409

Chen X, Zhang L, Zheng M et al (2015) Quantitative nanomechanical characterization of the van der Waals interfaces between carbon nanotubes and epoxy. Carbon 82:214–228. https://doi.org/10.1016/j.carbon.2014.10.065

Cheng Q, Wang S, Rials TG, Lee S-H (2007) Physical and mechanical properties of polyvinyl alcohol and polypropylene composite materials reinforced with fibril aggregates isolated from regenerated cellulose fibers. Cellulose 14:593–602

Cicala G, Cristaldi G, Recca G et al (2009) Properties and performances of various hybrid glass/natural fibre composites for curved pipes. Mater Des 30:2538–2542

Claramunt J, Ventura H, Fernández-Carrasco LJ, Ardanuy M (2017) Tensile and flexural properties of cement composites reinforced with flax nonwoven fabrics. Materials 10:215

Clemons C (2016) Nanocellulose in spun continuous fibers: a review and future outlook. J Renew Mater 4:327–339

Coleman JN, Cadek M, Blake R et al (2004) High performance nanotube-reinforced plastics: understanding the mechanism of strength increase. Adv Func Mater 14:791–798. https://doi.org/10.1002/adfm.200305200

Cunniff PM, Fossey SA, Auerbach MA et al (1994) Mechanical and thermal properties of dragline silk from the spider Nephila clavipes. Polym Adv Technol 5:401–410

da Luz FS, Ramos FJHTV, Nascimento LFC et al (2018) Critical length and interfacial strength of PALF and coir fiber incorporated in epoxy resin matrix. J Mater Res Technol 7:528–534. https://doi.org/10.1016/j.jmrt.2018.04.025

Dalton AB, Collins S, Munoz E et al (2003) Super-tough carbon-nanotube fibres. Nature 423:703–703

Dasgupta S, Hammond WB, Goddard WA (1996) Crystal structures and properties of nylon polymers from theory. J Am Chem Soc 118:12291–12301. https://doi.org/10.1021/ja944125d

Derbali I, Terekhina S, Guillaumat L, Ouagne P (2016) Rapid manufacturing of composite structures made of fabric flax/polypropylene. In: ECCM17—17th European conference on composite materials. Munich, Germany, pp 1–9

Di J, Hu D, Chen H et al (2012) Ultrastrong, foldable, and highly conductive carbon nanotube film. ACS Nano 6:5457–5464

Diallo AK, Jahier C, Drolet R et al (2019) Cellulose filaments reinforced low-density polyethylene. Polym Compos 40:16–23

Dong Z, Ding Z, Zhang S et al (2017) Natural fibres from the bark of mulberry branches for textile application. Fibres Text Eastern Europe 25:20–25. https://doi.org/10.5604/10.5604/01.3001.0010.1683

Du N, Yang Z, Liu XY et al (2011) Structural origin of the strain-hardening of spider silk. Adv Func Mater 21:772–778

Du S, Li J, Zhang J, Wang X (2015) Microstructure and mechanical properties of silk from different components of the Antheraea pernyi cocoon. Mater Des 1980–2015(65):766–771

Dufresne A (2017) Cellulose nanomaterial reinforced polymer nanocomposites. Curr Opin Coll Interf Sci 29:1–8

Eichhorn SJ, Davies GR (2006) Modelling the crystalline deformation of native and regenerated cellulose. Cellulose 13:291–307. https://doi.org/10.1007/s10570-006-9046-3

Eichhorn SJ, Young RJ, Davies GR (2005) Modeling crystal and molecular deformation in regenerated cellulose fibers. Biomacromol 6:507–513. https://doi.org/10.1021/bm049409x

EPA (2019) Advancing sustainable materials management: 2017 fact sheet assessing trends in material generation, recycling, composting, combustion with energy recovery and landfilling in the United States. United State Environmental Protection Agency (EPA), Washington, DC

EPA (2020) containers and packaging: product-specific data. In: facts and figures about materials, waste and recycling. https://www.epa.gov/facts-and-figures-about-materials-waste-and-recycling/containers-and-packaging-product-specific-data. Accessed 10 Mar 2020

Etcheverry M, Barbosa SE (2012) Glass fiber reinforced polypropylene mechanical properties enhancement by adhesion improvement. Materials 5:1084–1113

Everitt NM, Aboulkhair NT, Clifford MJ (2013) Looking for links between natural fibres' structures and their physical properties. UK, Hindawi

Fahma F, Hori N, Iwata T, Takemura A (2017) PVA nanocomposites reinforced with cellulose nanofibers from oil palm empty fruit bunches (OPEFBs). Emirates J Food Agric 29(5):323–329

FAO (2017) FAO yearbook of forest products 2017. Food And Agriculture Organization Of The United Nations, Rome

Franciszczak P, Merijs-Meri R, Kalniņš K et al (2017) Short-fibre hybrid polypropylene composites reinforced with PET and rayon fibres–effects of SSP and interphase tailoring. Compos Struct 181:121–137

Fu Q, Medina L, Li Y et al (2017) Nanostructured wood hybrids for fire-retardancy prepared by clay impregnation into the cell wall. ACS Appl Mater Interf 9:36154–36163

Ganster J, Fink H-P (2006) Novel cellulose fibre reinforced thermoplastic materials. Cellulose 13:271–280. https://doi.org/10.1007/s10570-005-9045-9

Ganster J, Fink H-P, Pinnow M (2006) High-tenacity man-made cellulose fibre reinforced thermoplastics–injection moulding compounds with polypropylene and alternative matrices. Compos A Appl Sci Manuf 37:1796–1804

Gavillon R, Budtova T (2008) Aerocellulose: new highly porous cellulose prepared from cellulose− NaOH aqueous solutions. Biomacromol 9:269–277

Geethamma V, Joseph R, Thomas S (1995) Short coir fiber-reinforced natural rubber composites: effects of fiber length, orientation, and alkali treatment. J Appl Polym Sci 55:583–594

Geyer R, Jambeck JR, Law KL (2017) Production, use, and fate of all plastics ever made. Sci Adv 3:e1700782

Gibson LJ (2012) The hierarchical structure and mechanics of plant materials. J R Soc Interf 9:2749–2766

Gindl W, Keckes J (2006) Strain hardening in regenerated cellulose fibres. Compos Sci Technol 66:2049–2053

Gindl W, Keckes J (2007) Drawing of self-reinforced cellulose films. J Appl Polym Sci 103:2703–2708

Gindl W, Reifferscheid M, Adusumalli R-B et al (2008) Anisotropy of the modulus of elasticity in regenerated cellulose fibres related to molecular orientation. Polymer 49:792–799

Gojny FH, Wichmann MH, Fiedler B, Schulte K (2005) Influence of different carbon nanotubes on the mechanical properties of epoxy matrix composites–a comparative study. Compos Sci Technol 65:2300–2313

González I, Alcalà M, Chinga-Carrasco G et al (2014) From paper to nanopaper: evolution of mechanical and physical properties. Cellulose 21:2599–2609

Gonzalez JS, Ludueña LN, Ponce A, Alvarez VA (2014) Poly (vinyl alcohol)/cellulose nanowhiskers nanocomposite hydrogels for potential wound dressings. Mater Sci Eng, C 34:54–61

Gosline J, Guerette P, Ortlepp C, Savage K (1999) The mechanical design of spider silks: from fibroin sequence to mechanical function. J Exp Biol 202:3295–3303

Hamedi MM, Hajian A, Fall AB et al (2014) Highly conducting, strong nanocomposites based on nanocellulose-assisted aqueous dispersions of single-wall carbon nanotubes. ACS Nano 8:2467–2476

Han JS (1998) Properties of nonwood fibers. The Korean Society of Science and Technology Seoul, Korea, pp 3–12

Heijenrath R, Peijs T (1996) Natural-fibre-mat-reinforced thermoplastic composites based on flax fibres and polypropylene. Advanced Compos Lett 5:096369359600500303

Henriksson M, Berglund LA, Isaksson P et al (2008) Cellulose nanopaper structures of high toughness. Biomacromol 9:1579–1585

Huan S, Bai L, Cheng W, Han G (2016) Manufacture of electrospun all-aqueous poly (vinyl alcohol)/cellulose nanocrystal composite nanofibrous mats with enhanced properties through controlling fibers arrangement and microstructure. Polymer 92:25–35

Hubbe MA, Grigsby W (2020) From nanocellulose to wood particles: a review of particle size vs. the properties of plastic composites reinforced with cellulose-based entities. BioResources 15:2030–2081

Huque QM, Islam R, Islam MM et al (2012) Preparation of rayon fiber-reinforced polypropylene composites by extrusion techniques. Polym Plast Technol Eng 51:116–121

Inai R, Kotaki M, Ramakrishna S (2005) Structure and properties of electrospun PLLA single nanofibres. Nanotechnology 16:208–213. https://doi.org/10.1088/0957-4484/16/2/005

Isogai A (2017) Determination of length and width of nanocelluloses from their dilute dispersions. In: Advances in Pulp and Paper Research, Oxford 2017, Trans. of the XVIth Fund. Res. Symp. Oxford, 2017, (W. Batchelor and D. Söderberg, eds), pp 801–811, FRC, Manchester, 2018

Iwamoto S, Lee S-H, Endo T (2014a) Relationship between aspect ratio and suspension viscosity of wood cellulose nanofibers. Polym J 46:73

Iwamoto S, Yamamoto S, Lee S-H, Endo T (2014b) Solid-state shear pulverization as effective treatment for dispersing lignocellulose nanofibers in polypropylene composites. Cellulose 21:1573–1580

Jalal Uddin A, Araki J, Gotoh Y (2011) Toward "strong" green nanocomposites: polyvinyl alcohol reinforced with extremely oriented cellulose whiskers. Biomacromol 12:617–624

Jitjaicham M, Kusuktham B (2016) Preparation of paper mulberry fibers and possibility of cotton/paper mulberry yarns production. Indian J Mater Sci 2016:1498967. https://doi.org/10.1155/2016/1498967

Johnson RK, Zink-Sharp A, Renneckar SH, Glasser WG (2008) Mechanical properties of wetlaid lyocell and hybrid fiber-reinforced composites with polypropylene. Compos A Appl Sci Manuf 39:470–477

Jonoobi M, Harun J, Mathew AP, Oksman K (2010) Mechanical properties of cellulose nanofiber (CNF) reinforced polylactic acid (PLA) prepared by twin screw extrusion. Compos Sci Technol 70:1742–1747. https://doi.org/10.1016/j.compscitech.2010.07.005

Jonoobi M, Mathew AP, Abdi MM et al (2012) A comparison of modified and unmodified cellulose nanofiber reinforced polylactic acid (PLA) prepared by twin screw extrusion. J Polym Environ 20:991–997

Kannan TG, Wu CM, Cheng KB, Wang CY (2013) Effect of reinforcement on the mechanical and thermal properties of flax/polypropylene interwoven fabric composites. J Ind Text 42:417–433

Khonsari A, Taghiyari HR, Karimi A, Tajvidi M (2015) Study on the effects of wood flour geometry on physical and mechanical properties of wood-plastic composites. Maderas Ciencia y tecnología 17:545–558

Khoshkava V, Kamal M (2014) Effect of drying conditions on cellulose nanocrystal (CNC) agglomerate porosity and dispersibility in polymer nanocomposites. Powder Technol 261:288–298

Kim HC, Kim D, Lee JY et al (2019a) Effect of wet spinning and stretching to enhance mechanical properties of cellulose nanofiber filament. Int J Precis Eng Manuf Green Technol 6:567–575

Kim S-H, Kim E-S, Choi K et al (2019b) Rheological and mechanical properties of polypropylene composites containing microfibrillated cellulose (MFC) with improved compatibility through surface silylation. Cellulose 26:1085–1097

Kinloch IA, Suhr J, Lou J et al (2018) Composites with carbon nanotubes and graphene: an outlook. Science 362:547–553

Kiziltas EE, Kiziltas A, Gardner DJ (2016) Rheological and mechanical properties of ultra-fine cellulose-filled thermoplastic epoxy composites. BioResources 11:4770–4780

Koga H, Saito T, Kitaoka T et al (2013) Transparent, conductive, and printable composites consisting of TEMPO-oxidized nanocellulose and carbon nanotube. Biomacromol 14:1160–1165

Kreze T, Malej S (2003) Structural characteristics of new and conventional regenerated cellulosic fibers. Text Res J 73:675–684. https://doi.org/10.1177/004051750307300804

Kunchi C, Venkateshan KC, Adusumalli RB (2018) Effect of scalp position on tensile properties of single hair fibers. Int J Trichol 10:218

Kunugi T, Kawasumi T, Ito T (1990) Preparation of ultra-high modulus polyvinyl alcohol fibers by the zone-drawing method. J Appl Polym Sci 40:2101–2112

Landel RF, Nielsen LE (1974) Mechanical properties of polymers and composites. In: Mechanical properties of polymers and composites, 2nd edn. Marcel Dekker, New York, p 380

Larsson PT, Lindström T, Carlsson LA, Fellers C (2018) Fiber length and bonding effects on tensile strength and toughness of kraft paper. J Mater Sci 53:3006–3015

Lee C, Wei X, Kysar JW, Hone J (2008) Measurement of the elastic properties and intrinsic strength of monolayer graphene. Science 321:385–388

Lee D, Ryu S (1999) The influence of fiber aspect ratio on the tensile and tear properties of short-fiber reinforced rubber. ICCM12, Paris

Lee K-Y, Aitomäki Y, Berglund LA et al (2014) On the use of nanocellulose as reinforcement in polymer matrix composites. Compos Sci Technol 105:15–27. https://doi.org/10.1016/j.compscitech.2014.08.032

Lee K-Y, Blaker JJ, Bismarck A (2009) Surface functionalisation of bacterial cellulose as the route to produce green polylactide nanocomposites with improved properties. Compos Sci Technol 69:2724–2733

Lee WJ, Clancy AJ, Kontturi E et al (2016) Strong and stiff: high-performance cellulose nanocrystal/poly (vinyl alcohol) composite fibers. ACS Appl Mater Interf 8:31500–31504

Lin J, Bang SH, Malakooti MH, Sodano HA (2017) Isolation of aramid nanofibers for high strength and toughness polymer nanocomposites. ACS Appl Mater Interf 9:11167–11175

Liu D, Sun X, Tian H et al (2013) Effects of cellulose nanofibrils on the structure and properties on PVA nanocomposites. Cellulose 20:2981–2989

Liu FP, Wolcott MP, Gardner DJ, Rials TG (1994) Characterization of the interface between cellulosic fibers and a thermoplastic matrix. Compos Interf 2:419–432

Liu J, Gong W, Yao Y et al (2018) Strengthening carbon nanotube fibers with semi-crystallized polyvinyl alcohol and hot-stretching. Compos Sci Technol 164:290–295

Liu L, Barber AH, Nuriel S, Wagner HD (2005) Mechanical properties of functionalized single-walled carbon-nanotube/poly (vinyl alcohol) nanocomposites. Adv Func Mater 15:975–980

Lossada F, Jiao D, Guo J et al (2019) Outstanding synergies in mechanical properties of bioinspired cellulose nanofibril nanocomposites using self-cross-linking polyurethanes. ACS Appl Polym Materi 1:3334–3342

Lu L, Hou W, Sun J et al (2014) Preparation of poly (vinyl alcohol) fibers strengthened using multiwalled carbon nanotubes functionalized with tea polyphenols. J Mater Sci 49:3322–3330

Lundahl MJ, Klar V, Ajdary R et al (2018) Absorbent filaments from cellulose nanofibril hydrogels through continuous coaxial wet spinning. ACS Appl Mater Interf 10:27287–27296

Madsen B, Lilholt H (2003) Physical and mechanical properties of unidirectional plant fibre composites—an evaluation of the influence of porosity. Compos Sci Technol 63:1265–1272

Mahjoub R, Yatim JM, Sam ARM, Hashemi SH (2014) Tensile properties of kenaf fiber due to various conditions of chemical fiber surface modifications. Constr Build Mater 55:103–113

Manchado ML, Valentini L, Biagiotti J, Kenny J (2005) Thermal and mechanical properties of single-walled carbon nanotubes–polypropylene composites prepared by melt processing. Carbon 43:1499–1505

Mander L, Liu H (2010) Comprehensive natural products II: chemistry and biology. Elsevier

Martínez-Sanz M, Lopez-Rubio A, Lagaron JM (2012) Optimization of the dispersion of unmodified bacterial cellulose nanowhiskers into polylactide via melt compounding to significantly enhance barrier and mechanical properties. Biomacromol 13:3887–3899

Martone A, Faiella G, Antonucci V et al (2011) The effect of the aspect ratio of carbon nanotubes on their effective reinforcement modulus in an epoxy matrix. Compos Sci Technol 71:1117–1123

Masuda J, Torkelson JM (2008) Dispersion and major property enhancements in polymer/multiwall carbon nanotube nanocomposites via solid-state shear pulverization followed by melt mixing. Macromolecules 41:5974–5977

Matuana L, Stark N (2015) The use of wood fibers as reinforcements in composites. In: Biofiber reinforcements in composite materials. Elsevier, pp 648–688

Matveeva AY, Pyrlin SV, Ramos MM et al (2014) Influence of waviness and curliness of fibres on mechanical properties of composites. Comput Mater Sci 87:1–11

McIntosh D, Khabashesku VN, Barrera EV (2007) Benzoyl peroxide initiated in situ functionalization, processing, and mechanical properties of single-walled carbon nanotube−polypropylene composite fibers. J Phys Chem C 111:1592–1600

McIntosh D, Khabashesku VN, Barrera EV (2006) Nanocomposite fiber systems processed from fluorinated single-walled carbon nanotubes and a polypropylene matrix. Chem Mater 18:4561–4569

Meng Q, Li B, Li T, Feng X-Q (2017) A multiscale crack-bridging model of cellulose nanopaper. J Mech Phys Solids 103:22–39

Mercader C, Denis-Lutard V, Jestin S et al (2012) Scalable process for the spinning of PVA–carbon nanotube composite fibers. J Appl Polym Sci 125:E191–E196

Mercer BS (2016) Molecular dynamics modeling of PPTA crystals in aramid fibers. Lawrence Livermore National Lab.(LLNL), Livermore, CA (United States)

Mertaniemi H, Escobedo-Lucea C, Sanz-Garcia A et al (2016) Human stem cell decorated nanocellulose threads for biomedical applications. Biomaterials 82:208–220

Miao M, Shan M (2011) Highly aligned flax/polypropylene nonwoven preforms for thermoplastic composites. Compos Sci Technol 71:1713–1718

Migneault S, Koubaa A, Perré P (2014) Effect of fiber origin, proportion, and chemical composition on the mechanical and physical properties of wood-plastic composites. J Wood Chem Technol 34:241–261

Mihalic M, Sobczak L, Pretschuh C, Unterweger C (2019) Increasing the impact toughness of cellulose fiber reinforced polypropylene composites—influence of different impact modifiers and production scales. J Compos Sci 3:82

Mittal N, Ansari F, Gowda.V K, et al (2018) Multiscale control of nanocellulose assembly: transferring remarkable nanoscale fibril mechanics to macroscale fibers. ACS Nano 12:6378–6388. https://doi.org/10.1021/acsnano.8b01084

Mohammadi P, Toivonen MS, Ikkala O et al (2017) Aligning cellulose nanofibril dispersions for tougher fibers. Sci Rep 7:1–10

Mokshina N, Chernova T, Galinousky D et al (2018) Key stages of fiber development as determinants of bast fiber yield and quality. Fibers 6:20

Molnár G, Rodney D, Martoïa F et al (2018) Cellulose crystals plastify by localized shear. Proc Natl Acad Sci 115:7260–7265. https://doi.org/10.1073/pnas.1800098115

Moon RJ, Martini A, Nairn J et al (2011) Cellulose nanomaterials review: structure, properties and nanocomposites. Chem Soc Rev 40:3941. https://doi.org/10.1039/c0cs00108b

Motamedian HR, Halilovic AE, Kulachenko A (2019) Mechanisms of strength and stiffness improvement of paper after PFI refining with a focus on the effect of fines. Cellulose 26:4099–4124

Muiruri JK, Liu S, Teo WS et al (2017) Highly biodegradable and tough polylactic acid–cellulose nanocrystal composite. ACS Sustain Chem Eng 5:3929–3937

Nadler M, Werner J, Mahrholz T et al (2009) Effect of CNT surface functionalisation on the mechanical properties of multi-walled carbon nanotube/epoxy-composites. Compos A Appl Sci Manuf 40:932–937

Nair SS, Dartiailh C, Levin DB, Yan N (2019) Highly toughened and transparent biobased epoxy composites reinforced with cellulose nanofibrils. Polymers 11:612

Nairn J (2011) Aspect ratio requirements for nanotube-reinforced, polymer–matrix composites. Compos A Appl Sci Manuf 42:1850–1855

Nakamae K, Nishino T, Gotoh Y (1995) Temperature dependence of the elastic modulus of the crystalline regions of poly (ethylene 2, 6-naphthalate). Polymer 36:1401–1405

Napper IE, Thompson RC (2016) Release of synthetic microplastic plastic fibres from domestic washing machines: Effects of fabric type and washing conditions. Mar Pollut Bull 112:39–45

Nechyporchuk O, Håkansson KM, Gowda VK et al (2019) Continuous assembly of cellulose nanofibrils and nanocrystals into strong macrofibers through microfluidic spinning. Adv Materi Technol 4:1800557

Neufeld L, Stassen F, Sheppard R, Gilman T (2016) The new plastics economy: rethinking the future of plastics. Ellen MacArthur Foundation, Cowes, United Kingdom, Report, pp 1–120. https://www.ellenmacarthurfoundation.org/assets/downloads/EllenMacArthurFoundation_TheNewPlasticsEconomy_Pages.pdf

Nishiyama Y, Kim U-J, Kim D-Y et al (2003) Periodic disorder along ramie cellulose microfibrils. Biomacromol 4:1013–1017

NOKIAN TYRES PLC (2015) REINFORCING MATERIALS IN RUBBER PRODUCTS

Northolt MG, Boerstoel H, Maatman H et al (2001) The structure and properties of cellulose fbres spun from an anisotropic phosphoric acid solution. Polymer 42:8249–8264

Nunez AJ, Sturm PC, Kenny JM et al (2003) Mechanical characterization of polypropylene–wood flour composites. J Appl Polym Sci 88:1420–1428

OCDE (2019) Global material resources outlook to 2060: economic drivers and environmental consequences. https://doi.org/https://doi.org/10.1787/9789264307452-en.

Oksman K (2000) Mechanical properties of natural fibre mat reinforced thermoplastic. Appl Compos Mater 7:403–414

Oksman K, Skrifvars M, Selin J-F (2003) Natural fibres as reinforcement in polylactic acid (PLA) composites. Compos Sci Technol 63:1317–1324

Okuda H, Young RJ, Tanaka F et al (2016) Tensile failure phenomena in carbon fibres. Carbon 107:474–481. https://doi.org/10.1016/j.carbon.2016.06.037

Oliva JM, Manzanares P, Ballesteros I, et al (2005) Application of Fenton's reaction to steam explosion prehydrolysates from poplar biomass. In: twenty-sixth symposium on biotechnology for fuels and chemicals. pp 887–899

Osong SH, Norgren S, Engstrand P (2014) Paper strength improvement by inclusion of nano-lignocellulose to Chemi-thermomechanical pulp. Nord Pulp Pap Res J 29:309–316

Osorio L, Trujillo E, Lens F et al (2018) In-depth study of the microstructure of bamboo fibres and their relation to the mechanical properties. J Reinf Plast Compos 37:1099–1113

Österberg M, Vartiainen J, Lucenius J et al (2013) A fast method to produce strong NFC films as a platform for barrier and functional materials. ACS Appl Mater Interf 5:4640–4647. https://doi.org/10.1021/am401046x

Paiva M, Zhou B, Fernando K et al (2004) Mechanical and morphological characterization of polymer–carbon nanocomposites from functionalized carbon nanotubes. Carbon 42:2849–2854

Panthapulakkal S, Sain M (2012) Preparation and characterization of cellulose nanofibril films from wood fibre and their thermoplastic polycarbonate composites. Int J Polym Sci 2012: https://doi.org/10.1155/2012/381342

Park Y, You M, Shin J et al (2019) Thermal conductivity enhancement in electrospun poly (vinyl alcohol) and poly (vinyl alcohol)/cellulose nanocrystal composite nanofibers. Sci Rep 9:1–10

Peelman N, Ragaert P, Verguldt E et al (2016) Applicability of biobased packaging materials for long shelf-life food products. Packag Res 1:7–20. https://doi.org/10.1515/pacres-2016-0002

Peng J, Ellingham T, Sabo R et al (2014) Short cellulose nanofibrils as reinforcement in polyvinyl alcohol fiber. Cellulose 21:4287–4298

Peng J, Ellingham T, Sabo R et al (2015) Oriented polyvinyl alcohol films using short cellulose nanofibrils as a reinforcement. J Appl Polym Sci 132:1–10

Peng Y, Gallegos SA, Gardner DJ et al (2016) Maleic anhydride polypropylene modified cellulose nanofibril polypropylene nanocomposites with enhanced impact strength. Polym Compos 37:782–793

Peresin MS, Habibi Y, Zoppe JO et al (2010) Nanofiber composites of polyvinyl alcohol and cellulose nanocrystals: manufacture and characterization. Biomacromol 11:674–681

Plastics Europe. (2019) Plastics-The Facts 2019, An analysis of European plastics production, demand and waste data. https://www.plasticseurope.org/application/files/9715/7129/9584/FINAL_web_version_Plastics_the_facts2019_14102019.pdf. Accessed 21 Aug 2020

Prashantha K, Soulestin J, Lacrampe M-F et al (2009) Masterbatch-based multi-walled carbon nanotube filled polypropylene nanocomposites: assessment of rheological and mechanical properties. Compos Sci Technol 69:1756–1763

Qi JQ, Xie JL, Huang XY et al (2014) Influence of characteristic inhomogeneity of bamboo culm on mechanical properties of bamboo plywood: effect of culm height. J Wood Sci 60:396–402

Qiu W, Zhang F, Endo T, Hirotsu T (2003) Preparation and characteristics of composites of high-crystalline cellulose with polypropylene: effects of maleated polypropylene and cellulose content. J Appl Polym Sci 87:337–345

Radkar SS, Amiri A, Ulven CA (2019) Tensile behavior and diffusion of moisture through flax fibers by desorption method. Sustainability 11:3558

Risnasari I, Herawati E, Sirait E (2018) Characterization of Polypropylene Composite Reinforced with Wood Flour or Cellulose Fiber. IOP Publishing p 012002

Robles E, Urruzola I, Labidi J, Serrano L (2015) Surface-modified nano-cellulose as reinforcement in poly (lactic acid) to conform new composites. Ind Crops Prod 71:44–53

Rohatgi A (2018) WebPlotDigitizer Version: 4.1. Austin, TX

Rusch F, Ceolin GB, Hillig É (2019) Morphology, density and dimensions of bamboo fibers: a bibliographical compilation. Pesquisa Agropecuária Trop 49:1–12

Rusli R, Eichhorn SJ (2008) Determination of the stiffness of cellulose nanowhiskers and the fiber-matrix interface in a nanocomposite using Raman spectroscopy. Appl Phys Lett 93:033111

Saba N, Mohammad F, Pervaiz M et al (2017) Mechanical, morphological and structural properties of cellulose nanofibers reinforced epoxy composites. Int J Biol Macromol 97:190–200

Saito T, Uematsu T, Kimura S et al (2011) Self-aligned integration of native cellulose nanofibrils towards producing diverse bulk materials. Soft Matter 7:8804–8809

Salajkova M, Valentini L, Zhou Q, Berglund LA (2013) Tough nanopaper structures based on cellulose nanofibers and carbon nanotubes. Compos Sci Technol 87:103–110

Sanders JE, Han Y, Rushing TS, Gardner DJ (2019) Electrospinning of cellulose nanocrystal-filled poly (Vinyl Alcohol) solutions: material property assessment. Nanomaterials 9:805

Schaider LA, Balan SA, Blum A et al (2017) Fluorinated compounds in U.S. fast food packaging. Environ Sci Technol Lett 4:105–111. https://doi.org/10.1021/acs.estlett.6b00435

Sehaqui H, Ezekiel Mushi N, Morimune S et al (2012) Cellulose nanofiber orientation in nanopaper and nanocomposites by cold drawing. ACS Appl Mater Interf 4:1043–1049

Sehaqui H, Zhou Q, Ikkala O, Berglund LA (2011) Strong and tough cellulose nanopaper with high specific surface area and porosity. Biomacromol 12:3638–3644

Shamsuddin S-R, Lee K-Y, Bismarck A (2016) Ductile unidirectional continuous rayon fibre-reinforced hierarchical composites. Compos A Appl Sci Manuf 90:633–641

Shojaeiarani J, Bajwa DS, Stark NM (2018) Green esterification: a new approach to improve thermal and mechanical properties of poly (lactic acid) composites reinforced by cellulose nanocrystals. J Appl Polym Sci 135:46468

Shrestha S, Montes F, Schueneman GT et al (2018) Effects of aspect ratio and crystal orientation of cellulose nanocrystals on properties of poly(vinyl alcohol) composite fibers. Compos Sci Technol 167:482–488. https://doi.org/10.1016/j.compscitech.2018.08.032

Sixta H, Michud A, Hauru L et al (2015) Ioncell-F: a high-strength regenerated cellulose fibre. Nord Pulp Pap Res J 30:43–57

Song M, Yu H, Gu J et al (2018) Chemical cross-linked polyvinyl alcohol/cellulose nanocrystal composite films with high structural stability by spraying Fenton reagent as initiator. Int J Biol Macromol 113:171–178

Sun S, Mitchell JR, MacNaughtan W et al (2010) Comparison of the mechanical properties of cellulose and starch films. Biomacromol 11:126–132

Suzuki A, Koide C (2000) High-performance poly(ethylene-2,6-naphthalate) fiber prepared by high-tension annealing. J Polym Sci, Part B: Polym Phys 38:61–67. https://doi.org/10.1002/(SICI)1099-0488(20000101)38:1%3c61::AID-POLB7%3e3.0.CO;2-C

Suzuki K, Okumura H, Kitagawa K et al (2013) Development of continuous process enabling nanofibrillation of pulp and melt compounding. Cellulose 20:201–210

Suzuki K, Sato A, Okumura H et al (2014) Novel high-strength, micro fibrillated cellulose-reinforced polypropylene composites using a cationic polymer as compatibilizer. Cellulose 21:507–518

Takakura A, Beppu K, Nishihara T et al (2019) Strength of carbon nanotubes depends on their chemical structures. Nat Commun 10:1–7

Textile Exchange (2020) Preferred fiber & materials market report

Thistlethwaite T, Jakeways R, Ward I (1988) The crystal modulus and structure of oriented poly (ethylene terephthalate). Polymer 29:61–69

Toivonen MS, Kaskela A, Rojas OJ et al (2015) Ambient-dried cellulose nanofibril aerogel membranes with high tensile strength and their use for aerosol collection and templates for transparent, flexible devices. Adv Func Mater 25:6618–6626

Toivonen MS, Onelli OD, Jacucci G et al (2018) Anomalous-diffusion-assisted brightness in white cellulose nanofibril membranes. Adv Mater 30:1704050

Tomczak F, Sydenstricker THD, Satyanarayana KG (2007) Studies on lignocellulosic fibers of Brazil. Part II: morphology and properties of Brazilian coconut fibers. Compos A Appl Sci Manuf 38:1710–1721

Tomé LC, Pinto RJ, Trovatti E et al (2011) Transparent bionanocomposites with improved properties prepared from acetylated bacterial cellulose and poly (lactic acid) through a simple approach. Green Chem 13:419–427

Torres-Rendon JG, Schacher FH, Ifuku S, Walther A (2014) Mechanical performance of macrofibers of cellulose and chitin nanofibrils aligned by wet-stretching: a critical comparison. Biomacromol 15:2709–2717

Tummala GK, Joffre T, Rojas R et al (2017) Strain-induced stiffening of nanocellulose-reinforced poly (vinyl alcohol) hydrogels mimicking collagenous soft tissues. Soft Matter 13:3936–3945

Tze WT, O’Neill SC, Tripp CP et al (2007) Evaluation of load transfer in the cellulosic-fiber/polymer interphase using a micro-Raman tensile test. Wood Fiber Sci 39:184–195

Uddin AJ, Watanabe A, Gotoh Y et al (2012) From “Strong” to “Much Stronger”: utilization of green tea extract dispersant for SWCNT-reinforced polymer composites. Macromol Mater Eng 297:1114–1123

Unterweger C, Brüggemann O, Fürst C (2014) Synthetic fibers and thermoplastic short-fiber-reinforced polymers: properties and characterization. Polym Compos 35:227–236

USDA A (2001) The classification of cotton. Agricultural handbook 566

Usov I, Nyström G, Adamcik J et al (2015) Understanding nanocellulose chirality and structure–properties relationship at the single fibril level. Nat Commun 6:1–11

Vallejos ME, Felissia FE, Area MC et al (2016) Nanofibrillated cellulose (CNF) from eucalyptus sawdust as a dry strength agent of unrefined eucalyptus handsheets. Carbohyd Polym 139:99–105

Van den Oever M, Bos H (1998) Critical fibre length and apparent interfacial shear strength of single flax fibre polypropylene composites. Adv Compos Lett 7:096369359800700303

Van den Oever M, Bos H, Van Kemenade M (2000) Influence of the physical structure of flax fibres on the mechanical properties of flax fibre reinforced polypropylene composites. Appl Compos Mater 7:387–402

Vigolo B, Penicaud A, Coulon C et al (2000) Macroscopic fibers and ribbons of oriented carbon nanotubes. Science 290:1331–1334

Virk AS, Hall W, Summerscales J (2012) Modulus and strength prediction for natural fibre composites. Mater Sci Technol 28:864–871

Virtanen S, Vartianen J, Setälä H et al (2014) Modified nanofibrillated cellulose–polyvinyl alcohol films with improved mechanical performance. RSC Adv 4:11343–11350. https://doi.org/10.1039/c3ra46287k

Wakabayashi M, Fujisawa S, Saito T, Isogai A (2020) Nanocellulose film properties tunable by controlling degree of fibrillation of TEMPO-oxidized cellulose. Front Chem 8:37

Walther A, Timonen JV, Díez I et al (2011) Multifunctional high-performance biofibers based on wet-extrusion of renewable native cellulose nanofibrils. Adv Mater 23:2924–2928

Wanasekara ND, Michud A, Zhu C et al (2016) Deformation mechanisms in ionic liquid spun cellulose fibers. Polymer 99:222–230

Wang L, Roach AW, Gardner DJ, Han Y (2018) Mechanisms contributing to mechanical property changes in composites of polypropylene reinforced with spray-dried cellulose nanofibrils. Cellulose 25:439–448

Wang S, Jiang F, Xu X et al (2017) Super-strong, super-stiff macrofibers with aligned, long bacterial cellulose nanofibers. Adv Mater 29:1702498

Wang W-J, Wang W-W, Shao Z-Q (2014) Surface modification of cellulose nanowhiskers for application in thermosetting epoxy polymers. Cellulose 21:2529–2538

Wei Y, Lai D, Zou L, et al (2015) Gel spinning of PVA composite fibers with high content of multi-walled carbon nanotubes and graphene oxide hybrids. IOP Publishing, p 012052

Wernik J, Meguid S (2014) On the mechanical characterization of carbon nanotube reinforced epoxy adhesives. Mater Des 59:19–32

Wu C-M, Lai W-Y, Wang C-Y (2016) Effects of surface modification on the mechanical properties of flax/β-polypropylene composites. Materials 9:314

Wu D, Wu L, Zhou W et al (2010) Relations between the aspect ratio of carbon nanotubes and the formation of percolation networks in biodegradable polylactide/carbon nanotube composites. J Polym Sci, Part B: Polym Phys 48:479–489

Xia H, Wang Q, Li K, Hu G (2004) Preparation of polypropylene/carbon nanotube composite powder with a solid-state mechanochemical pulverization process. J Appl Polym Sci 93:378–386

Xu X, Uddin AJ, Aoki K et al (2010) Fabrication of high strength PVA/SWCNT composite fibers by gel spinning. Carbon 48:1977–1984

Yan L, Chouw N, Jayaraman K (2014) Flax fibre and its composites–a review. Compos B Eng 56:296–317

Yang H-S, Gardner DJ (2011) Mechanical properties of cellulose nanofibril-filled polypropylene composites. Wood Fiber Sci 43:143–152

Yang Q, Fujisawa S, Saito T, Isogai A (2012) Improvement of mechanical and oxygen barrier properties of cellulose films by controlling drying conditions of regenerated cellulose hydrogels. Cellulose 19:695–703. https://doi.org/10.1007/s10570-012-9683-7

Yang Y, Ramirez C, Wang X et al (2017) Impact of carbon nanotube defects on fracture mechanisms in ceramic nanocomposites. Carbon 115:402–408

Yao J, Chen S, Chen Y et al (2017) Macrofibers with high mechanical performance based on aligned bacterial cellulose nanofibers. ACS Appl Mater Interf 9:20330–20339

Yee MJ, Mubarak N, Khalid M et al (2018) Synthesis of polyvinyl alcohol (PVA) infiltrated MWCNTs buckypaper for strain sensing application. Sci Rep 8:1–16

Yin W, Shan L, Lu H et al (2016) Impact resistance of oil-immersed lignum vitae. Sci Rep 6:30090

Zhang JM, Hua Q, Reynolds CT et al (2017) Preparation of high modulus poly(Ethylene Terephthalate): influence of molecular weight, extrusion, and drawing parameters. Int J Polym Sci 2017:1–10. https://doi.org/10.1155/2017/2781425

Zhang X, Liu T, Sreekumar T et al (2004) Gel spinning of PVA/SWNT composite fiber. Polymer 45:8801–8807

Zhang X, Liu T, Sreekumar T et al (2003) Poly (vinyl alcohol)/SWNT composite film. Nano Lett 3:1285–1288

Zhao M, Ansari F, Takeuchi M et al (2018) Nematic structuring of transparent and multifunctional nanocellulose papers.

Nanoscale Horiz 3:28–34. https://doi.org/10.1039/C7NH00104E

Zhao P, Wang K, Yang H et al (2007) Excellent tensile ductility in highly oriented injection-molded bars of polypropylene/carbon nanotubes composites. Polymer 48:5688–5695

Zhu J, Zhu H, Njuguna J, Abhyankar H (2013) Recent development of flax fibres and their reinforced composites based on different polymeric matrices. Materials 6:5171–5198