



Review

Recent advancement in isolation, processing, characterization and applications of emerging nanocellulose: A review

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ABSTRACT

The emergence of nanocellulose from various natural resources as a promising nanomaterial has been gaining interest for a wide range application. Nanocellulose serves as an excellent candidate since it contributes numerous superior properties and functionalities. In this review, details of the three main nanocellulose categorised: cellulose nanocrystal (CNC), cellulose nanofibril (CNF), and bacterial nanocellulose (BNC) have been described. We focused on the preparation and isolation techniques to produce nanocellulose including alkaline pre-treatment, acid hydrolysis, TEMPO-mediated oxidation, and enzymatic hydrolysis. The surface modification of nanocellulose through esterification, silylation, amidation, phosphorylation, and carboxymethylation to improve the diverse applications has also been reviewed. Some invigorating perspectives on the applications, challenges, and future directions on the relevant issues regarding nanocellulose are also presented.

1. Introduction

Over the past few years, the nature-derived such as cellulose, lignin, starch, chitosan, protein, triglycerides, natural gums, and polyphenols have constituted an interesting renewable feedstock for advanced material applications [1–8]. Among these, production of novel nanomaterials, such as nanocellulose, and cellulose with nano-scale structural dimensions, has been proven to be one of the most prominent green materials. These materials have gained growing interests owing to their excellent characteristics such as being the most abundant polymer on the Earth, high aspect ratio, better mechanical properties, renewability, biodegradability, and non-toxicity [9]. Nanocellulose is described as the product or extract from native cellulose (found in plants, animals, and bacteria) composed of the nanoscaled structure material, relying on simple, scalable, and efficient isolation techniques [10]. Furthermore, nanocellulose, with size range of 10 nm–70 nm, are constructed from constituents of microfibrils (diametre of 0.1 μm–1.0 μm), whereby bundle of microfibrils are made up the single cellulose fibre [11]. The hierarchical structure of cellulose from tree to nanometre scale is shown in Fig. 1.

There are wide range of cellulose's sources including bacteria, plants, algae, and tunicates. The source of cellulose influences not only its size and qualities, but also the amount of energy used in the extraction

process to make nanocellulose [13]. Microorganisms can also manufacture nanocellulose on their own. Bacterial cellulose, also known as microbial cellulose, is produced extracellularly by gram-negative bacteria of various genera, such as *Acetobacter*, *Achromobacter*, *Aerobacter*, *Agrobacterium*, *Alkaligenes*, *Azotobacter*, *Pseudomonas*, *Rhizobium*, *Rhodobacter*, *Salmonella*, *Sarcina*, and most notably *Gluconacetobacter*. This bacterial of *Gluconacetobacter* for nanocellulose is made up of a form stable water-containing nanofiber network that contains 1% cellulose and 99.9% water [14]. In addition, trees, shrubs, diverse herbs, grasses, flowers, root vegetables, succulents, and other plants provide vast supplies of plant nanocellulose. The trees include the woody-derived (birch, poplar, eucalyptus, douglas fir and pine) and non-woody (hemp, cotton, fax, jute, sisal, tomato bean, and etc.) [15–17]. Various algae from macro and microalgae possess cellulosic cell walls like any other plants. Nanocellulose prepared from algae shows a higher cellulose crystallinity than prepared from forest wastes and agricultural residues [18]. Nanocellulose from *Eremosphaera viridis* was used for investigation toxicity of zinc oxide (ZnO), *Cladophora* sp. for the potential leachables and extractables in nanocellulose for biomedical uses and *Cystoseira myrica* in removal of heavy metals [19–21]. Nanocellulose sources from animals are from tunicates and exoskeleton (prawns and crabs). In addition, forest production of crops and agriculture residues are considered as cellulose and nanocellulosic

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materials production such as maize straw, wheat straw, coconut husk, pineapple leaf and others [22–24].

Primarily, Technical Association of the Pulp and Paper Industry (TAPPI) and multiple concerned bodies have recommended that nanocellulose can be categorised into three main groups depending on its morphology: 1) short and rigid crystalline rod-like cellulose nanocrystals (CNC), 2) more entangled cellulose nanofibres (CNF) which are long and flexible (occasionally denoted as microfibrillated cellulose (MFC)) that exists as interconnected nano-fibrillar structures, and 3) bacterial nanocellulose (BNC) that are high in purity and very crystalline [13,25–28,270]. CNF and CNC exhibit excellent capabilities in comparison to the native cellulose fibres. CNF possess mechanical robustness which exhibit high specific strength, modulus, and high surface area while CNC has unique liquid crystalline properties, high permeability and reactivity [29]. Both CNF and CNC have unique properties such as high specific area and sustainability that make them ideal candidates for reinforcing various polymeric matrices [30].

BNC is a biomaterial that has unique properties such as high crystallinity, high specific surface area, excellent permeability, good mechanical strength, high flexibility, high absorbency, and possibility of forming any shape and size. However, the overall production process of BNC still requires major improvement to become more competitive, primarily due to the low BNC productivity of the known strains, use of fine and production cost, high capital investments, and lack of well-organised scale-up processes, resulting in low BNC production [31]. Presently, the nanocellulose industry is developing rapidly, with gradually increasing industrialised production, changing consumption patterns associated with growing economies and prosperity. Thus, contributing to increase in the demand of nanocellulose in the market [32,33]. Advantages of the use of nanocellulose materials are related not only to its properties, but also in the dimensions and nanometre scale, which open a wide range of possible properties to be discovered along with potential applications across the several industrial sectors and addresses a lot of issues related to the environmental concern [34].

Based on Mordor Intelligence report, the nanocellulose market was valued at USD 570.45 million in 2020, and the market is anticipated to register a CAGR of more than 22% during the forecast period. COVID-19 pandemic has detrimentally influenced nanocellulose industry landscape. Producers are resolving the challenges caused by the pandemic and adapting to the changing dynamics of the industry. The severe impact on demand of end-use products due to the shutdown of day-to-day operational activities has limited the market opportunities for nanocellulose producers [35]. According to Silva et al., the price of nanocellulose is still volatile due to the production cost (related to the production method adopted), the marketplace, and the specific format and characteristics. Although current market prices are higher, he estimated CNC, CNF, and BNC production costs to be 15, 2.15, and 22.11 €/Kg, respectively (in dry equivalent). Despite the benefits of nanocellulose, its manufacturing costs (like those of PLA, PHA, and starch) are still higher than those of petroleum-based plastics [36].

In addition, many reports have recently been published that focus on the creation of robust nanocellulose, with research ranging from preparation of the materials to potential applications [37,38]. However, this material continues to show surprisingly high challenges to master its interactions and tailorability to allow well-controlled assemblies for functional materials [27].

Rapid advances in nanotechnology and material sciences have facilitated investigation of nanocellulose, giving rise to the emergence of various fields such as renewable energy, electronics, environment, food agriculture, biomedical, and healthcare [39–44]. This paper presents the general overview of nanocellulose from cellulose, nanocellulose properties and morphology, isolation techniques, its potential application and challenges. Compared to other review papers, the essential elements of our review paper are the section on preparation and isolation procedures, as well as surface modification of nanocellulose. This is crucial to be reviewed as in nanocellulose studies, both sections are the critical part of analysis. Overall, we covered the top down issues regarding the nanocellulose.

2. Structure and morphology of cellulose

Cellulose is the most abundant, renewable natural polymer in the world and has long been used as a sustainable building block of conventional paper [45,46]. In 2020, the annual global production of cellulose pulp is approximately 180 million tons and the demand for this material is rapidly growing due to high demand [47]. In 1838, Payen, A. first identified a major insoluble residue called cellulose. Since the discovery, thousands of scientific papers, patents, and books have been published concerning the importance of natural polymer [31]. Briefly, cellulose is a polysaccharide that composed of a linear chain of β -1,4 linked D-glucose of repeating units called anhydroglucose units (AGU), with a degree of polymerisation range from several hundreds to over ten thousand as illustrated in Fig. 2 [48]. Then, it is packed into microfibrils that are held together by intra and inter molecular hydrogen bonds as well as intermolecular van der Waals forces, with both non-reducing and reducing ends [49]. Consistently, an AGU is pivoted 180 degrees in the plane to encourage the favoured position for making an acetal bond

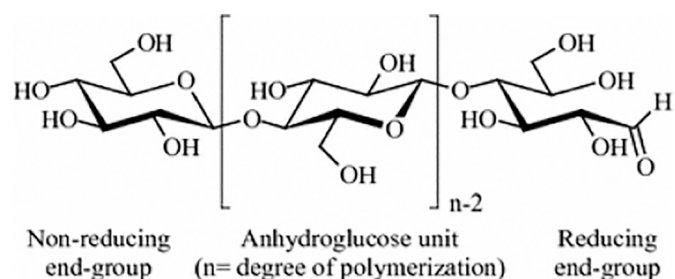


Fig. 2. Schematic representation of cellulose [27].

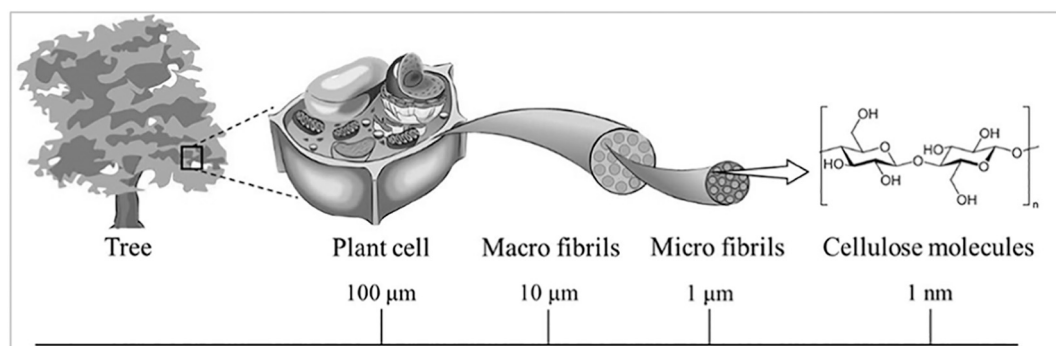


Fig. 1. Hierarchical structure of cellulose from tree to nanometre [12].

between two neighbouring glucopyranosyl rings. In this way, the two neighbouring deposits shape the auxiliary unit called cellobiose [27,50]. These intra- and inter-chain non-covalent attractions are vital for the stability and firm structure of cellulose, the latter of which is essential for plants and some marine creatures [51]. Each AGU has three hydroxyl (OH) groups accessible for chemical reaction that serve an important role in the compactness of the crystalline structure, determine the physical properties of cellulose, and create an extensive hydrogen bonding network that makes cellulose insoluble in most aqueous and organic solvents [52]. Therefore, the hydrogen bonding network must be disrupted to allow dissolution of the cellulose in cellulose homogeneous reactions [53].

Cellulose can be obtained from a broad range of sources including algae, bacteria, plants, and tunicates [54]. The cellulose has several polymorphs, namely cellulose I, II, III and IV and their varieties I α , I β , IIII, IVI, IIIII and IVII which each of these forms can be identified by its characteristic X-ray diffraction pattern [55]. Principally, cellulose is a branched homopolysaccharide of a β -D-glucose linear chain linked with β -(1-4)-glycosidic bonds in a 4C_1 conformation with the molecular formula $(C_6H_{10}O_5)_n$, with each glucose subunit of hydrogen bonds with neighbouring glucose monomers within its chain [31]. Furthermore, cellulose exists in nature as nanoscale microfibrils that are arranged in larger fibril aggregates and fibre structures [56].

Pure cellulose has a key drawback such as difficult to process due to its low solubility in typical organic solvents caused by the strong intramolecular and intermolecular interactions. Because of their superior dissolving capabilities, cellulose derivatives are a potential alternative to pure cellulose. Ethanolic or aqueous hydrolysis can also be used to convert them to cellulose. Cellulose esters (cellulose acetate), ethers (carboxymethyl cellulose, methyl cellulose, and ethyl cellulose), cellulose sulphate, and cellulose nitrate are the most well-known cellulose derivatives [57]. Cellulose acetate is a renewable substance that is gradually attracting researchers' interest due to its potential benefits as a biodegradable, non-corrosive, non-toxic, and biocompatible material. It's commonly used as a dispersant and to equally distribute nanoparticles in a solution [58].

3. Classification and properties of nanocellulose

Taking advantage of the structure and properties of nanocellulose, the current science of biomaterials aims at developing new and formerly non-existing materials with novel and multifunctional properties, with an attempt to meet the current requirements in different fields such as biomedicine, the environment, energy, pharmaceuticals, agriculture, and food. All types of nanocelluloses have a relatively similar chemical composition, there are major differences in their degrees of crystallinity, particle size, and morphological characteristics [13]. Table 1 shows the features of different types of nanocellulose.

Despite of various advantages of nanocellulose, there are some drawbacks involve with this nanomaterial. In contrast to CNF, CNC has

limited flexibility because of the removal of amorphous regions. In addition, because of random cleaving of cellulose chains during hydrolysis, the diameters of CNC are not uniform [59]. Apart from that, nanocellulose has a large amount of reactive hydroxyl group on its surface make it feasible to modify the surface properties by various chemical modifications. However, the presence of a hydroxyl group on the surface encourages agglomeration and prevents correct dispersion, so chemical modification is required to improve dispensability and compatibility with the solvent or matrix used to make polymer nanocomposite. As a result, numerous studies have focused on modifying the CNC surface to minimise the amount of hydroxyl groups per area, which reduces the intensity of particle–particle interactions and improves compatibility with hydrophobic polymer matrices [60]. The low contrast in TEM images of nanocellulose is a key limitation for morphological characterization and size determination. Da Silva et al. protocol presents an optimisation of the standard uranyl acetate protocol commonly used for biological specimens in order to suit cellulose nanomaterials. Moreover, the fabrication of CNC and CNF involve some disruption of their structure and so efforts to reduce damage during extraction are of paramount importance [61].

CNC is also known as nanocrystalline cellulose (NCC), cellulose nanowhiskers (CNW), or cellulose crystallites [62]. Normally, CNC can be obtained through acid hydrolysis of cellulose, where the cellulose is exposed to sulphuric acid (H_2SO_4) under controlled temperature and time. The acid hydrolysis destroys the amorphous regions in the crystal and between microfibrils, resulting in the remaining the crystalline sections to stick together [65]. Acids like hydrochloric (HCl), phosphoric (H_3PO_4), and hydrobromic acids (HBr) have been used for the isolation of CNC [66].

CNF is also called microfibrillated cellulose, nanofibrils and microfibrils, or nanofibrillated cellulose (NFC). It is a cellulose with a diameter less than 100 nm, consisting of both amorphous and crystalline parts [13]. CNF displays two main drawbacks that are associated with its intrinsic physical properties: (1) a high number of –OH groups, which leads to strong hydrogen interactions between the two nanofibrils and results in a gel-like structure once it is produced, and (2) high hydrophilicity, which limits its uses in several applications such as in paper-coating (increase of dewatering effect) or composites (tendency to form agglomerates in petro-chemical polymers). Therefore, the most feasible solution to overcome these disadvantages is chemical surface modification to reduce the number of hydroxyl interactions and increase the compatibility with several matrices [31]. However, CNF offers several unique advantages, whereby it can be produced in industrial scales with a variety of functional groups and by a multitude of industrially-attractive processes for several industrial applications [67].

BNC is a nanoscale form of cellulose that is bio-technologically produced as an exopolysaccharide by some aerobic non-pathogenic bacteria (for example, *Komagataibacter* (formerly *Acetobacter*), *Agrobacterium*, *Aerobacter*, *Achromobacter*, *Azotobacter*, *Rhizobium*, *Sarcina*, *Salmonella*, and *Escherichia*), which combines the properties of cellulose

Table 1
Features of different types of nanocellulose.

Nanocellulose	Length (nm)	Diameter (nm)	Morphology	Properties	Applications	References
Cellulose nanocrystal (CNC)	100–250	5–70	Cylindrical, elongated, less flexible, and rod like nanoparticles	Hyperfine structure, high transparency, high purity, high crystallinity, high strength and Young's modulus, and high reactivity	Biomedicine, pharmaceuticals, electronics, barrier films, nanocomposites, membranes, and super capacitors	[62,63]
Cellulose nanofiber (CNF)	>1000	5–60	Entangled network structure with flexible, long, and wide nanofibers	With 50–70% crystallinity, possessing electrostatic charge, extremely high surface, uniform dispersion, high chemical tunability, exceptional hydrophilicity	Pharmaceuticals, electronics, paper industries, cosmetics, food, and biomedical sectors	[13,56,63]
Bacterial nanocellulose (BNC)	0.001	100	Dense and well-organised three-dimensional network of fibrils that forms porous sheets	Hydrophilicity, relatively large specific surface area, broad potential of surface chemical modification	Textiles, cosmetics, and food products, and it has a high potential for medical applications	[31,64]

with the features of nanomaterials [68]. The production of bacterial cellulose consists of two important processes: fermentation and purification [69]. BNC is characterised by considerably higher crystallinity (84%–89%), excellent chemical purity (lack of impurities such as hemicellulose, lignin, or pectin), high tensile strength in comparison to other biomaterials, high water absorption capacity, high biodegradability, good moldability, and smaller cross-section of fibres that contributes to high porosity of this material [70,71].

4. Preparation and isolation techniques

The methods of preparation of nanocellulose vary based on its sources and final applications. The pre-treatment and preparation techniques used to produce nanocellulose must be conducted under controlled conditions in order for nanocellulose to be obtained at not only high yield but also with the desired size and properties [72]. Fig. 3 illustrated the various techniques for isolation of nanocellulose.

The plant fibres are usually employed in preparing CNC in abundance for low prices [69]. The methods for obtaining the CNC and CNF are the processes of decreasing the size of natural cellulose, which includes the process of mechanical, chemical, biological hydrolysis processing, or a combination of all [74–76]. Preparation of CNC via mechanical method is a physical method that consists of grinding, homogenisation under high pressure, ultrasonication, ball-milling, microfluidization and cryogenesis [77]. The advantages of using mechanical methods are that it is a simple operation, it uses relatively inexpensive equipment, has broad applicability to most types of biomass, and can easily produce nanocellulose in large quantities at room temperature and pressure [66]. Supian et al. characterised CNF by isolating it from empty fruit bunch (EFB) through nano-grinding treatment. They found that the nano-grinding treatment does not give significant changes on chemical and thermal behaviour compared to cellulose. The crystallinity of the fibre showed a minimum increment after the mechanical treatment [78]. CNF were prepared from sugarcane bagasse pulp by mechanical grinding mixed with nisin to prepare CNF/nisin nanohybrid films. This novel material exhibited good light transmission, relatively high tensile strength, low oxygen permeability, and low water vapour transmission rates [79]. In the research work by Lopes et al., they analysed the nanofibrillated cellulose of Eucalyptus sp. obtained by different grinding intensities. The nanofibrils presented average

diameter of approximately 30 nm and reduced crystallinity index and viscosity of the nanofibrillated cellulose compared to cellulose before processing [80]. For instance, Zheng et al. obtained CNF by a combined process of enzymatic hydrolysis and grinding (EG-CNF), grinding and microfluidisation (GH-CNF), and TEMPO-mediated oxidation and grinding (TE-CNF). Results showed that CNF fibrillation was promoted as times of passes increased in microfluidisation, and CNF that was pre-treated by enzyme possessed shorter length. Crystallinity of CNF was related to CNF manufacturing methods, while CNF film's transparency was correlated to CNF diameter distributions [81].

High-pressure homogenisation generates several disruptive forces that can partially disintegrate the structural organisation of cellulosic materials, including cavitation, turbulence and shear effects, and break down cellulosic materials by generating intense cavitation forces. Wang et al. isolated nanocellulose from pre-treated wood and bamboo wastes by high-pressure homogenisation (HPH) in homogeneous media. The results showed that the crystallinity indices increased with good stability of suspension. However, the thermal stability of nanocellulose was lower than that of raw fibre [82]. Furthermore, Huang et al. prepared nanocellulose from cotton linter using ionic liquid (IL) 1-butyl-3-methylimidazolium hydrogen sulphate (HSO_4) as both swelling agent and catalyst, followed by high-pressure homogenisation. The nanocellulose had a uniform rod-like shape that was 50 nm to 100 nm in diameter and 500 nm to 800 nm in length [83]. Susanto et al. investigated the influence of high-pressure homogeniser process on the crystalline structure and morphology of the BNC membrane. This study showed that the crystalline structure of BNC membrane changed and the crystalline index decreased from 88% to 86%, while the particle size reduced after applying high-pressure homogenisation [84].

In addition, Tan et al. studied the effectiveness of using ultrasonification and cryocrushing methods for extraction of nanocellulose from kapok husk. It was found that when the ultrasonication time was increased, the size of the samples (viewed under the optical microscope) became more invisible. Besides, nanocellulose produced from 80 min of ultrasonication with higher degradation temperature has more applications in the field of engineering compared to nanocellulose produced from 20 min of ultrasonication and frozen for 24 h and 48 h of cryocrushing [85]. Moreover, Szymańska-Chargot et al. investigated the effect of ultrasonication on physicochemical properties of apple-based nanocellulose-calcium carbonate composites. They concluded that

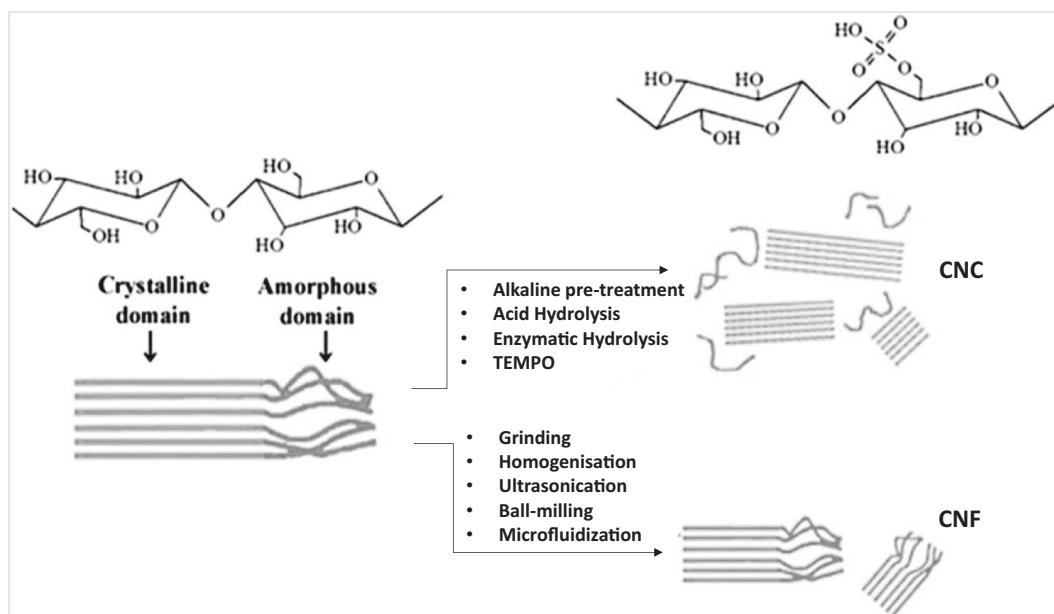


Fig. 3. Various isolation techniques of nanocellulose [73].

ultrasonication improved the composite rigidity [86]. A correlation on ultrasonication with nanocrystalline cellulose characteristics was constructed by Ishak et al. They revealed that the ultrasonic pre-treatment improved the hydrolysis process of cellulose in ionic liquids with increasing yield and crystallinity of nanocellulose [87]. In another work, Jimat et al. isolated nanocellulose fibres from Cocoa Pod (*Theobroma cacao* L.) via chemical treatment combined with ultrasonication. The result proved that the fibres had nano-dimension of diameter of 20 nm–30 nm [88].

Ball milling is a simple, fast, and cost-effective green technology with enormous potential [89]. Research reported by Souza et al. involved soybean straw to prepare nanocellulose via a ball mill, and in-situ modified with an anionic surfactant. The modified-nanocellulose showed slightly superior thermal stability with lower crystallinity and crystal size than unmodified nanocelluloses due to the surfactant chains' addition and influence during the milling process [90]. Ferreira et al. developed a combination of ball milling, followed by a high-intensity ultrasound irradiation to obtain a suspension of nanocellulose from Eucalyptus sawdust. The resulted nanocellulose suspension exhibited reduction in diameter, increase in homogeneity, crystallinity, electrostatic and thermal stability [91].

A microfluidizer composed of an intensifier pump accelerates the slurry into the interaction chamber under high pressure. Using a high shear rate and impact forces against colliding streams and channel walls, this approach defibrillates the fibres to nanoscale dimensions. The homogenous diameters of these nanoparticles are fewer than 100 nm. However, because this procedure uses a lot of energy, pre-treatments have been recommended as alternative way to save energy [92]. According to Kassab et al., they produced carboxylated-CNF with diameter only 2.5 nm via microfluidization techniques. The small diameter and negatively charged surfaces of CNF help it disperse in a variety of mediums. In contrast to cellulose microfibrils (CMF), which flocculates in water, CNF has great colloidal stability in water [93]. Further, it is possible to lower the amount of chemicals used for bleaching and the energy used in the mechanical microfluidization process during the manufacturing process [94].

In case of biological treatments, cellulosic materials are treated with cellulolytic enzymes like cellulase that cleave the fibre structures to simpler ones, but since biological treatment takes more time, they are often coupled with mechanical or chemical methods to reduce the process time and get better CNF [95]. Apart from that, enzyme endoglucanase-assisted mechanical grinding was able to produce CNF with controllable length of 298 nm–4500 nm and diameter of 9 nm–26 nm, while improving CNF yield and energy-efficiency [96].

4.1. Chemical method

Due to great energy consumption from mechanical methods, chemical process was focused for obtaining nanocellulose. Besides, chemical hydrolysis offers great advantages to produce CNC by partially breaking the glucosidic bonds [97]. Chemical hydrolysis involves alkaline pre-treatment, acid hydrolysis, enzymatic hydrolysis, and TEMPO hydrolysis with other substances, e.g. phthalimide-N-oxyl (PINO) radical, deep eutectic solvents (DES), organic acids, ionic liquids, choline chloride and other substances [98,99]. Acidic and alkali pre-treatments have shown great promise in biomass solubilisation because of their multiple advantages. For example, it requires a simple device, it has ease of operation, high methane conversion efficiency, and low cost [100]. Acidic hydrolysis is performed using acids such as sulphuric acid (H₂SO₄), hydrochloric acid (HCl), phosphoric acid (H₃PO₄), and others while alkali pre-treatment usually employs several alkaline solutions, inducing sodium hydroxide (NaOH) and potassium hydroxide (KOH).

4.1.1. Alkaline pretreatment

The composition of natural fibre is divided into three main components which are cellulose (30–80%), lignin (3–33%), and hemicellulose

(7–40%) [101]. The difficulties to break the inner hydrogen bonds of the supramolecular structure of cellulose were overcome by using alkali. If the cellulose is linked to the fibrillar structure that is present in the fibre naturally, it would make it difficult for the acid to diffuse through the amorphous regions of the cellulose and produce reasonable amounts of insoluble lignin, thus generating secondary products or precipitation during the process hydrolysis [102]. This was proved by the report from Septevani et al., which stated that the cellulose content after simultaneous pre-treatment processes increased significantly compared to the raw material of oil palm empty fruit bunches [103]. Furthermore, Rui et al. also reported that after treatment with KOH, CNF contained majority of cellulose with a small portion of residual xylan, lignin, and extractives [104]. An alkaline treatment involved NaOH was also reported by Bongao et al., where they did extraction of nanocellulose from waste pili pulp as a potential anti-aging ingredients for cosmetics formulations. The infrared spectra of the extracted sample showed relevant peaks of OH, CO, and CH, confirming the presence of cellulose [105].

Indeed, alkali treatment and bleaching steps are essential to remove impurities, especially lignin and hemicelluloses when starting directly from biomass or even biowaste, thus caused the cellulose content of raw material to be drastically increased [106]. After the first treatment with alkali, bleaching is usually performed, which involves single or multiple stages depending on the end-use applications. However, alkaline treatment and bleaching process can be pre-treated simultaneously prior to hydrolysis process [107]. According to Chieng et al., they first treated oil palm mesocarp fibre (OPMF) with alkali and then bleached it before the production of CNC by acid hydrolysis (H₂SO₄). The changes in the peaks of FTIR spectrum indicated that the alkali treatment completely removed hemicelluloses and lignin from the fibre surface [9]. Zheng et al. utilised walnut shell to obtained nanocellulose. After subjected to NaOH treatment, the lignin and hemicelluloses contents decreased, whereas the cellulose concentration of walnut shell increased. Meanwhile, after bleaching treatment, the cellulose content increased to 87.9% due to the removal of remaining lignin and hemicelluloses, resulting in highly purified cellulose [108]. Additionally, CNC was isolated for the first time from pine-cones (PC) by alkali and bleaching treatments and subsequently, H₂SO₄ hydrolysis (64%) at 45 °C, as studied by Garcia-Garcia et al. The removal of non-cellulosic constituents during the alkaline and bleaching treatments resulted in high pure cellulosic fibres [109].

The presence of alkali also served the following functions: (1) swelling agent to cellulose, (2) leading to an increase of internal surface area, (3) leading to a decrease in the degree of polymerisation and crystallinity of cellulose, (4) leading to partial solvation of hemicellulose, (5) destroying the structural linkages between lignin and carbohydrate by saponification of intermolecular ester bonds, and (6) disrupting the lignin structure by breaking its glycosidic ether bond. Lignin fails to act as protective shield to the cellulose after lignin solubilisation step, thus making extracted cellulose more susceptible to nanosynthesis [110]. In another aspect, *alkali pre-treatment* is capable of modify the surface of particles and improve the thermal stability by eliminating the lignin and hemicellulose from fibre [111]. The basic principles of alkali hydrolysis are based on solvation and saponification, which induce depolymerisation and cleavage of lignin-carbohydrate linkages, accordingly rendering the uneasily-biodegradable substances to become more accessible to the extracellular enzyme. In addition, it solubilises the xylan hemicellulose by saponifying the intermolecular ester bonds (for example, acetyl and uronic acid substitutions), though to a less degree than the acidic pre-treatment [100]. Table 2 displays some alkaline pre-treatments prior to hydrolysis for nanocellulose productions.

Among the alkaline reagents, NaOH is the most effective pre-treatment agent. NaOH solutions cause the breakage of intermolecular α - and β -aryl ether linkages between hemicellulose and lignin, causing swelling of the biomass pores, disruption of the lignin structure, and an increase in available surface area [117]. In terms of concentration,

Table 2
Alkaline pre-treatments prior to hydrolysis for nanocellulose production.

Plant source	Conditions	Results	References
Pinecone	4.5% NaOH for 2 h at 80 °C under vigorous mechanical stirring	Resulted in a sample with a lighter brown appearance, indicating the partially removal of lignin, pectins and hemicelluloses	[109]
Walnut	2% NaOH, stirred for 4 h at 100 °C	The lignin and hemicelluloses contents decreased to 30.98% and 7.6%, respectively, whereas the cellulose concentration of increased to 56.6%.	[108]
Sugarcane bagasse pulp	1 L of 100 g/L, 300 g/L, and 500 g/L NaOH solution, frozen at –20 °C	Adhesion levels becomes more severe, the number of pores formed through fibre interweaving increases, and fibre dispersion levels decrease	[17]
Cotton	5% NaOH, stirring constantly at room temperature for 4 h	Minimised the hydrogen bonding by eliminating hydroxyl groups by a reaction with NaOH	[16]
Pineapple crown	5% NaOH, at 70 °C at room temperature, under magnetic stirring for approximately 1 h	The disruption of the binding of lignin, hemicellulose, and other components with cellulose during treatment	[99]
Ramie fibres	2% NaOH, stirred at 90 °C for 2 h	Indicated for the elimination of hemicellulose and lignin.	[112]
Corn cob residue	16% KOH dosage at 70 °C for 90 min	Efficient removal of lignin and extractives	[113]
Corn cob	16% KOH at 70 °C for 90 min	Contained majority of cellulose with a small portion of residual xylan, lignin, and extractives	[104]
Sugarcane bagasse	2% KOH solution, stirring at 85 °C in a water bath for 2 h	Increased its cellulose and hemicellulose content	[114]
Cassava peel	5% KOH solution at 25 °C, stirred for 14 h	Processes used were not enough to remove all existing fractions of lignin	[115]
Licorice residue	5% KOH solution at 25 °C for 5 h.	Removed most of lignin in licorice residues	[116]

NaOH concentration of 4% is sufficient and optimum for the alkali treatment and cellulose extraction. Meanwhile, at NaOH concentration of 2%, the cellulose fibres still looks closely packed, which suggests that the expected ineffectiveness of the 2% NaOH treatment due to the stronger hydrogen bonding remains between the adjacent fibres [118]. According to Nie et al., as alkali concentrations increased, adhesion levels became more severe. Then, as the number of pores formed through fibre interweaving increases, fibre dispersion levels decrease. This is attributed to the phenomenon that changes occur in the crystal structure of cellulose that in turn spur changes in the entropy elastic force, causing the axial shrinkage of fibres and adhesion between fibres [17]. In another work by Chi et al., it was proven that the increase of KOH dosage declined the solid recovery, mainly due to the efficient removal of lignin and extractives, thus leading to the increase of glucan content in the pre-treated corn cub residue [113].

The aim of KOH treatment is to remove residual lignin and hemicelluloses. Besides, it can improve swelling ability of pulp fibres, which can help to improve the purity of cellulose, and further enhance the enzymatic accessibility of pulp fibres. Therefore, using a high dosage of effective alkali leads to degradation of cellulose and hemicelluloses rather than improvement of delignification intensity. So, it is not

necessary to input additional alkali into the pulping process [116].

Furthermore, the addition of base avoids the necessity of high temperature, and thus the process can be operated at ambient or moderate temperatures, possessing zero physical hazard, has low risk of inhalation danger, is not carcinogenic, persistent, or bioaccumulative [70,100,268]. According to the results reported by Faria et al., the mercerisation secondary process with NaOH at 70 °C shows greater efficiency. They suggested that the disruption of binding of lignin, hemicellulose, and other components with cellulose can be considered from the colour aspect of the fibre after mercerisation and bleaching, and pulp formation at the end of the process [99]. Moreover, alkaline treatment at temperature –20 °C converts cellulose I to cellulose II. The cellulose II CNF has the opposite polarities to the others, and the neighbouring CNF would intermingle, resulting in irregular aggregation [17]. Other than that, the low-concentration cold alkali treatment has strong solubility to hemicellulose. After low-concentration cold alkali pre-treatment, the crystallinity of CNF significantly reduced, and the crystal structure of the cellulose changed from type I to type II. Cold alkali destroyed the original crystalline structure of cellulose, changed the lattice of cellulose, and converted some cellulose from type I to type II, reducing the crystallinity of CNF [119]. On the other hand, KOH pre-treatment could be implemented under relatively mild conditions due to the strong basicity of KOH. Therefore, KOH pre-treatment has a high potential of feasibility [113].

The significant changes of fibre morphologies after treatment with alkali could be observed after it became cleaner, and roughness was reduced due to the removal of impurities from the fibre surface. Impurities such as wax and cuticle on the fibre surface were removed by the interaction with sodium during alkaline treatment using NaOH. Besides, based on study by Awang et al., the surface morphology of the alkali-treated waste office paper was smoother than the untreated ones. The yield had the highest crystallinity index and crystallite size of 36.35% and 32.92 nm, respectively [120]. Furthermore, the microstructure of cellulose fibre consisted of discontinuous regions of crystalline cellulose and amorphous cellulose, whereby the unconfined amorphous region was between two inerratic crystalline regions. Therefore, the amorphous regions needed to be removed quickly while keeping the crystalline regions [74].

However, the main disadvantages related to this pre-treatment are the black generated hydrolyzate and processing time. These problems can be reduced by using this pre-treatment in combination with others, which results in requirement of lower concentration alkaline solutions and processing times [117]. Chieng et al. stated that the change in colour of fibre after treatments is related to the degradations of cellulose, hemicellulose, and lignin substances, while the degradation process is normally accompanied by weight loss and alteration of chemical compositions of fibre [9].

4.1.2. Acid hydrolysis

The acid hydrolysis is performed to the chemically purified cellulose (CPC) to remove amorphous domains [112]. In general, an acid hydrolysis process involves the conversion of cellulose and hemicellulose polymers to their monomers. After this treatment, the cellulose that is released can be degraded to glucose (saccharification) or carried out to another series of processes to take further advantage of it [70]. Acid pre-treatment is a process to break the rigid structure of lignocellulosic material, in which hydronium ions breakdown and attack intermolecular and intra-molecular bonds among cellulose, hemicellulose, and lignin in the biomass hierarchy structure. The acid hydrolysis includes concentrated and dilute acid solutions where different levels of acid severity contribute to various biomass fractionated products [78]. Table 3 shows the acid hydrolysis from strong (sulphuric acid (H₂SO₄), hydrochloric acid (HCl)) acids and weak acids (phosphoric acid (H₃PO₄), citric acid (CA) and formic acid (FA)). The vital requirement in hydrolysis process is to keep in the basic structure of cellulose backbone. In the acid hydrolysis process, oxygen (cyclic or glucosidic) of cellulose

Table 3
Acid hydrolysis from strong and weak acids.

Acids	Conditions	Raw materials	Crystallinity Index (%)	Production yield (%)	Size (nm)	References
H ₂ SO ₄	40 °C, 1 h stirring,	Cotton fibre	91.2			[16]
H ₂ SO ₄	45 °C for 30 min with 58% H ₂ SO ₄	Ramie fibre	90.77		145.61	[112]
H ₂ SO ₄	40% H ₂ SO ₄ at 45 °C for 3 h	Extremophile spinifex grass	73	42	497 ± 106	[122]
H ₂ SO ₄	45 °C and stirred for 60 min, using 10%, 30%, 60% H ₂ SO ₄	Pinecone flower waste		22.22	110–125 185	[123]
H ₂ SO ₄	41 min, 50 °C, 51% H ₂ SO ₄	Brewery's spent grain	76.3	43.24	309.4	[124]
H ₂ SO ₄	123.35 min, 45 °C, 62.2% H ₂ SO ₄	Tea stalk	61.32	49.87	4–8	[125]
H ₂ SO ₄	64% H ₂ SO ₄ , 45 °C, 1.5 h	Corn husk	83.51		26.9	[126]
H ₂ SO ₄	hydrolysis reaction for 1 h at 45 °C	Walnut shell	40.1		55–82	[108]
H ₂ SO ₄	65%, temperature 55 °C, 60 min	Pineapple peels			18 ± 6	[127]
HCl	6 M HCl at 45 °C for 70 min	Ramie fibre	89.61		158.51	[128]
HCl	3 M HCl at 80 °C	Oil palm empty fruit bunches (OPEFB)		21, 18, and 19		[129]
HCl	3.5 M HCl at 60 °C, 20 h	Water hyacinth fibre	84.87		147.4	[130]
HCl	3.5 M HCl at 50 °C for 12 h		82.7		88–1100	[12]
HCl	stirrer at 450 rpm at 80 °C, centrifuge at 4500 rpm for 30 min	Sugarcane bagasse	68.09 66.76	68.72 66.76	4.12 4.24	[131]
HCl	4 M HCl in the hydrothermal kettle at 110 °C for 3 h	Commercial microcrystalline cellulose		73 (FeCl ₃), 77 (AlCl ₃), 78 (CuCl ₂), 80 (MnCl ₂)	222 ± 33 (MnCl ₂), 206 ± 16 (CuCl ₂), 178 ± 34 (AlCl ₃), and 152 ± 39 (FeCl ₃)	[132]
HCl	80 °C for 4 h, centrifuged at 20,000 rpm	Microcrystalline cellulose			195 (average radii)	[133]
CA	80% acid concentration, 100 °C, 4 h	Sugarcane bagasse pulp	77.3	>95	250–450	[134]
CA	10%, 30%, and 60% acid, 45 °C, 60 mins	Pine flower waste		95.6%, 94.6%, and 93.4%, 80.28		[135]
CA	FeCl ₃ -catalysed, 85% CA at 80 °C for 6 h	Bleached eucalyptus kraft pulp	80		6–12	[136]
H ₃ PO ₄	H ₃ PO ₄ 62%, at 20,000 rpm for 40 min	Oil palm empty fruit bunch			12.5 ± 2.6	[137]
H ₃ PO ₄	stirred at 800 rpm at room temperature for 120 min	Cassava residue				[138]
H ₃ PO ₄	3.5 h, 50 °C, centrifuged at 10000 rpm for 15 min	Oil palm empty fruit bunch	86		>500	[139]
FA	80–98% 70–100 °C, 24 h	Bleached eucalyptus kraft pulp	79			[140]
FA	150 rpm stirring, at 25 °C for 30 min	Microcrystalline cellulose		95		[141]
FA	Stirred at 300 rpm for 12 h at 95 °C	Bleached softwood kraft pulp	12.46	75	50–200	[142]

is rapidly protonated by the action of H⁺ acid. Subsequently, glucosidic bonds are scissored tardily and motivated by the addition of H₂O molecules [121].

H₂SO₄, HCl, and H₃PO₄ have been used on an industrial scale due to their low cost, and lower toxicity, and is therefore, commonly used to break the glycoside bonds in cellulose [68]. Initially, the lignin and other compounds like proteins were removed using during pre-treatment with NaOH and temperature before treatment with acid hydrolysis. In this step, the lignin ester bond could be broken, and the carboxyl groups were oxidised. After the hydrolysis with acid, this treatment promoted the partial hydrolysis of the cellulose, mainly by breaking the glycosidic bonds, and decreased the size [100].

The structural complexity and composition of the raw materials determine the type of treatment in nanocellulose extraction. Most lignocellulosic biomass include agricultural residue (for example, rice husk, and bagasse), and terrestrial plant (for example, bamboo, and wood) require delignification prior to the acid hydrolysis. The amorphous region is more accessible to acid and subsequently degraded during hydrolysis process. The glycosidic bonds cleave, and the degradation continues during the hydrolysis process. The repulsion forces, contributed by the electrical double layers, allow nanocellulose to disperse in water and form stable colloidal suspension. It has fewer tendencies to aggregate compared to the neutral surface charged hydrochloric acid-hydrolysed nanocellulose [143].

This method is easy and fast to produce nanocellulose that has better properties. One of the benefits of acid isolation is the abundance of hydroxyl (OH) groups in CNC, which makes CNC more reactive and

hydrophilic [144]. In addition, chemical hydrolysis, particularly acid hydrolysis, has been proven as an effective solution for generating highly pure CNC [145].

Negatively charged nanocellulose can be produced through H₂SO₄ hydrolysis [146]. In the synthesis of nanocrystalline cellulose, chemical isolation with acid hydrolysis is an often-used technique. The surface charges' repulsion forces from acid hydrolysis enable CNC to disperse in an aqueous medium, creating a stable colloidal suspension [144]. The characteristics of gained CNC are strongly influenced by several parameters including type and concentration of acid, temperature, time, and acid-to-pulp ratio [112]. Amorphous domains are randomly oriented and arranged, inducing a lower density of these domains, and are thus more vulnerable to acid hydrolysis, especially to the infiltration of hydronium ions H₃O⁺ leading to hydrolytic cleavage of glycosidic bonds [106].

Recently, attention has been given to nanocellulose produced by bacteria due to the unique properties and environmentally-friendly nature of the product when compared with plant cellulose [96]. Bacterial cellulose (BNC) is synthesised in highly crystalline and high-water content form by many bacteria, such as *Gluconacetobacter* (formerly *Acetobacter*), *Agrobacterium*, *Rhizobium*, and *Sarcina* [147]. *Gluconacetobacter xylinus* is the most widely-studied producer of extra-cellular pure cellulose [148]. For isolation and purification of BNC, no pre-treatments are required due to the presence of pure cellulose [149]. In other words, BNC does not require any pretreatment to remove the noncellulosic components as the synthesised cellulose has a distinct superior purity and crystallinity [150]. During BNC production, the

microbial cells polymerise various carbon sources into single linear β -1,4-glucan chains that protrude out through the pores located on the cell membrane. Then, the synthesised β -1,4-glucan chains get assembled in the culture medium and form protofibrils, which further crystallise into micro- and macro-fibrils, bundles, and ultimately form ribbons-like structures [151].

In recent research, BNC tends to be reproduced as nano-sized particles with high crystallinity, through acidic or enzymatic known as nanofication of BNC or bacterial cellulose nanocrystals (BCNC) [31]. The negative charges of BCNC prevent its aggregation of nanocrystals driven by hydrogen bonding, and then, the stable well-dispersed nanocrystal suspension can be obtained [73]. The general isolation process for BCNC from BNC is founded upon acid hydrolysis. The amorphous (disordered) and crystalline (ordered) regions of BNC transform BNC into a stable suspension of cellulose nanocrystals, adding new functionality to the material through hydrolysis with strong acids [96]. Amorphous or disordered sections of cellulose are better hydrolysed by hydrogen ions, enhancing hydrolytic cleavage of the glycosidic bonds, which releases individual crystallites of the remaining intact crystalline regions that have a higher acid resistance [152]. It was observed that BCNC obtained by enzymatic hydrolysis leads to particles of similar length of 250 nm, but in a greater amount of nano-sized particles [153].

4.1.2.1. Acid hydrolysis by strong acids. Acid sulphuric (H_2SO_4) hydrolysis has become the most extensively-used method to isolate cellulose nanocrystal (CNC) because it has been proven effective in the elimination of amorphous components to release crystalline part, resulting in stable CNC suspensions [9,119]. The other advantages of using H_2SO_4 in CNC production are that a more uniform and shorter with a narrow polydispersity of CNC can be produced, which has a higher crystallinity of more than 90%, and better stabilisation of solution against flocculation. However, the residual sulphate groups (SO_4^{2-}) can trigger dehydration, causing the decompositions reaction, thus leading to lower thermal stability that limits its application in high-temperature processing polymer production, such as, extrusion, and injection moulding. To overcome these problems, many studies have been done by using various kinds of acid, for example HCl, and H_3PO_4 , to improve the thermal stability properties of nanocellulose [70]. In the case of H_2SO_4 hydrolysis, the surface of cellulose attains half ester sulphate groups, resulting in high negative charges. Hydrogen bonding and polar bonds between surface OH groups of CNC and protein molecules create non-covalent interactions and electrostatic interaction of positive charge moieties (for example, lysozyme) of protein with CNC negatively-charged surface of SO_4^{2-} groups [144]. Moreover, H_2SO_4 hydrolysis not only removes the amorphous region but also dissolves the crystalline region partially. The decomposition at lower temperature originates from its highly-sulphated regions, while the degradation at higher temperature results from the decomposition inside unsulphated crystalline domains. Briefly, Vanderfleet et al. Cranston explained that CNC are produced by H_2SO_4 hydrolysis, whereby cellulose chains undergo two reactions: hydrolysis of glycosidic bonds and esterification of surface hydroxy (OH) groups. The hydrolysis of glycosidic bonds, which occurs rapidly within the less-ordered regions of cellulose, decreases the length of cellulose chains until mostly crystalline regions remain. Simultaneously, a fraction of the surface OH groups is esterified to form sulphate half-ester groups that are anionic under practical working solution conditions [154].

Morphology is imperative to study the obtained nanocellulose to understand the sizes, smoothness, and fibrillation [102]. Determination of the fundamental properties such as morphology and dimensions of CNF and CNC is a prerequisite for improving the structure–property and structure-processing relationships through a wide range of applications [150]. According to Kusmono et al., the chemical purification increased the crystallinity owing to the dissolving of hemicellulose and lignin. The crystallinity index of CNC was higher compared to that of both untreated

ramie fibres and CPC because the removal of amorphous components (due to H_2SO_4 hydrolysis) and structural changes in organisation and alignment produced highly ordered crystal bundles. However, at the higher temperature, acid hydrolysis removed amorphous components and also some parts of crystalline, accelerating the hydrolytic cleavage of the glycosidic bonds, and finally resulting in a decreased crystallinity [112]. More recently, Matebie et al. synthesised CNC using acid hydrolysis of H_2SO_4 with further alkali treatment of 2% NaOH from barley brewery spent grain. They reported that CNC had rod-like whisker shape with crystallinity of 76.3% and an average particle size of 309.4 nm [124]. Other than that, Shaheen et al. isolated CNC from wood sawdust as a new resource for CNC. The crystal structure of CNC obtained is obeyed cellulose type I with crystallinity index about 90% and average dimensions (diameter \times length) of $35.2 \pm 7.4 \text{ nm} \times 238.7 \pm 81.2 \text{ nm}$ after subjected with 65% H_2SO_4 [121]. In another research, Theivasanthi et al. used H_2SO_4 -mediated synthesis for nanocellulose production. They found that the nanocellulose with nearly 91.2% crystalline had a structure of proper order. They also reported that the rod shape of nanocellulose increased the area of the surface and created fibre to be more reactive [16].

Acid hydrolysis separates the cellulose fibres by destroying the hydrogen bonds between the cellulose polymers, hence reducing the diameter of fibres. The forces produced during sonication were sufficient to break some of the bonds between the glucose unit, resulting in depolymerisation and shorter fibre lengths in the nanometre range [130]. H_2SO_4 hydrolysis resulted in nanoparticle size in the range of 100 nm–1000 nm. H_2SO_4 is known as a very strong hydrolytic agent that offers nanocellulose with a short and uniform nano size compared to other acid hydrolysis [137]. The nanoscale rod-like cellulose structures with the diameter of CNC ranging from 50 nm to 100 nm CNC were obtained by Gunathilake et al. This was caused by more densely packed fibrous cellulose networks due to the intermolecular hydrogen bonding and strong hydrophilic interaction between the cellulosic chains [144]. Lusiana et al. produced CNC using 10%, 30%, and 60% of H_2SO_4 solutions from the waste of pine flower afford the dimension of CNC in about $15 \text{ nm} \times 185 \text{ nm}$. They revealed that the amorphous nanocellulose was produced from hydrolysis using 60% of H_2SO_4 , but 10% and 30% H_2SO_4 provided similar crystallinity of nanocellulose [123].

As reported by Amiralian et al., under optimised conditions, relatively long and thin nanocellulose particles (consisting of individual fibres, as well as some agglomerated bundles) still exhibited a slightly curved appearance like CNF rather than presenting a more typical flat, straight, and rigid structure. This was due to the controlled acid hydrolysis process, whereby the amorphous hemicellulose near the surface was readily hydrolysed and lignin slowed down the cellulose hydrolysis process and was condensed by H_2SO_4 . Therefore, this acid could not easily penetrate into the crystalline structure of fibril bundles [122]. On the other hand, Guo et al. proved that the nanocellulose from tea stalk, wood, and microcrystalline cellulose obtained rod-like particles in the range of 4 nm–8 nm width and average widths of 6.36 nm, 7.02 nm, and 6.24 nm, respectively, from hydrolysis with H_2SO_4 . At higher hydrolysis temperatures greater than 45 °C, the amorphous domains and some parts of the crystalline domains were eliminated, which then lead to reduced diameters and lengths of CNC [125]. Furthermore, acid hydrolysis is a well-known harsh process, which caused most of the disordered regions of cellulose to be hydrolysed during the acid hydrolysis process, leaving behind the rod-like cellulose crystallites, and resulting in a small aspect ratio of acid-hydrolysed nanocellulose [126].

The yield of CNC is dependent not only on the source of cellulose, but also on the conditions for the isolation of CNC. In general, longer reaction times yield shorter nanocellulose particles. In the study by Le Gars et al., a higher yield of CNC was obtained by the combined use of mechanical pre-treatment of cellulose, acid hydrolysis with controlled hydrolysis conditions (such as acid concentration, hydrolysis time, and temperature), and post-treatment (such as homogenisation and ultrasonication) [106]. The hydrolysis process needs the H_2SO_4

concentration to be 60%–65%, reaction temperature to be 40 °C–50 °C, and reaction time to be 30 min–60 min. However, the yield of CNC was very low (less than 30 wt%) due to the excessive degradation [45]. In another research work, Yang et al. developed a novel method for the simultaneous preparation of CNC and CNF through dilute H₂SO₄ hydrolysis. They claimed that during the hydrolysis process, acid content had a significant effect on the production of both nanocelluloses. Then, they found that higher acid content can promote the hydrolysis degree of cellulose until it reaches at one point that was beneficial to the production of CNC, while the yield of CNF decreased as the hydrolysis degree of cellulose was increased [75]. In another work, Peng et al. reported a new protocol for efficient and high yield preparation of CNF in a short time, using less water during acid hydrolysis treatment. They reported that the acid-assisted CNF yield was 91.6% from dissolving pulp when 55 wt% H₂SO₄ was applied at 45 °C for 40 min with diameter of 10 nm–20 nm, while the length varied from 40 nm to 800 nm. According to them, the main reason that nanocellulose yield was about 30%–40% from traditional acid hydrolysis was that the higher concentration H₂SO₄ leads to excessive degradation, and cellulosic solid residues from acid hydrolysis were underutilised. Therefore, it may be possible that a milder acid-assisted treatment may be combined with a subsequent alkali neutralisation process to overcome degradation and low yields. Hydrogen bonds between fibres and β-1,4 glycoside linkages can be hydrolysed, but much less degradation would occur [155].

It is crucial to mention that long contact times and variation of temperature decompose the nanocellulose to small organic molecules and producing a dark brown colour [127]. In their research work, Lin et al. investigated the applicability in CNC fabrication with a variety of hydrolysis conditions from dry hardwood pulps. They claimed that impurities other than silicon and magnesium were almost fully removed by H₂SO₄ hydrolysis, especially with a high acid concentration. However, silicon and magnesium were not removed through alkaline extraction nor acid hydrolysis [145].

H₂SO₄ is mainly used for hydrolysis because it produces stable nanocellulose suspension in water, but H₂SO₄ is more expensive than HCl. Nanocellulose hydrolysed with HCl does not have good dispersion in organic solvents and tendency to agglomerate due to the lack of charges at the surface of CNC [106]. The agglomerate phenomenon may be attributed to the intermolecular hydrogen bonding and a strong hydrophilic interaction in between the cellulosic chains [128]. H₂SO₄ is strongly oxidising, and sometimes causes degradation of cellulose because sulphate groups behave as a catalyst for oxidative decomposition, which has a negative effect on large-scale production and applications of CNC. Therefore, strong hydrolysing acids, other than H₂SO₄, and particularly HCl, have been widely used to prepare CNC. Unlike H₂SO₄ hydrolysis, HCl hydrolysis affords unmodified charge-neutral CNC. In addition, CNC isolated by HCl hydrolysis shows some superior features compared with sulphated CNC; CNC prepared by HCl treatments form stable oil/water interfaces called Pickering emulsions, whereas sulphated CNC do not show this interfacial behaviour [129]. Furthermore, due to the lack of charge on the surface of CNC, the production of CNC prepared by HCl are easily flocculation in water but the thermal stability of CNC by HCl is higher than that by H₂SO₄ [45]. Besides, HCl does not result in stable suspensions due to the absence of surface charge, and thus, the length of CNC fibres decreased, while increasing the hydrolysis time by increasing the surface charge [119]. The treatment with HCl also promoted partial hydrolysis of the cellulose, mainly by breaking the glycosidic bonds to decrease size. Then, HCl hydrolysed samples exhibited more intermolecular hydrogen bonding [145].

The crystallinity study of produced nanocellulose is necessary to understand the effect of production methods on the crystal structure of cellulose [102]. Asrofi et al. successfully isolated and characterised nanocellulose from water hyacinth fibre (*Eichhornia crassipes*) (WHF) in 3.5 M HCl at 60 °C and 500 rpm for 20 h. The resulted nanocellulose crystallinity index and crystalline domain area significantly increased

after chemical treatment. The highest crystallinity index was 84.87% after acid hydrolysis process, which leads to good thermal stability [130]. Apart from that, Aprilia et al. utilised sugarcane bagasse to isolate nanocellulose as filler in polymer based on a combination process between acid hydrolysis and continued with the ultrasonication process. They confirmed that the combined process of HCl hydrolysis and homogeneous with ultrasonication process could be utilised as an effective chemical and mechanical process for nanocellulose production from various cellulosic sources [131]. In addition, different extraction protocols lead to different properties of length, crystallinity, aspect ratio, and amount, and type of surface functional groups (such as hydrolysis with HCl gave hydroxyl CNC) that were selected as a toughening agent in epoxy adhesives. In other work, Cheng et al. introduced four inorganic chlorides into HCl hydrolysis to extract CNC from microcrystalline celluloses (MCC) under hydrothermal conditions. They demonstrated that the introduction of inorganic chlorides enhanced the hydrolysis process through faster degradation of the disordered region of cellulose and the use of salt-catalysed hydrolysis, especially ferric chloride (FeCl₃) [132].

Moreover, nanocellulose can be obtained from hydrolysis of mixture from H₂SO₄ and HCl with greater crystallinity compared to the original cellulose, and possessed a higher density of OH groups [156].

4.1.2.2. Acid hydrolysis by weak acids. Acid hydrolysis using concentrated mineral acids is an efficient method to prepare nanocellulose, specifically cellulose nanocrystals CNC. However, the method exhibits several critical defects, namely it is hazardous to the environment and human body, causes corrosion to the process equipment and over-degradation of raw cellulose material, is high cost, and has large water usage [77]. Therefore, the method that have potential for commercial success and lower toxic risks are required to make applications environmentally-friendly, sustainable, and create new bio-based nanomaterials in high-tech fields, such as biomaterials [150].

While CNC produced with H₃PO₄, a weak acid, possesses phosphate half-ester groups, the increase in colloidal stability over uncharged CNC from other mineral acid hydrolyses is small [154]. According to Wang et al., H₃PO₄ is a medium-strong acid (pK_a = 2.12) and tends to ionise, releasing H⁺, in aqueous solutions. The glycosidic bonds in cellulose have a low stability in acidic media and are prone to multiple hydrolysis processes. The accessible area of cellulose was hydrolysed first, and the area with a lower accessibility was then hydrolysed. Hemicellulose was degraded because the glycosidic bonds in hemicellulose could be cleaved by the water molecules in an acidic medium, which is similar to the acid hydrolysis of cellulose [157].

Accordingly, H₃PO₄ hydrolysed samples created low surface charge of hydrolysed nanocellulose [106]. On the other hand, Septevani et al. successfully prepared oil palm empty fruit bunch (EFB)-based nanocellulose as a super-adsorbent for water remediation via mixture of H₂SO₄ and H₃PO₄ hydrolysis. They discovered that the long-entangled nanofibre cellulose obtained from the phosphoric acid hydrolysis had a diameter of 12.5 ± 2.6 nm and a length that exceeded 500 nm due to the less aggressive nature of mild acid in attacking amorphous regions other than stronger acids such as H₂SO₄. They also stated that nanocellulose from phosphoric acid hydrolysis had a rougher surface area and agglomerate than that obtained from H₂SO₄ [137]. In other publication constructed by Septevani et al. in 2019, they synthesised nanocellulose obtained from oil palm empty fruit bunches via two different methods, which were strong (H₂SO₄) and mild acid (H₃PO₄) hydrolysis. They claimed that strong acid hydrolysis produced a rod-like shape and lower aspect ratio with unprecedented crystallinity index. Meanwhile, long-entangled nanofibrils were produced from mild acid hydrolysis [103].

Surprisingly, while most studies show lower thermal stability of sulphuric-based nanocellulose compared to mild acid based nanocellulose, Hernandez et al. studied the thermal stability of the H₂SO₄ hydrolysis and reported that it was relatively similar to its counterpart at

the temperature of 260 °C [139]. The thermal property of nanocellulose was indispensable to be used in the composites [102]. The lower thermal stability may be due to the smaller dimensions of CNC, which provide a larger surface area accessible to heat treatment [9]. In 2018, Huang et al. prepared nanocellulose by a mechanochemical method using cassava residue as a raw material and H₃PO₄ as the auxiliary agent. They noted that nanofibres still had the basic structure of cellulose, indicating that mechanochemical processing did not destroy the cellulose structure, whereby the lower thermal stability may be due to the smaller dimensions of CNC, which provide a larger surface area accessible to heat treatment [138].

The use of organic acids such as citric acid (CA) to obtain nanocellulosic materials with good colloidal and thermal stability appears to be a promising strategy since they have multiple advantages such as being biodegradable, relatively inexpensive, and environmentally-friendly. This is attributed to the COOH and OH groups in amorphous cellulose chains that provide hydrophilic characteristics, while the crystalline regions confer hydrophobic behaviour and the replacement of OH with COOH groups in the cellulose can reduce the hydrogen network and increase the repulsion and absorption capacities of the CNC [158]. Nasihin et al. derived bioplastic isolated nanocellulose from pine flower (*Pinus merkusii*) waste by hydrolysing CA using concentrations of 10%, 30%, and 60%. They revealed that the increase of CA concentration in the hydrolysis process damaged the crystalline area of nanocellulose [135]. Liu et al. stated that CA is much weaker than most of inorganic acids and intended to enhance the hydrolysis efficiency for highly efficient preparation of CNC. The addition of FeCl₃ significantly enhanced the hydrolysis efficiency even though with a small loading amount [136].

Another type of weak acid such as formic acid (FA) hydrolysis was demonstrated as an effective pre-treatment for the preparation of cellulose nanomaterials because the cellulose fibres were swelled and broken down during FA hydrolysis that benefited the subsequent fibrillation. Therefore, FA hydrolysis is a clean and sustainable route in the production of thermally-stable and functional CNC [140]. In a study conducted by Wu et al., integrated and sustainable preparation of functional nanocellulose via FA and choline chloride solvents pretreatment produced yield of nanocellulose over 95%. They stated that the surface ester groups had better ability to stabilise the oil/water interface for further preparation of oil-in-water Pickering emulsion [141]. According to Liu et al., the production yield of CNC was lowered due to insufficient hydrolysis, as the acidity of FA was weaker than mineral acids such as H₂SO₄, but the CNC yield was comparable with other results using other organic acids. They also pointed that FA had a high selectivity in hydrolysing hemicellulose and cellulose could react with FA at room temperature without using a catalyst [142].

4.1.3. TEMPO-mediated oxidation

2,2,6,6-Tetramethylpiperidin-1-oxyl (TEMPO) is a series of reagents used to oxidise the alcohol and ether, including TEMPO and its ramifications with the structure of 1-oxyl, an oxygen radical [11,159]. In principle, the TEMPO radical helps introduce additional negatively carboxylic (COOH) groups to the fibres. This allows individual fibres to form dispersions in water, as opposed to the removal of non-crystalline (amorphous) sections of the cellulose material by acid hydrolysis, which then releases the nanosized elements [160]. Various pre-treatment methods such as TEMPO-mediated oxidation have been proposed to reduce the energy required for the mechanical deconstruction process by reducing the negative or positive charge on the fibre surfaces and enhance the colloidal stability of final cellulose nanofibrils [108]. Although the energy consumption could be remarkably reduced in this case, there are still many drawbacks in the existing such as low processing efficiency, the use of hazardous reagent, and difficulty in chemical recovery [140]. Furthermore, this method exhibited some shortcomings, such as toxic reagents, tedious steps, and limited oxidation positions [161]. Table 4 displays TEMPO-mediated oxidation from

Table 4
TEMPO-mediated oxidation from natural sources.

Source	Nanocellulose	Findings	Conditions	References
Sugar cane bagasse	CNF	Length distributed between 60 and 1820 nm	TEMPO/NaBr/NaClO system in water at pH 10	[114]
Oil palm empty fruit bunches (OPEFB)	Nanocellulose	72% crystallinity degree	TEMPO/NaBr/NaClO system in water at pH 10	[160]
Aspen wood	CNF	The yield of CNFs after oxidation was determined to 47.4 ± 0.7%	TEMPO/NaBr/NaClO-system at pH 6.8	[162]
Lemon seeds	CNC	344.5 nm in size	TEMPO/NaBr/NaClO system in water at pH 10	[148]
Walnut shell	Nanocellulose	Exhibited an irregular block structure, low thermal stability	pH 5 using 0.1 M NaOH and 0.1 M HCl, 5 h at room temperature under magnetic stirring at 1000 rpm	[108]
Bleached bagasse pulp	CNF	Carboxylic content increased, finer in shape, higher adsorption capacity	TEMPO/NaBr/NaClO system in water at pH 10	[163]
Dissolving pulp	CNF	Used in low-fat food products due to their availability and excellent viscosifying and gel forming abilities	TEMPO/NaBr/NaClO system in water at pH 10.5	[164]
1) Corn stalk pulp, 2) rape stalk pulp	CNF	Increased the amounts of COOH groups	TEMPO/NaClO system	[165]
Wood pulp	CNC	TEMPO oxidation did not induce any significant change in terms of CNC dimensions.	TEMPO/NaBr/NaClO system in water at pH 10	[166]
Fique tow	Nanocellulose	Exhibited a network-like morphology and high values of carboxylate content	TEMPO/NaBr/NaClO system in water at pH 10	[167]
Bleached bisulte pulp	CNF	Stabilized the negative charges on the CNF surface	TEMPO/NaBr/NaClO system in water at pH 10	[168]
Peelings (inner peel and bark) of cassava roots	CNF	5–16 nm diameter	TEMPO/NaBr/NaClO system in water at pH 10	[115]
Banana pseudo-stem	Nanocellulose	Reduced the thermal stability of nanocellulose	TEMPO/NaBr/NaClO system react for 3 h under constant stirring	[104]
	CNF			[116]

(continued on next page)

Table 4 (continued)

Source	Nanocellulose	Findings	Conditions	References
Licorice residue		Stretched rod-like structures, which may be due to the negatively charged groups such as –COOH	TEMPO/NaBr/NaClO system in water at pH 10	

various natural sources.

The mechanism of oxidation reaction of TEMPO generates nitronium ions (+N=O) in situ during the reaction of TEMPO radicals with the oxidants and they are used to oxidise the cellulose fibres. Consequently, the primary alcohol groups are first oxidised to aldehydes and further converted to COOH groups. At the same time, the depolymerisation phenomenon of cellulose appears. There are two phenomena which may be caused by cellulose depolymerisation: (1) β -elimination could occur, owing to C6 aldehyde groups presented in an alkaline environment, and (2) the cleavage of a hydroglucose unit takes place, owing to the existing hydroxyl radicals [101]. CNC can be obtained using TEMPO-mediated oxidation, followed by mechanical disintegration, which can position-selectively oxidise C6-primary OH groups of cellulose to sodium C6-carboxylate groups. Then, the introduction of COOH by the TEMPO-mediated oxidation of C6 primary OH groups on the cellulose surface could result in a significant decrease in thermal degradation [121]. Additionally, in the analysis carried out by Kumar et al., it was revealed that the TEMPO oxidising treatment induced a modification of the C6 carbon, resulting in 24% carboxylate groups in CNF, with 74% of C6 carbons remaining unmodified. Overall, the sum of unmodified and oxidised C6 contributions was 99%. This proved the self-consistency and accuracy of the analysis but interestingly, no aldehydes were observed within their detection limit [168].

Besides, TEMPO-mediated oxidation did not affect cellulose polymorph. The increase of carboxylate content by increasing the amount of primary oxidant (NaClO) indicated that more carboxylate and aldehyde groups were formed in the water-insoluble fractions by the oxidation [169]. On the other hand, Balea et al. proved that TEMPO-mediated oxidation increased the amounts of COOH groups because bleaching pretreatment removed the lignin content, leaving cellulose to be more accessible for the TEMPO-mediated oxidation. The presence of lignin and impurities in refined pulp consumed part of the NaClO and, therefore, the oxidation of cellulose happened to a lower extent [165]. Zheng et al. stated that the original crystal structure of the cellulose was unchanged after the TEMPO-oxidation of nanocellulose. This showed an indication that the carboxylate groups formed by the TEMPO-mediated oxidation were selectively introduced on the surfaces of the cellulose microfibril rather than the internal cellulose crystallites [108]. Additionally, Abou-Zeid et al. prepared CNF through sequence oxidation steps with TEMPO and later, CNF-TEMPO-oxidised with periodate-chlorite oxidation. They reported increase of carboxylic content from 0.9 to 3.5 as repulsion forces were increased among the fibres [163]. According to Czaikoski et al., a minor increase in crystallinity index (45%) nanocellulose was obtained after TEMPO-mediated oxidation was associated with chemical treatment that only transformed the surface OH into carboxylate groups, without interfering with the internal conformation of cellulose crystals. Besides, they concluded that the removal of amorphous regions resulted in the formation of higher crystallinity. Principally, it was due to the introduction of COOH groups into the primary alcohol groups on the surface of cellulose after TEMPO oxidation [161].

The study carried out by Lusiana et al. briefly explained that after oxidation of periodate-chlorite, the CNF-TEMPO-oxidised was finer in shape, compared to CNF-TEMPO at the first stage while after TEMPO-oxidation treatment. Then, the diameter of the fibres is around 20 nm, with different lengths up to a few micrometres, but after periodate/

chlorite oxidation, the fibres become smaller in diameter and length [103]. Additionally, after TEMPO pre-treatment of the cellulose fibres, followed by disintegration via a mild magnetic stirring, the CNF can be easily obtained with a native elementary width of 3 nm–5 nm [11]. In contrast, CNC-carboxylated from TEMPO reaction has been reported by Jonasson et al., who involved only subfraction of raw cellulose materials to be oxidised to CNC, but the rest of oxidised cellulose had much larger dimensions. It indicated that TEMPO was not as effective as other cellulose oxidation methods. However, CNC from TEMPO oxidation was highly carboxylated which provided opportunity for functionalisation [170]. Furthermore, Faradilla et al. conducted an extensive study to characterise a film produced from banana pseudo-stem nanocellulose. They reported that nanocellulose obtained from TEMPO-mediated oxidation of banana pseudo-stem (BP) had diameter of 10 nm–25 nm with length in several micrometres. They explained that the changes in average crystal sizes of CNF could be associated to a removal of amorphous hemicellulose and lignin fractions as a result of applied chemical treatments of the biomass and a consequent recrystallisation of cellulose fibrils into larger crystallites [114]. Besides, as reported by Zhang et al., CNC obtained by TEMPO oxidation showed a larger size than that of H₂SO₄ hydrolysis and ammonium persulfate (APS) oxidation [161].

The reduced stability of samples by the introduction of sodium carboxylate groups in CNF during TEMPO-mediated oxidation leads to decarbonation during heating process and decreases the thermal stability [114]. In addition, the TEMPO-mediated oxidation is also known to reduce the thermal stability of nanocellulose due to the COOH groups that are introduced to the cellulose C6 of cellulose [104]. For instance, nanocellulose produced using walnut shell reported by Zheng et al. exhibited lower thermal stability using TEMPO than nanocellulose-hydrolysed H₂SO₄ because it had fewer crystalline regions, which have shown to provide thermal stability [121].

Different from H₂SO₄-hydrolysed nanocellulose, TEMPO-oxidised nanocellulose have long flexible fibre networks and consist of alternating crystalline and amorphous cellulose domains as concluded by Shen et al. The CNF obtained by TEMPO-mediated oxidation are more uniform and can be well dispersed in an aqueous phase [171]. In addition, Czaikoski et al. explained that acid hydrolysis produced more dispersed (separated) nanofibres than the samples obtained by TEMPO-mediated oxidation. The pre-treatment together with acid hydrolysis or TEMPO-mediated oxidation must cause the superficial hydrolysis that favour the ultrasonic treatment to penetrate the amorphous inner region, facilitating the formation of smaller cellulose fragments, such as cellulose nanospheres [115]. As for TEMPO-oxidised CNF, although cellulose can be degraded in alkaline environment of TEMPO-mediated oxidation, the oxidation mainly happened on the surface of amorphous and crystalline regions and had little effect on the morphology of pulp fibres [116]. Besides, Aaen et al. investigated the fundamental rheological properties of TEMPO-oxidised CNF, both in pure water and in electrolyte solutions. Low electrolyte additions strengthened the CNF network by allowing stronger interactions, while higher additions led to fibril aggregation, and loss of viscosity, especially under shear. The fibrils of the sample CNF seemed to be somewhat more aggregated than the samples with higher charge densities [164].

Jonasson et al. reported that the yield of CNF after oxidation was expected to be high considering the hemicellulose sparing treatment coupled with an overall minor degradation effect. The yield efficiency of these oxidative treatments has been acknowledged even in the case of the harsher treatments where yields corresponding to retention of all cellulosic solids [172]. Furthermore, the isolation of CNF from the hardwood pulp by Kafy et al. was done using TEMPO method that resulted in the fabrication of long fibre that exhibited maximum tensile modulus of 23.9 GPa with the maximum tensile strength of 383.3 MPa, and high strain at break (indicating high toughness) [173].

4.1.4. Enzymatic hydrolysis

Acid hydrolysis and mechanical treatments are the most widely

employed methods for obtaining cellulose nanocrystal (CNC) and nanofibril (CNF), respectively. However, these procedures have some drawbacks related to economic and environmental aspects such as the energy demand of the process and the high amount of water required in the neutralisation steps. Therefore, another possibility for producing nanocellulose is the use of enzymatic hydrolysis, which is a promising environmentally-friendly and sustainable route due to the advantages of enzymatic treatments, although the cost of enzymes may be an inconvenience [117]. Other disadvantages are related to the common method used for CNC production that uses concentrated mineral acid hydrolysis with sulphuric acid (H₂SO₄) and hydrochloric acid (HCl), which if compared to enzymatic hydrolysis such as acid hydrolysis, requires higher capital investment for reactors, tanks, and piping. Besides, on account of the corrosive nature of reactants, the effluent produced is harmful to the environment and the operational costs may increase due to the necessity of intensive effluent treatments [174].

Recently, utilisation of enzymes for the isolation of cellulosic bundles has served as a new chance to achieve high yield and almost pure cellulose. Furthermore, it offers potentially high saccharification efficiency, less energy consumption, high penetration power, and avoidance of pollution. These active molecules are in their high specificity, which makes them suitable candidates for the degradation of cellulosic biomass [169]. Generally, the enzymatic treatment does not modify the cellulose because it only attacks the amorphous regions of the cellulose microfibrils, thus reduces the energy requirements during the mechanical fibrillation by a homogeniser or microfluidiser [11].

As an emerging method, enzymatic hydrolysis may be an interesting pathway to CNF production since enzyme hydrolysis does not generate toxic residues as mineral acid hydrolysis does. Enzymatic hydrolysis conditions are normally held in mild thermal and pressure conditions, resulting in a lower energy-intensive process. Therefore, nanocellulose produced by enzymes normally has more high-valued applications due to its morphologies [174]. In recent years, several trade companies (for example, Celluclast, Accelerase, Spezyme CP, and Viscoferm) have made efforts to increase the efficiency of cellulolytic enzymes by using a process that is economically viable to improve the enzymes' resistance to operational conditions (such as temperature and pH), or by increasing their speed of production and decreasing their prices [73]. Nanocellulose production from enzymatic hydrolysis is shown in Table 5.

Biological enzymes catalyse the hydrolysis of cellulose fibres that makes the fibrillation much easier [102]. Cellulase is a multicomponent

enzyme system composed of its active components included endoglucanases (EG), cellobiohydrolases (CBH), and β -glucosidase (GB). EG mainly acts on the amorphous region of cellulose to randomly hydrolyse the β -1,4-glycosidic bond, so the long chain cellulose molecules are truncated into small molecular cellulose in the interior of cellulose. CBH mainly acts on the end of linear cellulose molecule to destroy the crystalline region of cellulose, while GB is used to hydrolyse cellulose into glucose [175]. When CNC are prepared by enzymatic hydrolysis, the destruction of crystalline area of cellulose by CBH should be avoided as much as possible. Therefore, three components of cellulose should be separated from each other. EG is used to hydrolyse cellulose amorphous regions, and many more crystalline regions are retained in the process [45]. However, as far as the selectivity target is concerned, cellulase enzymes are not selective, as cellulose is a mixture EG, exoglucanases or CBH, and GB each of which performs a specific hydrolytic action [153].

Kumari et al. isolated the CNF from lignocellulosic waste of lemon grass using enzymatic hydrolysis, along with ultrasonication through mild operating. The obtained CNF had a web-like structure along with long entangled cellulosic segments, with homogeneity of the nanofibres [175]. However, CNF isolated by Aprilla et al. from enzymatic treated bamboo fibres via mechanical homogenisation exhibited short rod-like structure due to the combination of mild enzymatic hydrolysis and mechanical homogenisation processes, which promoted fibre cutting [176]. In a study carried out by Rossi et al., much longer and more thermostable CNF from enzymatic reaction of recombinant enzymes were obtained as compared to the CNF prepared by TEMPO-mediated oxidation. This result showed that the actions of hydrolytic and oxidative enzymes can be used as a fine treatment prior to the sonication step to produce CNF cellulose with advanced properties [114]. For instance, Cesare et al. performed an enzymatic hydrolysis for BC production hydrolysed by using two enzymes: endo-1,4- β -glucanases (EG) and cellulase. The authors revealed that the hydrolysis mediated by EG yielded big particles, and for EG added with cellulase. Moreover, the number of big (micron-sized) particles was much higher for the enzymatic hydrolysis mediated by EG, whereas the combined hydrolytic process led to a higher number of nanoparticles [153]. It can be seen from the research reported by Wang et al., the average diameter of CNF was reduced at higher intensity of enzymatic pre-treatment. Under the combined action of enzymatic hydrolysis and homogenisation, the diameter was gradually reduced until nanometre scale, meanwhile a large amount of polar hydroxyl groups on the surface of fibres was exposed [178].

Nie et al. proved that after pre-treatment with xylanase, the crystallinity index of CNF increased. This shows that some loosely structured hemicellulose was removed through the xylanase treatment, which reduced the volume of amorphous content, while increasing the crystallinity index [17]. In another research work, Alonse-Lerma et al. generated CNC using a single enzyme offered special capabilities towards crystalline nanocellulose obtention. The CNC showed a needle-like morphology (due to its high aspect ratio), and an opaque dispersion in which aggregates and sedimentation were apparent due to the absence of charged groups [177]. Based on a study by Chu et al., the use of cellulase and low-concentration cold alkali pre-treatments (combined with ultrafine grinding and high-pressure homogenisation) is an environment-friendly method for preparing nanocellulose. According to the findings, the pre-treatment with cellulase did not change the crystal structure of cellulose but increased the crystallinity of cellulose. The fibres were also smaller and resulted in greater regeneration of cellulose, thereby reducing the thermal stability of CNF [119].

In the research of Chi et al., the effect of KOH dosage of pre-treatment on glucose yield was conducted after enzymatic hydrolysis with a high solid loading (15%) and a low enzyme loading from corncob residue (CCR). It was found that the glucose yield of raw CCR was the same whether with low enzyme loading or with the high enzyme loading [113].

Moreover, Xu et al. studied the effect of enzymatic time hydrolysis

Table 5
Nanocellulose production from enzymatic hydrolysis.

Source	Enzyme	Size (nm)	Yield (%)	References
Lemon grass	Viscozyme®L	105.7 (length)	57	[175]
Sugarcane bagasse	Recombinant enzymes: endoglucanase, xylanase, and lytic polysaccharide monoxygenase	31 (average diameter)		[114]
Sugarcane Bagasse pulp	Xylanase	80–100 (diameter)		[17]
Bamboo culms	<i>G. scortechinii</i>	15.75 (diameter)	86	[176]
<i>Thermobifida halotolerans</i>	Endo-1,4- β -glucanases (EG)	0.0018–0.002 (length)		[153]
<i>Bacillus subtilis</i>	Cel5A bacterial endoglucanase	408 (average length)		[177]
Bagasse pulp	Cellulase	30 (diameter)		[119]
Corncob residue	Cellulase		91	[113]
Corncob residue	Cellulase	Less 50 (diameter)		[104]
Licorice residue	Cellulase	137–415 (diameter)		[116]

for enzymatic hydrolysis residues (HER) from corncob. They concluded that with the increase of enzymolysis time, the relative proportion of lignin significantly increased the content of xylan, and extractives did not change greatly with enzymolysis time due to the strong specificity of enzyme [104].

5. Surface modification of nanocellulose

Despite various advantages, nanocellulose exhibits limitations that restrict its widespread applications, such as poor thermal stability, incompatibility with hydrophobic polymers, and absorption of moisture. The presence of large number of hydroxyl groups (OH) within the nanofibre's structure may act as a structured platform through different substituents to employ nanocellulose for high value materials, by providing additional functionalities via surface modification [179]. Tortorella et al. stated that the OH group at Position 6 was 10 times more reactive than the other OH groups. The reactivity of the OH group at Position 2 was found to be twice that of the OH group at Position 3. Therefore, chemical modification should be mild in order to preserve the other useful properties of pristine nanocellulose [180]. Diverse surface-modification routes are now being developed to improve the miscibility and interfacial compatibility of nanocellulose with hosting matrices and confer new functionalities [181]. Table 6 shows some surface modification of nanocellulose.

Conventional surface modification of CNC can include covalent attachment of functional groups (derivatisation), molecules, polymers, and/or nanoparticles on the surface of the CNC, which provides various properties and functions for the development of CNC with high-value-added applications [182]. These modifications of nanocellulose aim to overcome the problems such as: (1) hydrogen bonding between the three hydroxyl groups comprising the repeat unit of CNC that occurs in aggregates, and (2) differences in the corresponding polarities, whereby

CNC is poorly dispersed in non-polar polymer matrices [183]. Chemical modifications of CNC can be classified into three groups: (1) native surface chemistry during the isolation/purification process or as a result of similar methods of surface treatment, (2) physical adsorption to the surface, and (3) covalent bond formation or derivatisation of the surface [181].

Furthermore, after acetylation, hydrophobic cationisation, polymer grafting and surface alkylation, cellulose III nanocrystals (CNC-III) keep the original crystalline morphology, but the crystallinity decreases, while dispersibility and thermal stability of CNC-III samples changes [184].

A common problem in producing BNC is that the BNC is usually obtained with a random fibril, which prevents the BNC from being utilised to its full potential in biomedical field applications [185]. Due to the lack of appropriate functionalities to trigger the initial cell attachment and control the porosity, and very slow degradation, BNC has been modified by chemical and physical by means of applying versatile in situ and ex situ methods [186]. BNC modification with an amino acid, methanesulfonic chloride, and toluenesulfonyl chloride possesses high antibiotic activity against *Escherichia coli* and *Staphylococcus aureus*, and can be used in medicine as a long-acting wound dressing [187].

In many cases, chemical modification (such as esterification, silylation, and amidation) is used to generate a hydrophobic surface on the nanocellulosic materials that occurs on the nanocellulose [11].

5.1. Esterification

Esterification is the most well-known and simple approach for reducing the hydroxy density of nanocellulose. Acetylation of nanocellulose is the most widely studied esterification reaction that introduces an ester functional group COO on the surface of cellulose nanoparticles by condensation of a carboxylic acid group (COOH) and

Table 6
Surface modification of nanocellulose.

Modification types	Nanocellulose	Coupling agent	Application/findings	References
Esterification	CNC	Acid anhydride	PBS-matrix-nanocellulose composite exhibited the optimal overall properties (including dispersity and balanced mechanical strength)	[183]
Esterification	CNC	N-morpholino-3-methoxyazetidinium, N,N-dihexyl-3-methoxyazetidinium and N,N-diallyl-3-methoxyazetidinium	Pronounced changes in rheological behaviour due to the surface modification	[189]
Esterification	CNC	ϵ -caprolactone and δ -decalactone monomers	Acted as fillers or reinforcements of polymeric materials	[190]
Esterification	CNC	Acid anhydride	Keep the original crystalline morphology but the crystallinity decreased, the dispersibility and thermal stability of modified samples have been changed.	[184]
Esterification	CNF	Acetic anhydride	Acetylated CNF and PLA formed smooth surface composite films with good transparency, mechanical properties, tensile strength, thermal stability, and higher hydrophobicity	[191]
Silylation	CNF	Methacryloxypropyltrimethoxysilane (MPS)	Improved the compatibility with the PSf matrix.	[195]
Silylation	CNF	Polypropylene homopolymer	Wide improvement of the mechanical properties of silylated cellulose nanofibers-reinforced composite series	[192]
Silylation	Nanocellulose-TEMPO	3-Aminopropyltriethoxysilane	Used as bionanocomposite reinforcing elements	[160]
Phosphorylation	CNC	Phosphoryl chloride and hexachlorocyclotriphosphazene	Efficient as adsorbent for removal of textile pollutants	[194]
Phosphorylation	CNF	Diammonium hydrogen phosphate	A facile and reproducible strategy to generate phosphorylated cellulose nanofibrils from SCB.	[200]
Phosphorylation	CNF	H ₃ PO ₄	For Copper adsorption from aqueous solution	[198]
Carboxylation	CNC, CNF	Chloroacetic acid	Acted as nanofiller for carboxymethyl cellulose (CMC) film	[201]
Carboxylation	CNC, CNF	Citric acid	Environmentally friendly, sustainable, and new bio-based nanomaterials in high-tech fields, such as biomaterials	[12]
Carboxylation	CNC	Citric acid	Highly efficient production of carboxylic CNCs with great dispersibility and high thermal stability	[136]
Amidation	CNC	1-Methyl-3-phenylpropylamine via EDC, NHS	Directly graft aromatic molecules on the surface of the CNC through amide linkage	[166]
Amidation	Nanocellulose	Octadecylamine via EDC, NHS	Product became more hydrophobic as more hydrophilic of the starting material	[167]
Amidation	CNF	Furfurylamine via EDC, NHS	Enables a precise estimation of the drug loading for the development of drug delivery application	[168]

alcohol group (OH) [181]. Anionic groups, such as sulphate (SO_4^{2-}), COOH, and phosphate groups, are commonly introduced to the surface of nanocellulose to improve the charge repulsion [119]. The cellulose esterification process basically uses acid anhydrides or acyl chlorides as acetylating agents in the presence of a small amount of either sulphuric or perchloric acid [183]. The first stage of acetylation occurs with the most accessible OH groups located on the surface and/or in the disordered/amorphous regions of nanocellulose. The acetylation reaction then progresses into less accessible OH groups, such as those inside the cellulose crystals [133,188,270].

For instance, He et al. modified CNC with different length chains of acid anhydrides. They confirmed that through an esterification reaction, hydrogen in the OH of CNC was substituted by a hydrophobic ester group. OH reacted with acid anhydride in the presence of H_2SO_4 , and the oxygen atoms of CNC underwent acid-induced deprotonation to O^- ions, and then reacted with the acid anhydride through an esterification reaction [183]. In the typical process of preparing CNCs by H_2SO_4 hydrolysis, SO_4^{2-} groups were introduced into C6 of cellulose via esterification, thus significantly increasing the negative charge density on the surface. CNC obtained via H_2SO_4 hydrolysis were able to form stable aqueous suspensions, consisting mostly of individual fibrils and few aggregates [119]. Besides, the attachment of anionic sulphate-ester groups to the surface of CNC particles by Mahsuli et al. resulted in electrostatic repulsion between particles and prevented aggregation [71].

Besides that, acetylation increased the thermal stability due to lower water content by the presence of acetyl groups, which minimised the water absorption. Therefore, it reduced the degradation by hydrolytic cleavage, and conferred greater thermal stability to the nanoparticles. This behaviour has a significant great advantage in terms of processability in the production of nanocomposites [133]. In other work by Sahlín-Sjökvöld et al., functionalisation of the CNC produced by H_2SO_4 hydrolysis with azetidinium salts caused the azetidinium reagents to react with the negatively charged SO_4^{2-} groups, lowering the negative charge on the surface. The hydrogen was removed through grafting of the azetidinium substituents and replacing the hydrogen ion (associated with the SO_4^{2-} groups) with a sodium ion. The acid catalysed dehydration was inhibited, thus improving thermal resistance [189]. Besides, Wu et al. investigated some typical modification routes included in the acetylation of CNC. They demonstrated that after modification the thermal stability of the samples showed a lower onset decomposition temperature. They concluded that the externally-attached polymer and long chain alkane coating generated a new cross-linked material in the thermal crosslinking reaction [184].

Díaz de León et al. proposed plasma-induced polymerisation as a strategy to modify nanocrystals at different plasma power intensities using ϵ -caprolactone and δ -decalactone to improve their compatibility with polymeric matrices. In this work, the surface modification of cellulose nanocrystals via plasma-induced polymerisation was studied using the monomers caprolactone and decalactone, and applying different plasma input powers in the plasma reactor [190]. Jamaluddin et al. confirmed that the average diameter of CNF after the chemical modification acetylation of CNF with acetic anhydride was below 100 nm. The average diameters of CNF fibres in solvents were maintained at 50 ± 8 nm and 72 ± 13 nm for chloroform and acetone, respectively, and showed some aggregation in the chloroform. However, after sonication, the dispersion was improved and continued to use for composite preparation with poly(lactic)acid (PLA) [191].

5.2. Silylation

Silylation is the most common coupling method used to combine CNF and non-polar matrixes due to its simplicity, inexpensiveness, and availability of the chemicals compared to acetylation [192]. Silane surface modification is a simple way to increase the hydrophobicity of a hydrophilic cellulose surface [193]. Silylation introduces hydrophobic

alkyl moieties, such as chlorosilanes, onto nanocellulose [188]. It should be noted that whereas the surface modification was increased directly proportional to silane ratio on the reaction, the aggregation of nanofibrils was also increased. However, in certain applications, the increment of silane modification also had substantial repercussions on the crystallinity of the CNF by the addition of amorphous components to the crystalline unit but clearly, silane surface modifications enhanced the hydrophobic character of the nanofibrils [194].

Silylated cellulose was successfully synthesised by Indarti et al. using TEMPO-oxidised nanocellulose (TEMPO-NC) from oil palm empty fruit bunch (OPEFB) and 3-aminopropyltriethoxysilane used as bio-nanocomposite reinforcing elements. The researchers confirmed that the silylation of cellulose generally involves three stages: (1) production of silanols, (2) absorption of the silanol and the cellulose hydroxyl groups via H-bonding, and (3) covalent bonding through the formation of Si-O bridges of Si-O-cellulose and Si-O-Si bonds on the cellulose surface. They also mentioned that silylation not only occurs on the OH, but more importantly on the TEMPO-NC carboxylic moiety of C6', which is postulated as being the primary factor for this successful modification [160].

Zhang et al. studied direct surface modification of CNF in the presence of methacryloxypropyltrimethoxy silane (MPS). From the report, they concluded that MPS's OH was powerfully hydrogen united with that of CNF. The existence of silanol groups which were featured to silanol, siloxane or something similar that came from the acid-condensation reactions with MPS able to improve the compatibility between polysulfone (PSf) and CNF. They also presented a mechanism for MPS on the CNF surface: (1) hydrolysis of methoxy group ($-\text{OCH}_3$) on MPS: three methanol molecules were yielded during the interaction of one MPS molecule and three water molecules, (2) self-condensation of $-\text{Si}-\text{OH}$ which forms $-\text{Si}-\text{O}-\text{Si}-$ bearing oligomers with different status: linear chain, branched chain, endless chain, or even three-dimensional polymer, and (3) formation of hydrogen bonds between the hydroxyl groups of CNF surface and oligomers at room temperature: the hydrophobic groups were introduced into the outermost surface of CNF in this process, which was beneficial to the compatibility between CNF and PSF [195].

5.3. Amidation

A carbodiimide-mediated reaction is the most common way to amidate cellulosic surfaces. The reaction usually targets the carboxylic (COOH) groups of pre-oxidised nanocellulose substrates [10]. The most common conditions involve the use of N-(3-dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride (EDC) and N-hydroxysuccinimide (NHS), whereby EDC activates the carboxylic acid to form an O-acylisourea or carboxylic ester [196]. Since the O-acylisourea is prone to hydrolysis, the addition of NHS creates a more stable NHS-ester that is resistant to hydrolysis. As the amine is added, the amidation reaction proceeds with less undesired products since NHS is a better-leaving group [193].

Le Gars et al. directly grafted aromatic molecules on the surface of the CNC via a 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO)-mediated oxidation process, which introduced carboxylate groups ($-\text{COO}^-$) at the surface. Then in the second step, an amidation was carried out between these carboxylate groups ($-\text{COO}^-$) and the amine functions ($-\text{NH}_2$) borne by a reactive molecule through peptidic linkages using EDC and NHS. They claimed that no significant change in size was induced after the amidation with crystallinity index was substantially the same and it was suggested that the morphology of CNC did not change after amidation with 1-methyl-3-phenylpropylamine due to the mild conditions (pH and temperature) of the amidation reaction [166].

Other than that, Calderón-Vergara et al. investigated the effect of post-oxidation reactions in TEMPO-nanocellulose (obtained from Fiquetow) on the hydrophobisation of these materials via amidation reactions. After the reaction, they observed that the more hydrophilic the

starting material, the more hydrophobic the amidated product was able to form, which had stable dispersions in toluene and able to act as wetting or spreading agents with poor dispersibility in water [167]. For instance, Akshay et al. reported on the implementation of CNF-based complex as a smart drug carrier by functionalised CNF through amidation with furfurylamine, then coupled with pro-drug molecule metronidazole-maleimide through Diels–Alder reaction. This approach resulted in efficient surface chemistry routes and had direct implication for the development of drug delivery applications [168]. Alternatively, amine modification aided in the chelating action on the desired pollutant anions, thereby enhancing the adsorption capacity of the nanocellulose adsorbent materials. The amino groups possess a lone pair of electrons that is attributed to the nitrogen atoms and may form a covalent bond with metal ions. This adsorption of metal ions is coordinated by amino and OH groups found in the functionalised cellulose [13].

5.4. Phosphorylation and carboxymethylation

The second routes for functionalisation to provide an ionic surface on the nanomaterials include phosphorylation and carboxymethylation. Phosphorylated cellulose is capable of binding metal ions [10]. The amalgamation of phosphate (PO_4^{3-}) ester groups on the nanocellulosic surface results in enhancement of its original properties [197]. Phosphorylated CNF using fibre sludge (a paper industry waste stream) as a cheap and abundant raw material in production of nanopaper ion-exchanger represents a step forward in tackling the problem of heavy metal such as copper, accumulation in water resources [198]. The removal efficiency of CNF towards a broad range of contaminants was enhanced by phosphorylation [163]. Blilid et al. conducted a comparative study on phosphorylated CNF and CNC for the removal of chemical pollutants. They found that the presence of phosphorus fragments provided a way for anchoring ultrastable metal oxide clusters on the nanofibrillar surface through metal-phosphonate (P-O-Ti) bridges. They further illustrated the efficiency for scavenging representative anionic and cationic dye pollutants compared to native, non-modified cellulose and its carboxymethylcellulose analogues [199].

Phosphorylation is the most common mechanism of regulating protein function and transmitting signals throughout the cell. A hexokinase (EC 2.7.1.1) is an enzyme that phosphorylates a six-carbon sugar (a hexose) to a hexose phosphate. In most tissues and organisms, glucose is the most important substrate of hexokinases and glucose 6-phosphate the most important product [169]. Recently, Messa et al. reported a facile strategy to convert sugarcane bagasse cellulose into phosphorylated CNF, whose gelation is pH-dependent. They claimed that the optimum pH value was 12, and the most charged phosphate (PO_4^{3-}) to have disintegrated to generate 84.3% CNF by means of high yield of CNF production [200].

Carboxymethyl groups are introduced to cellulosic surfaces via carboxymethylation process rendering the surface negatively charged. These charges create electrostatic repulsions and facilitate the degradation of lignocellulosic fibres into nanosized particles [10]. In principle, carboxymethylation usually introduces charges to the cellulose fibrils (modifies its surface), which helps the fibre to swell more by electrostatic repulsion, thereby facilitating nanofibrillation [49,197]. Based on an experiment by Wei et al., carboxymethylation caused some changes, such as reduction of length and appearance of rough surface with gaps and cracks, that exhibited good water solubility for CNC [201].

6. Applications

Cellulose is the main component of paper, cardboard, and textiles made of cotton, flax, or other plant fibres, and is used in the production of fibres, films, and cellulose derivatives [202]. Moreover, the mechanical properties, reinforcing capabilities, abundance, low density,

and biodegradability of nanosized cellulose make it an ideal candidate for polymer nanocomposites processing [203]. The usage of cellulose as a raw material is intended to help conserve the environment while also addressing issues like biocompatibility and stretchability, which are required by flexible sensing devices in electronic skin, sports equipment and medical equipment [204]. Besides, cellulose extensively used as flexible substrate or template of electrodes to enhance performance of flexible supercapacitors, potential nanocomposite aerogel for organic and solvent adsorption, substrate and green binder to improve electrical conductivity [205–207]. Fig. 4 presents the synthesis of nanocellulose to applications of these materials in various fields.

6.1. Food industry

Nowadays, nanocellulose is being used in a variety of practical applications such as in food sector as materials and reinforced agents in the development of active packaging, food additives and food stabilisers [164,208–210]. The high surface area, aspect ratio, rheological behaviour, water absorption, and absence of cytotoxic and genotoxic properties of nanocellulose facilitate its use in food applications [62]. In food packaging, a regenerated and biodegradable cellulose like the cellophane has long been used as a thin and transparent sheet for its high barrier against gases, oils, greases, and bacteria [194]. Adel et al. prepared an active packaging from ternary biocomposite of chitosan (CS), β -cyclodextrin citrate (β -CD_{cit}) and oxidised nanocellulose (ONC) biopolymers with clove essential oil (CEO) that was incorporated into biocomposite films to enhance their activity. The results gave an indication of possible impacts on the strength, transparency, and water vapour permeation occurring after loading different concentrations of ONC into the ternary biocomposite films [210]. The coating layers with excellent gas-barrier properties are assembled on polypropylene films through layer-by-layer (LbL) deposition of biorenewable nanocellulose and nanochitin as reported by Nguyen et al. The resulting coated film is highly transparent, unfavourable to bacterial adhesion, and thermally recyclable, hence it is promising for advanced food packaging applications [211]. Recently, Atta et al. developed silver decorated BC nanocomposite-based food packaging material through the reduction of silver nitrate with sodium chloride. This product proved for providing protection and extending the shelf-life of different foods [212]. Moreover, report, a sonication and dip coating method were developed to deposit a polyvinyl alcohol (PVA)/montmorillonite (MMT) nanocoating on regular paper and cotton paper. The results suggest that applying nanocoating is a promising approach to improve the barrier properties of paper [213]. Apart from that, electrospinning also used as efficient nanofiber method for production of materials for food packing [214].

Nanocellulose is also extensively used as a food additive to improve food homogeneity and food such as flour substitutes, fat substitutes, frozen food and beverage additives, thickeners, suspension agents, foam stabilisers, high temperature stabilisers [215,265]. Aaen et al. studied the suitability of CNF as rheology modifiers and fat replacers since CNF have been proposed for use in low-fat food products due to their availability and excellent viscosifying and gel forming abilities. They stated that the CNF-to-xanthan ratio, as well as the presence of electrolytes was shown to be the key factors in determining whether the viscosity and storage modulus of CNF dispersions increased or decreased when xanthan was added [164]. In other work, Corral et al. evaluated whether the addition of BNC improved the baking quality of wheat flours, making a change in the viscoelastic behaviour of the mass. They found that BNC increased specific volume and moisture retention, decreasing the browning index. However, both raw and heat-treated doughs with more elastic characteristics were produced, but the addition of BNC reduced firmness of bread crumb [209].

The effect of three nanocellulose from corn oil-in-water emulsions on lipid in vitro gastrointestinal digestibility was investigated by Li et al. The free fatty acid (FFA)-released test results indicated that both crystalline allomorph and morphology of nanocellulose affected the degree

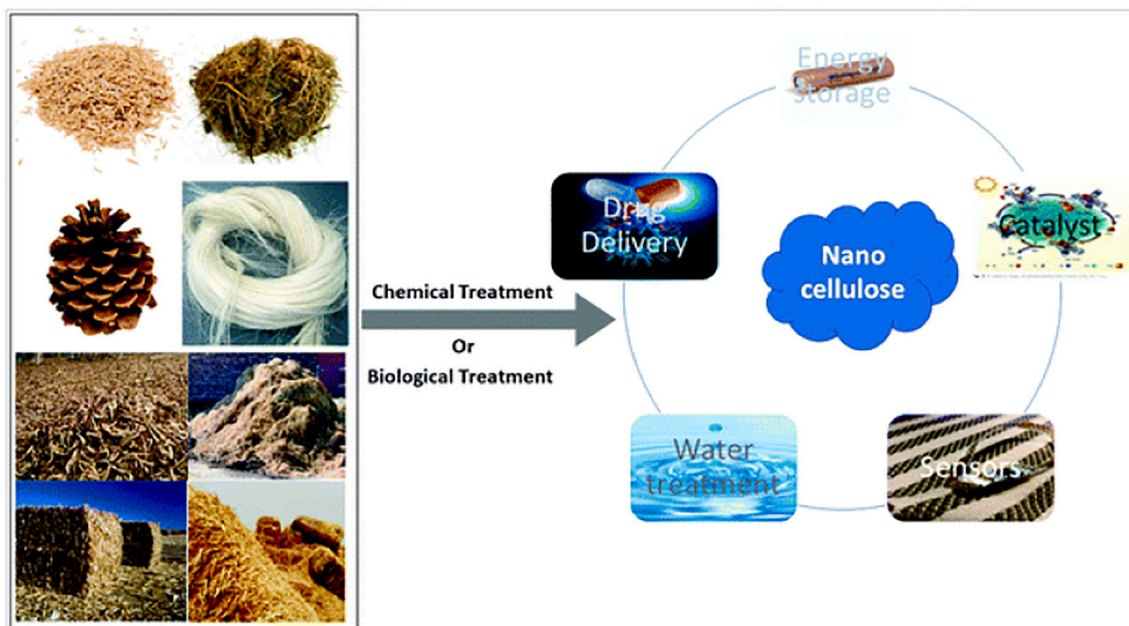


Fig. 4. Applications of synthesis of nanocellulose [197].

of lipid digestion, especially the morphology. Their findings provided an innovative solution that using nanocellulose as food-grade particle stabiliser to modulate the digestion of Pickering emulsified lipids would benefit the development of given functional foods [216]. Besides, Ni et al. produced nanocellulose from ginkgo seed shells to stabilise Pickering emulsions. The finding showed that the small amount of nanocellulose could stabilise emulsions containing high oil phase, including nanocellulose stabilised emulsions which had good ability to resist on environment change [217].

6.2. Biomedical

Nanocellulose-based platforms can be considered as an emerging technology for several biomedical applications such as biosensor, tissue engineering, and biotechnology [25,62,218,219]. Today, it serves as promising materials in various sensor fields such as optical, electrochemical, and piezoelectric sensors [220–223]. On most occasions, nanocellulose is modified along its surface with other superficial groups, other polymeric chains, or even other nanoparticles to achieve different properties and functions to acquire low-cost smart materials with responsiveness in variety configurations [224]. The electrode is composed of nanocellulose, and *f*-MWCNTs are fabricated for the detection of diclofenac sodium (DCF), a non-steroidal anti-inflammatory drug (NSAID), as reported by Shalauddin et al. The developed sensor showed good sensitivity and selectivity for the determination of DCF with good recoveries [156]. Furthermore, a novel and sensitive electrochemical cholesterol biosensor was developed based on immobilisation of cholesterol oxidase (ChOx) on the polyaniline/crystalline nanocellulose/ionic liquid modified Screen-Printed Electrode (PANI/CNC/IL/SPE) by Abdi et al. It was observed that the proposed biosensor showed a high sensitivity value and an acceptable reproducibility and repeatability with minimal interference from the coexisting electroactive compounds such as ascorbic acid, uric acid, and glucose [225]. Furthermore, Yuen et al. demonstrated microbially-grown nanocellulose to build functional medical sensors as an interesting alternative to plastics for applications in medical sensing such as heart rate monitoring and temperature sensing [226].

Nanocellulose presents an extensive review of research over the past years on the promising pharmaceutical and medical applications of nanocellulose-based materials such as wound-healing, tissue

engineering, and imaging applications in therapeutic platform for cancer treatment and drug delivery system [28,56,227,264]. BNC has attracted increasing attention as a novel wound dressing material, but it has no antimicrobial activity. Therefore, to overcome this problem, Liyaskina et al. studied the BNC saturated with antibiotic fusidic acid (FA). The resulting biocomposites had high antibiotic activity against *Staphylococcus aureus* and can be used in medicine as a wound dressing [228]. Moreover, aminoalkylsilane was chemically grafted on a BNC membrane (A-g-BNC) and used as a bridge to combine BNC with Pululan zinc oxide nanoparticles (Pul-ZnO-NPs) hybrid electrospun nanofibres, as reported by Khariari-Khalji et al. The new functional dressing, fabricated with a cost-effective and easy method, showed excellent antibacterial activity, and thus could accelerate wound healing [229]. Boni et al. combined silk sericin for surface micropatterns in BC for dressings to control fibrosis and enhance wound healing. The findings show improvement in cell growth, while preventing the anarchic deposition of collagen responsible for fibrosis and scar formation through cell orientation [230].

In another aspects, Osorio et al. developed a novel methodology on biomimic soft extracellular matrix (ECM) chemistry to generate 3D BNC using the bioengineering of fibroblasts (the cells responsible for producing and regenerating the ECM) to immobilise adhesion proteins, such as collagen and fibronectin. The novel methodology was robust and able to biomimic the chemical surface of soft ECM and immobilise cell-derived adhesion proteins from fibroblast [231]. Hydrogels of alginate and gelatin were prepared with and without CNC and printed using a CELLINK® BIOX 3D bio-printer. It was observed that the 3D-printed scaffolds exhibited superior properties such as improved mechanical strength, superior swelling potential, better cell viability, healthy cells that adhered appropriately to the surface of the scaffolds, enhanced mineral deposition, and upregulation of osteogenic-associated genes was observed in the scaffold-treated groups [232]. Fig. 5 illustrates some applications of nanocellulose in biomedical fields.

Moreover, the potential use of cellulose CNC in cancer diagnosis have been greatly explored [227]. Curvello et al. investigated the suitability of collagen-nanocellulose hydrogels to mimic the extracellular matrix of PDAC and to promote the formation of tumour spheroids and multicellular 3D cultures with stromal cells. The blended 3D matrices with tuneable mechanical properties consistently maintained the growth of PDAC cells and its cellular microenvironment, and this would

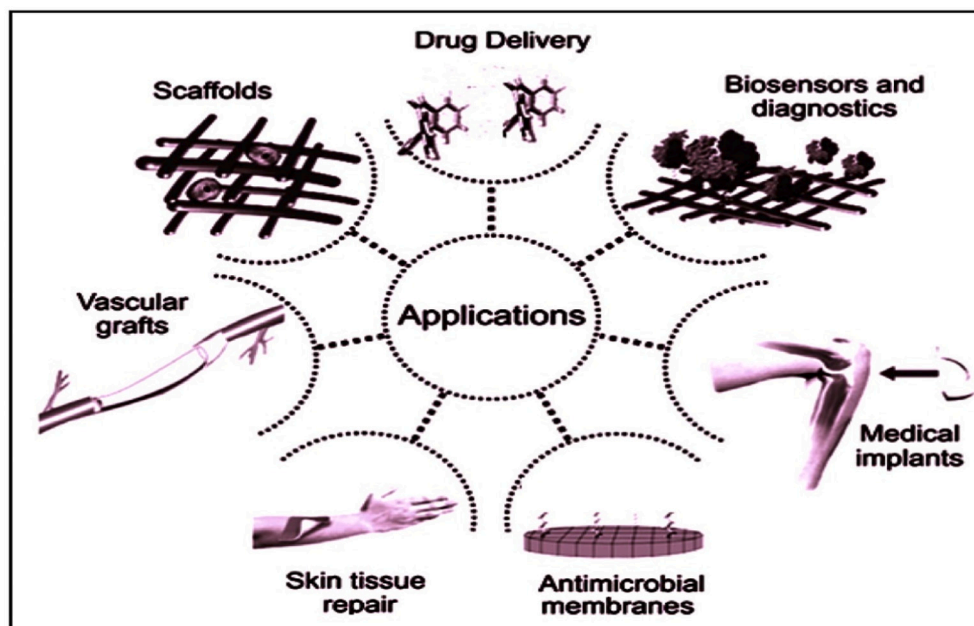


Fig. 5. Application of nanocellulose in biomedical fields [218].

enhance the screening of anti-cancer treatments [233]. Additionally, Wong et al. reacted functionalised CNC (fCNC) with radiometal loaded-chelates or fluorescent dyes as tracers to evaluate the pharmacokinetic profile of fCNC in vivo. According to their findings, these nanoscale macromolecules can be covalently functionalised and yield water-soluble and biocompatible fibrillar nanoplatforms for gene, drug and radionuclide delivery in vivo [234].

6.3. Water treatment

Water contamination is a major concern today [235]. Nanocellulose has demonstrated properties that makes it an efficient adsorbent, flocculant, and membrane in water and wastewater treatment [54,66,236,237,267]. The high specific surface area, broad possibility of surface modification, and high mechanical strength, nanocellulose has emerged as a new class of bio-based adsorbent with promising potential application in environmental remediation [238,239]. In this case, the specialties of nanocellulose for this application are due to their high aspect ratio and accessibility of plenty of –OH groups for binding with dyes, heavy metals, and other pollutants [240–243]. Recently, Mo et al. developed a wood-inspired nanocellulose aerogel adsorbent with excellent selective capability, superfast adsorption, and easy regeneration. The premise for the design is that the biomimetic honeycomb architecture and specific covalent bonding networks can provide the adsorbent with structural and mechanical integrity, yet superfast removal of target contaminants. They found that the aerogel adsorbent had better affinity for Pb(II) than other coexisting ions in wastewater and could be regenerated for at least five cycles [244]. In another research conducted by Mo et al., a compressible cellulose nanofibre (CNF)/carboxymethyl cellulose (CMC)/branched polyethyleneimine (BPEI) aerogel was synthesised via electrostatic-modulated interfacial covalent crosslinking and freeze-drying process. The porous BPEI@CNF/CMC aerogel possessed excellent mechanical compression and high-density metal-chelating groups, which exhibited fast adsorption kinetics and high adsorption capacity in static copper adsorption process [245].

In order to increase the sustainability of wastewater treatment, and avoid contamination, Blockx et al. developed the first cellulose nanocrystalline flocculant with a grafted cationic point charge, that is glycine betaine (*N,N,N*-trimethylglycine) grafted cellulose nanocrystals (CNC)

effective for the flocculation of kaolin (a model system for wastewater treatment), the freshwater microalgae *Chlorella vulgaris*, and the marine microalgae *Nannochloropsis oculata*. They successfully grafted glycine betaine onto CNC using a one-pot reaction with tosyl chloride-activated esterification reaction acting as bio-based flocculants to replace synthetic polymer flocculants or metal salt coagulants [246]. Campano et al. presented a paper to investigate, for the first time, the potential of cationic hairy CNC to induce the flocculation of a model suspension of kaolinite. They claimed that CNC acted through charge neutralisation mechanism and the model used to estimate flocs fractal dimension (D_f) was found very successful to describe the flocculation process [247].

Nanocelluloses have emerged as an alternative approach from conventional wastewater treatment materials to fulfil diverse functions. Gholami et al. demonstrated a novel membrane composed of polydopamine (PDA) particles and BNC, which can efficiently remove a variety of metal ions and organic dyes from contaminated water. They showed that the PDA/BNC membrane can effectively remove heavy metal ions (such as lead and cadmium), and organic dyes as surrogate markers of organic pollutants (such as rhodamine 6G (R6G), methylene blue (MB), and methyl orange (MO) [248]. In the study, a unique class of low fouling nanocellulose-enabled thin film nanofibrous composite (TFNC) ultrafiltration (UF) membranes was fabricated by coating of negatively charged TEMPO-oxidised CNF on the porous electrospun polyacrylonitrile (ePAN) substrate. The results indicated the practicality of using charged CNF as a barrier layer for antifouling ultrafiltration membranes in wastewater treatment [249].

6.4. Nanocomposite

CNC, CNF and BNC have been emerging as a new class of nanomaterials that can be used as a reinforcing component for the preparation of high performance nanocomposites due to their unique properties such as biodegradability, renewability, non-toxicity, high modulus and mechanical strength, high specific surface area, and interesting surface chemistry and optical properties [250,265]. The high-strength nanocomposite hydrogels were synthesised from CNC core, and polyacrylamide shell led to the increase of energy dissipation, viscoelastic properties, and extensibilities and fracture stresses [13]. Yuen et al. prepared nanocomposite films from a suspension of cellulose nanocrystals as the filler and a polycaprolactone-based waterborne

polyurethane (WPU) as the matrix. The material resulted in an improvement of microphase separation between the soft and hard segments of the WPU matrix, and a significant increase in Young's modulus and tensile strength [226,262]. Additionally, in the study by Noguchi et al., nanocellulose-reinforced polyethylene composite sheets were prepared with an in situ surface-hydrophobised, TEMPO-oxidised-CNF (TEMPO-CNF) and polyethylene by thermal kneading with high shear forces (i.e., under 'elastic' kneading conditions). They successfully reinforced polyethylene with TEMPO-CNF using oven-drying and elastic kneading method with the yield stress and modulus of the sheets linearly increased with increasing TEMPO-CNF content up to ~8 vol% [251]. In another work, Rani et al. described a nanocomposite polymer electrolyte that was prepared using a solution casting technique. Polyethylene oxide (PEO), lithium iodide (LI), and nanocellulose were extracted from palm oil empty fruit bunch (EFB) to prepare electrolyte films. A photovoltaic response efficiency of 2.12% was achieved using the highest-conducting film in a dye-sensitised solar cell (DSSC). Therefore, this showed that the film is suitable for DSSC applications [252]. Vilela et al. prepared nanocomposites consisting of cross-linked poly([2-(methacryloyloxy)ethyl]trimethylammonium chloride) (PMETAC) and BNC for fighting fungal infections. It is proved that this functional material potentially an effective system for the treatment of *C. albicans* infections [253].

Further, the advantages of nanocellulose such as lightweight and high electromagnetic shielding ability provides them as alternative materials in electromagnetic shielding applications. This is crucial for both human health and proper operation [254]. Besides, the performance of microwave-absorbing material was developed to reduce electromagnetic pollution. The study used cornstalks as precursor to construct microwave-absorbing nanocomposites with iron nanoparticles dispersed in carbon matrix impregnation-carbonization process [255]. The same study was successfully designed by ultralight high-performance microwave absorption material utilised cobalt/carbon nanocomposites [256].

6.5. Carbon-based materials

Other application of cellulose is to produce carbon materials. Cellulose has a beneficial molecular structure which makes it possible to produce carbon fibre with mechanical properties comparable to those of carbon fibre from polyacrylonitrile (PAN) and mesophase pitch [257]. Besides, Pan et al. prepared carbonized cellulose as a thermally conductive bridge to connect boron nitride to synthesize 3D carbonized cellulose/boric acid ball mill modified boron nitride aerogel. This structure proved to enhance the thermal conductivity and flame retardancy of epoxy-based composites [258]. In other case, carbon nanofiber was prepared by Guo et al. in microwave frequency band. The negative permittivity has great significant for the development of new lightweight microwave absorbing and shielding materials, electronic equipment, and stealth materials [259].

7. Challenges and future directions

Despite the extraordinary properties of nanocellulose, as confirmed through two decades of exhaustive research addressing an array of potential applications, the nanocellulose market is still far from reaching its full potential. The main cause is the lack of process-adapted measuring tools capable of characterising nanocellulose, at acceptable speed and reliability, to meet the industrial demands in a cost-effective way [260]. The performance issues of nanocellulose-based films, coatings, and layers for packaging rises due to film performance is compromised by high humidity or moisture, films crack (due to bending stresses and inadequate flexibility), and films failing to block wetting or leakage of a liquid [261,263]. The challenges in the reducing end-modification of CNC still remain. Regarding the modification of CNC, the aim is regulate their properties for various potential applications by:

(1) the accurate measurement of the effective amount of reducing end aldehyde groups is a prerequisite for any reducing end modification of CNC, (2) reducing end-modification strategies of CNC that are required to improve grafting efficiency, particularly milder conditions, fewer preparation steps and lower reaction time, (3) characterisation techniques (especially lack of powerful and direct characterisation) for reducing end-modification of CNC that are still insufficient because of low grafting density, and are therefore low detectability, and 4) combination of reducing end-modification and surface-modification of CNC that deserves further investigations to design CNC with diverse structures and functionalities based on different regio-selective and local modifications [182].

Most of the laboratories and small pilot-scale efforts have processed fibres with homogenisers, but due to limited capacity, and poor scale-up prospects, high maintenance under the very high pressures is needed to produce CNF. These small-scale efforts were very useful and effective at the laboratories and pilot scales, but may not provide a useful method for multi-ton-scale plants [159]. Since human beings do not have specific enzymes (such as, cellulases) that can break down the nanocellulose in vivo, it is not well-accepted to engage the nanocellulose itself or the composite material with nanocellulose as the main constituent in implant manufacture, where it is highly desired that the biomaterials are bioresorbable after tissue healing or organ repair [179]. Additionally, the energy consumed during nanocellulose fabrication may cause it to be commercially impracticable in its utilisation for certain applications. Therefore, it is reasonable to assume that the use of nanocellulose for more complex and sophisticated applications can compensate its actual price and not impose any constraints in the production of high-value products on a large-scale [250]. Commercial manufacturing of nanocelluloses adapted to different end-user applications would have a promising future in the context of expanding global technology development, with strong collaboration between industries and the academic world [117,269].

8. Conclusions

Nanocellulose materials such as cellulose nanocrystal (CNC), cellulose nanofibril (CNF), and bacterial nanocellulose (BNC) known as the most prominent green materials. The promising materials of nanocellulose has provided interest due to their multiple superior properties such as high aspect ratio, better mechanical properties, renewability, biodegradable, non-toxic and others. The properties of nanocellulose depend on the raw materials and methods that are used to obtain nanocellulose. Surface modifications of nanocellulose provided excellent platform for additional functionalities with other material to employ nanocellulose for high value materials i.e., acetylation, silylation, amidation, phosphorylation and carboxymethylation. These superior properties give nanocellulose wide range applications including in food industry, bio-medical, water treatment and nanocomposites. However, many issues arise make nanocellulose facing challenges for commercial applications. Regarding this, strong collaboration between all parties needed for promising future of nanocellulose.

Declaration of competing interest

The authors state that they have no known conflicting financial interests or personal ties that may seem to have influenced the work described in this study.

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