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Cellulose based materials to accelerate the transition towards sustainability

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ABSTRACT

Several significant sectors use cellulose, including paper manufacturing, cellophane, textiles (including rayon and viscose), and food and medicine additives. It can be utilised as a raw materials in production of fuel sources like cellulosic ethanol. Crystalline cellulose possesses a tensile strength of about 7.5 GPa, Young's modulus of 110–220 GPa and as the most abundantly available natural polymer, with excellent biocompatibility, good degradation and regeneration properties, it is considered as a remarkable biomaterial. Cellulose-based materials can be fabricated with tuneable magnetic properties, electrical conductivity, photosensitivity, sensing abilities, catalytic activity, and other specific properties by incorporating nanoparticles. These qualities make cellulose a sustainable multifunctional material. To harness such properties, strenuous efforts are being made to manufacture cellulose based materials through a wide number of manufacturing processes. This review provides an overview of the current readiness in producing cellulose-based functional materials by surveying the manufacturing procedures, characteristics and their potential applications for the end users. Future directions and opportunities of work are suggested and the limitations inherent with every process and the challenges that needs to be overcome in scalable manufacturing of cellulose-based materials are also discussed.

1. Introduction

A major issue that has concerned mankind and the world post-COVID-19 is the problem of climate change which has alarmed the need for achieving sustainability in all aspects of life. This is the only way to tackle the growing issue of climate change which poses a greater threat to mankind (Subbotina et al., 2022; Kan et al., 2023; Chandel et al., 2023). In particular, the use of materials and the process by which they are manufactured needs to be sustainable as otherwise the reliance on oil and other fossil fuels to make plastic is not sustainable. To accomplish this goal, we need to accelerate the transition to use materials that are sustainable, degradable, renewable, and at the very least, capable of being recycled in a closed-loop system, thus enabling a system of bio based circular economy. These are some of the reasons why the use of cellulose is heavily advocated. Cellulose is a material derived from plants, a renewable resource that can be returned to the planet at the end of its useful life and it can also consume carbon dioxide from the atmosphere for its biological creation (Eichhorn et al., 2022). Recent efforts are directed at developing methods and processes. Several processes are proposed to modify the surface by functionalisation for transforming the cellulose from a low value conventional material to a high value functional and intelligent material. Cellulose-based materials can be tailored to possess tuneable properties such as sensing abilities, photosensitivity, electrical conductivity, catalytic activity, magnetic properties, and other unique properties by incorporating nanoparticles or other functional components (Qi and Qi, 2017). Reviewing the recent developments in this direction is very timely so that the research in this direction can excite the new breed of researchers to pursue further research to grow this direction. In this aspect, we first discuss the basic anatomy of cellulose.

1.1. Anatomy of cellulose - molecular and structural characteristics

Given its extensive industrial use not just in the present but also in the past for the manufacture of ropes, paper sails, and wood, cellulose is undoubtedly one of the most pervasive and plentiful polymers on the

Abbreviations: BC, Bacterial cellulose; CNCs, Cellulose nanocrystals; CNFs, Cellulose nanofibers; CNSs, Cellulose nanosheets; CNTs, Carbon nanotubes; LCC, Hemicelluloses/cellulose carbohydrates; PEG, Polyethylene glycol; PLA, Polylactic acid; PVA, Polyvinyl alcohol; PPy, Polypyrrole; PEE, Polyethyleneimine; PL, Photoluminescent; RCM, Regenerated cellulose membrane; TENG, Thermoelectric nanogenerators; $TiO_{2,}$, Titanium dioxide.

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planet. Wood is by far the most widely used cellulose-containing natural resource in commerce. In actuality, "material" comes from the Latin meaning "stem of the tree" (Einchhorn et al., 2010). Hemp, flax, jute, ramie, and cotton are just a few of the other plants that have significant cellulose content. In addition to this, cellulose can also be found in non-plant sources, such as cellulose made by bacteria and tunicates. The gram-negative bacterium Acetobacter xylinum (or Gluconacetobacter xylinum) produces bacterial cellulose (BC), which appears under special cultivation conditions as a fine fibrous network of fibres (Klemm et al., 2005). Sea organisms (such as Microcosmus fulcatus) create tunicate cellulose as rod-like, nearly flawless crystals of the substance (Favier et al., 1995). It is important to note that although cellulose is categorised as a carbohydrate (a material made up of carbon, oxygen and hydrogen), it is distinct from other organic compounds in that it can be synthesised from or hydrolyzed to monosaccharides (El Khadem, 2012). According to Klemm et al (Klemm et al., 1998)., the repeat unit of the cellulose polymer (shown in Fig. 1) is known to consist of two anhydroglucose rings connected by β − 1,4 glycosidic bond which is referred to as cellobiose. Two allomorphs of cellulose, cellulose I and II, commonly known as native cellulose β (Qi and Qi, 2017). Although the ratio of these allomorphs varies from one plant species to another, the bacterial and tunicate forms are Iα and I β -rich (Einchhorn et al., 2010; Klemm et al., 2005). The crystal structures of cellulose allomorphs have been determined experimentally which involves an intricated hydrogen bonding system (Einchhorn et al., 2010; Klemm et al., 2005). Due to this hydrogen network, cellulose is a highly durable polymer that has no melting point, and it is difficult to dissolve in ordinary aqueous solutions. The cellulose chains receive a considerable degree of axial rigidity from this network (Favier et al., 1995) (O'sullivan, 1997) (Fig. 2a).

In plants, initial fibrils with cross-sections of 3–5 nm and lengths of over several hundred nanometers are formed by the parallel stacking of macromolecular cellulose chains and van der Waals interactions between neighbouring glucose molecules (Klemm et al., 2005; Donaldson, 2007). Thereafter, tens of these basic fibrils combine into rectangular arrays bordered by lignin and hemicellulose to create microfibrils with widths between 10 and 30 nm (Fernandes et al., 2011; Qin et al., 2016). The plant cell walls as a result of the hierarchical packing of these microfibrils. The microfibrils have exceptional structural stability and 1D nanomaterial characteristics thanks to the intricate integration structure between lignin and hemicelluloses/cellulose carbohydrates (LCC) (Chen et al., 2023). According to a top-down approach, lignin and hemicellulose can be depolymerized and/or dissolved using pretreatments such as diluted acid prehydrolysis, (Qin et al., 2016), recyclable organic acid treatment, (Bian et al., 2017; Chen et al., 2017; Cai et al., 2020), deep eutectic solvent fractionation, (Liu et al., 2017), and enzyme-assisted hydrolysis, (Zhu et al., 2011) to extract cellulose from plants. This may then be processed to produce cellulosic materials with the greatest structural and performance merits for various applications. Certain varieties of acetic acid bacteria, in addition to plants and algae, can also generate cellulose (Kramer et al., 2006; Iguchi et al., 2000). Bacterial cellulose (BC) is made up of nanofibrils with widths of 2–4 nm, a polymerization degree of up to 8000, and a crystallinity of over 70% (Foresti et al., 2017), giving it ideal mechanical toughness in comparison to cellulose produced from plants (Kamalesh et al., 2023). BC materials typically consist of transparent, interconnected porous network structures such as hydrogels made of cellulose microfibrils and water (Fig. 2b).

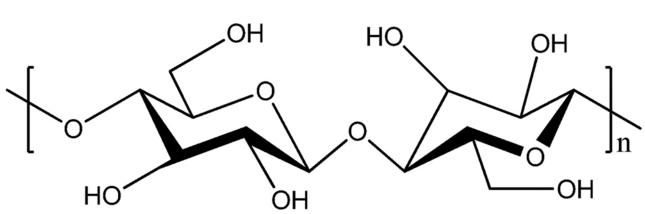


Fig. 1. Unit of cellulose (Willgert et al., 2014).

1.2. Elemental forms of cellulosic materials

Purified cellulose, which often appears as micron-sized cellulose fibres, can be created by chemically eliminating lignin and hemicelluloses from plants (Chen et al., 2011; Qi et al., 2019). Within some sections of cellulose fibres, the macromolecular chains are aligned in a crystalline structure, and there are also sections where these chains are disorganised and unkempt (Fig. 2c). These crystalline and disordered sections have different shapes and distributions depending on the starting materials and processing techniques. Native cellulose is typically found in trees, plants, bacteria, algae, etc. (cellulose I polymorph). With fewer restrictions and good processability, this cellulose polymorph can be processed to create a variety of morphologies, including cellulose nanofibers (CNFs) (Abe et al., 2007), cellulose nanocrystals (CNCs) (Chen et al., 2014) and cellulose nanosheets (CNSs) (Zhou et al., 2014) (Fig. 2d). CNFs are primarily produced by mechanical nanofibrillation techniques such high-pressure homogenization, high-intensity ultrasonication, high-speed stirring and ultrafine grinding. CNFs feature high aspect ratios (over one hundred), web-like entangled morphologies, and great specific moduli. Contrarily, Cellulose nanocrystals (CNCs) are made from disordered and paracrystalline areas of cellulose and are extremely crystalline with a low aspect ratio. The cellulose supply, the acid content, the hydrolysis temperature, and duration can affect the length, aspect ratio, and specific surface area of the CNCs. Typically, CNCs made of wood have a width of 3–20 nm and a length of 100–200 nm, whereas CNCs made from cotton have a width of 5–10 nm and a length of 100–300 nm (Habibi et al., 2010; Roman and Winter, 2004). More than three times longer than wood or cotton CNCs, BC CNCs can reach lengths of upto 1 m (Roman and Winter, 2004). Although having strong resistance to conventional acid/alkaline solutions, cellulose can be dissolved in some environmentally friendly solvents such as ionic liquids and alkali hydroxide-urea solutions (Zhao et al., 2017). By submerging in a water bath, the dissolved cellulose can be regenerated through a phase-conversion process. This makes a thin CNS or thick cellulose hydrogel. Using a solvent evaporation drying approach on CNSs or a freeze-drying method on cellulose hydrogels, cellulose films (also known as cellulose nanopaper) or cellulose aerogels can be created (Chen et al., 2014). The 2D CNSs and regenerated films differ from 1D CNCs and CNFs in that they contain the cellulose II polymorph (lower crystallinity), which gives them a more homogenous appearance and precise dimensions. They have potential uses as layered structural materials and useful membranes. For instance, Zhao et al (Zhao et al., 2017). created a mesoporous cellulose membrane through controlled regeneration and dissolution. Such a cellulose membrane can have a porosity of 71.8%, good mechanical flexibility and an abundance amount of mesopores, making it an ideal flexible separator for energy storage systems (Qi and Qi, 2017).

2. Applications of cellulose-based functional-smart materials

Cellulose derived materials can be tailored to possess desirable magnetic properties, electrical conductivity, photosensitivity, sensing abilities, catalytic activity, and other specific properties by incorporating nanoparticles or other functional components. It has been shown in Table 1, how these characteristics have so far been imparted to develop tailored cellulose derived materials.

2.1. Cellulose-based electrically conducting materials and applications in flexible electronics

In its native form, cellulose is an insulating material. However, the inclusion of particular conducting materials like nanocarbons and conducting polymers, can make cellulose electro-conductive. Carbon nanotubes (CNTs) stand out as a material of choice since CNTs possess

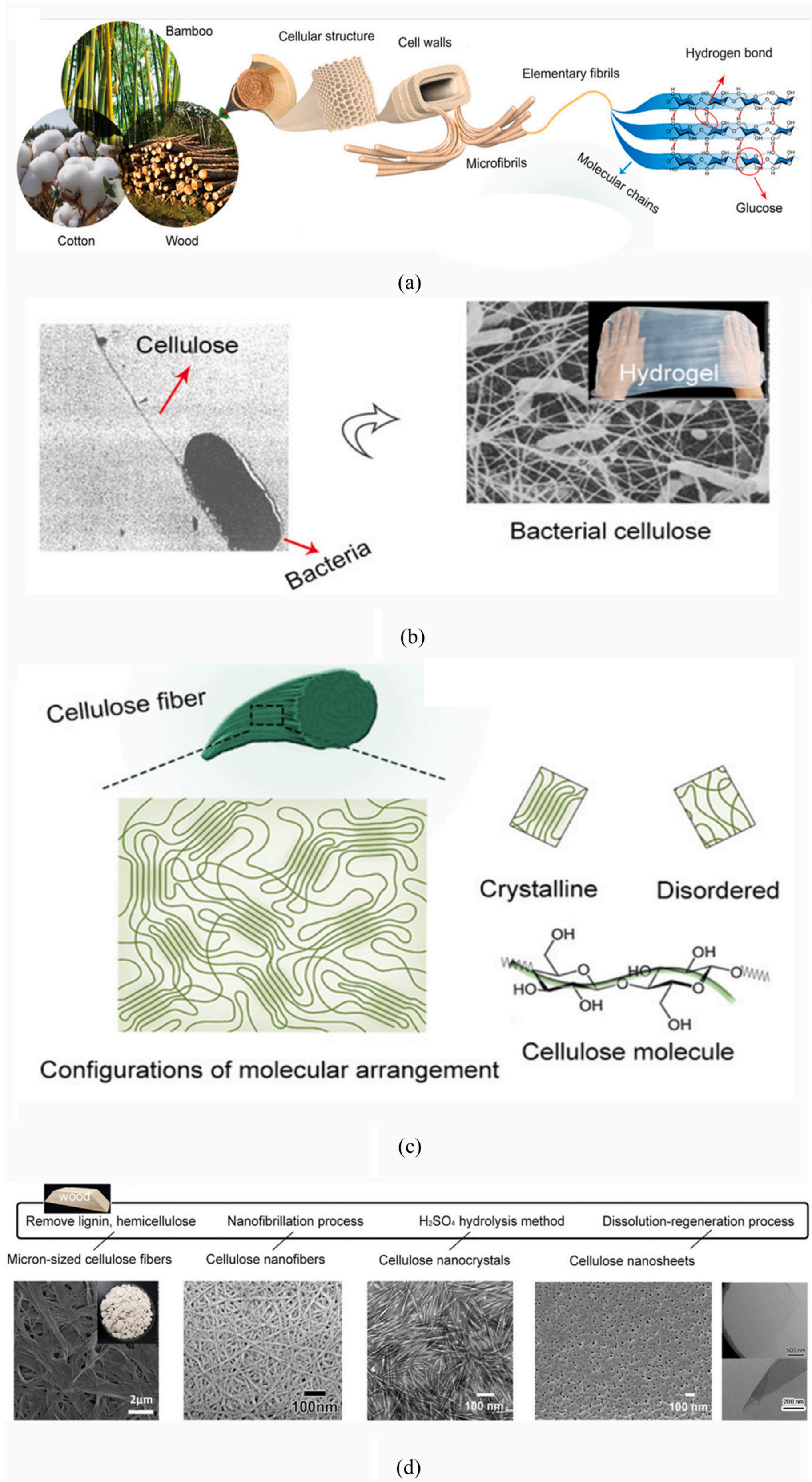


Fig. 2. Schematic representation of hierarchical fibril structure and morphology of cellulose and derived cellulosic materials: (a, b) Cellulose can be collected from plant cell walls, such as cotton, bamboo and trees as well as from the bacteria (BC), (c, d) The representative cellulosic materials (micron sized cellulose fibres) are shown in schematics of the crystalline and disordered regions, images of bacteria and BC (Zhao et al., 2021).

Table 1
Several uses for cellulose in various form.

Material forms	Applications	References
Fiber	Magnetic paper, fiber, biomaterial, reinforcement material, etc.	(Bledzki and Gassan, 1999; Kalia et al., 2009; Mashkour et al., 2011; Belgacem and Gandini, 2005; Eichhorn et al., 2010)
Nanocomposite	Biomaterials, conductive material, reinforcement material, drug delivery, barrier film, membrane, adhesion, etc.	(Huber et al., 2012; Eichhorn, 2011; Siqueira et al., 2010)
Film/ membrane	Adsorption, drug delivery, separation, package, water treatment, optical media, biomembrane, etc.	(Bledzki and Gassan, 1999; Kalia et al., 2009; Mashkour et al., 2011; Belgacem and Gandini, 2005; Eichhorn et al., 2010)
Polymer	Polymer drug delivery, biomaterial, water treatment, stabilizer, thickener, etc.	(Hubbe et al., 2008; Khalil et al., 2012; Gardner et al., 2008)

distinct electrical, mechanical, and thermal properties. Several cellulose composites with CNT integration have been reported over the last decade. Qi and Qi (2017). developed a CNT/cellulose composite paper having electrical conductivity by using direct mechanical mixing techniques. However, through such mixing, poor dispersion leads to uneven electrical conductivity (Qi and Qi, 2017). A biodegradable and extremely robust conductive macrofiber was described by Han et al (Hu et al., 2022). by wet-stretching and wet-twisting strip-shaped BC combined with conductive CNTs and PPy. CNTs are physically doped into the BC network using a surfactant, unlike PPy, which is produced by in-situ oxidation using the ions of iron. The cellulose-based conductive macrofiber developed through this route was seen to have a dense fibre structure with a tensile strength of 449 MPa (capable of lifting weights of 2 kg). The nanofiber's homogeneous distribution of conductive CNTs and PPy allows for a maximum electrical conductivity of 5.32 S cm^{-1}. The degradation experiment demonstrated that the BC/CNT/PPy conductive macrofiber can completely disintegrate in 108 h and that any CNTs or PPy that are still conducting can be recovered and used again. After soaking in water for a day, the macrofiber can still maintain its dense fibre structure with just a 6.7% and an 8.1% drop in tensile strength and conductivity, respectively. The fabric-based Thermoelectric nanogenerators (TENG) developed had a maximum open-circuit voltage of 170 V, a short-circuit current of 0.8 A and an output power of 352 W, which is adequate to power industrial electronics. The conductive fibre was also used to construct BC/CNT/PPy electrodes. The fabric-based TENGs attached to the body of a person can also serve as self-powered sensors to precisely track the movement of body parts including running, jumping, lifting an arm or leg and bending an arm. The idea of developing fabric-based TENG for energy harvesting and biomechanical monitoring is due to the advantage of having required biodegradability durability and washability (see Fig. 3).

There are numerous applications for cellulosic derived materials in flexible electronics due to their diverse micromorphology and types (whisker, fibre, film, hydrogel, ionic gel, aerogel, 3D scaffold etc.) (Chandel et al., 2023). Hence, it's crucial to have a comprehensive understanding of how structure, property and performance interact to govern the functionality of the cellulosic materials. The composition, functionality, and processing methods of cellulosic materials for applications in flexible electronics are summarised in Fig. 4. Specifically, these materials possess the following salient features:

(i) Due to biocompatibility and biodegradability cellulose and its derivatives are green biomaterials that can be used to make disposable and biodegradable electrical devices.
(ii) Piezoelectric and unique dielectric properties of cellulosic materials (such as flexible substrates, separators and gel electrolytes) make them important structural materials.
(iii) Due to its high intra- and intermolecular hydrogen bonding concentration, cellulose possess exceptional mechanical properties including flexibility and durability, which makes it a good choice for flexible substrates in solar cells, sensors, TENGs and field-effect transistor (FET) devices.
(iv) Due to cellulose's large aspect ratio, as well as its good wettability and thermal stability in a variety of electrolytes, cellulosic materials are ideal gel electrolytes for flexible OLEDs, foldable batteries and printed solar cells.
(v) Moreover, Cellulose can be customised to possess desirable microstructure and properties in healthcare applications

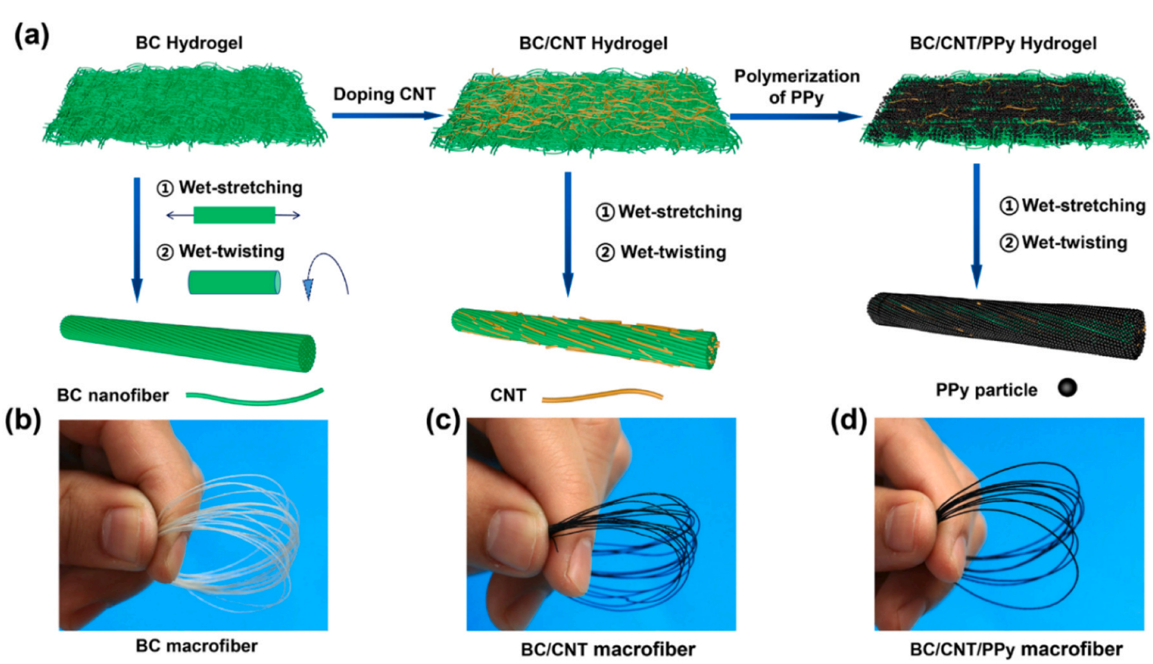


Fig. 3. Biodegradable, super-strong and conductive cellulose macrofibers for fabric-based TENG (Hu et al., 2022).

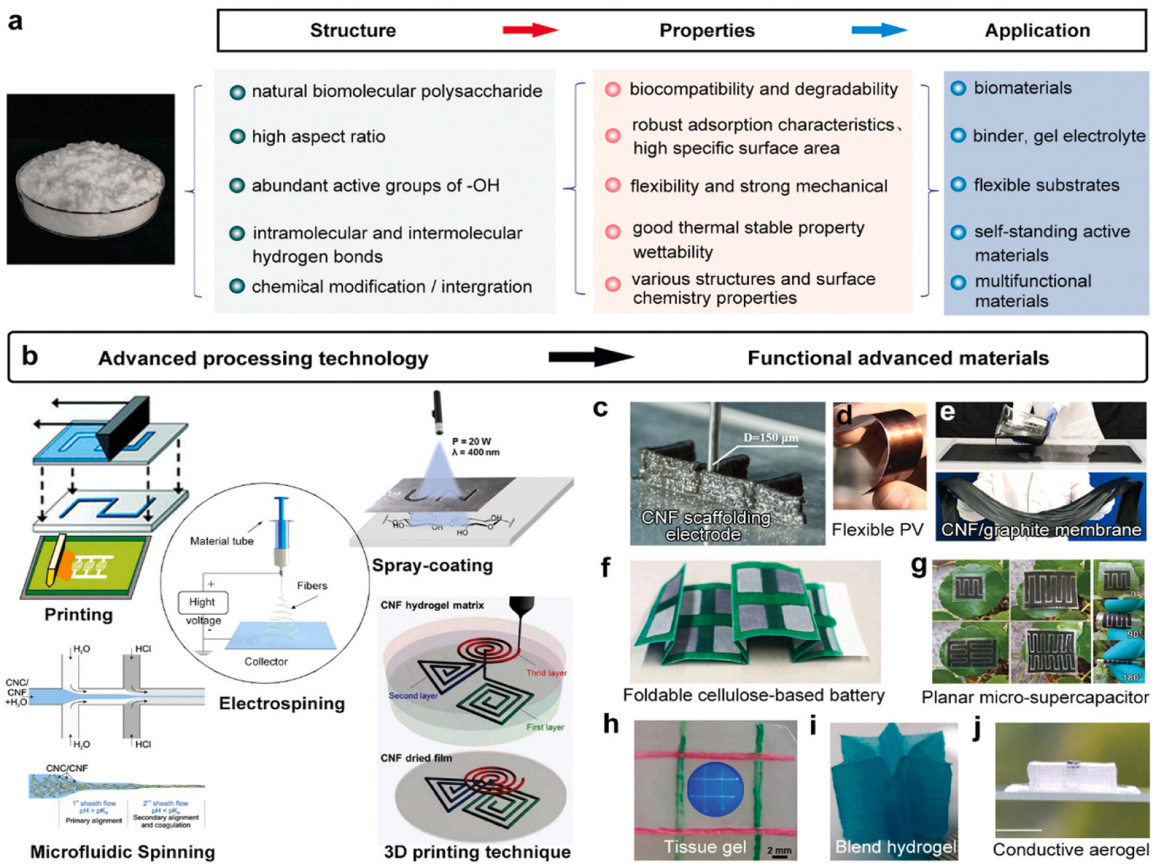


Fig. 4. Cellulosic materials' composition, functionality, and processing methods for usage in flexible electronics (a) A summary of the links between the structure, properties, and prospective applications of cellulose (b) A few manufacturing techniques for creating flexible functional materials based on cellulose, such as electrospinning, microfluidic spinning, spray coating, and 3D printing (c–j) Lithium iron phosphate electrodes, conductive composite membrane, photovoltaic cells (PV), foldable batteries, planar microsupercapacitors, mix hydrogels, tissue gels, and conductive aerogels are all made of various cellulose-based innovative materials (Qi and Qi, 2017).

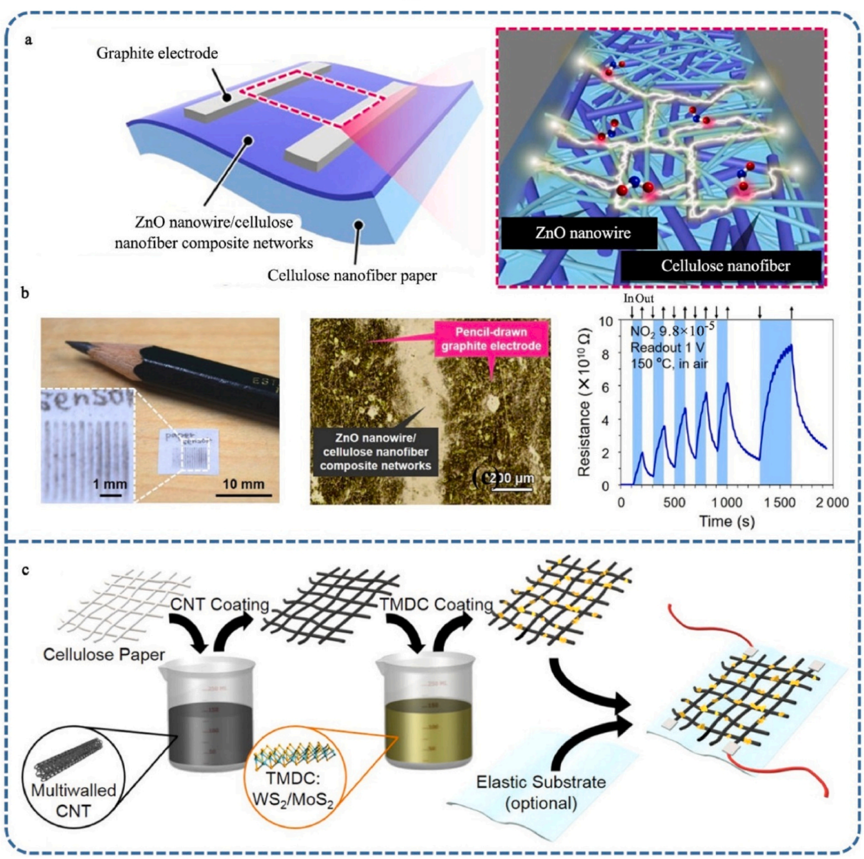
including piezoelectric films, skin and tissue regions that are inspired by biological systems.

The cellulose used to manufacture the sensor is typically a nanocellulose, some of which is made from cellulose nanofibers (CNFs) (Miao et al., 2020) and some of which are built from cellulose nanocrystals (CNCs) (Heinze and Liebert, 2001; Tang et al., 2017; Amin et al., 2022). Moreover, cellulose has demonstrated promise as a component and useful material for sustainable sensors by utilising its microscopic morphology (Jelinek and Kolusheva, 2004; Ates et al., 2020). Its inherent qualities as building blocks can increase the mechanical strength of flexible electronic devices and improve the biocompatibility of a sensor which makes it ideal for working with living being (Amin et al., 2022). Moreover, the naturally microporous characteristics of the cellulose-based aerogels or films can enhance analyte penetration and boost sensor's sensitivity (Jelinek and Kolusheva, 2004). The distinctive chiral nematic structure of nanocellulose, which makes it a functional building block material make it to respond well to humidity and circularly polarised light (CPL) (Bumbudsanpharoke et al., 2018; Peng et al., 2020). Cellulose's dielectric effect also makes it a great candidate for use in flexible electronics such as supercapacitors (Ates et al., 2020; Bumbudsanpharoke et al., 2018; Peng et al., 2020; Ummartyotin et al., 2015). More crucially, under specific circumstances, the discarded sensors can be successfully degraded (Ummartyotin et al., 2015). Nanocellulose's diverse microscopic morphological categories and special qualities have enabled the development of a wide range of new sensing materials, including anisotropic aerogel strain sensors made with CNFs (Zhang et al., 2018), self-powered sensors made of bacterial cellulose (BC) for bionic electronic skin (Song et al., 2020), and non-irritating, self-healing, robust and ultra-sensitive hydrogel sensors made of CNCs. The structural and practical characteristics of cellulose are further illustrated in Fig. 5 (i).

The literature on cellulose functional materials (CFMs) for sensing has grown rapidly because of cellulose's quick development as a green sensing material and its wide range of potential applications. The term "CFMs" refers to composite materials made from CNFs, CNCs, and BCs through chemical cross-linking or straightforward doping. Also, some recent papers have reviewed the development of CFMs' research in many subdivision sectors (such as medical biomaterials (Khine and Stenzel, 2020; Yang et al., 2021), flexible electronic devices (Zhao et al., 2021; Gong et al., 2020) and bionic actuators (Le et al., 2019; Spinks, 2020). However, very few studies (Thomas et al., 2018; Dai et al., 2020; Liu et al., 2021; Zhang et al., 2022) have examined the CFMs' recent advancements in the realm of sensing research. Newer research papers on cellulose composite sensors have yet to be summarised and there is a lack of suitable theory and convincing explanation about the CFMs' internal structure and interrelationships.

Multi-walled carbon nanotubes (MWCNTs) and cellulose have also been used to create chemical vapour sensors (Yun et al., 2010). To create a gas-sensitive cellulose membrane, the MWCNTs were covalently grafted with cellulose. The chemical sensor had good biodegradation and biocompatibility properties, and it could detect a variety of analyte molecules such as methanol, 1-butanol, ethanol, and 1-propanol while evaporation. Koga et al (Koga et al., 2019). described disposable molecular sensor based on a paper that uses biodegradable cellulose nanofiber paper derived from wood, ZnO nanowires and affordable

(i)



(ii)

Fig. 5. (i) Structure and practical properties of cellulose (ii) (a) Cellulose functional materials employed in a variety of sensing applications (b) Graphite electrodes are shown in pencil along with how well they detect NO_2 in a schematic illustration of a paper sensor made of ZnO nanowires and cellulose nanofiber paper and (c) carbon nanotubes (CNT) and transition metal dithio compounds (TMDC) are integrated into cellulose paper in a schematic illustration of a paper sensor for chemical detection (Ma et al., 2022).

graphite electrodes (Fig. 5(ii)). The NO_2 gas sensor performance was at par with that of the noble metal sensor, offering a wide range of potential applications, thanks to the straightforward pencil sketching method employed to produce the graphite electrode. Wang et al (Wang et al., 2020). created a CFMs formaldehyde gas sensor using spin coating and sol-gel techniques. Due to the BC and polyethyleneimine (PEI) double-layer nano-strong membrane's ability to adsorb formaldehyde molecules, the sensor showed an excellent stability, selectivity and the detection limit for formaldehyde was as low as 1×10^{-7}.

2.2. Magnetic cellulose materials

One of the most fascinating characteristics investigated in materials over time is magnetism. It offers a method for controlling the positioning and alignment of materials at the microscopic and macroscopic levels the control on this aspect is crucial for the development of many gadgets and appliances (Nypelö, 2022). The application of cellulose is limited in devices which rely on electrical or magnetic signals. Ferromagnetic particles are frequently used to modify cellulose to use it in such applications. Ferromagnetic cellulose fibres have been used in optical and medical equipment, electrical fabrics, loudspeakers, magnetic shielding and separation of biomolecules and environmental accumulations. Through addition of magnetic nanoparticles, cellulose-based materials can become a new class of smart materials that can adapt to changing external magnetic fields. As previously indicated, these magnetically responsive materials are anticipated to show magnetic field-dependent mechanical behaviour. One of the frequent procedures employed in the creation of magnetic cellulose materials is lumen-loading technology (Zakaria et al., 2004a, 2004b). The major steps in this procedure include dispersing the cellulose pulp in a concentrated suspension of iron oxide particles such as magnetite and maghemite particles, which is then followed by a gentle wash to eliminate unbound magnetic particles (Rioux et al., 1992; Marchessault et al., 1992). Based on this technique, magnetic papers can be created by introducing readily available magnetic pigments into the lumens of softwood fibres. Iron oxide particles are synthesised within the cellulosic matrix itself as a preferential

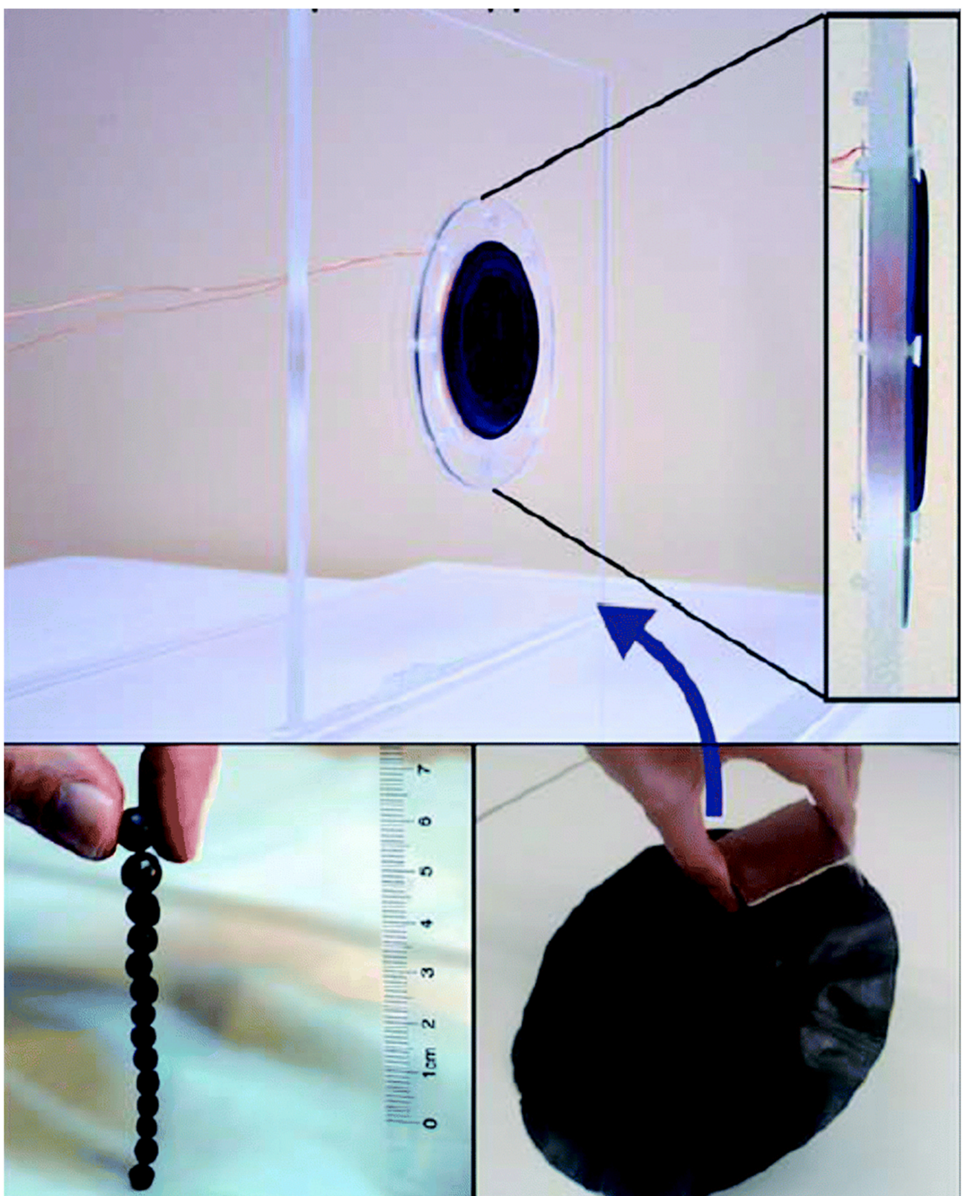


Fig. 6. Application of cellulose in making of a loudspeaker: A thin prototype loudspeaker without an external magnet that produces a magnetic membrane (bottom right) by vacuum filtering a suspension of magnetic nanofibrils. Figure bottom left shows hard spherical beads that are permanently magnetised (Nypelö, 2022).

method of creating magnetic cellulose materials. By adding an excessive amount of NaOH solution, iron ions in the cellulose matrix are converted to iron oxide particles using the process called as in-situ co-precipitation. It was discovered that the size distribution of magnetic nanoparticles is significantly influenced by the processing conditions. Using an ammonia gas enhances the co-precipitation approach in a closed system without oxygen and through this approach, magnetic nanoparticles in the cellulose matrix were made to disperse uniformly to control their crystalline phase (Katepetch and Rujiravanit, 2011). This technique produces magnetic particles with an average particle size of 20–39 nm in the crystal form of magnetite (Fe_3O_4). Also, by varying the concentration of an aqueous iron ion solution, magnetic nanoparticles' particle size and particle size distribution were controlled.

2.2.1. *Cellulose in magnetic devices*

Examples of devices that make use of the cellulose and magnetic materials combination are highlighted in this section.

2.2.1.1. Loudspeakers. The use of magnetic cellulose films in loudspeakers is an example of a practical application of cellulose in magnetic devices. A thin prototype loudspeaker was created using a static coil to drive a cellulose-$CoFe_2O_4$ membrane to amplify sound (Galland et al., 2013). A membrane linked to the voice coil of a conventional loudspeaker moves in a magnetic gap, causing the membrane to vibrate and emit sound. A magnetic cellulose membrane was used in the setup by Galland et al. (2013) in order to eliminate the need of an external magnet. Fig. 6 show the magnetic membrane used in the prototype loudspeaker. Using CNFs as a template for magnetic nanoparticles and in the membranes has the advantage of ensuring uniform particle distribution because the nanoparticles were connected to the fibrils. At 60 wt % $CoFe_2O_4$ loading, the membranes showed outstanding strength of 100 MPa with a stiffness of 5 GPa. The CNF matrix gives strength to the material which explains why the CNF films consistently showed tensile strengths of more than 200 MPa (Nypelö, 2022; Fukuzumi et al., 2013).

2.2.1.2. Optical devices. It is frequently stated that the transparency of nanocellulose films with magnetic capability is advantageous for magneto-optic applications (Hosoya et al., 2014; Sadasivuni et al., 2016). However, the magneto-optic effect has not been exploited in the optical usage considerations of magnetic cellulose; instead, the cellulose light-interactions in combination with the material's magnetic properties have been used. The creation of cholesteric phase using CNCs has stimulated research into chiral photonic materials. Forcing interactions between light and cellulose requires both the chirality and the distance between the rod-shaped nanoparticles. Future magnetic cellulose materials for optical devices could rely on the combination of magnetic functionality and optically active structure. Iron oxide particles were integrated into CNCs by Chen et al. (2020) by co-precipitation in the cellulose matrix. When these particles were combined with clean CNCs, films with liquid crystalline cholesteric ordering could be created. The

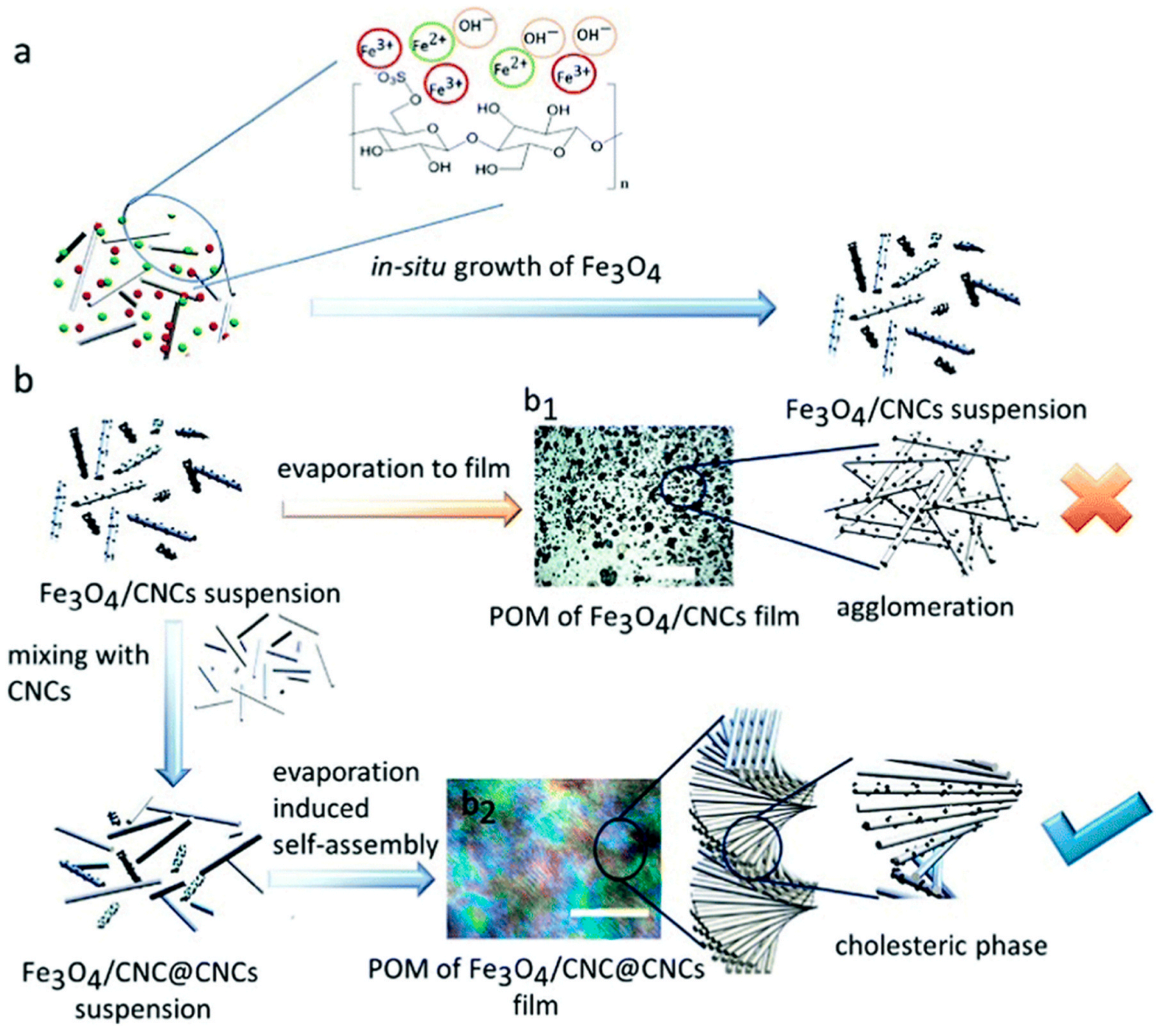


Fig. 7. (a) schematic showing the development of Fe_3O_4 nanoparticles on CNCs in situ; (b) evaporation of Fe_3O_4/CNC suspensions to produce Fe_3O_4/CNC films with the agglomeration of cholesteric order using Fe_3O_4/CNC suspension or Fe_3O_4/CNC suspension with neat CNCs, respectively (Nypelö, 2022).

tuning of the chiral nematic pitch of the cholesteric phase required a magnetic field strength of 7 mT. (Fig. 7). The idea of sophisticated multimodal protection of garments against counterfeiting has been proven through the use of magnetic functionality mixed with optical activity (Skwierczyńska et al., 2018, 2020). By adding magnetic and luminous nanoparticles made of magnetite and lanthanide-doped fluorides to cellulose fibres, the multifunctionality was achieved. The fibres' suitability for use in textiles remained due to the preservation of their mechanical qualities.

2.2.1.3. Storage systems. Phase transition material matrices with magnetic CNC-Fe_3O_4 hybrids added produced materials with the potential to store solar or magnetic heating energy (Abdalkarim et al., 2021). Such phase change materials are shown to operate between 44 °C and 61 °C and have an energy storage capacity of up to 83 J g^{-1} which is beneficial in agricultural applications to dry the products.

Zhang et al. (2021) developed an all-cellulose energy harvesting device, which was composed of pure and conductive bacterial cellulose as both the friction layers and electrodes. Due to the widely distributed -OH groups and the porous cellulose network structure, conductive bacterial cellulose could be fabricated by modifying the surface with polypyrrole and carbon nanotubes. With a maximum open-circuit voltage of 29 V, short-circuit current of 0.6 μA, and output power of 3 μW, the sandwich-structured device prepared was capable of driving commercial electronics (Teng et al., 2023).

2.2.1.4. Magnetic shielding. By installing a second magnetic field, one can shelter items from a magnetic field. This process is known as magnetic shielding. The extra field produced by a ferromagnetic material redirects the magnetic field. Iron oxide (Yang et al., 2020) and cobalt ferrite (Nypelö et al., 2014) adorned cellulose composites successfully demonstrated this effect of magnetic shielding. By measuring the current in the film applied with and without an external magnetic field in a configuration where the magnetic field-generating films are placed atop a magnet, the shielding effect can be measured (Yang et al., 2020; Qin et al., 2021). Also, depending on the alignment within the film and of the magnetic field, the magnetic domains in such films permitted a certain response. When a second magnetic field was added, the current immediately dropped from 7.4 nA to 5.1 nA along the direction perpendicular to the magnetic orientation. The shift went from 3.8 nA to 3.2 nA in a perpendicular direction, showing that the directionality of the material controls the magnetic shielding property. Electromagnetic interference shielding has also made use of the chiral nematic structural characteristics of mesoporous photonic cellulose and the magnetic properties of $CoFe_2O_4$. The mesoporous cellulose template was created by cholesteric CNCs and urea-formaldehyde templating (Nypelö, 2022).

2.3. Light-responsive cellulose-based materials

Cellulose is a colourless polysaccharide due to the lack of groups that absorbs light in the visual range. Hence, transparent paper can be produced utilising cellulose-based materials such as regenerated cellulose and cellulose fibre with a diameter of micro/nanoscale size through casting, filtering, extrusion and other processes (Liu et al., 2014; Zhu et al., 2014). By incorporating different fluorescent or luminous inorganic compounds or other functional compounds, photofunctional cellulosic materials with tailored properties can be made for a wide range of applications. To develop fluorescent cellulose films, fluorescent dyes that showed intense fluorescence when exposed to UV light were applied to never-dried regenerated films (Qi et al., 2009). The creation of luminous cellulose composite sheets with prolonged after-glow emission was made possible by the regeneration of a solution containing photoluminescent (PL) pigments (alkaline earth aluminates) (Qi et al., 2009). The advantages of the PL pigments incorporated in the cellulose matrix include a quick activation time, a protracted afterglow and great brightness. It can absorb light (sunlight, fluorescent, incandescent, etc.) for a period of 10 to 15 min before emitting visible light. Moreover, they are chemically stable and free from radioactive and hazardous elements. As a result, the new PL films can be used to create PL signs, photographs, etc., which have a wide range of applications in the fields of information technology, anti-counterfeiting methods and sustainable packaging (Fig. 8).

2.4. Creating eco-friendly materials using cellulose nanofibers in epoxy composites

Innovative studies have been carried out by researchers from Thailand, France and Singapore to develop stronger materials employing long pineapple leaf fibres (PALF) and small cellulose nanofibers (CNF). They experimented with several CNF addition rates and discovered that 1% CNF significantly boosted impact strength. Significant gains in flexibility and strength were seen in PALF-epoxy composites. When CNF and PALF were combined, impact strength increased noticeably. The discoveries might completely alter the development of stronger materials. In their study (Klinthoopthamrong et al., 2023), the researchers investigated cellulose nanofibers (CNF), which are exceedingly small fibres that have the potential to strengthen materials (Fig. 9). They altered the CNF content at 1%, 3%, and 5% of the epoxy mixture while always including 20% PALF to maintain consistency. The materials were manually layered while the composites were meticulously put together (Surajarusarn et al., 2020).

They benchmarked the materials against the composites reinforced with CNF alone, solely PALF and a combination of both CNF and PALF to observe the effects of various combinations. Surprisingly, the epoxy's flexibility and strength were barely impacted by the addition of these microscopic CNF fibres. However, compared to utilising epoxy alone, adding just 1% CNF caused the impact strength of the epoxy to be well over 115% (Rudich et al., 2023).

A powerful microscope was used to expand the investigation by looking at the composites' fractured surfaces. They found that the surface of the composites varied from being smooth to significantly rougher when the materials broke. When the epoxy with 20% PALF was used, things became extremely interesting because both flexibility and strength significantly increased. When compared to utilising epoxy alone, the flexibility improved by roughly 300% and the strength by 240%. However, compared to utilising neat epoxy, the impact strength of this composite material increased by an astounding 700%. Researchers found no appreciable differences in flexibility and strength between employing PALF alone and CNF in the composite materials, albeit the impact strength did see a noticeable improvement. They found that using epoxy with just 1% CNF increased impact strength by as much as 220% when compared to using epoxy with 20% PALF or a staggering 1520% when compared to using clean epoxy alone. This outcome demonstrates unequivocally that the combined effects of CNF and PALF were responsible for the impressive increase in impact strength (Vinod et al., 2021; Jayan et al., 2020; Chaturvedi et al., 2022; Agustina et al., 2023).

Possible Toughening Mechanism has been shown in Fig. 10. In the case of the CNF-reinforced EP composite material (Fig. 10, top panel), when an external load is applied in the direction as indicated, the EP matrix deforms. The deforming matrix shears over the interface between the EP and CNF, causing shear stress to be generated at the interface. This in turn caused the CNF to bend, and bending stress is generated. For a sufficiently high external load, the interfacial shear stress peaks at the ends of the CNF and this leads to matrix yielding and finally cracking; i.e., cracks are initiated in the matrix adjacent to the CNF ends. The cracks propagate into the matrix. In the absence of PALF, i.e., the stronger phase (compared to the EP matrix), there is nothing standing in the way of the path of the crack propagation. In other words, the crack propagation could not be halted (Goh, 2016). The crack propagates cleanly throughout the specimen cross section until the specimen fractures into

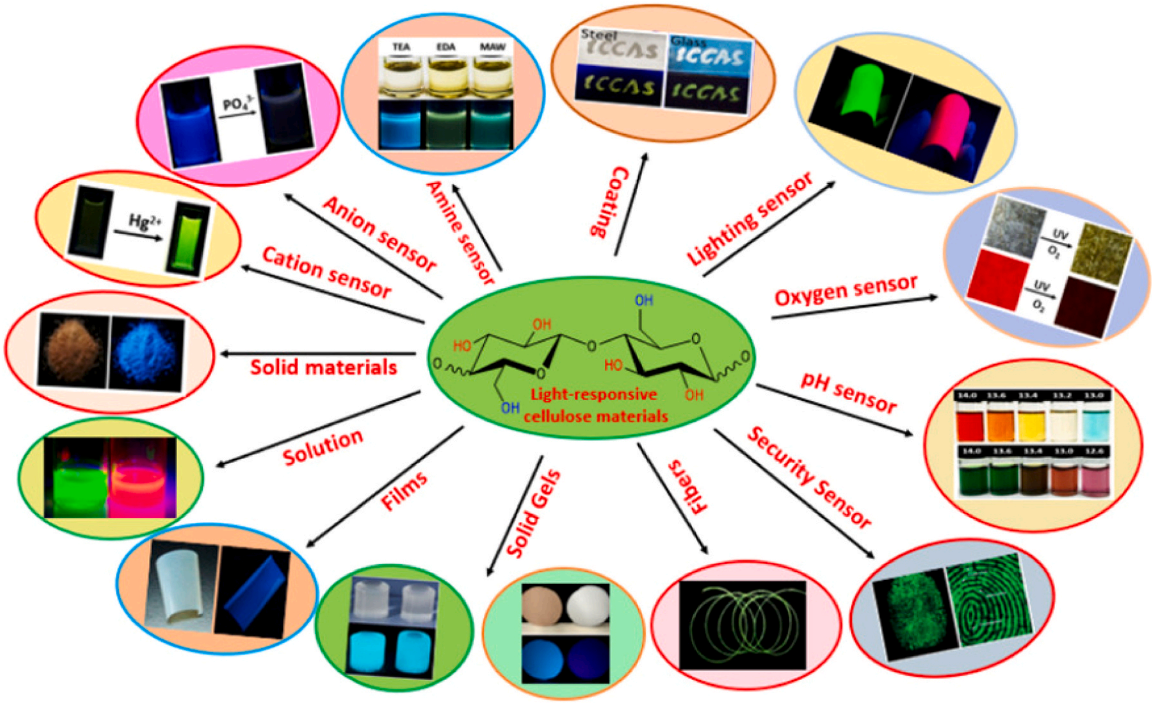


Fig. 8. Multifunctional applications of light-responsive cellulose-based materials (Nawaz et al., 2021).

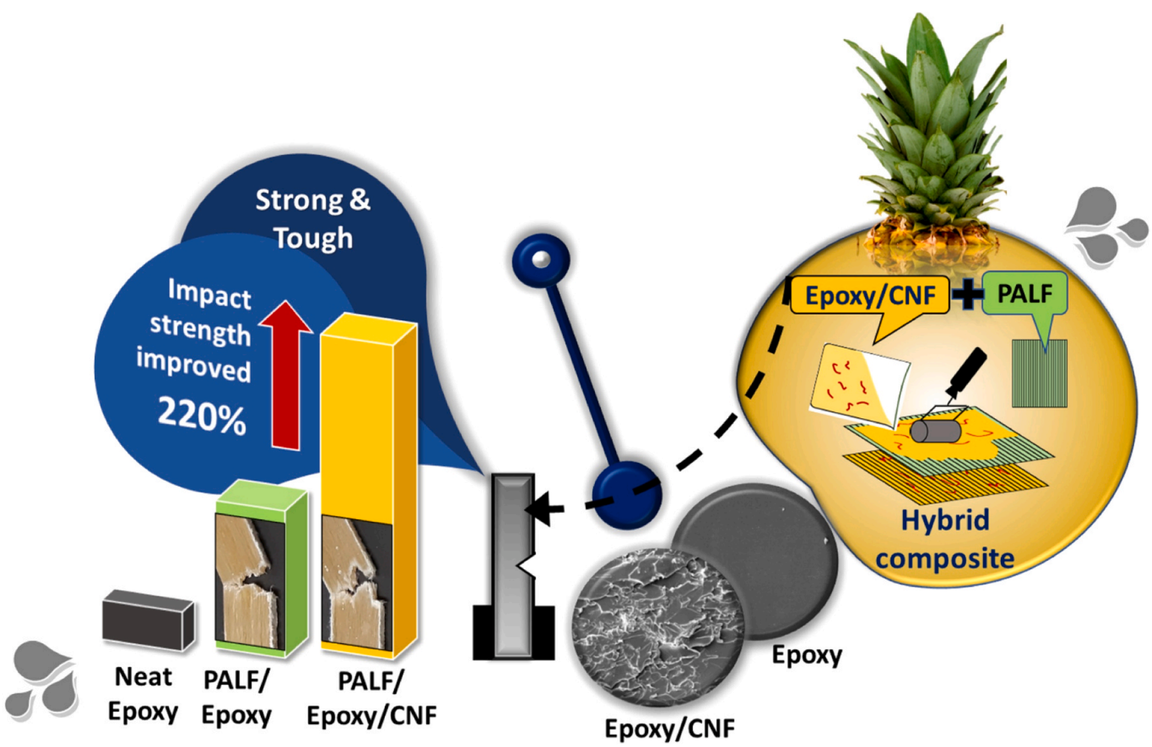


Fig. 9. Synergistic Toughening of Epoxy Composite with Cellulose Nanofiber and Continuous Pineapple Leaf Fiber as Sustainable Reinforcements (Klinthoopthamrong et al., 2023).

two.

In the case of the hybrid (CNF/PALF-reinforced) epoxy composite material (Fig. 10, bottom panel), in the presence of the external load, the EP matrix deforms. The deforming matrix shears over the interface between the EP and CNF, causing shear stress to be generated at the interface. This in turn caused the CNF to bend, and bending stress is generated. In addition, the deforming matrix also compresses the interface between the EP and PALF. Compressive stresses generated at the interface lead to bending of the PALF. Thus, both the PALF and CNF take up stress from the matrix in this way. For a sufficiently high external load, similar to the previous case, the matrix crack appears as the CNF end propagates, but on reaching the PALF, the propagation is halted

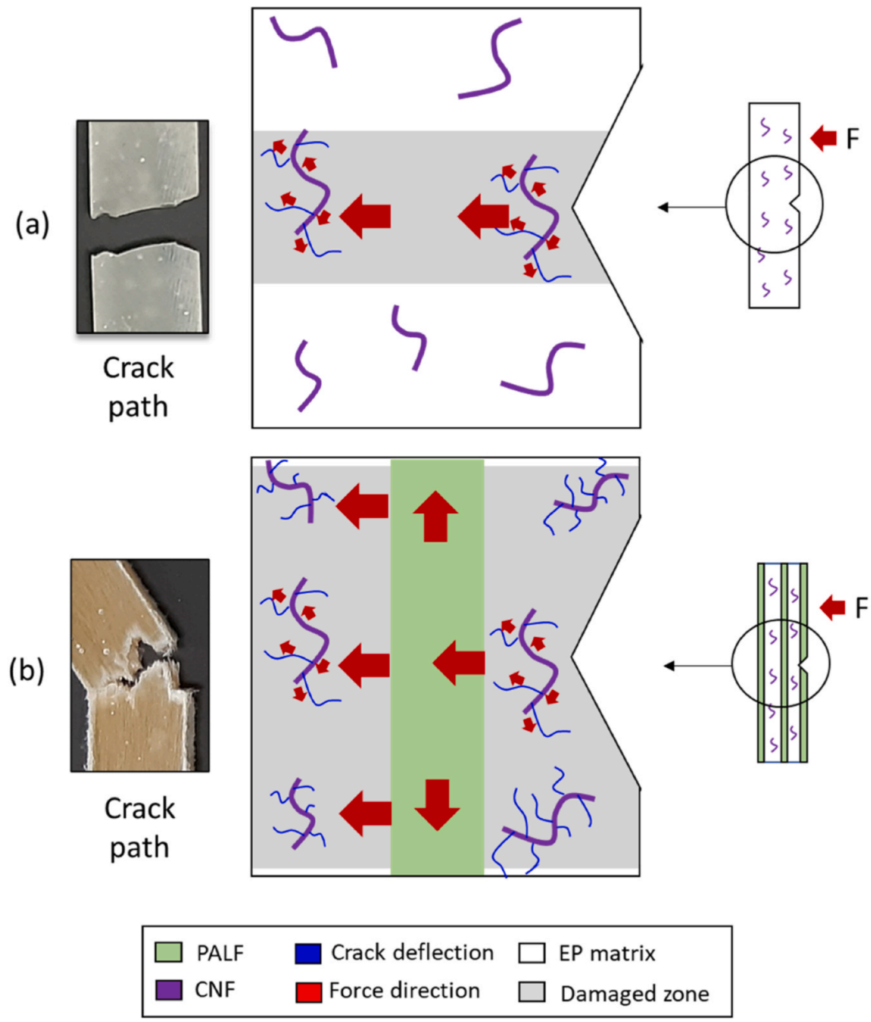


Fig. 10. Possible reinforcing mechanism after impact loading of (**a**) CNF-reinforced EP and (**b**) hybrid epoxy composite (CNF/PALF) (Klinthoopthamrong et al., 2023).

(Goh, 2016). If the load is much higher, eventually, the stress taken up by the PALFs (as they all bend) could exceed the fracture strength of the PALF, and thus the PALFs break. The crack in matrix material continues to propagate past the broken ends of the PALFs, leading to a complete fracture of the composite material.

By combining both CNF and PALF together, spectacular improvement in toughness is obtained. This is a result of the synergistic effect of CNF and PALF. This could lead to greener, more sustainable, and more cost-effective materials for demanding applications (Klinthoopthamrong et al., 2023).

2.5. Cellulose-based biomaterials for biomedical applications

Cellulose has a long history of being used as a biomaterial. Innovative and practical biomaterials have been created by altering cellulose physically or chemically (Orlando et al., 2020), derivatizing cellulose (Yang et al., 2021), or combining cellulose with other materials to create composites (Aris et al., 2019; Wahid et al., 2019). These cellulose-based polymers are increasingly helpful in biomedicine, including disease analysis, prevention, diagnosis and treatment (Fig. 11). Few biomedical applications of cellulose based biomaterials have been discussed in this section (Yuan et al., 2023).

2.5.1. Skin and wound dressings

For wound healing, cellulose-based materials have been utilised to resemble skin, speed up skin cell regeneration, and reduce scarring (Fatema et al., 2022). The materials created through bioprinting are some of the most cutting-edge ones used in wound dressings. Nanocellulose may be the perfect ingredients for bio-ink. For instance, in bioprinting, nanocellulose fibrils produced by TEMPO-mediated oxidation can lower viscosity and produce beneficial rheological qualities (Rees et al., 2015). Furthermore, porous nanostructures can be created through bioprinting using bio-ink based on nanocellulose to stabilise functionalized molecules. In bioprinting, for instance, carboxymethylated-periodate nanocellulose has been utilised to create 3D porous structures that can hold and release microbicides (Rees et al., 2015). A practical technique for creating 3D porous matrices that resemble the skin's natural layering is electrospinning. In the creation of scaffolds by electrospinning, mixtures of cellulose acetate and hydrogel (such as gelatin and poly urethane) have been employed (Vatankhah et al., 2014).

2.5.2. Bone tissue engineering

Since cellulose fibres resembles the collagen fibres found in bone tissue and are compatible with the rigid, mechanical environment present in bone systems, cellulose has been employed in bone tissue engineering (Torgbo and Sukyai, 2018; Vallejo et al., 2021). Nanocellulose is frequently used to strengthen hydrogels because their mechanical characteristics cannot resist mechanical pressures found in bone (e.g., NFC). Polylactic acid or polyvinyl alcohol hydrogel electrospun matrices

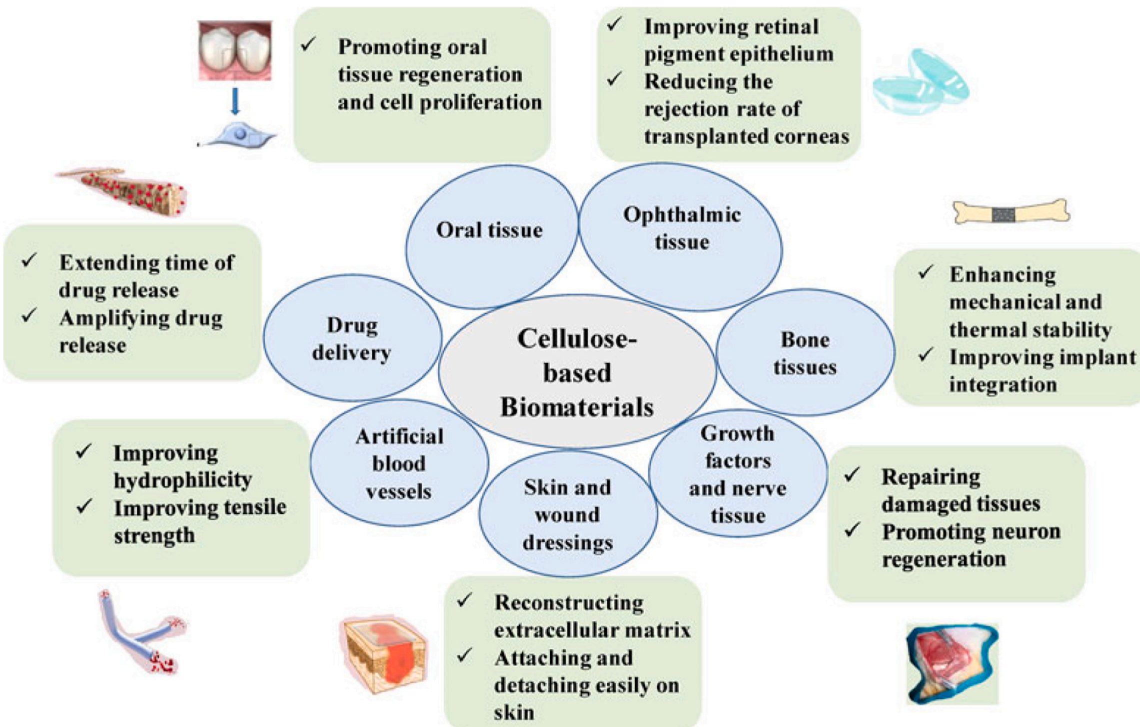


Fig. 11. Advantages and applications of cellulose-based materials in biomedical field (Fatema et al., 2022).

contain cellulose nanocrystals (such as NCC) as support (Chahal et al., 2014; Rescignano et al., 2014; Zhang et al., 2015). It has been shown that maleic anhydride grafting, sodium dodecyl sulphate and polyethylene glycol grafting can all improve the adhesion between PLA and cellulose in electrospinning (SDS). Through this procedure, the nanocrystals are modified to create matrices with smaller diameters and more polydispersity (Zhou et al., 2013). Moreover, it improves thermal and mechanical stability.

2.5.3. *Artificial blood vessels*

Vasculature replacement and regeneration have both been used as cellulosic biomaterials. During its manufacture, BC can be shaped into a variety of shapes to provide substrates that are best for promoting cell adhesion and proliferation (Mohite et al., 2014; Picheth et al., 2017). Research (Fink et al., 2010) show that materials created with BC in vascular grafting cause a decrease in thrombin at target surfaces, preventing the formation of clots. This distinguishes it from other materials that are frequently utilised for vascular grafting in a significant way (e.g., PET and PTFE). Furthermore, composites made from BC have become a significant replacement option for atherosclerotic blood arteries. The creation of such hydrophilic BC composites promises to significantly enhance the creation of innovative artificial blood vessel implants, bacterial adhesion-resistant coatings for cardiovascular stents, and artificial heart valve replacements.

3. Limitations, challenges and opportunities for future

The most crucial type of polymer composite materials are nanocomposites and biocomposites, which have attracted a lot of interest. Polymers that are capable of natural degradation are crucial for addressing climate change issues, lowering dangers associated with polymeric products and to maintain ecological equilibrium. Performance, processing and affordability are the three factors that are connected to the promotion and popularisation of biopolymers. By utilising cutting-edge technology and procedures, it is possible to eliminate the inherent drawbacks of some biodegradable polymers, such as their poor mechanical qualities, limited processing window, and poor thermal, electrical, and barrier properties (Chen et al., 2016; Sha et al., 2016). As a result, there is a lot of study being done on composites made of natural fibres that are predominantly made of organic substances including cellulose, hemicellulose and lignin. Phase boundary interactions, which can be challenging when a hydrophobic matrix and hydrophilic filler are used are key determinants in the properties of polymer composites manufactured from plant-based raw materials (Chen et al., 2015). The right choice of the additive's ratio in relation to the matrix is also crucial. Hence, it appears that the ongoing search for new, effective fillers, both inorganic and derived from renewable plant sources, as well as performing various modifications on them and adding additional additives, are especially crucial for the development of these materials. Hence, recent advancements in the creation of polymer biocomposites frequently centre on the creation of strategies or techniques to enhance the compatibility of the composition's constituent parts (Wei et al., 2015; Gandini et al., 2016; Vaidya et al., 2016; Lenfeld et al., 2020). Chemical or physical modification, such as corona discharge, heat or plasma treatment, or impregnation of fibres with a matrix-compatible polymer, graft copolymerization, acetylation, or mercerization, stand out as the most often employed techniques. These changes are largely meant to decrease water absorption and boost the product's dimensional stability, in addition to adhesion improvement.

The study conducted often comprises the assessment of mechanical, rheological, and thermal parameters as well as the structural analysis of composites made with plant fillers of diverse origins (Masek and Kosmalska, 2022). These tasks are done in addition to formulation. Moreover, experiments are carried out to ascertain the stability of the generated materials under the effect of environmental and microbial (biodegradation) conditions. The creation of efficient technologies to produce homogeneous composites with desirable functional qualities is a crucial topic. There is still a great deal for improvement in both the manufactured composites and the effectiveness of their production.

3.1. Raw materials of cellulose

Plant, animal, and mineral fibers make up the three groups of natural fibers. The most common use of plant fibers in traditional composite processing is only as reinforcement. Animal fibers are formed of proteins, whereas all plant fibers contain cellulose (Diyana et al., 2021). One of the most promising alternatives to raw materials based on petroleum is plant-based cellulose. The plant species, age, growing location, climate, and portion of the plant all affect the fiber structure (Karimah et al., 2021) and properties (Wu et al., 2020). Because of this, one of the biggest problems with plant fibers is the wide range of their characteristics. The qualities of wood fiber are largely determined by the anatomy of the wood material in combination with the processing technique in the particular tree species. Softwoods, like spruce and pine, and hardwoods, like birch and eucalyptus, can be used to categorize wood fibers. Both can be utilized to dissolve new pulp. Dissolving pulp is utilized more frequently in upgraded and high-quality products because it has more cellulose and less hemicellulose than paper-grade pulp. Pre-hydrolysis kraft (PHK) and acid sulfites synthesis are two frequently employed commercial manufacturing techniques (Ramage et al., 2017) for pulp dissolution (Lehr et al., 2021). With the aid of cutting-edge technologies, the pulp may also be made from cellulosic waste, which supports the circular economy by employing a productive closed-loop system. Another innovative pulp-based wood fiber with comparable sustainable qualities is kuura-fiber, which is now being produced on a pilot-scale bioproduct plant. The infinite fiber company developed a fiber technology that uses recycled textiles and agricultural waste, and Haksa Textiles has converted recycled cotton fibers into sustainable yarn. New varieties of wood fiber have lately been created mechanically without the use of hazardous solvent systems. The more environmentally friendly plant-based fibers are used in Spinnova's cutting-edge fiber process to satisfy the requirements of current standards with an emphasis on waste reduction, production efficiency, durability, water conservation (Felgueiras et al., 2021), recycling, and closed-loop manufacturing (Ho and Leo, 2021). The industry leader in producing sustainable fibers. The pulp is mechanically improved and converted into a suspension of fibers ready for spinning using Spinnova's ground-breaking fiber technology without dissolution or regeneration. These innovative textile fibers provide flexible options for reinforcing the environmentally friendly composite industry (Uusi-Tarkka et al., 2021). Recently, denim waste has also been used to generate bio composites and cellulose composites (Aziz et al., 2023).

3.2. Flax fiber-based composite processing

The composite can be used in flax fiber in many ways. For reinforcement (Aziz et al., 2023), it can take the shape of flax yarn, and pulp etc. The short flax fiber, bidirectional (BD), unidirectional (UD) and cross-ply fiber arrangements are recently investigated. Researchers are trying the various unidirectional laminates (0 °−90 °C). Polymer composites can be produced using a variety of processing methods, such as vacuum-assisted compression molding, filament winding, and compression molding (Rajak et al., 2019). In the fiber hybridization process, compression molding can be used to create the layers. The hand lay-up technique used to create fiber laminates (Li et al., 2022). The hot press molding technique used to create the plates and composites (More, 2022). Another technique for creating the composite is hot pressing. The fiber-polymer nonwovens are created by the wet-laid technique and then hot-press molded. Nonwoven is created using the air-laying method. The prepregs are created using thermal consolidation, and composites are created using compression molding. The process of creating paper can be used to create the fiber and polymer mat layers. Another technique for manufacturing the composite is vacuum-assisted resin transfer molding (VARTM). Direct impregnation of fiber in a matrix is a tight mold method for creating fiber-reinforced composites. Several investigations have found that after melting down fiber bundles, the length of the fiber might shorten. The coextrusion technology can be used to treat the granulated polymer and flax fiber. A fiber composite product can be created by rotational molding extrusion. A twin-screw extruder is a processing tool. Before sending the fiber into an extruder, it might be palletized. The fibers are divided into their simplest forms during the palletization process (Navada et al., 2022). An extruder can operate at low constant flow to the pelletization process. For creating composites, traditional and microcellular injection molding processes were used. A shot flax fiber and poly (3-hydroxybutyrate) (PHB) matrix, compression molding had a higher impact strength than injection molding (Santamala et al., 2016). Another method for composite creating is pultrusion. It is important to maintain processing temperatures at a very low level to prevent fiber deterioration (Jorda et al., 2022).

4. Concluding remarks

As an abundantly available organic sustainable polymer from plant sources, cellulose offers myriad of opportunities as reviewed in this work. It has been shown in this review as to how one can use eco-friendly solvents to exploit dissolution and regeneration processes for achieving tailored functionality and intelligent manufacturing in cellulosic materials. The development of cellulose-based electrically conducting materials is useful in producing many engineering components such as wearable electronics, conductors, supercapacitors, sensors, actuators and so on. As highlighted by this review, it is possible to create magnetic nanoparticles incorporated cellulose-based materials that can adapt to changes in their physical properties caused by an external magnetic field. It has been found out that functional materials such as QDs, PL pigments, metals or other functional components can be used to create photofunctional and other functional cellulosic materials. Application areas for those materials include anti-counterfeiting methods, information technology, packaging, sensing, plasmonic, and catalytic applications. Overall, the synergistic interaction of the precursors created by the combination of cellulose and organic/inorganic elements offers an avalanche of opportunities to synthesise newer materials that not only preserve the inherent qualities of their component precursors but also show novel capabilities. Future studies are projected to concentrate on nanofillers in composite materials and investigate commercial prospects. Artificial intelligence and other technologies should also be explored to increase production and predict the productivity and properties of various cellulose-based materials.

CRediT authorship contribution statement

Goel Saurav: Funding acquisition, Supervision, Writing – review & editing. **Petru Michal:** Writing – review & editing. **Verma Jaya:** Project administration, Writing – original draft.

Declaration of Competing Interest

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

Data Availability

No data was used for the research described in the article.

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