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Emerging technologies for the production of nanocellulose from lignocellulosic biomass

Dileswar Pradhan

Amit Jaiswal

Swarna Jaiswal

Review

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Dileswar Pradhan, Amit K. Jaiswal [*], Swarna Jaiswal

School of Food Science and Environmental Health, College of Sciences and Health, Technological University Dublin - City Campus, Central Quad, Grangegorman, Dublin D07 ADY7, Ireland
Environmental Sustainability and Health Institute, Technological University Dublin - City Campus, Grangegorman, Dublin D07 H6K8, Ireland

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ABSTRACT

Nanocellulose is a unique and promising natural nanomaterial and has gained significant attention due to its applications in several important areas. Thus, researchers are continuously looking for the most efficient, sustainable, economically viable, and environmentally friendly production technologies to fulfil its growing demand. Conventional production technologies, which include various physical, chemical, and physicochemical methods, are currently inadequate for this purpose and have several limitations such as long processing time, high energy consumption, low recovery of nanocellulose, and many others. To overcome these shortcomings, scientists have investigated the prospect of utilizing emerging processing technologies such as microwave irradiation, deep eutectic solvent, enzymatic processing, cold plasma, electron beam irradiation, and pulsed electric field in nanocellulose preparation. In general, studies have shown that the application of emerging technologies enhances the extraction yield and properties of nanocellulose. This article presents a review of the most recent works reported on the application of emerging technologies in nanocellulose production.

1. Introduction

Nanotechnology is now widely recognized as one of the most important factors behind a new industrial revolution in various interdisciplinary sectors such as the pharmaceuticals industry, automotive industry, food industry, and so on. Nanomaterials have at least one dimension in the nanometer scale, i.e., around 100 nm or less, and have different chemical, physical and biological characteristics than bulk material (Pires et al., 2019). Nanocellulose is a natural nanomaterial having several advantageous characteristics, such as high surface area, nanoscale dimension, unique morphology, specific high strength and modulus, renewability, and good optical properties (Phanthong et al., 2016). Thus, the utilization of nanocellulose in the development of a variety of sustainable and renewable materials has drawn considerable interest in recent years. However, one of the crucial factors in achieving the wide application of nanocellulose is to develop sustainable and economically feasible techniques to produce nanocellulose (Song et al., 2018).

Based on preparation techniques, nanocellulose can be classified into three categories such as cellulose nanocrystals (CNC), cellulose nanofiber (CNF), and bacterial nanocellulose (BNC) (Nasir et al., 2017). These three types of nanocellulose possess similar chemical compositions; however, they exhibit different physical characteristics like particle size, morphology, crystallinity etc. (Phanthong et al., 2018). The preparation techniques primarily determine their structure, dimensions, and properties. The preparation of CNF and CNC is a top-down process, while BNC preparation is a bottom-up process. In the top-down process, nanocellulose is prepared by disintegrating the cellulose fibres from lignocellulosic sources to their nano size. Whereas, in the bottom-up process, nanocellulose is produced by a buildup of nanofibers from low molecular weight sugars by bacteria (Nechyporchuk et al., 2016; Teo & Wahab, 2020). For this article, the authors have focused only on the production of CNC and CNF.

CNC is also known as nanocrystalline cellulose (Ilyas et al., 2018), cellulose nano-whiskers (Motta Neves et al., 2020), and crystalline nanocellulose (Lenfant et al., 2017). CNC has a whisker shape or a short-rod-like shape with a length in the range of 100–500 nm and a diameter of 2–20 nm. Besides, it contains a 100% chemical composition of cellulose, primarily in crystalline regions (Phanthong et al., 2018). In addition, it also possesses some attractive properties like large surface

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area (~150 m^2/g), high crystallinity index (>70%), high tensile strength (7500 MPa), and big aspect ratio (~70) (Tang et al., 2014). Thus, CNC can be utilized in various industrial sectors such as healthcare (Favatela et al., 2021; Thomas et al., 2020), electronics (Nyamayaro et al., 2020; Yao et al., 2020), construction (Ghahari et al., 2020; Lee et al., 2019), food packaging (Alvarado et al., 2018; He et al., 2021), inks for 3D printing (Dai et al., 2019; Li et al., 2021a; Wang et al., 2018), oilfield servicing fluids (Li et al., 2021a; Li et al., 2015), polymer composites (Kargarzadeh et al., 2018) and many others.

CNF is also known as nano fibrillar cellulose (Baati et al., 2017), cellulose nanofibrils (Li et al., 2020b), and nano fibrillated cellulose (Ilyas et al., 2019). Generally, it has a diameter of about 1 to 100 nm and a length of about 500 to 2000 nm (Phanthong et al., 2018). It contains both amorphous and crystalline cellulose domains within the single fibres (Nasir et al., 2017). CNF exhibits some attractive properties such as non-toxic and non-abrasive nature, very high elastic modulus, large specific surface area, and low thermal expansion (Deepa et al., 2015). Besides, it possesses a high aspect ratio and forms gels in water with shear-thinning and thixotropic behavior (Nechyporchuk et al., 2016). Thus, CNF has found applications in several fields such as food packaging (Balasubramaniam et al., 2020; Deng et al., 2017; Van Hai et al., 2020), drug delivery (Bhandari et al., 2017; Wang et al., 2020c), water treatment (Gopakumar et al., 2017; Soyekwo et al., 2017), tissue engineering (Chakraborty et al., 2019; Huerta et al., 2020; Maharjan et al., 2021), polymer composites (Kargarzadeh et al., 2018), oilfield servicing fluids (Li et al., 2020a; Li et al., 2021a), 3D printing inks (Dai et al., 2019; Håkansson et al., 2016; Li et al., 2021a), and many others.

The production of CNFs generally occurs through mechanical disintegration, resulting in particles having both crystalline and amorphous regions. The primary objective during the production of CNF is to retain the crystalline structure and the degree of polymerization of the delaminated CNF filaments as much as possible (Klemm et al., 2018). Conventionally, for isolating CNFs, a dilute suspension of cellulose fibres is usually subjected to high shear forces and different mechanical techniques, including high-intensity ultrasonication (Dilamian & Noroozi, 2019; Syafri et al., 2019), cryocrushing (Alemdar & Sain, 2008; Thiripura Sundari & Ramesh, 2012), grinding (Berglund et al., 2016; Ghaderi et al., 2014), and high-pressure homogenization (Hongrattanavichit & Aht-Ong, 2020; Wu et al., 2021). Although efficient in producing CNFs, these mechanical methods have several disadvantages such as high cost of the process, low efficiency, and requiring extensive energy to disintegrate highly ordered hydrogen bonds and dense network structure of cellulose (Yan et al., 2021). Thus, various pretreatment technologies are utilized before mechanical operations to minimize the high energy consumption and make the surface hydrophobic so CNF can be an appealing material for commercial uses. Examples of pretreatment techniques are mild enzymatic or acidic hydrolysis, phosphorylation, periodate oxidation, carboxymethylation, and TEMPO (2,2,6,6-Tetramethylpiperidine 1-oxyl) oxidation (Pires et al., 2019).

On the other hand, CNCs are produced by selectively removing the amorphous portion of cellulose (Liyanage et al., 2021). Although acid hydrolysis is widely utilized for this purpose (Seta et al., 2020; Wang et al., 2020b), studies have shown that other conventional chemical techniques such as ammonium persulfate oxidation (Khanjanzadeh & Park, 2021; Liu et al., 2020d), and TEMPO-mediated oxidation (Pacheco et al., 2020; Zhou et al., 2018) can also be utilized for CNCs production (Liyanage et al., 2021). These chemical methods require a long processing time, expensive chemicals, an extensive amount of energy, and also generate wastewater causing harm to the environment. Despite that, the industrial production of nanocellulose still involves harsh chemical treatment (Nasir et al., 2017). Thus, there is a need to develop process technologies for preparing nanocellulose that are sustainable, cost-effective, eco-friendly and can be used on an industrial scale. Several researchers have used commercially available novel processing technologies for the preparation of nanocellulose in recent years. Some

examples are microwave irradiation (Harini et al., 2018; Jiang et al., 2018; Liu et al., 2020a), electron beam irradiation (Kim et al., 2016; Kim et al., 2019) and production using enzymes (Michelin et al., 2020; Ribeiro et al., 2019; Squinca et al., 2020).

In recent years, many articles have reviewed the production of nanocellulose involving various conventional and eco-friendly technologies (Nasir et al., 2017; Nechyporchuk et al., 2016; Phanthong et al., 2018; Salimi et al., 2019; Teo & Wahab, 2020); however, to the best of the authors' knowledge, a comprehensive review on all emerging technologies used in nanocellulose production is not available in the literature. Therefore, this article aims at compiling various emerging technologies such as microwave irradiation, cold plasma, electron beam irradiation, pulsed electric field, enzymatic processing, and deep eutectic solvent that have been used for the production of CNC and CNF from lignocellulosic waste.

2. Feedstock for nanocellulose production

Nanocellulose is the natural fibre that can be produced from cellulose (Phanthong et al., 2018). Cellulose is the most abundant renewable organic biopolymer in the world, with an estimated production of 10^{11}–10^{12} tons per year (Nechyporchuk et al., 2016). It is a linear polysaccharide and consists of D-glucose subunits linked by β-(1,4)-glycosidic bonds (Baruah et al., 2018). It does not exist as an isolated individual molecule in nature; rather, it is found as assemblies of individual cellulose chain-forming fibres (Brinchi et al., 2013). The cellulose chains are bound together by hydrogen bonds and contained in an insoluble long-chained compound known as microfibrils (Ravindran & Jaiswal, 2016). Within the cellulose fibrils, cellulose chains are arranged in highly ordered regions (crystalline structure) and disordered regions (amorphous structure). It is these crystalline regions that are extracted, resulting in CNC (Brinchi et al., 2013). Besides, the mechanical defibrillation of cellulose fibrils to nanometer size results in CNF (Phanthong et al., 2018).

Lignocellulosic biomass (LCB) refers to plant biomass, which is the largest promising resource for the sustainable production of cellulose. The LCB can be mainly classified into two categories such as wood and non-wood biomass (agricultural residues and industrial wastes) (Nechyporchuk et al., 2016). Wood-based biomass has been cellulose's primary source over the past century, constituting about 90–95% of all cellulosic pulp produced during this period (Pennells et al., 2020). On the other hand, agricultural residues are the secondary source of cellulose which have various advantages over wood, such as annual renewability, high biomass yield, fast biomass generation, and high carbohydrate content (Pennells et al., 2020). Further, the tertiary sources of cellulose include the byproducts of the food and beverage industry, bagasse, municipal waste, and papermaking sludge (García et al., 2016; Pennells et al., 2020). These wastes can be used as a feedstock for preparing nanocellulose, which is not only advantageous from the environmental point of view but also beneficial in economic aspects (García et al., 2016).

Apart from cellulose, the other major components of LCB are hemicellulose and lignin. Generally, the cellulose, hemicellulose, and lignin contents in a typical LCB fall within the range of 30–60, 20–40, and 15–25%, respectively. However, the proportion of these major components varies depending on the source (Baruah et al., 2018). Thus, the different abundance of these components is a crucial parameter in evaluating the suitability of feedstock for nanocellulose production (Ghaemi et al., 2019). The composition of different types of biomass can be found from the ECN Phyllis2 database (www.phyllis.nl) (Hassan et al., 2018). Besides, the composition of various LCBs that have been used as a feedstock for nanocellulose production is provided in Table 1.

3. Conventional technologies in nanocellulose production

Nanocellulose production is a two-stage process; one is the

Table 1
Chemical composition of different lignocellulosic feedstocks used for nanocellulose production (% dry basis).

Feedstock	NC Type	Cellulose	Hemicellulose	Lignin	References
Rice straw	CNF	36.5 ± 2.1	38.0 ± 1.6	22.0 ± 2.7	(Oun & Rhim, 2018)
Corn cobs	CNC	45.01 ± 0.9	33.12 ± 1.1	13.81 ± 1.3	(Louis & Venkatachalam, 2020)
Tea stalk	CNC	35.01	20.45	28.01	(Guo et al., 2020)
Pinecones	CNF	43.8 ± 2.0	27.2 ± 0.1	21.5 ± 1.2	(Rambabu et al., 2016)
Industrial orange bagasse	CNF	21.04 ± 7.43	9.75 ± 1.07	3.50 ± 0.09	(Mariño et al., 2018)
Orange bagasse in natura	CNF	11.85 ± 2.73	15.58 ± 2.07	1.67 ± 0.88	(Mariño et al., 2018)
Corn stover	CNF	44.4 ± 0.4	27.8 ± 0.3	19.6 ± 0.2	(Xu et al., 2018)
Coconut coir fibres	CNF	44	12	33	(Wu et al., 2019)
Soy hulls	CNC	48.2 ± 2.1	24.0 ± 3.0	5.78 ± 1.06	(Flauzino Neto et al., 2013)
Apple pomace	CNC	32.48 ± 0.33	29.06 ± 0.17	22.56 ± 0.24	(Melikoğlu et al., 2019)
Pineapple leaf	CNC	36.3 ± 3.8	22.9 ± 2.0	27.53 ± 1.94	(Dos Santos et al., 2013)
Grape pomace	CNC	19.30 ± 0.67	7.20 ± 0.50	15.60 ± 0.28	(Coelho et al., 2018)
Pistachio shell	CNC	38.1 ± 1.9	31.4 ± 2.7	25.6 ± 3.0	(Kasiri & Fathi, 2018)
Garlic straw residues	CNC	41	18	6.3	(Kallel et al., 2016)
Soybean straw	CNF	39.8	22.6	12.8	(Martelli-Tosi et al., 2016)
Wheat straw	CNF	45.70 ± 0.18	37.12 ± 0.9	17.43 ± 2.1	(Kaushik et al., 2010)
Barley straw	CNC	56.2	7.0	9.2	(Fortunati et al., 2016)
Barley husk	CNC	45.7	22.4	7.2	(Fortunati et al., 2016)
Oat hull	CNF	31.16 ± 1.15	28.72 ± 0.25	18.12 ± 0.63	(Debiagi et al., 2021)
Empty fruit bunch fibres	CNC	59.14	12.07	25.33	(Azrina et al., 2017)

pretreating lignocellulosic feedstock for cellulose fibre isolation, and the other is the breakdown of isolated cellulose fibre to the nanoscale. Several types of conventional technologies have been reported in the literature for the preparation of nanocellulose. However, the choice of technology depends on the type of nanocellulose being prepared, i.e., CNF or CNC. The production of different types of nanocellulose from various lignocellulosic feedstocks using conventional technologies has been provided in Table 2. Besides, various chemical and mechanical conventional technologies used in nanocellulose production have been briefly discussed.

Acid hydrolysis is the easiest and oldest chemical method for the production of nanocellulose from cellulosic materials. Here, the acid dissolves and removes the amorphous cellulose region while the crystalline parts are retained (Nasir et al., 2017). Generally, different acids including HCl, H_2SO_4, HBr, and H_3PO_4 are utilized; however, H_2SO_4 is mainly used because it can strongly isolate CNC as well as make the nanocellulose dispersed as a stable colloid system because of the esterification of the hydroxyl group by sulfate ions (Phanthong et al., 2018). In this process, the reaction temperature and duration, along with the type of acid and its concentration, influence the size and morphology of nanocellulose (Salimi et al., 2019). This method has several limitations such as high-water usage and generation of acidic wastewater, long

processing time, high operational and maintenance costs, risk of equipment corrosion, the formation of inhibitors, and not being environmentally friendly (Teo & Wahab, 2020).

TEMPO oxidation is another chemical method for producing nanocellulose from lignocellulosic materials. The main goal of utilizing TEMPO is to reduce the energy required for mechanical disintegration by diminishing the negative or positive charge on the surfaces of fibre and by improving the colloidal suspension's stability of the produced nanocellulose (Rana et al., 2021). TEMPO-mediated oxidation treatment is generally carried out in the presence of bleaching agents such as NaClO and catalysts such as sodium bromide (NaBr) under alkaline conditions (pH between 9 and 11) (Dhali et al., 2021). For instance, Zhang et al. (2016) prepared CNCs by performing TEMPO hydrolysis of bleached sugarcane bagasse pulp using TEMPO/NaBr/NaClO system in an aqueous medium at pH 10 for 5 h followed by washing, sonication, and centrifugation. Similarly, Zhang et al. (2020) obtained a CNC yield of 52.01% using TEMPO hydrolysis of lemon seeds using TEMPO/NaBr/NaClO system in an aqueous medium at pH 10 followed by centrifugation, dialysis, and sonication. The TEMPO oxidation method has low energy consumption, simple operation, and mild reaction conditions (Wang et al., 2019). However, it has several drawbacks, such as limited oxidation position and toxic reagents (Zhang et al., 2020).

Ammonium persulfate (APS) oxidation is another chemical method that is utilized to produce nanocellulose. Because of its low toxicity and high solubility in water, APS has been recognized as a viable candidate to be utilized for producing H_2O_2 and SO_4^{2-} free radicals at acidic medium and high operating temperature, which are effective in solubilizing the amorphous portion of cellulose, and lignin content (Ng et al., 2021). In a study, Zhang et al. (2020) utilized the APS oxidation method to prepare CNCs from lemon (*Citrus limon*) seeds. They obtained a CNC yield of 13.02% (*w/w*) using the reported method, which involved the utilization of APS solution (1 mol/L) under continuous stirring for 14 h at 70 °C followed by centrifugation, dialysis, and sonication. The concentration of APS as well as other processing conditions such as temperature and treatment time influence the yield and characteristics of nanocellulose in this method (Zhang et al., 2016). The longer processing time is a major disadvantage in this method, limiting its utilization in the industrial-scale production of nanocellulose.

Ball milling is a mechanical method for the preparation of nanocellulose. In this process, a cellulose suspension is kept in a hollow cylindrical container that is partially filled with balls (e.g., zirconia, ceramic, or metal). The high-energy collision between the balls disintegrates cellulose fibres as the container rotates (Nechyporchuk et al., 2016). For nanocellulose production using this process, milling in a wet state is desirable for maintaining the fibrous state and preventing defibrillation to an amorphous state (Phanthong et al., 2018). The ball size, ball-to-cellulose weight ratio, grinding time, and moisture content are factors of ball milling that affect nanocellulose production (Nasir et al., 2017). High power and energy consumption and the generation of a large amount of heat energy during processing are some of the limitations of ball milling (Teo & Wahab, 2020).

Cryocrushing is another mechanical technique used for the production of CNF. In this process, fibres are kept in water, and cellulose absorbs water in its cavity. Water-soaked cellulose is immersed in liquid nitrogen, which solidifies the water content, and is subsequently crushed by mortar and pestle (Nasir et al., 2017). Application of high impact forces to the frozen cellulosic fibres leads to rupture of cell wall due to exerting pressure by ice crystals and thus, liberating nanofibers (Abdul Khalil et al., 2014). This process's drawbacks are high cost, high energy consumption, low recovery, and low uniformity of nanocellulose (Teo & Wahab, 2020).

High-shear grinding is another mechanical method for CNF preparation. In this process, pulp passes through a couple of stones, where one stone is fixed while the other stone rotates (Nasir et al., 2017). The distance between these stones can be adjusted, which enables avoiding the problem of clogging (Nechyporchuk et al., 2016). In the grinder, the

Table 2
Extraction of nanocellulose (NC) from various lignocellulosic sources using different conventional technologies.

Source	Type of NC	Pretreatment	NC extraction	Crystallinity (%)	Dimension	Other properties	References
Tea stalk	CNC	Chemical treatment with H_2O_2 and acetic acid solution	Acid hydrolysis (H_2SO_4)	61.32	Width between 4 and 8 nm	Short rod-like structure, zeta potentials: −33.39, 49.87% yield	(Guo et al., 2020)
Pineapple leaf (PL)	CNC	Alkali treatment with aqueous NaOH solution + bleaching	Acid hydrolysis (H_2SO_4)	73	Length: 249.7 ± 51.5 nm Diameter: 4.45 ± 1.41 nm	Needle-shaped nature, aspect ratio of around 60, high thermal stability (225 °C)	(Santos et al., 2013)
Empty fruit bunch	CNF	Steam explosion + alkali treatment (NaOH) + bleaching	Nano grinding	85.09	Average diameter: 17.85 nm	Crystallinity domain size: 2.55 nm, high char yield (31.19%)	(Supian et al., 2020)
Sugarcane bagasse	CNC	Alkali treatment + bleaching	Acid hydrolysis	72.5	Diameter: 20–60 nm Length: 250–480	Rod-like structure	(A. Kumar et al., 2014)
Maize straw	CNC	Alkali treatment (NaOH) + bleaching	Acid hydrolysis (H_2SO_4)	75.5	Diameter: 19 nm Length: 388 nm	Aspect ratio of 20	(Rehman et al., 2014)
Softwood pulp	CNF	Blending + alkali treatment (Na_2CO_3)	Ball milling	>70 but <95	Maximum diameter: 139 nm	Arithmetic average diameter: 57 nm	(L. Zhang et al., 2015)
Pinecones	CNF	Alkali treatment (NaOH) + acidified sodium chlorite treatment	Mechanical grinding	70	Diameter between 10 and 20 nm	Peak thermal degradation temperature: 400 °C, Tensile strength: 73 MPa, elastic modules: 17 GPa	(Rambabu et al., 2016)
Cotton linter	CNC	–	Acid hydrolysis (H_2SO_4)	90.45	Length: 177 nm Width: 12 nm	Aspect ratio of 19, High hydrophilicity	(Morais et al., 2013)
Soy hulls	CNC	Alkali treatment (NaOH) + bleaching + blending	Acid hydrolysis (H_2SO_4)	73.5	Average length: 122.66 ± 39.40 nm, average diameter: 2.77 ± 0.67 nm	Aspect ratio of around 44, good thermal stability (around 200 °C)	(Flauzino Neto et al., 2013)

fibrillation mechanism is to disrupt the hydrogen bond and cell wall structure by shear forces and individualization of pulp to nanoscale fibres (Abdul Khalil et al., 2014). The disadvantages of the process are high energy consumption, overheating of raw materials, low recovery and low uniformity of nanocellulose, and reduction in CNF's crystallinity (Teo & Wahab, 2020).

4. Green mechanical techniques in nanocellulose production

Ultrasound irradiation and high-pressure homogenization are known and relatively well established green mechanical methods that have been utilized extensively to produce nanocellulose. Recent application of these technologies in nanocellulose production from different lignocellulosic materials has been discussed in this section.

4.1. Ultrasonication

Ultrasonication is a mechanical method for extracting nanocellulose with the hydrodynamic forces of the ultrasound. During ultrasound treatment, the ultrasonic energy is absorbed by the liquid molecules, leading to the generation of mechanical oscillating power, which results in the formation, expansion, and implosion of microscopic gas (Phanthong et al., 2018; Abdul Khalil et al., 2014). A huge amount of heat is generated during the ultrasonication process; thus, it is generally carried out in a water pool for controlling the heat transfer. Major independent parameters in this process are cellulose concentration, processing time, and ultrasonication power (Salimi et al., 2019). Besides, ultrasonication has also been used during the pretreatment stage of lignocellulosic feedstock during nanocellulose production. After the ultrasonic treatment, the lignocellulosic feedstock undergoes several chemical and physical changes such as structural disorientation of the cell wall, increased specific surface area, and decreased degree of polymerization of cell wall components (Teo & Wahab, 2020).

The production yield and quality of CNC can be improved by employing ultrasonic treatment during acid hydrolysis. Azrina et al. (2017) reported the utilization of ultrasound treatment during the acid hydrolysis process to produce CNC from oil palm empty fruit bunch pulp (EFBP). They compared the properties of prepared CNC with EFBP and

raw empty fruit bunch fibre (REFB). Based on the FESEM analysis, they reported that the obtained CNCs have the morphology of spherical shapes, which might be because of the application of ultrasound during the hydrolysis process. The prepared CNC had higher thermal stability and a higher crystallinity of 80%, compared to 42.0 and 73.0% for REFB and EFBP. In another study, Gibril et al. (2018) used ultrasonication as a pretreatment method to extract CNC from dissolving wood pulp fibres. The ultrasonic treatment was done to induce cavitation of cellulose in suspension prior to acid hydrolysis. The CNC yield increased significantly with the increase of ultrasonic treatment time, and a maximum yield of 79.54% was obtained after 45 min of ultrasonication. The increase in yield was due to the enhancement of acid accessibility by the ultrasonic pretreatment. SEM analysis showed cracks on the fibre's surface for a short treatment time (<5 min), probably due to the collapse of bubbles and pressure variation induced by cavitation. However, beyond 5 min of treatment, cellulose fibres' surface morphology was changed entirely into nanofibrils. XRD analysis showed a slight increase in the crystallinity index, while the TGA analysis indicated that the ultrasonic pretreatment did not influence the thermal characteristics of CNC.

Ultrasonication in combination with high shear homogenization (HSH) has been reported for the production of CNF from Pineapple leaf fibres (PLF). Ultrasonic treatment was carried out at 400 W and 60 °C for 30 and 60 min. High purity CNFs with an average diameter of 68 nm, length of 88–1100 nm, and crystallinity of 61.7% were obtained after 1 h of sonication. TGA analysis showed that CNF prepared using ultrasonication had a higher degradation temperature (320 °C) than untreated fibre (215 °C), indicating superior thermal stability of CNF (Mahardika et al., 2018). In another study, Dilamian and Noroozi (2019) reported the use of high-intensity ultrasonication (HIUS) combined with homogenization for conversion of high-quality cellulose fibrils derived from rice straw to CNF. SEM images of the CNF produced using HIUS treatment (output power of 560 W for 40 min) showed a web-like network of long and interconnected CNFs with median width of 71 (±15) nm. The diameter distribution from the SEM images data revealed that almost 65% of CNFs had a diameter less than 75 nm. TEM images confirmed that the individual CNFs with long entangled cellulosic fibres were extracted from rice straw with nanoscale dimension. The diameter

distribution from TEM images data revealed that around 70% of CNFs obtained using HIUS treatment had a diameter less than 11 nm. The CNF yield increased after HIUS treatment; however, the thermal stability, viscosity, and crystallinity slightly decreased in the ultrasonic treated fibres, confirming a reduction in cellulose chain lengths.

Recently, ultrasonication has been utilized for the production of Lignocellulosic nanofibers (LCNF), which is prepared from unbleached lignocellulosic material. Huerta and Saldaña (2019) developed a novel green process technology by combining ultrasonication with pressurized aqueous ethanol (PAE) treatment to produce LCNF from canola straw. They applied ultrasonic treatment at a theoretical specific energy (TSE) of 4–20 kJ/g for producing LCNF from the PAE treated fibre. The ultrasound TSE significantly influenced the diameter, nanofibril content, optical transmittance, and swelling capacity of the LCNF. LCNF prepared at ultrasonication TSE of 20 kJ/g had an average diameter of 21 nm and a swelling capacity of 1.9 g water/g LCNF. They concluded that the combination of ultrasound and PAE has excellent potential in LCNF production from biomass.

High energy consumption is one of the drawbacks of ultrasonic treatment; however, it is an eco-friendly, and cost-effective process. The ultrasonication treatment has proven to be a highly efficient technology in nanocellulose production.

4.2. High-pressure homogenization

High-pressure homogenization (HPH) is a mechanical method that is mainly used for the production of CNF. It was used for the first time in 1983 for extracting CNF from wood pulp (Herrick et al., 1983; Turbak et al., 1983). The cellulosic pulp is passed through a tiny nozzle at very high pressure. The high velocity and pressure, along with shear and impact forces, influence the fluid to produce shear rates in the stream and reduce the fibres' size to the nanometer range (Nasir et al., 2017).

Hongrattanavichit and Aht-Ong (2020) used HPH in the development of a chemi-mechanical method to isolate CNF from sugarcane bagasse waste. The steam explosion pretreatment combined with alkaline treatment and bleaching was conducted before the HPH treatment at a pressure of 20,000 psi for 12 passes (each cycle took about 150 s per 200 mL of cellulose solution). The extracted CNFs had a crystallinity index in the range of 65.13 ± 2.80 to 72.76 ± 1.09%, a very fine diameter of 3–7 nm with a very low density of approximately 0.8989 ± 0.0113 g/cm³. The CNFs showed weight loss at a thermal degradation temperature range of 296.62 ± 1.59 to 305.83 ± 2.71 °C. In another study, HPH (80 MPa for 30 cycles) of cellulose fibre extracted from sugarcane bagasse resulted in 90% recovery of nanocellulose with a diameter of 10–20 nm. The nanocellulose exhibited the lowest thermal stability with the decomposition temperature of 238 °C and lowest crystallinity index of 36%, which was attributed to the breakage of hydrogen bonds between celluloses by ionic liquid homogenous treatment and the high-pressure shearing of HPH (Li et al., 2012).

The influence of HPH process parameters, i.e., pressure and number of cycles on the yield, crystallinity, and diameter of CNF extracted from kenaf bast were evaluated using response surface methodology. The linear terms for the pressure and homogenization cycles in the regression model significantly influenced the CNF yield, crystallinity, and diameter, while the interaction between the pressure and homogenization cycles had a significant effect on the CNF crystallinity. The optimum HPH process parameters to extract CNF was reported to be a pressure of 56 MPa, 44 P homogenization cycles, and a 0.1 wt% fibre suspension concentration. The CNF yield was 89.9%, with a diameter of 8 nm and crystallinity of 56.5% when extracted under optimum conditions. Further, the morphologies of CNFs produced using HPH at 50 MPa and 40 P were not entirely like web-like nanofibers or classical networks; instead, the CNFs were long entangled networks, having a length of around 500 nm (Davoudpour et al., 2015). The HPH disrupted the hydrogen bond network of celluloses to isolate nanocellulose from the eucalyptus pulp. The optimum conditions to produce nanocellulose was

50 MPa of homogenization pressure and 10 HPH cycles. The prepared nanocellulose had the lowest weight average molecular weight (111,420 Da), which might be due to the strong elongational flow at the homogenizing valve entrance, and the resulting frictional forces encountered by the fluids during HPH induced mechanical degradation of long molecules. The nanocellulose also had the lowest crystallinity index (34.43%) and lowest decomposition temperature (307.9 °C) (Wang et al., 2017).

Although the application of HPH in nanocellulose production has been widely reported, the process has few drawbacks, such as high operational cost, extreme mechanical damage to the crystalline structure of CNF, and its large size distribution (Teo & Wahab, 2020). Another major disadvantage is the clogging issue because of its very small orifice size. However, reducing the size of fibre using mechanical treatment prior to the HPH process can solve the clogging issue (Abdul Khalil et al., 2014). In addition, for a long period, the biggest obstacle to the commercial success of the HPH was the requirement of a high amount of energy, which could reach 70 MW h/t. However, the energy consumption has decreased to nearly 2 MW h/t with the development of highly efficient pretreatment methods (Nechyporchuk et al., 2016). On the other hand, the HPH process has several advantages, making it an important technology in nanocellulose production. The HPH is a rapid process with a simple setup and does not require solvent, so no wastewater generation. Besides, the process can be utilized for the large-scale production of CNF in the industry (Teo & Wahab, 2020).

5. Emerging technologies in nanocellulose production

Emerging processing technologies that have been utilized in nanocellulose production in recent years include an enzymatic method, deep eutectic solvent, microwaves, electron beam irradiation, cold plasma, and pulsed-electric field. Recent application of these technologies in nanocellulose production from different lignocellulosic feedstock is provided in Table 3.

5.1. Microwave irradiation

Microwaves (MW) are non-ionizing waves having a frequency in the range of 300 MHz to 300 GHz and a wavelength from 1 mm to 1 m. MW has been used widely in several areas because of its efficacy and easy operation and is considered as an alternative method to conventional heating (Aguilar-Reynosa et al., 2017). In conventional heating, heat transfer occurs through conduction, convection, and radiation. Whereas in MW, the electromagnetic energy is directly converted into heat at the molecular level of the material (Hassan et al., 2018). In nanocellulose production, MW has been primarily used during the pretreatment stage as a non-conventional heating source for lignocellulosic biomass fractionation. In most cases, researchers have combined MW with other conventional, green, or emerging technologies for pretreating the lignocellulosic material. The combined technique integrates the advantages of MW energy to overcome the shortcomings of other technologies, thus creating a win-win situation for both technologies (Chizoba Ekezie et al., 2017).

MW energy can be utilized in the liquefaction of lignocellulosic biomass during the production of CNFs and CNCs. MW-assisted liquefaction has several advantages over conventional liquefaction, such as low consumption of energy and chemicals, shorter treatment time, and cost-effectiveness (Xie et al., 2016). MW-assisted liquefaction catalyzed by acid can effectively remove hemicellulose and lignin from biomass while producing a liquefied residue with a high cellulose content that can be utilized for nanocellulose production (Huang et al., 2017). In a study, Xie et al. (2016) isolated the CNF from bamboo by combining MW liquefaction with chemical treatment and ultrasonic nano fibrillation processes. They reported that almost all the lignin content could be removed within 7 min using MW liquefaction. The quantity of chemical reagents and time required during chemical treatment for obtaining

Table 3

Application of emerging technologies in nanocellulose (NC) production.

Source	Type of NC	Pretreatment	NC production	Crystallinity	Dimension	Other properties	References
Bamboo	CNF	Microwave liquefaction + chemical treatment	Ultrasonication	67.4%	Diameter: 2–30 nm	Improvement in thermal stability	(Xie et al., 2016)
Wheat straw	CNF	Steam explosion + microwave assisted alkali hydrolysis	Microfluidization	58.62%	Average diameter: 5.42 nm, 10–40 nm wide	Long and loose nanofiber bundles, improvement in thermal stability	(Qi Liu et al., 2017)
Corn cob	CNC	Microwave assisted chemical treatment + ultrasound assisted chemical treatment	Acid hydrolysis + ultrasonication	72.36%	Average diameter: 131.4 nm	Crystallite size: 0.156 nm	(Louis & Venkatachalam, 2020)
Cotton powder	CNC	–	Microwave-assisted ammonium persulfate method under pressurized conditions + ultrasonication	–	Width 7 nm, and an average length of 153 nm	45% yield, and zeta potential of −0.04 V	(Amoroso et al., 2020)
Sugarcane bagasse	NC	Bleaching + ionic liquid treatment under microwave heating	High pressure homogenization	36%	Diameter: 10–20 nm	Low thermal stability	(Li et al., 2012)
Eucalyptus pulp	CNF	Acid treatment	Ionic liquid treatment under microwave heating + high pressure homogenization	34.43%	Diameter: 20–100 nm	Low thermal stability, narrower molecular weight distribution	(Wang et al., 2017)
Wheat straw	CNF	Alkaline treatment	Enzymatic hydrolysis (FiberCare®) + Twin-screw extrusion	58%	Diameter of 15 nm and length of 991 nm	Yield of 42.31%, aspect ratio of 66	(Espinosa et al., 2019)
Bleached eucalyptus kraft pulp	CNC	–	Endoglucanase treatment + sonication	88%	Diameter of 6–10 nm	Formation of uniform CNC	(Siqueira et al., 2019)
Eucalyptus cellulose kraft pulp	CNC	Ball milling	Cellulolytic enzymatic (endoglucanase) + sonication	77.9–78.3%	Lengths: 294 nm and diameter: 24.0 nm	Highest yield of 24.6%	(Squinca et al., 2020)
Curauá fibres	CNF	Alkaline treatment + bleaching	Enzyme (FiberCare® R + Viscozyme® L) treatment + sonication	73–78%	55–109 nm diameter	Formation of rod like single crystals and nanofibers with larger diameters	(de Campos et al., 2013)
Soybean straw	CNF	Alkaline treatment + bleaching	Enzyme (Optimash™VR) treatment + homogenization + sonication	50%	9.4 nm diameter	High aspect ratio and thermal stability	(Martelli-Tosi et al., 2018)
Sugarcane bagasse	CNC	Alkaline treatment + bleaching	Enzyme hydrolysis (Cellic CTec3)	~70%	Diameter of 8.4–12.2 nm	11.3% yield, maximum degradation temperature of 346.3 °C	(De Aguiar et al., 2020)
Sugarcane straw	CNC	Alkaline treatment + bleaching	Enzyme hydrolysis (Cellic CTec3)	~70%	Diameter of 8.7–14.1 nm	12% yield, maximum degradation temperature of 351 °C	(De Aguiar et al., 2020)
Banana peel	CNF	–	Enzymatic hydrolysis using xylanase	61.5–66.2%	Diameter of 3.7–8.8 nm	Around 170.2 to 404.5 of aspect ratio	(Tibolla et al., 2019)
Bleached bagasse kraft pulp	CNF	–	Endoglucanase (FiberCare R) treatment + ultrafine grinding	–	Diameter of 9–26 nm	Maximum yield of 75.1%, high thermal stability	(Liu et al., 2020c)
Bleached kraft birch pulp	Cationic CNF and CNC	DAC production using sodium periodate oxidation	DES (aminoguanidine hydrochloride + glycerol) treatment of DAC + microfluidization	63.2–64.9% after DES treatment	Diameter of around 4.6 nm and CNC around 5.7 nm	5 times reutilization of DES	(Li et al., 2018a)
Moso bamboo	NC	DES (ChCl + LA) treatment	High speed homogenization + microfluidization	60.43% after DES treatment	Width of 20–80 nm	Aspect ratio of 67–101	(Liu et al., 2019)
Bleached birch pulp	CNF	DES (betaine hydrochloride + glycerol) treatment	Microfluidization	67.7–74.4%	Diameter of 17–20 nm	High CNF mass yield of up to 72.5%	(Hong et al., 2020b)
Softwood thermomechanical pulp	LNC	–	DES (ChCl + OAD) treatment + blending	55–61%	Width of 7.1 nm and thickness of 3.7 nm	57% yield and high thermal stability	(Jiang et al., 2020a)
Softwood bleached kraft pulp	CNC	Electron beam irradiation + alkali treatment	High pressure homogenization	71–81%	Width: 23–30 nm, Length: 128–747 nm	Aspect ratio of 6 to 27, improvement in thermal stability and surface charge	(Lee et al., 2018)

Table 3 (*continued*)

Source	Type of NC	Pretreatment	NC production	Crystallinity	Dimension	Other properties	References
Kenaf's core	NC	Alkali treatment + bleaching	Electron beam irradiation + acid hydrolysis	68.21–69.38%	–	Decrease in NC yield and thermal stability	(Kim et al., 2016)
Tall goldenrod plant	CNF	Alkali cooking + bleaching	Electron beam irradiation + manual grinding	–	Diameter: 160 nm (300 kGy treated sample)	Lower thermal stability of finely separated CNFs	(Kim et al., 2019)
Cotton linter	CNF	Electron beam irradiation	Grinding using super masscolloider	65.5–75.3%	Width of 30–70 nm	Low thermal degradation temperature	(Le & Seo, 2016)
Bleached softwood kraft pulp	CNC	Electron beam irradiation	Acid hydrolysis	50–55.8%	Width of 10–30 nm	Thermal degradation at around 150 to 250 °C	(Van Hai & Seo, 2017)
Cotton linter	CNC	Electron beam irradiation	Acid hydrolysis	64–70.2%	Width of around 33 nm	Thermal degradation at around 230 to 300 °C	(Van Hai & Seo, 2017)
Microcrystalline cellulose	CNF	–	Submerged liquid plasma + ultrasonication	83.6%	Width of 40–60 nm	Slight decrease in thermal stability	(Vizireanu et al., 2018)
Wheat straw	CNF	Phase one air plasma activation + mild alkali treatment	Phase two air plasma activation + TEMPO oxidation	–	Width of 30 ± 10 nm	Yield of 97.6%, energy consumption reduced by 90.4%	(Shaghaleh et al., 2021)

NC: nanocellulose, CNC: cellulose nanocrystals, CNF: cellulose nanofibers, LCNF: lignin containing nanocellulose, DAC: dialdehyde cellulose, DES: deep eutectic solvent, ChCl: choline chloride, LA: lactic acid, OAD: oxalic acid dihydrate.

purified cellulose reduced to 1/8 of traditional methods because of MW treatment. Further, CNF obtained from MW liquefied and chemically treated residue had a diameter of around 2 to 30 nm, a crystallinity index of 67.4% with high thermal stability. Similarly, Huang et al. (2017) carried out MW-assisted liquefaction of rape straw and treated the residue with dilute alkali (2% NaOH) and hydrogen peroxide (5% H$_2$O$_2$) followed by high-intensity ultrasonication (15 min) to produce CNCs. The hemicellulose and lignin contents from the biomass were effectively removed after liquefaction at 180 °C for 7.5 min. Further, CNCs obtained from liquefied, and chemically purified residue had an average diameter of around 12.59 nm with good thermal stability.

MW can be utilized to assist chemical pretreatment of biomass prior to production of CNFs using mechanical disintegration. In a recent study, Liu et al. (2020a) prepared CNFs from energy cane bagasse using MW-assisted NaOH/NaClO$_2$ treatments followed by wet-grinding and microfluidization. A delignification of 88.9% occurred while a cellulose fibres yield of 34.2% was obtained after MW treatment. The prepared CNFs showed shear-thinning behaviours with solid-like viscoelastic characteristics because of their entangled network structure. In another study, Louis and Venkatachalam (2020) reported an energy-efficient process technology using MW irradiation and ultrasound technology to produce nanocellulose from corn cob. Using microwave in the process, 97.31% delignification was achieved at optimum conditions of 180 W power, 12.86% sodium chlorite, and 16 min reaction time. Further, ultrasound-assisted alkali extraction resulted in a yield of 0.445 g of cellulose/g of the corn cob. Finally, acid hydrolysis reduced the cellulose's particle size from 894 nm to 131.4 nm. The prepared nanocellulose was reported to have a crystallinity index of 72.36%.

MW can be utilized in combination with other technologies to develop environmentally friendly processes for CNFs production with minimal utilization of chemicals. In a study, high purity CNFs from wheat straw were obtained using an eco-friendly method comprised of steam explosion (100 °C, 3 MPa for 2 min) and MW-assisted alkaline hydrolysis process (140 ± 2 °C for 20 min) followed by microfluidization (150 to 159 MPa). The obtained CNFs had an individual diameter of 5.42 nm with a width between 10 and 40 nm. The cellulose content increased from 44.81% in the raw wheat straw to 94.04% in CNFs. Besides, the crystallinity index increased with each treatment, and a maximum crystallinity index of 62.15% was observed after MW-assisted alkali treatment, whereas the CNFs had a crystallinity index of 58.62% (Liu et al., 2017). Recently, Impoolsup et al. (2020) utilized MW for pretreatment (in water at 850 W for 10 min and up to 3 rounds) of lime

residue to assist a zero-waste chemical-free CNFs production process that was comprised of high-shear and high-pressure homogenization. The produced CNFs had diameters in the range of 3 to 46 nm and a crystallinity index of 36 to 41%. In addition, the yield of CNFs reduced after each round of MW treatment. A maximum yield of around 43.17% was obtained when the sample treated with only one round of MW was used for CNFs production. On the other hand, the CNF yield reduced to approximately 37.85% and 36.70% after the second and third round of MW treatment, respectively.

The production of CNCs with the assistance of MW irradiation has been reported in many recent studies. MW-assisted dilute acid pretreatment combined with enzyme hydrolysis was used to prepare CNC from pure microcrystalline cellulose. The utilization of MW heating (at 300 W for 10 or 30 min) during acid hydrolysis pretreatment significantly increased the CNC yield of subsequent enzymatic hydrolysis. The highest CNC yield of 84.4 wt% and highest peak decomposition temperature of 378.1 °C were measured in the sample extracted in 5 wt% sulfuric acids under MW heating. The acid consumption in the process reduced significantly due to MW-assisted pretreatment. Hence, the product security improved while the process became energy-efficient and eco-friendly (Qian et al., 2020). Most recently, Amoroso et al. (2020) developed an MW-assisted ammonium persulfate (APS) method to extract CNC from cotton powder under pressurized conditions in a closed reaction system. The MW-assisted APS method needed only 90 min for the hydrolysis of cellulosic amorphous regions, while the conventional heating required 16 h. Further, CNCs extracted at optimum reaction conditions had average particle width of 7 nm, an average length of 153 nm, 45% yield, and zeta potential of −0.04 V. In addition, MW-assisted APS method gave a CNCs yield of 45.81 ± 3.79% which was comparable to those extracted using the conventional method (CNCs yield of 48.85 ± 11.99%) in much longer processing time. Besides, because of the rigorous control of the reaction conditions allowed by the microwave reactor, CNCs yield results were more reproducible in the MW-assisted method, with standard deviations much lower than the conventional production technique. Thus, the developed method proved to be energy-saving and highly efficient in the extraction of CNCs.

MW and ultrasound irradiation can be used together simultaneously as a process intensification technology for other chemical methods of nanocellulose production to improve yield and reduce reaction time. In the hybrid process, MW can improve the heat transmission rate as well as the reaction activity of cellulose, while ultrasound can enhance the efficiency of mass transfer among cellulosic fibrils (Lu et al., 2019). In a

study, Lu et al. (2019) utilized MW (500 W) and ultrasound (800 W) simultaneously for assisting oxalic acid hydrolysis (115 °C, 15–75 min) of bamboo pulp to produce CNCs. With MW and ultrasound-assisted acid hydrolysis for 30 min, the CNCs yield increased substantially to 80%. On the other hand, when only acid hydrolysis was used for 30 min without the assistance of MW and ultrasound, the CNCs could not be produced; however, when the hydrolysis time was increased to 360 min, a CNCs yield of only 13.8% was obtained. These findings revealed that MW – ultrasound hybrid processing technique considerably reduced reaction time and was very effective in nanocellulose production.

Although the utilization of MW required a higher initial investment, it has lower operational costs than conventional heating, allowing rapid recovery of capital investment. Apart from this, MW has other benefits such as short processing time, lower maintenance costs, energy conservation, and being environmentally friendly (Aguilar-Reynosa et al., 2017; Chizoba Ekezie et al., 2017). All these benefits make MW a promising emerging technology in nanocellulose production. However, various factors need to be considered during the development of a process involving MW to produce nanocellulose using a particular type of biomass at an industrial scale. Overall, the chemical and thermal processes that specific lignocellulosic biomass undergoes when subjected to MW treatment are complex. Thus, a thorough investigation of the morphological and chemical changes as well as the thermal degradation process of the lignocellulosic material and its relationship with electromagnetic energy and MW process parameters is required.

Besides, the major challenges of MW treatment on lignocellulosic biomass for nanocellulose production is establishing and understanding the relationship among MW process parameters (radiated power, treatment time, and temperature), the conditions of the reaction medium (e.g., additives, catalyzers, nonpolar or polar solvents selection, design of the reactor, and stirring intensity) and the lignocellulosic material's dielectric properties as well as its chemical composition, shape and size. Therefore, future studies should consider these factors while developing a MW-assisted process for nanocellulose production, which will enable the fabrication of intended products with sufficient repeatability data and industrial process scale-up (Romero-Zúñiga et al., 2021). In addition, future studies should also focus on developing eco-friendly and sustainable processes by combining MW with other emerging mechanical, chemical, and biological methods to produce nanocellulose with a higher yield, desirable characteristics, and at a lower cost.

5.2. Enzymatic production method

The enzymatic processing technology is an emerging route to obtain nanocellulose from lignocellulosic biomass, which involves several phases. The first phase is the pretreatment of lignocellulosic biomass to fractionate and recover the main components such as cellulose, hemicellulose, and lignin by generally using physical or chemical pretreatment methods. The second phase involves the controlled enzymatic
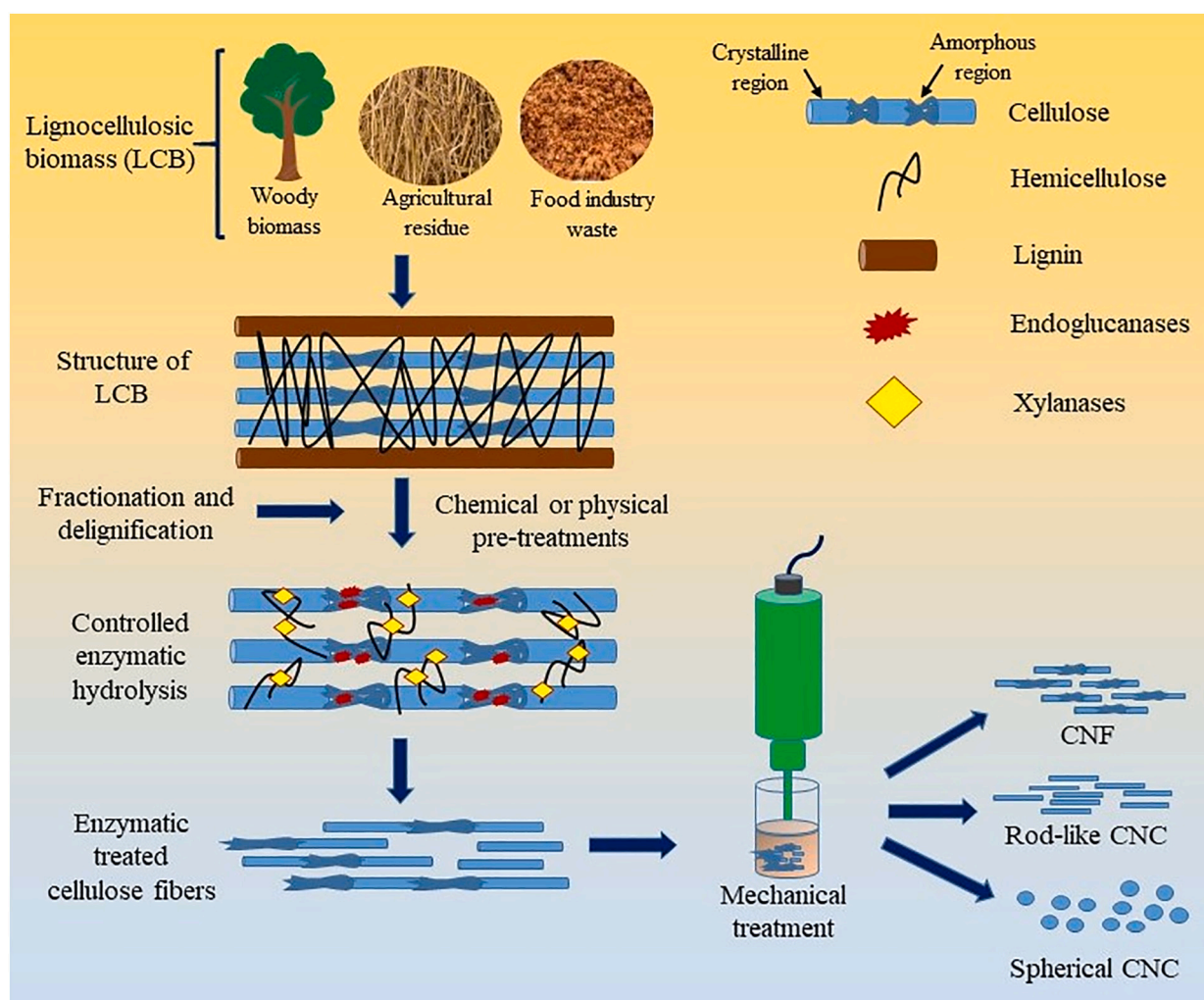

Fig. 1. A typical mechanism of production of nanocellulose from lignocellulosic biomass using enzymatic treatment (endoglucanases and xylanases) followed by mechanical treatment. [Figure is drawn using Microsoft PowerPoint 2016 (Microsoft Corporation) and EdrawMax (Version: 11.5.2; EdrawSoft, Wondershare Group]

hydrolysis of fibre samples. An enzymatic cocktail is used to solubilize the pretreated material in the buffer solution. The enzymatic cocktail's role is to break down the cellulose polymer into a smaller size. The third phase involves homogenizing the enzymatically treated fibres, generally using a microfluidizer, ultrasonicator, or ultrafine grinder, among others (Ribeiro et al., 2019). The typical mechanism involving all three phases of enzymatic production of nanocellulose from lignocellulosic biomass is depicted in Fig. 1.

The pulp obtained after the first phase of the enzymatic production mechanism is primarily comprised of cellulose and, in certain cases, residual hemicellulose. Thus, possible biocatalysts for application in nanocellulose isolation include carbohydrate-active enzymes, mainly cellulases, which are responsible for cleaving the β-1,4 glycosidic bonds in cellulose chains. However, depending on the morphological and physicochemical characteristics of the cellulose-rich pulp, additional carbohydrate-active enzymes, such as hemicellulases and other auxiliary enzymes (e.g., lytic polysaccharide monooxygenases) that work synergistically with carbohydrate-active cellulases, may be relevant as well (Arantes et al., 2020).

Although various enzymes have been utilized for nanocellulose production, cellulases account for the vast majority of these cases. Cellulases are a class of enzymes that directly catalyzes cellulose's breakdown into simpler sugars. They are normally produced by cellulolytic organisms, including those from the genera *Aspergillus*, *Trichoderma*, and *Clostridium*, among others (Michelin et al., 2020). The cellulases are classified into endoglucanase, cellobiohydrolase, and β-glucosidase. Endoglucanases cleave random internal β-1,4-glycosidic bonds of the cellulose chains, usually in amorphous areas, forming new cellulose chain ends. Cellobiohydrolase then progressively hydrolyzes these ends of the chain, yielding cellobiose as the major product. Finally, cellobiose is hydrolyzed into glucose by β-glucosidases (Michelin et al., 2020; Ribeiro et al., 2019).

The production of nanocellulose through the enzymatic route using cellulases enzyme has been investigated in many recent studies. Beltramino et al. (2018) obtained a CNC yield of around 82% from cotton linter treated with a cellulase enzyme (from *Cerrena* sp. fungus) at optimized conditions (hydrolysis 2 h using an enzyme dose of 20 U g^{-1} odp) followed by sulfuric acid hydrolysis (62% wt. H$_2$SO$_4$, 47 °C, 25 min). This yield was 21 percentage points higher than the CNC yield obtained from fibres not treated with the enzyme. Further, the surface charge reduced while the crystallinity of the CNC increased due to enzymatic pretreatment. Chen et al. (2019) prepared ribbon-like CNCs by conducting enzyme hydrolysis (50 °C, 5 to 11 h) of cotton pulp fibres using cellulase (from *Aspergillus niger*, the enzyme activity of 1.10×10^4 μ/mL). When a lower cellulase concentration was used, the endoglucanase truncated the cellulose chains at their amorphous zone and disintegrated them to produce ribbon-like CNCs with a diameter of around 45 nm. On the other hand, granular CNCs were noticed in the enzymolysis product when an enzyme concentration of 100 μ/mL was used. However, further increasing the concentration to 300 μ/mL resulted in completely granular CNCs. This meant that endoglucanase at a higher concentration truncated the cellulose chains at both crystalline and amorphous regions.

Mono-component endoglucanases have been utilized in the production of both CNCs and CNFs from different lignocellulosic biomass. Endoglucanase's efficiency is due to its mechanism of action, which targets the amorphous portions of cellulose fibres, selectively disrupting accessible glycosidic bonds and forming new reducing and non-reducing ends. Because of its characteristics, endoglucanase has the ability to stimulate specific alterations, which can help in the defibrillation process without affecting the crystalline portions. Besides, the wide utilization of endoglucanases is due to their defibrillation efficiency, biosafety, commercial availability, and relatively low cost (Berto et al., 2021). In a study, Liu et al. (2018) utilized mono-component endoglucanase (FiberCare R; cellulolytic activity of 2036 U/mL) for enzymatic pretreatment (enzyme dosage of 3% based on the weight of dried pulp,

5% pulp consistency, 12 h, 50 °C, pH 7, at 200 rpm) of bleached bagasse (BBK) and softwood kraft pulp (BSK) prior to grinding for CNFs production. Enzymatic treatment significantly enhanced the yield of CNFs, and a maximum yield of 70.56% was obtained in enzyme-treated BBK after 2 h of grinding. Besides, the specific net energy consumption during defibrillation was reduced by 59.71% and 42.98% for BBK and BSK, respectively, after enzymatic pretreatment. In another study, Squinca et al. (2020) obtained a maximum CNCs yield of 24.6% by pretreating the eucalyptus cellulose kraft pulp using ball milling (90 min) followed by enzyme hydrolysis using a cellulolytic enzymatic complex (production at on-site using *Aspergillus niger*) with high endoglucanase specific activity (17.09 IU/mg$_{protein}$) and 5 min of sonication. The obtained CNCs had a diameter of 24.0 nm and a length of 294 nm with a crystallinity index of 78.3%. These results indicated that nanocellulose could be successfully isolated from biomass using on-site produced enzymes.

Many recent studies have reported the use of xylanases in the enzymatic production of nanocellulose. The glycosidic linkages of hemicelluloses can be hydrolyzed, and the xylan chains from the surface of cellulose can be peeled off by xylanases. Furthermore, using xylanases allows xylans to be converted into renewable fuels and other value-added products. In addition, xylanases synergistically work with cellulases to improve the properties of fibre by enhancing its porosity and swelling capacity (Rossi et al., 2021). In a study, xylanase (activity of 2980 IU/g) utilized for enzyme hydrolysis of (5 and 30 IU/g, 50 °C for 2 h) unbleached eucalyptus pulp followed by superfine grinding and microfluidization (20 times, 20,000 psi) resulted in CNFs having high dispersion stability, uniform particle size distribution, low hemicellulose content, and high crystallinity index (64% using 30 IU/g) (Nie et al., 2018). Similarly, Zhang et al. (2018) used xylanase (enzyme activity 2980 IU/g) in a concentration of 0 IU/g (control), 5, 10, and 30 IU/g for enzyme hydrolysis of unbleached bagasse pulp (50 °C for 2 h) followed by superfine grinding and microfluidization to prepare CNFs. With the increase in enzyme concentration, there was a reduction in particle diameter and thermal stability of CNFs, while there was an increase in carboxyl group content and zeta potential.

Xylanases and cellulases can be utilized in preparing a suitable enzyme mixture for the preparation of nanocellulose. In a study, Tong et al. (2020) reported that using an enzyme mixture comprising of cellulase (*Trichoderma reesei*, activity of 1.1×10^4 U mL^{-1}) and xylanase (*T. reesei*, activity of 2.2×10^4 U mL^{-1}) in a ratio of 9:1, enzyme concentration of 10 U mL^{-1} for 12 h hydrolysis of bleached eucalyptus pulp resulted in rod-like CNCs with a width of 30 nm and length of 600 nm. On the other hand, spherical CNCs were produced when enzyme concentration was increased to 500 U mL^{-1} and hydrolysis time reduced to 5 h. The study indicated that by varying the process parameters of enzyme hydrolysis and by using the cellulase-xylanase mixture, the morphology of CNCs could be controlled.

Commercial enzymes showing both xylanase and endoglucanase activity have been reported for nanocellulose production. In a study, Martelli-Tosi et al. (2018) produced CNFs from mercerized soybean straw (MSS) by treating it with Optimash™ VR enzyme (446 U of xylanase and 134 U of endoglucanase per gram of MSS) followed by mechanical homogenization (5 min) and ultrasonication (3 min). They also prepared CNCs from MSS using sulfuric acid hydrolysis (at 70 °C for 40 min) followed by the same mechanical treatment. The CNCs had a crystallinity index of 57%, with a length of around 300 nm and a thickness of 10 nm. On the other hand, the CNFs had a lower crystallinity index (50%), higher thermal stability, length greater than 1 μm, and diameters similar to CNCs. In addition, a CNFs yield of around 13.3% was achieved using enzymatic processing, which was slightly higher than the yield of CNCs (around 12.4%) obtained using acid hydrolysis.

Lytic polysaccharide monooxygenases (LPMOs) are a prominent class of enzymes that have been utilized in nanocellulose production. LPMOs initiate a very selective oxidation process on cellulose at either

the C1 or C4 position, resulting in the formation of aldonic acids or gem-diols, respectively (Koskela et al., 2019). The formed aldonic acids possess an ionizable carboxyl group that improves the electrostatic repulsion of cellulose, thereby accelerating the fibrillation process while reducing the energy requirements of mechanical treatment. Besides, LPMOs might partially or completely eliminate the requirement for harsh chemical treatments like bleaching that cause fibre alteration and swelling (Karnaouri et al., 2020).

In a study, Moreau et al. (2019) used fungal LPMO from AA9 family (PaLPMO9E) for pretreatment (for 24 h at 50 °C) of bleached birchwood Kraft fibres followed by homogenization and microfluidization. Without any further mechanical treatments, the enzyme-treated samples were successfully processed through the microfluidizer and resulted in a CNF yield of around 60–65% with a crystallinity index of around 47.65 to 53.71%. On the other hand, the control samples treated under the same pretreatment conditions but without using enzyme could not be homogenized, as they blocked the system at the entrance to the cell. In another research, Koskela et al. (2019) reported the use of two C1-active LPMOs from the fungus *Neurospora crassa* (NcLPMO9E and NcLPMO9F) for enzymatic treatment (25 $\pm$ 2 °C for 2 days) of delignified softwood fibres followed by homogenization for CNFs production. They also investigated the influence of the carbohydrate-binding module (CBM) on nanofibrillation of cellulose. The CBM-lacking LPMO (NcLPMO9F) introduced the carboxyl groups (0.53 mmol g^{-1}) on the surface of cellulose more effectively and produced CNFs with a thinner width (4.3 $\pm$ 1.5 nm) compared to that of the CBM-containing LPMO (NcLPMO9E) which resulted in carboxylate content of 0.38 mmol g^{-1} and CNFs having width of around 6.7 $\pm$ 2.5 nm. In addition, the CNFs yield obtained using CBM-lacking LPMO (81.8 $\pm$ 5.0%) was higher than that of CBM-containing LPMO (65.4 $\pm$ 0.5%). Similarly, the production of CNCs using CBM-containing LPMO (NcLPMO9E; 30 °C and pH 6) and CBM-lacking LPMO (NcLPMO9F; 55 °C and pH 5) from microcrystalline cellulose has been reported by Koskela et al. (2021). The carboxylate content in CBM-lacking LPMO (0.70 $\pm$ 0.09 mmol g^{-1}) was higher compared to that in CBM-containing LPMO sample (0.40 $\pm$ 0.07 mmol g^{-1}). In addition, the CNCs yield obtained using CBM-lacking LPMO treated sample was 13.3%, which was higher than the yield obtained using CBM-containing LPMO sample (12.5%) and control sample (no enzyme treatment; yield of 7.4%). These results indicated that utilization of LMPO without a CBM is more appropriate for production of both CNFs and CNCs.

Enzyme mixture comprising of LPMO, xylanases, and endoglucanases can improve the accessibility of the enzymes to the cellulosic component (Hu et al., 2018). Thus, several studies have reported the application of this enzyme mixture in nanocellulose production. In a study, Hu et al. (2018) enzymatically pretreated the hardwood kraft pulp using the different combinations of endoglucanase (EG), LPMO (AA9), and endoxylanase (EX) at an enzyme loading of 1 mg of each enzyme per g substrate (dry weight). The ternary mixture (EG + EX+AA9) hydrolyzed 7% of cellulose and 30% of xylans and reduced the degree of polymerization significantly higher than the samples treated with EG, EG + EX, and EG + AA9. Further sonication (30% amplitude, 150 W for 20 min) of the treated sample resulted in CNFs with high zeta potential and high transmittance value. The transmittance of CNFs obtained from the sample treated with ternary mixture increased 7 times (35%) compared to that of the control sample (5%). In addition, the synergistic coordination of EG, AA9, and EX improved fibre breakdown, resulting in non-aggregated, individual CNFs that were clearly visible. Most recently, Rossi et al. (2021) used a mixture comprising of AA9 LPMO from *Thermothelomyces thermophilus* (TtLPMO9H), GH10 xylanase from *Thermobacillus composti* (TcXyn10A), and GH7 EG from *Trichoderma harzianum* (ThCel7B) at an enzyme loading of 1 mg of each enzyme per g of chemically pretreated sugarcane bagasse (dry weight) followed by ultrasonication for preparation of CNFs. Compared to the CNFs prepared using TEMPO oxidation, the CNFs obtained through the enzymatic route were more thermostable (resisting up to 260 °C for the

initial degradation temperature) and significantly longer. Thus, an enzymatic route could be preferred where highly thermostable CNFs are required, such as moulded nanocomposite and electronic components manufacturing applications.

Prior to nanocellulose production, cellulose fibres can be isolated from lignocellulosic biomass through enzymatic route with the assistance of enzyme mixture comprising of multiple commercial enzymes with different enzyme activities. In a recent study, Perzon et al. (2020) demonstrated that cellulose fibres could be isolated from sugar beet pulp by enzyme hydrolysis (pH 5, 40 °C, 2 h) using an enzyme cocktail that comprised of Aquazym 240 L (Alpha-amylase, 10 μL/g pulp), Fibercare R (Cellulase, 5 μL/g pulp), Pulpzyme HC (Endo-xylanase, 10 μL/g pulp), Pectinex Ultra Clear (Polygalacturonase, 10 μL/g pulp) and Viscozyme L (Beta-glucanase, 10 μL/g pulp) and subsequent NaClO$_2$ treatment (pH 5, 70 °C, 2 h). Using enzyme hydrolysis, the non-cellulosic polysaccharides were removed to the same extent as the chemical treatment by pre-incubating the pulp at pH 9. Further microfluidization of the fibre samples resulted in CNFs. The developed process consumed 67% less water and generated less toxic effluent than the chemical production method (NaOH and NaClO$_2$ treatment followed by microfluidization). The authors further suggested that if the bleaching (NaClO$_2$) step could be removed, the water consumption could be completely avoided. This study indicated that a more sustainable process can be developed using enzymes in nanocellulose production.

Process intensification technologies such as ultrasound can be utilized in combination with enzyme hydrolysis to improve the efficiency of nanocellulose production as well as reduce the reaction time. Ultrasound can increase the contact area of substrate and enzyme as well as the temperature, potentially accelerating the enzyme-substrate reaction (Cui et al., 2016). In a study, ultrasonication in combination with enzyme hydrolysis (Celluclast 1.5 L having enzyme activity of 700 endoglucanase unit per g) was utilized for the preparation of CNC from wheat microcrystalline cellulose (MCC). During the 120 h of enzyme hydrolysis period, the use of ultrasonic treatment for 60 min at every 12 h interval led to an increase in CNC yield from 15.76% to 22.57%. The prepared CNCs have a rod-like structure with a width of less than 10 nm and a length between 50 and 80 nm. Ultrasonic treatment also increased relative crystallinity from 60.37% of MCC to 87.46% of CNC (Cui et al., 2016).

Immobilized enzymes can be utilized in nanocellulose preparation which can reduce the overall cost of production as the enzyme can be reused over multiple cycles. In a study, Yassin et al. (2019) produced CNFs from bleached bagasse pulp by conducting enzyme hydrolysis (50 °C for 6 h) by using a cellulase enzyme that was covalently immobilized onto the carrageenan/polyamidoamine/glutaraldehyde gel disk. The immobilized enzyme retained 85% of its activity even after six cycles. In addition, the immobilized cellulase successfully hydrolyzed the amorphous portion of cellulose which resulted in the breakdown of fibres to nanometer scale. The produced CNFs had a width of about 15 to 35 nm with a length of several micrometers. Besides, around 70% of the CNFs had a diameter between 20 and 30 nm. Moreover, the authors concluded that the developed method involving immobilize cellulase could be a promising green and economic route for the industrial production of CNFs.

The enzymatic pathways for the production of nanocellulose from lignocellulosic biomass offer several advantages compared to conventional methods. One advantage is that the enzyme hydrolysis can be conducted under milder conditions, including moderate pressure, temperature, and pH. This can offer various advantages from an operational aspect, including a safer operation, lower energy consumption, no requirement of corrosion-resistant processing equipment and reduced cost. Another benefit is that the use of chemicals during the process may be reduced or avoided entirely, reducing the quantity of effluents produced and thereby ensuring environmental protection. The reduction or elimination of the utilization of chemicals will allow the use of a final stream of sugars to produce other products, such as biofuels, assisting in

the execution of a biorefinery model (Michelin et al., 2020). However, regardless of various advantages over chemical methods, the high cost of enzymes remains a significant obstacle that needs to be addressed. As a result, cost-cutting measures aimed at their reuse, such as enzyme immobilization as well as advances in production and purification of enzymes, can help overcome this hurdle (Arantes et al., 2020).

When enzymes are used in their free form, they can be used only once as it is difficult to separate the enzyme from the reaction mixture. Besides, extra energy is consumed during the boiling process to quench the enzyme's catalytic activity at the end of the reaction. When enzymes are immobilized, they can be easily separated from the product by filtration. On the one hand, utilization of immobilized enzyme could reduce the energy consumption as there is no need for boiling to quench the enzyme activity at the end of the reaction. On the other hand, the immobilized enzyme could be retrieved and reused across numerous cycles, lowering the overall cost of employing fresh enzymes. Thus, using immobilized enzymes in nanocellulose production has the potential to reduce costs. As a result, this method offers a promising eco-friendly and cost-effective route for industrial nanocellulose production (Yassin et al., 2019).

Further research can be carried out to evaluate the effectiveness of using different immobilized enzymes to produce nanocellulose from a range of lignocellulosic materials. Besides, the potential of reducing production time and achieving desirable nanocellulose properties by utilizing immobilized enzymes with the assistance of process intensification technologies such as ultrasound and others can be investigated. In addition, the efficiency of using process intensification technology for assisting the multi-component enzyme mixture in nanocellulose production can be evaluated. Furthermore, the suitability of developing hybrid chemical-free production techniques by combining enzymatic processing with other emerging methods can be investigated. Moreover, the yield of CNCs and CNFs produced through the enzymatic route have been reported in a limited number of studies. As yield is one of the most critical factors in determining the efficiency of a production process, it is recommended to be determined and reported in all future studies.

5.3. Deep eutectic solvent

Deep eutectic solvents (DESs) are emerging green solvents that adhere to all the 12 principles of green chemistry (Ma et al., 2019). DESs are prepared by mixing two or more substances at a proper ratio, normally with moderate heating (Abad-Gil et al., 2021; Smith et al., 2014a, 2014b). The substances used for making DESs can stabilize the interactions of hydrogen bonds to generate a eutectic mixture that has a lower melting point compared to the individual substances. Typically, quaternary ammonium salts, which act as hydrogen-bond acceptors, are mixed with hydrogen-bond donors to form DESs (Abad-Gil et al., 2021). The simple preparation method, eco-friendly characteristics, recyclability, and biodegradability are the major advantageous features of DESs. Besides, they possess various unique properties such as non-flammability, high thermal stability, relatively wide liquid range, and low vapour pressure (Ma et al., 2019; Smith et al., 2014a, 2014b). Due to their beneficial characteristic, DESs can be utilized in various chemical reactions at near-ambient conditions and in a range of industrial applications (Ma et al., 2019).

In recent years, the application of DESs as a green solvent in valorising lignocellulosic biomass has gained significant attention from the research community. DESs can disintegrate the recalcitrance structure of lignocellulosic biomass by cleaving the hydrogen bonding between the carbohydrates and lignin, and therefore, exposing cellulose. Among various value-added products, the DESs treatment of biomass for nanocellulose production has recently attracted considerable attention (Li et al., 2021b). In addition, DESs can also be utilized for treatment of bleached cellulosic pulp for nanocellulose production. A typical mechanism of applying DESs in nanocellulose production has been depicted in Fig. 2. Generally, a mechanical or chemical process is required following the DES pretreatment to convert the isolated cellulose into nanocellulose.

DESs treatment for the production of CNF from different lignocellulosic materials has been reported in several recent studies. For instance, Acidic DES prepared using choline chloride (ChCl), and oxalic
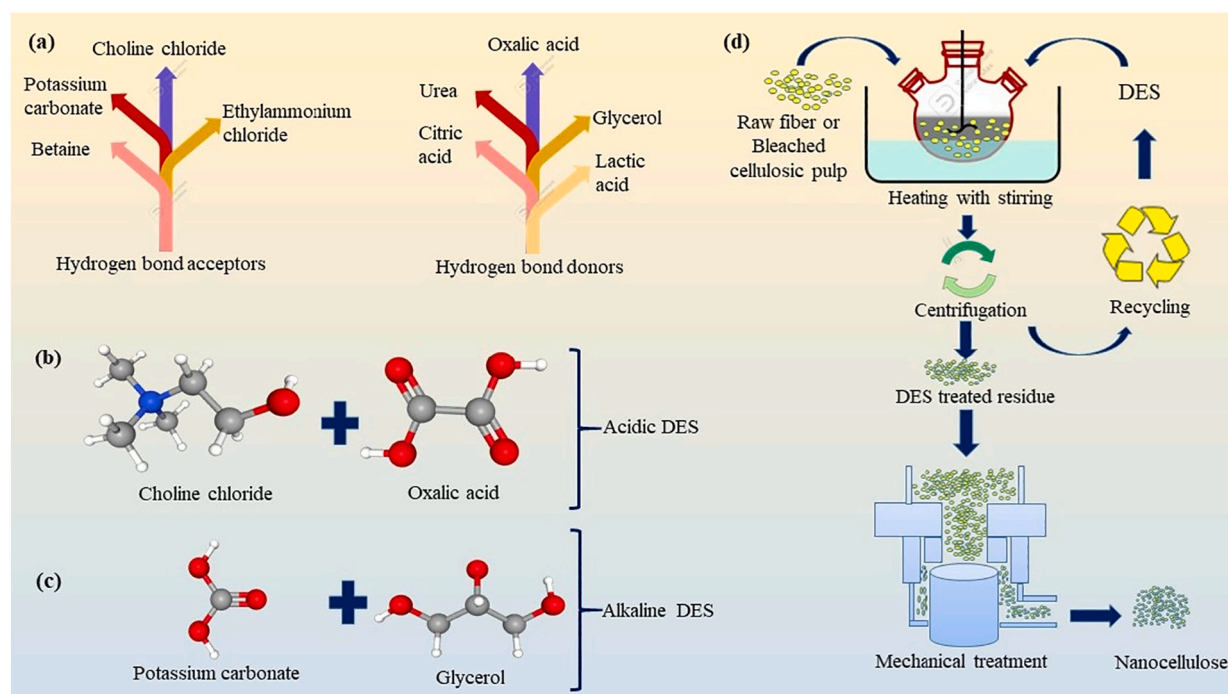

Fig. 2. (a) Commonly used hydrogen bond acceptors and hydrogen bond donors in DES preparation; (b) Example of an acidic DES formulation involving choline chloride (PubChem Identifier: CID 6209) and oxalic acid (PubChem Identifier: CID 971); (c) Example of an alkaline DES formulation involving potassium carbonate (PubChem Identifier: CID11430) and glycerol (PubChem Identifier: CID 753); (d) A typical mechanism of applying DESs in nanocellulose production. [Figure is drawn using Microsoft PowerPoint 2016 (Microsoft Corporation) and EdrawMax (Version: 11.5.2; EdrawSoft, Wondershare Group]

acid dehydrate (OAD) at a molar ratio of 1:1 was utilized for pretreatment of raw ramie fibres. The CNF obtained by ball milling of DES pretreated (for 4 h at 100 °C) fibres had an average width of 14.29 nm along with a high purity of glucan up to 90.31%. The produced CNF also exhibited high thermal stability (>316.7 °C) and high crystallinity (79.17%) (Yu et al., 2021). Similarly, Liu et al. (2019) prepared nanocellulose fibres from Moso bamboo pretreated with a DES prepared with ChCl and lactic acid (LA) (molar ratio of 1:9). The pretreatment carried out for 3 h at 120 °C with a solid-to-liquid ratio of 1:25 resulted in 94.39% delignification and 91% cellulose recovery. Further mechanical disintegration of pretreated samples resulted in nanofibers with 20 to 80 nm widths. Liu et al. (2021b) obtained a CNFs yield of 84.19% from bleached softwood kraft pulp using optimal DES (ChCl and anhydrous citric acid at a molar ratio of 1:1) pretreatment at 90 °C for 2 h followed by high-pressure homogenization. Furthermore, the CNFs had high crystallinity (81.36%) and narrow diameter distribution (21 ± 3 nm). In another research, Liu et al. (2021a) reported that the esterification of cellulose using carboxylic acid-based DESs prevented the over-hydrolysis and dissolution of cellulose during pretreatment. Further, the subsequent mechanical extrusion and colloidal milling of esterified cellulose resulted in a CNFs yield of 72 to 88%.

The production of CNCs with the assistance of DESs has been reported in many recent studies. For example, Douard et al. (2021) reported that a CNC yield of around 43.6% and crystallinity index of around 81% was obtained by using a DES (ChCl and OAD at a molar ratio of 1:1) treatment for a period of 6 h at 95 °C with a fibre concentration of 2%. Similarly, Lim et al. (2021) treated bleached rice straw pulp using oxalic acid – ChCl DES (molar ratio of 1:1) at 80 °C for 4 h under continuous stirring at 300 rpm and obtained a 55.1% CNCs yield. In addition, the DES-produced CNCs had higher thermal stability than the CNCs produced using the sulfuric acid hydrolysis method. Gan et al. (2020) reported the utilization of an alkaline DES (potassium carbonate and glycerol at a molar ratio of 1:7) with bleaching for dissolution of lignin and hemicellulose from oil palm empty fruit bunch prior to CNC production. A CNC yield of 37.1% with a crystallinity index of 65.3% was obtained by acid hydrolyzing the DES pretreated sample using 60.0 wt% acid concentration at 46.1 °C for 58.5 min.

The bleaching or pulping process is not required to produce lignin-containing nanocellulose (LNC) from lignocellulosic feedstock. Thus, the application of DESs in the production of LNC could increase the yield and reduce the requirement of energy and chemicals. In a study, Jiang et al. (2020b) used acidic DES prepared using OAD and ChCl (1:2, 1:1, 2:1, and 4:1 molar ratios) for pretreatment of softwood thermo-mechanical pulp to produce LNC. Under optimal pretreatment parameters (1:1 molar ratio, 6 h, and 90 °C), an LNC yield of 57% was achieved. Further, the LNC had a high lignin content of 32.6%. Besides, the lignin nanoparticles of 20–50 nm were uniformly distributed in LNC suspension. Similarly, Hong et al. (2020a) used acidic DES (ChCl and OAD at a molar ratio of 1:1) at 90 °C for 150 min for the pretreatment of non-wood biomass of luffa sponge and obtained a solid fraction which contained 76.4 wt% of cellulose and 10.7 wt% of lignin. Further, ultrasonication of DES-treated sample resulted in LNC yield of 59.1 wt% which was higher the LNC yield obtained using sulfuric acid hydrolysis method (50.5 wt%). In another research, Shu et al. (2022) pretreated Poplar using ternary DESs (prepared with ChCl, LA, and p-toluene-sulfonic acid) for isolation of LNC. A maximum LNC yield of 64.65% was obtained at optimized pretreatment conditions (molar ratio of 2:10:1 and 100 °C). Further, the LNC had high lignin content of 27.65% and showed good dispersion stability in water.

Compared to traditional physical heating techniques, the combination of various process intensification technologies such as microwave irradiation with DES pretreatment can considerably shorten the extraction/reaction time without affecting the ease of control (Ji et al., 2021). For instance, Liu et al. (2020b) used microwave-assisted DES (ChCl and LA with 1:10 molar ratio) (MV-DES) for pretreatment of energy cane bagasse (ECB) to produce lignin-containing cellulose

nanofibers (LCNFs). Using MV-DES treatment at 110 °C for 30 min, a delignification of 81.0% along with a lignocellulose yield of 45.2% was achieved. Further ultrasonication of the MV-DES pretreated lignocellulose resulted in LCNFs having a highly entangled network and a crystallinity index of 35.3%. Similarly, Ji et al. (2021) reported an ultrafast fabrication process of CNFs from sugarcane bagasse by combining microwave heating (20 min, 2800 W, and 80–120 °C) with the ternary carboxylic acid DES followed by sweep frequency ultrasonic (SFU) separation pretreatment. A cellulose yield of 56.2% was obtained when the biomass was pretreated using TCADES1 (ChCl, oxalic acid, and Aluminum chloride hexahydrate at a molar ratio of 1:1:0.2) while 62.6% yield was recorded when TCADES2 (ChCl, LA and Aluminum chloride hexahydrate at a molar ratio of 1:1:0.2). Further, high-intensity ultrasonication of the pretreated sample resulted in CNF having a height of about 6–7 nm and width of around 15–17 nm with high thermal stability.

The recycling of DESs can be beneficial in protecting the environment and minimizing the production cost of nanocellulose (Yang et al., 2019). Thus, the recycling and reuse potential of DESs during nanocellulose production has been investigated in many recent studies. In a study, Li et al. (2018b) prepared a recyclable DES using aminoguanidine hydrochloride and glycerol in a molar ratio of 1:2 for the production of cationic nanocellulose from bleached kraft birch (*Betula pendula*) pulp sheets. The DES was recyclable five times without reducing the reaction efficiency. Further, the cationic dialdehyde celluloses (CDACs) formed using DES under reaction conditions of 80 °C for 10 min had a high charge density of 2.48 mmol g^{-1}. In addition, the cationic CNCs and CNFs produced from CDACs had an average width of 5.7 ± 1.3 nm and 4.6 ± 1.1 nm, respectively. In another research, Wang et al. (2020a) utilized ChCl and OAD to prepare a recyclable DES for pretreatment of cotton fibre to produce CNC. The DES had high recyclability (>85%) and could be reutilized for at least three more pretreatment cycles without compromising the pretreatment efficacy. Further, subsequent high-pressure homogenization of the DES pretreated sample resulted in CNC having a 50–100 nm diameter with more than one month of storage stability. Yang et al. (2019) developed a recyclable FeCl$_3$-catalyzed DES (F-DES) using OAD, ChCl, and FeCl$_3$·6H$_2$O in a molar ratio of 4.43:1:0.1 for production of CNCs from bleached eucalyptus kraft pulp (BEKP) without any subsequent mechanical treatment. A CNCs yield of over 90% (based on the cellulose content in BEKP) was obtained after one step F-DES treatment for 6 h at 80 °C. In addition, a CNCs yield of around 75% was achieved when F-DES was recycled and reused for the third time. Furthermore, the CNCs produced using F-DES had superior dispersion stability in water and much higher thermal stability compared to the CNCs produced using the conventional sulfuric acid hydrolysis method.

Even while both binary and ternary DESs seem promising in nanocellulose synthesis, there are still difficulties in using them on an industrial scale for nanocellulose production. The most evident issues are high cost and high viscosity, which causes a low mass concentration of dissolution and a slower solvent-transfer process (Ma et al., 2019). Dilution with water is an effective approach to reduce viscosity and alter suitable DESs. One probable explanation is that water is a low-molecular-weight polar solvent that functions as a third type of ingredient in the DES system (Dai et al., 2015; Ma et al., 2019; Xie et al., 2014). Ma et al. (2019) used hydrated ChCl – OAD (1:1 molar ratio) DES for pretreatment of kraft pulp from poplar wood to produce nanocellulose. Water was mixed with DES at the volume concentrations of 10, 20, and 30% to synthesize hydrated DES. The lignin and hemicellulose content in the pulp was reduced after one pass through hydrated DESs, and minimum lignin content of 1.2% was recorded in the pulp pretreated with DES containing 30% water. Further ultrasonication (800 W, 20 min) of the 10% DES-pretreated pulp resulted in CNFs, while the ultrasonication of 20% DES and 30% DES-pretreated pulp resulted in CNCs. The widths of both CNCs and CNFs were mainly within 20 nm.

To the best of our knowledge, there is relatively little information in

the literature regarding the utilization of hydrated DESs in nanocellulose production. Therefore, more research is necessary to fully comprehend the applicability and efficiency of employing hydrated DESs in the large-scale production of nanocellulose. Research can be done by employing a range of hydrated DESs in various lignocellulosic materials and optimizing the process conditions such as reaction temperature, treatment time, and solid-to-liquid ratio. In addition, the molar ratio for preparing DESs, and the water to DESs ratio can be optimized to obtain the most suitable DESs for specific lignocellulosic feedstock. Furthermore, the use of hydrated DESs in conjunction with process intensification technologies such as microwave, ultrasonication, and others can be investigated to understand how effective the hybrid process is in reducing reaction time and making the process economically viable for nanocellulose production.

5.4. Electron beam irradiation

Electron beam irradiation (EBI) is an emerging technology in the field of nanocellulose production. The technology can be utilized both as a pretreatment method to isolate cellulose fibres from lignocellulosic feedstock and as a fibrillation method to reduce the cellulose fibres size to the nanometer range. The electron beam ionizing radiations are generated using a linear accelerator (Hassan et al., 2018). The EBI treatment leads to the generation of free radicals either by cleaving glycosidic bonds or by removing hydrogen from a glucose moiety followed by another degradation process. The free radicals then induce the degradation of cellulose by oxidation or chain scission. At higher doses of EBI, chain scission primarily occurs which might be because of the breakdown of glycosidic bonds, whereas a lower dose of EBI might cause a cross-linking reaction instead of chain scission (Lee et al., 2018).

In a study, Kim et al. (2016) utilized EBI in the production of nanocellulose from the kenaf core. Cellulose fibres isolated using alkali pretreatment and bleaching were subjected to EBI (50 to 200 kGy) followed by acid hydrolysis. With the increase in the EBI dose, there was a reduction in the molecular weight, crystallinity, polydispersity, and

decomposition temperature of cellulose. These results affected the yield and size distribution during acid hydrolysis. The size distribution of nanocellulose became narrower while the yield reduced with an increase in absorbed dose and acid hydrolysis reaction time. A maximum yield of 45.8% was achieved when the irradiated sample (treated at 50 kGy) was acid hydrolyzed for 30 min. However, acid hydrolysis (30 min) of the non-irradiated sample resulted in a higher nanocellulose yield of 50.7%.

EBI has been utilized to produce CNCs and CNFs from different cellulosic materials. In a study, Le and Seo (2016) prepared CNFs from cotton linter using EBI treatment (10 and 100 kGy) followed by grinding with a super masscolloider. CNFs from the 10 kGy treated sample had a crystallinity index of 75.3% and a width of 30–70 nm. On the other hand, CNFs with a width of 50–70 nm and a crystallinity index of 65.5% was obtained from samples treated with 100 kGy. Besides, the thermal decomposition temperature of raw cellulose and CNFs decreased with the increase in irradiation dose 10 kGy to 100 kGy. Similarly, Van Hai and Seo (2017) utilized cotton linter and bleached softwood kraft pulp (BSKP) to produce CNCs through EBI treatment (0, 10, 20, and 100 kGy) followed by sulfuric acid hydrolysis. The yield, crystallinity index, and thermal stability of CNCs (from cotton linter and BSKP) decreased with the increase in electron beam dosage. Maximum CNCs yields of 33.4% and 24.5% were obtained from a non-irradiated sample of cotton linter and BSKP, respectively. In the irradiated sample, maximum CNCs of yield 31.3% (cotton linter) and 20% (BSKP) were achieved when the samples were treated with a 10 kGy EBI dose.

EBI treatment in combination with other mechanical methods such as HPH could be a promising method in nanocellulose production. Lee et al. (2018) proposed a novel method to prepare CNC from once-dried softwood bleached kraft pulp by combining EBI pretreatment with HPH (Fig. 3). The 1st step was EBI pretreatment performed in the solid-state at atmospheric pressure and ambient temperature followed by mild and simple alkali treatment. The EBI treatment of native cellulose using an irradiation dose of up to 100 kGy (for downgradation) resulted in a quick reduction in the degree of polymerization (DP) from 998 to 156, a
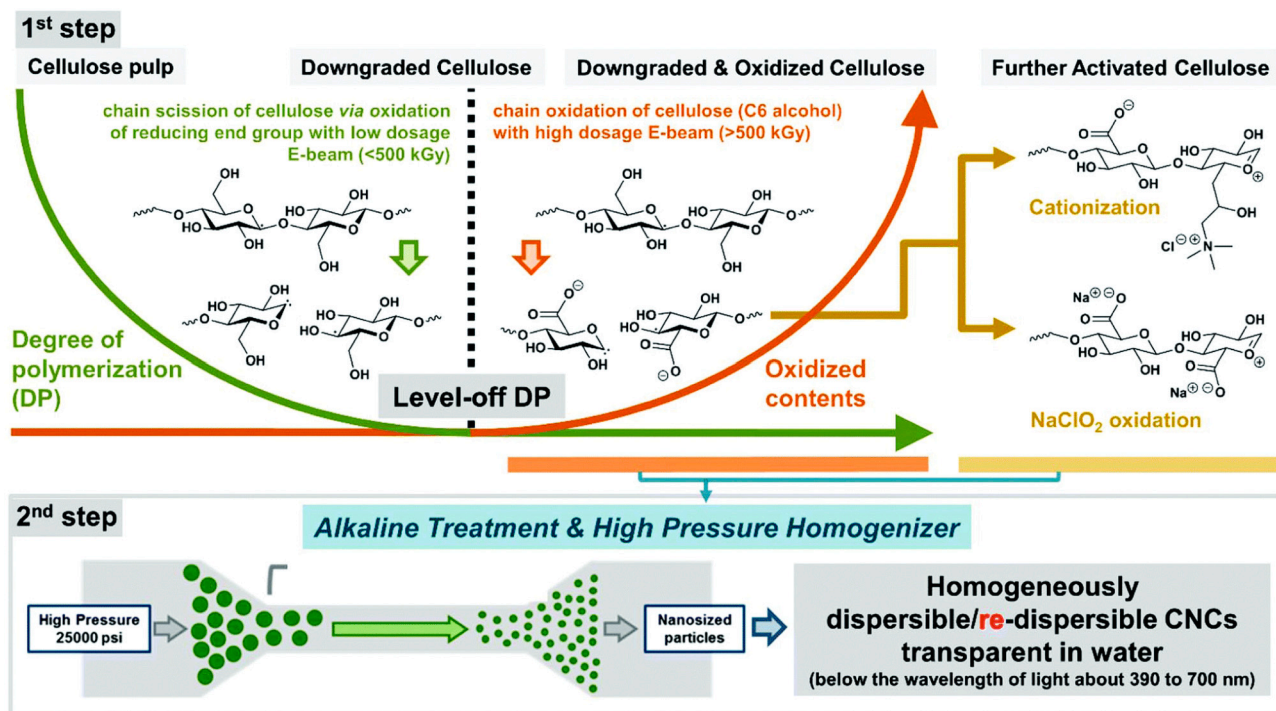

Fig. 3. EBI pretreatment combined with HPH for CNC production. Step-1: dissociation of cellulose pulp with EBI followed by treatment with alkaline treatment or/ and further oxidation/cationization; Step-2: HPH treatment for disintegration of the dissociated cellulose pulps. Reprinted with permission from the Royal Society of Chemistry (Lee et al., 2018).

so-called level-off DP by EBI. However, increasing the irradiation dose from 200 to 3000 kGy (for oxidation), the DP reduced very slowly from 126 to 59. Further oxidation (using $NaClO_2$) and cationization (using glycidyltrimethylammonium chloride) of the sample treated with 1500 kGy resulted in a DP of 72 and 63, respectively. The 2nd step involved the utilization of HPH (10 passes at 25,000 psi and 25 °C) for producing CNC from the EBI – alkaline treated sample. The extracted CNCs had a rod-like shape, a high crystallinity index of 71–81%, and high thermal stability. The severity of irradiation greatly affected the dimensions and surface charge of CNCs. With the increase in irradiation severity from 500 to 3000 kGy, the average length of CNC reduced from 747 to 128 nm, width reduced to 23 nm from 30 nm, and surface charge increased from −30.5 to −47.5 mV. In addition, a maximum CNC yield of 67.4% was achieved when an irradiation dose of 500 kGy was used. However, the CNC yield reduced to 35.3% with the increase in irradiation dose from 500 kGy to 3000 kGy. Based on these results, the authors concluded that the combination of the EBI and HPH technology provides a promising and effective approach with a low environmental impact for the sustainability of the CNC industry.

A recent study by Kim et al. (2019) showed the possibility of producing CNF from pulps of the tall goldenrod plant using only EBI, without combining it with any other technology such as HPH or acid hydrolysis. The EBI of cellulose fibres was conducted by keeping a 20 cm distance between the samples and window, and irradiation width of 110 cm under a scanned beam of 1.14 MeV accelerating voltage, 7.6 mA beam current. The EBI dose rate under an air atmosphere and at room temperature was 6.6 kGy/s. The treatment was carried out using absorbed doses of 50, 100, 200, and 300 kGy. The SEM images revealed that with the increase in EBI dose, the cellulose fibres became more finely separated and long CNF having a diameter of around 160 nm were obtained in the sample treated with an EBI dose of 300 kGy.

Hybrid nanocellulose (Hy-NCs) comprising of both CNCs and CNFs can be obtained from cellulosic materials with the assistance of EBI treatment combined with other chemical and mechanical methods. Heo et al. (2021) demonstrated the production of hybrid Hy-NCs by using EBI treatment of wet hardwood pulp (WP) followed by alkaline treatment (pH 11 using 0.5 M NaOH solution), HPH (5 passes at 15,000 psi and room temperature) and CO_2 neutralization. The Hy-NCs produced using 500 kGy irradiated WP sample had a mean width of 30 nm, length of 927 ± 512 nm, and an overall yield of 77.7% that contained 68% CNFs and 32% of CNCs. On the other hand, when a 1000 kGy irradiated WP sample was used, the Hy-NCs had mean widths of 6 nm, length of 653 ± 382 nm, and an overall yield of 71.3% that contained 33% CNFs and 67% CNCs. However, when the 1000 kGy treated dry hardwood pulp was used, the overall yield of Hy-NCs reduced to 39.5%, which was primarily comprised of CNCs (98%). Meanwhile, an overall nanocellulose yield of 40.4% and 94.2% were achieved using sulfuric acid hydrolysis and TEMPO oxidation of non-irradiated WP samples, respectively. These results showed that EBI and HPH could generate a significant yield of Hy-NCs from wet biomass without using any chemicals for cellulose degradation and oxidation.

The employment of EBI in nanocellulose production has several beneficial aspects such as low environmental impact, lower energy consumption, shot treatment time, convenient operation (exposure to an electron beam), and requirement of mild conditions (ordinary pressure, room temperature, and atmosphere) (Lee et al., 2018). Besides, EBI treatment is effective in reducing the length and width of nanocellulose while improving the dispersion stability. However, many studies have indicated that using EBI treatment during nanocellulose production has several negative impacts, such as lower yield, crystallinity, and thermal stability. Besides, to the best of our knowledge, a very limited research has been conducted on using EBI treatment in nanocellulose production. Thus, prior to the commercialization of EBI technology in the industry, extensive research is needed using a range of different lignocellulosic materials to investigate and understand the effects of EBI on the characteristics of nanocellulose. In addition, the hybrid process can be developed and analyzed by using EBI in combination with other mechanical, chemical, and enzymatic methods for nanocellulose production.

5.5. Plasma treatment

Plasma technology is one of the emerging green technologies which has recently been used for the production of nanocellulose. Plasma, referred to as the fourth state of matter, is an ionized gas and can be described as a medium comprised of photons, metastable molecules, radicals, and charged particles such as free electrons and ions (Abidi & Hequet, 2004). For plasma generation, an electric field is applied to electrodes with gas in between them, either at atmospheric or reduced pressure. The material and geometry of electrodes, electric power, and the nature of gases influence the characteristics of the generated plasma. The gases usually utilized are CH_4, NH_3, N_2, O_2, Ar, and He (Desmet et al., 2009; Liyanage et al., 2021).

Solution plasma processing (SPP) or plasma in the liquid phase is regarded as an advanced oxidation method because of its potential to generate highly active species, particularly hydroxyl radicals (Surov et al., 2018; Zakharov et al., 2007). In a study, Surov et al. (2018) reported a novel oxidation–hydrolysis method involving atmospheric pressure glow discharge SPP system for production of CNCs from MCC and filter paper (FP). The first treatment approach (mode 1) involved the application of plasma in sulfuric acid solutions, whereas the second approach (mode 2) involved the application of plasma in distilled water followed by sulfuric acid hydrolysis at 50 °C for 2 h. The produced CNC from MCC and FP using SPP had a width of 30–40 nm and length of 200–500 nm along with high colloidal stability. Besides, when FP was used as the raw material, a CNC yield of approximately 30% was obtained in both processing modes 1 and 2. However, processing mode 2 almost doubles the yield of CNC (up to 56%) when MCC was used as raw material. In distilled water, SPP causes cellulose oxidation and the production of surface carboxyl groups. On the other hand, the oxidation process in sulfuric acid solutions is accompanied by hydrolysis. The oxidized cellulose is mostly transformed to water-soluble compounds as a result of subsequent hydrolysis, which promotes the release of CNC particles.

In another research, Vizireanu et al. (2018) prepared CNFs from commercially available microcrystalline cellulose (MCC) by combining the submerged liquid plasma treatment with ultrasonication (Fig. 4). In the open air (5000 sccm argon, 150 W), the plasma jet was ignited and immersed in 80 mL MCC suspension for 30 or 60 min. Also, in the main argon flow, additional reactive gases (nitrogen or oxygen) were introduced for some experiments. Ar plasma – ultrasound treatment resulted in many nanofibers having a width of 40–60 nm and a high aspect ratio of around 80. Also, nanofibers having a width of 50–60 nm deposited as a network between microfibers were noticed in the sample treated with Ar/N_2 plasma. Further, the Ar plasma – ultrasonication and Ar/N_2 plasma – ultrasonication treatment increased the CNF yield to 49% and 25%, respectively, which were significantly higher than the yield measured in the only ultrasonicated sample (15%). Thus, the plasma treatment combined with ultrasonication is an effective green technology for cellulose defibrillation.

In a recent study, Shaghaleh et al. (2021) produced carboxyl- and amino-functionalized cellulose nanofibrils from wheat straw using a two-phase air plasma/mild alkaline (TPAP/MA) pretreatment method followed by TEMPO oxidation and amidation. Wheat straw fibres (WSFs) were subjected to first-phase air plasma and subsequently NaOH treatment followed by H_2O_2 bleaching to recover cellulose fibres (CFs). The CFs were then subjected to second-phase air plasma activation. In both phases of plasma treatment, the discharge cycle was 50 V at 0.11 A of electric current and 20–25 kHz of frequency, and the air gas flow rate was 1 L/min, held for 30, 90, or 300 s. Cellulose yield of 73.4% and nanofibrillation yield of 97.6% was achieved when 90 s-TPAP/MA treatment conditions (NaOH treatment at 50 °C for 30 min and H_2O_2
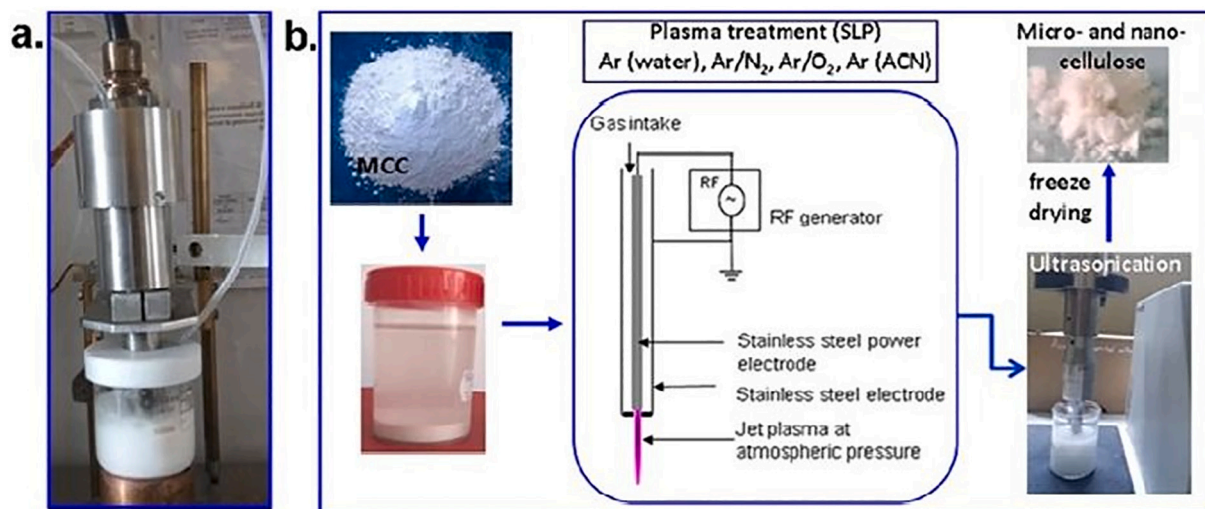
Fig. 4. (a) Image during the treatment with plasma torch immersed in the cellulose suspension; (b) Schematic representation of the experimental setup. Modified and reprinted with permission from Springer Nature (Vizireanu et al., 2018).

treatment at 25 °C for 10 min with 5 min of homogenization) were used. Moreover, the developed pretreatment process showed a 90.4% reduction in the total energy consumption compared to the baseline process.

The utilization of plasma technology for nanocellulose production offers several advantages. Since plasma is a clean, dry, and energy-efficient technology, it has lower acidification potential values, global warming potential values, and other life cycle analysis parameters than conventional production methods. However, the main hurdles for employing plasma on a big scale in the industry are the necessity of a continuous process as well as the huge surface area of the materials that need to be treated (Liyanage et al., 2021). Besides, to the best of our knowledge, the information regarding the utilization of plasma in nanocellulose production is very limited in the literature. Thus, the positive impact of plasma on fibrillation and characteristics of nanocellulose from a range of different raw materials is not fully understood to scale up the technology on an industrial scale. As a result, further research is needed to determine the suitability and effectiveness of adopting plasma technology to produce nanocellulose from various lignocellulosic feedstock. Moreover, the integration of plasma treatment sequentially with other technology such as microwave irradiation, HPH, or e-beam irradiation can be investigated for the production of nanocellulose. Besides, plasma-assisted production of nanocellulose using enzymes and emerging green solvents such as DESs can also be investigated as a novel eco-friendly process.

5.6. Pulsed electric field

The pulsed electric field is a non-thermal emerging technology that has been used for the pretreatment of lignocellulosic materials (Kumar et al., 2011). In PEF pretreatment, a simple device with two electrodes is used. The lignocellulosic feedstock is subjected to non-thermal voltage pulses for a very short period ranging from nano to milliseconds with 0.1–100 kV/cm of pulse amplitude. The PEF treatment leads to the disruption and structural changes in the biological membrane, which results in a loss of semi-permeability (Kumar et al., 2020; Hassan et al., 2018).

Recently, Suryanto et al. (2018) developed a PEF assisted alkali treatment method for the extraction of cellulose from Mendong (*Fimbristylis globulosa*) fibre. The PEF treatment was performed with an electric field of 1.3 kV·cm^{-1} for 30 s at a frequency of 20 kHz. As compared to the alkali (NaOH) extraction technique, the PEF assisted extraction process improved the crystallinity and crystalline index (from 83% to 86%) of the cellulose. With additional few seconds of PEF treatment, the extracted cellulose exhibited similar crystallinity as that of commercial cellulose. Besides, cellulose from the PEF treatment had a higher thermal decomposition temperature than the alkali-treated one. Moreover, the cellulose content increased from 72.1% (Mendong fibre) to 92.1% and 97.8% after using NaOH and PEF-assisted NaOH treatment, respectively. In addition, according to them, the amount of energy required for extracting cellulose from natural resources can be reduced by utilizing PEF treatment. Although they did not further prepare nanocellulose from the extracted cellulose fibres, from their study it can be interpreted that PEF could be utilized in the pretreatment of feedstock during nanocellulose production.

The PEF technique has several advantages, such as the treatment can be conducted at ambient, sub-ambient, or slightly above ambient temperature (Gómez et al., 2019). Besides, the pulse times are very short, so the energy consumption during the treatment is low. Therefore, hundreds to thousands of pulses can be applied over a relatively short period (Gómez et al., 2019; Kumar et al., 2011). In addition, the specific intensity of the treatment process can be adjusted depending on the conductivity of the material to be treated, the voltage delivered, and the distance and geometry of the working electrodes. However, various side effects such as the occurrence of electrochemical reactions or increase in temperature that depend on treatment conditions and processing parameters must be considered during PEF treatment (Gómez et al., 2019). Moreover, PEF treatment increases biomass porosity, which could be effective in improving the acid hydrolysis rate or enzymatic hydrolysis rate of biomass (Kumar et al., 2011). Thus, PEF may be utilized as process intensification technologies in enzymatic production of nanocellulose and combination with other conventional and emerging methods. However, further research is required to ascertain the effectiveness and suitability of using PEF combined with other chemical and mechanical techniques for nanocellulose production from lignocellulosic biomass.

6. Conclusion

In this article, a review of the most recent works on the utilization of emerging technologies in nanocellulose production is provided. Production of two types of nanocellulose (CNF and CNC) from various lignocellulosic feedstocks using emerging technologies have been reported. In most studies, two or more emerging technologies have been combined for enhancing the production efficiency and properties of nanocellulose. Besides, a very limited number of works have been reported on the application of hydrated deep eutectic solvent,

immobilized enzymes, plasma technology, electron beam irradiation, and pulsed electric field in nanocellulose production. Thus, the authors believe that using these technologies in developing sustainable, cost-effective, and eco-friendly methodologies is the future trend in nanocellulose production.

Declaration of competing interest

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

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