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Review Article

Recent advances in nanocellulose-based different biomaterials: types, properties, and emerging applications



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ABSTRACT

The research interest in sustainable and eco-friendly materials based on natural sources has increased dramatically due to their recyclability, biodegradability, compatibility, and nontoxic behavior. Nanocellulose contains chains of glucose residue and is an abundantly available green material. Recently, nanocellulose-based green composites are under extensive exploration and gained popularity among researchers owing to their lightweight, lost cost, low density, excellent mechanical and physical characteristics. These materials have also shown tremendous potential for applications in biomedical and numerous engineering fields. The mechanical properties of these materials play a vital role in effective utilization and their exploration for future applications. This review article comprehensively presents current developments, results, and findings in the arena of green and sustainable materials. Currently, the main problem is the large variability in their properties and qualities. Nanocellulose properties are influenced by various factors, including the fiber type, ecological conditions, manufacturing methods, and any alteration of the fiber surface. Finally, the review incorporates future challenges and opportunities in the field of nano cellulosic materials.

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1. Introduction

Cellulose, a ubiquitous and natural bio-based polymer material on the earth, is produced from plants, algae, tunicates, and

bacteria [1,2]. The total biomass production of the cellulose is 1.5×10^{12} tons/year that can be used as a raw material to form biocompatible and eco-friendly products. It is extracted from wool, hemp, flax, cotton, or sisal plants and can be used as a substitute for various polymers owing to lightweight,

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Nomenclature

ACC	All-Cellulose Composites
ACNF	Acetylated Cellulose Nanofiber
BC	Bamboo Cellulose
BNC	Bacterial Nanocellulose
CDA	Cellulose Diacetate
CLNP	Corn cob Lignin Nanoparticle
CNF	Cellulose Nanofiber
CNT	Carbon Nanotube
DGEBA	Diglyceryl Ether of Bisphenol
HDPE	High Density Polyethylene
GO	Graphene Oxide
L-CNC	Lignin-Coated Cellulose Nanocrystals
LED	Light Emitting Diode
MAPP	Maleic anhydride grafted polypropylene
MC	Methyl Cellulose
MCC	Microcrystalline Cellulose
MFC	Micro fibrillated Cellulose
MWCNT	Multiwalled Carbon Nanotubes
NF	Natural Fibers
NFC	Nano fibrillated Cellulose
NFRPC	Natural fiber reinforced polymer composites
OPEFB	Oil Palm Empty Fruit Bunch
PEI	Polyethyleneimine
PLA	Poly lactic Acid
PPG	Polypropylene Glycol
PVA	Polyvinyl Alcohol
PCB	Polysaccharide circuit board
PEVA	Poly ethylene-co-vinyl acetate
PBAT	Poly butylene adipate-co-terephthalate)
RGO	Reduced Graphene Oxide
rPP	recycled Polypropylene
SCB	Sugarcane Bagasse
SPCE	Screen-printed carbon electrodes
UV	Ultraviolet

renewability, natural abundance, biocompatibility, biodegradability, versatility, and excellent adaptability [3–5]. Cellulose and its derivatives have been applied for more than 150 years. It is estimated that the global market share for cellulose fibers will expand rapidly and reach 41.5 billion USD by 2025 [6]. They have recently been used as sustainable materials in various emerging fields like biomedicine, drug delivery, automotive, electronics, and structural engineering applications [7–9] owing to their non-toxicity and ecofriendly nature. Their usage can also be found in energy devices due to their appealing properties [10–12]. They also exist in the composite form containing lignin, polymers, and hemicelluloses. Lignin act as an adhesive media, holding hemicellulose and cellulose [13]. Cellulose is prominent and the principal part of the plant cell divider, allowing the entry of supplements and keeps up the cell shape with significant tensile strength [14,15]. Whereas hemicellulose, a secondary component, acts as a joining media between cellulose and lignin. Furthermore, pectin is also present in plant structures. However, it appears with a minor quantity than the other three components. The purification of cellulose is essential,

which will help to eliminate the hemicellulose, lignin, and other small impurities.

Cellulose in composite materials has also become an epicenter of attention and applied in natural fibers (NFs) [16,17]. Its utilization in polymer composites is due to excellent biodegradability, combustibility, non-abrasiveness, low cost, and low density [18]. However, cellulose-based green composites did not achieve industrial realization due to attached anomalies such as poor metallurgical adhesion and water absorption capability. Chemical treatments of the cellulose fibers are the remedies to these problems [19]. Nowadays, scientists are focusing on the extraction of nanocellulose-based materials from cellulose to manufacture nanocellulose-based reinforced green composites for diverse applications.

This study presents a brief knowledge and groundbreaking understanding of cellulose, its primary sources, and the classification of nano cellulosic materials for researchers in this field to unlock advanced applications. Different manufacturing processes and chemical modification techniques of these nanocellulose materials are also illustrated. Recent works (2015–21) about nanocellulose-based composites, their mechanical properties, and their applications are reviewed. The final section provides an insightful overview of the potential challenges and future opportunities in the arena of cellulose-based green composites.

2. Cellulose-based natural fibers reinforced composites

Appropriate exploration of easily accessible natural sources becomes an essential factor in establishing a sustainable industry [20–22]. Natural fiber-reinforced composites (NFRCs) fabricated with cellulosic materials have emerged as green and eco-friendly materials [23–26]. NFRCs are easy to manufacture and have minimal effect on the ecosystem compared to conventional composite materials [27–30]. NFRCs are sometimes referred to as “green composites”, which are fabricated by adding various types of NFs into biodegradable resins and starch as polymer matrix to improve the mechanical characteristics of the green composites [31,32]. The fabrication of these fibers is similar to the synthetic fibers and uses hand lay-up [33], extrusion [34], resin transfer molding [35], injection molding techniques [36], and compression molding [37]. Furthermore, these composites decrease the overall cost of materials and provide lightweight properties owing to the low density of NFs [38–40]. Classification of cellulose-based materials is presented in Fig. 1. These NFs can be classified based on their source and percentage of hemicellulose and lignin [41,42]. Fig. 2 illustrates the general attributes of cellulose-based materials.

NFRCs can be produced from sustainable cellulose materials that provide environmental and cost-related benefits over conventional composites. However, several critical drawbacks, like higher moisture absorption, inferior mechanical properties, incompatibility with the matrix, poor wetting characteristics, and low thermal resistance [43,44] are associated with these materials that influence their mechanical properties. These drawbacks lead to the failure of the

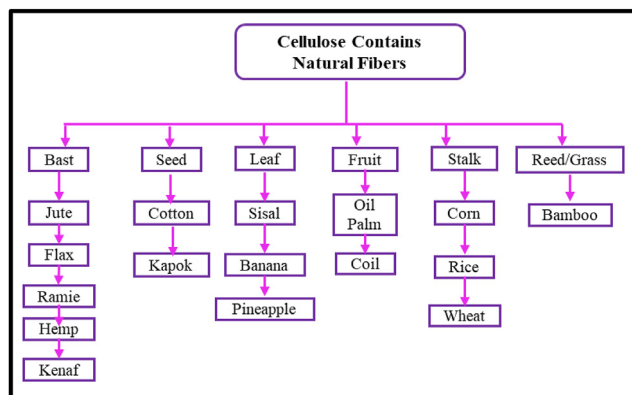


Fig. 1 – Different types of reinforcing bio fibers containing cellulose.

green composites [45–47]. Furthermore, it is challenging to fabricate composites due to the inconsistency of the NFs that also limits their industrial utilization. The characteristics of the fibers depend upon the harvesting season that ultimately affects the properties of the composites. Fig. 3 presents the changing percentage of cellulose contents in different NFs. Commercial production of cellulose heavily relies on the harvested source like wood or highly pure origin like cotton. Fig. 4 shows the interest shown by several countries in nanocellulosic materials over the past ten years [48,49].

3. Nanocellulose

Nanocellulose is an excellent material that occurs in the form of nano-scaled cellulose fibrils and nanocrystals separated from the cotton linters, plant cell walls, or bacteria through

mechanical, enzymatic, or chemical techniques [50,51]. Nanocellulose is extracted from natural cellulose fibers and contains few micrometer lengths and <100 nm diameter. It is mainly classified into three types of nanomaterials; Cellulose nanocrystal (CNC) [52], Nano fibrillated Cellulose (NFC), and Bacterial nanocellulose (BNC) [53].

A brief comparison between the properties of the different types of nanocellulose is provided in Fig. 5. The exceptional properties of nanocellulose include high modulus of elasticity (110–220 GPa), tensile strength (7.5–7.7 GPa), tailored aspect ratios, large specific surface area, easy surface functionalization, tunable crystallinity, high degree of polymerization, and high chemical resistance [54–56]. Due to these unique physicochemical properties, nanocellulosic sustainable materials are gaining importance in aerospace, automobile, packaging industries, energy devices, and other interdisciplinary fields [57]. Furthermore, synthetic materials drastically impact the climate and the consumption of fossil fuels is crucial to consider. Therefore, it is essential to shift towards these sustainable nanomaterials [28,58,59] in upcoming years.

Recent developments in engineering, biomaterials, and other high-end applications demonstrate the importance and capability of different celluloses, especially nanocellulose [60]. These materials have been frequently used in water purification [61], medical services, and veterinary medication [58,62,63]. The dimensions of nanocellulose extracted from different plants and NFs are important because the effectiveness of nanocellulose-based materials depends on the achieved accuracy in nanocellulose dimensions. The dimensions of the isolated nano cellulosic materials obtained from some recent works are summarized in Table 1.

3.1. Processing and synthesis

The primary route employed for the synthesis of nanocellulose is acid hydrolysis, including HCl and H₂SO₄ acids. Electrostatically stable CNC is produced through acid hydrolysis as a result of sulfuric sulfate half-esters. Sulfate half-esters are formed when the hydroxyl group of cellulose reacts with sulphuric acid. BNC is biosynthesized nanocellulose with an outstanding level of crystallinity [81]. The different processes adopted in the manufacturing of nanocellulose are presented in Fig. 6. After nanocellulose production, various surface treatments [82] are also adopted for improving their physical and mechanical properties. A generic flow chart of obtaining nanocellulose through different procedures is presented in Fig. 7. Among all the surface treatments, the most widely adopted technique is NaOH/urea-based. The various processes involved in NaOH/urea-based treatment are elaborated in Fig. 8.

3.2. Chemical treatments

The common issue in cellulose-based composites is the hydrophilic nature of the fibers and the hydrophobic nature of the matrix [83,84]. The intrinsic mismatch between these two natures results in poor adhesion at the interface. Different chemical treatments on NFs can decrease their hydrophilic inclination and improve their compatibility with the matrix

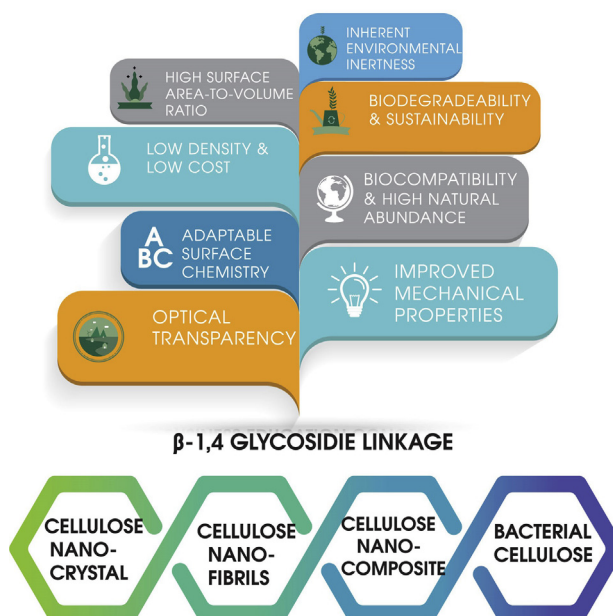


Fig. 2 – General attributes of cellulose-based materials.

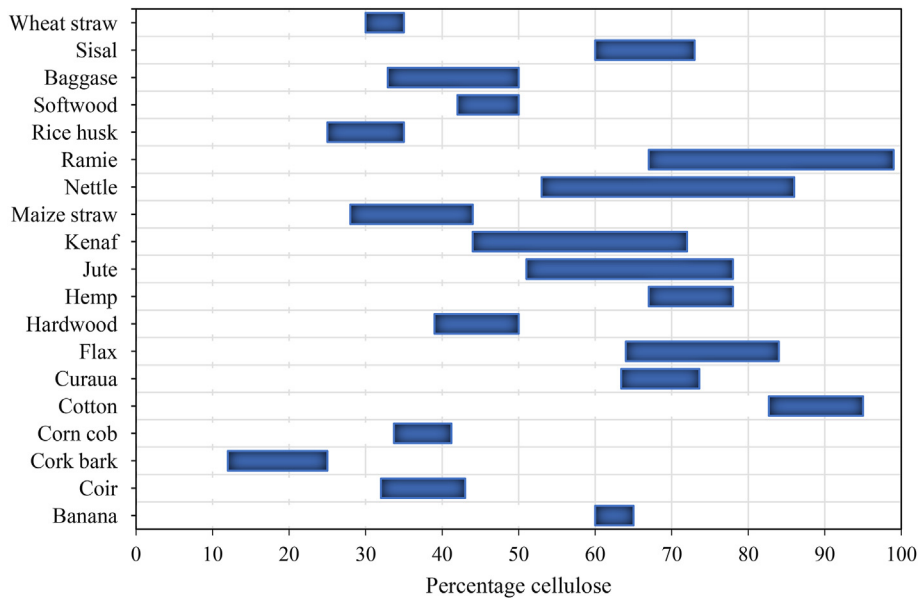


Fig. 3 – Percentage of cellulose in different natural fibers.

[85–88]. The most popular treatments on the fiber that have proved their significance in improving different cellulose-based composite properties [89] are; Alkali Treatment [90], Silane Treatment [91], Fungal Treatment, Maleated Couplings Agents, Benzoylation Treatment [92], Peroxide Treatment [93], and Permanganate Treatment [94]. One example related to these chemical treatments on NFCs is elaborated here. Liu et al. [95] studied the influence of the silane treatment on the mechanical properties of corn-stalk fiber-reinforced composites. As shown in Fig. 9, silane treatments produce a rough surface on the corn-stalk fiber surface and thus improves different mechanical properties due to the strong adhesion of fibers with matrix/epoxy.

Mostly different chemical modifications utilized on NFs improve the hydrophobicity [96–98]. The reactions in treatments involve swelling and non-swelling solvents. The resulting reactions from these treatments attach with the hydroxyl group of cellulose [97,99,100] and improve their

wettability, tune their microstructure and surface morphology. These changes have a positive effect on the connection between fibers and polymer [4,14,15,101]. Fig. 10 shows the cause-and-effect diagram of chemical treatments on different NFCs.

3.3. Nano fibrillated cellulose (NFC)

Numerous researchers [102–105] concentrated on various mechanical instruments, like ultrasonication, steam blast, and microfluidizers for obtaining cellulose [106–108]. Clogging is frequently seen with homogenizers, but it can be resolved by utilizing extruders and high consistency in milling processing, resulting in fiber delamination. Major drawbacks of this technique are high energy requirement and the presence of bigger fragments. The presence of delamination instruments relies upon the final utilization of NFCs [109].

The freeze-drying process is also used for producing permeable cryogels. The primary advantage associated with this technique is low water absorption. On the other hand, the principal disadvantage of freeze-drying is ice crystals during the pre-freezing step. The produced crystal extends and affects the structural qualities of cryogel. This can be reduced using sucrose, or fast freezing methods can also be adopted [110]. Apart from these processing techniques, some surface modification techniques were also applied to the NFCs. The advantages and drawbacks of various surface modification treatments adopted on NFCs are reported in Table 2.

3.4. Cellulose nanocrystals (CNC)

CNCs possess broader scope of properties, such as enormous surface zone, phenomenal strength, intriguing mechanical properties, and remarkable optical properties. Nano whiskers are sometimes also referred to as CNCs. It possesses higher flexibility than NFC and usually contains 54–88% of crystallinity.

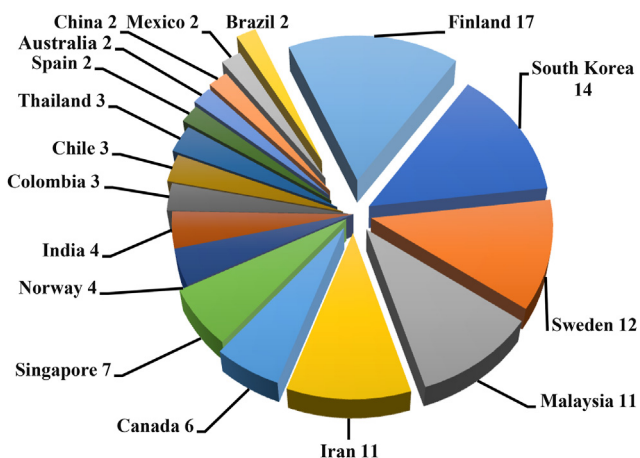


Fig. 4 – Country-wise interest in nanocellulose.

Properties	NFC	CNC	BNC
Dimensions & Aspect Ratio	dia - (1-10 μm) ; very high A.R	dia - (5-30 nm) ; Low A.R	dia - (20-100 nm) ; Low A.R
Chemical Requirements	Corrosive reagents	Corrosive reagents	Non- corrosive chemicals
Sustainability	Not very sustainable	Not very sustainable	Green Approach
Cost	Low cost	Low cost	High Cost
Energy	High Energy process	High Energy process	Green Process

Fig. 5 – Comparison of different properties against each type of nanocellulose.

Synthesis of CNC also involves acid hydrolysis technique. CNCs on reaction with naturally available or manufactured polymers yield valuable composites. The physical and mechanical properties of CNCs are improved through different surface modification procedures for their use in composites. Furthermore, these treatments also improve properties like low thickness and high surface zone [115].

3.5. Bacterial nanocellulose (BNC)

BNCs are produced by utilizing different microorganisms [116]. These microorganisms exhibit great significance because of their fast growth and availability around the year. Generally, two techniques, the static and stirred culture methods, are adopted to produce BNCs. In static culture, BNCs are formed due to the growth of a thick, cowhide-like white BC

pellicle at the air-fluid interface [117]. The prime factors for selecting these methods are the final applications of BNC, surface morphology, physical, and mechanical properties of the produced BNC [118]. A summary of some recent studies dedicated to produce different BNCs through different microorganisms is discussed in Table 3.

4. Mechanical properties

Nanocellulose-based materials proved to be excellent reinforcing media for improving the mechanical properties of different materials, especially composite materials [127–129]. These materials are most suited in replacements of different synthetic fiber-reinforced composites to fabricate eco-friendly green products [130]. The replacement of synthetic fibers with

Table 1 – Dimensions of nanocellulose extracted from different sources.

Natural Fibers/Plants	Type of Nanocellulose	Dimensions of extracted nanocellulose		Ref.
		Length	Diameter (nm)	
Eucalyptus pulp	CNC	150 nm	10–50	[64]
Pinecone	CNF	–	5–20	[65]
Soybean straw	CNF, CNC	>1 μm	10	[66]
Hyacinth Fiber (<i>Eichhornia crassipes</i>)	CNC	147.4 nm	15.6	[67]
Pineapple Leaf Fiber	NC	<(88–1100) nm	68	[68]
Rice hull	CNF	–	<100	[69]
Banana rachis, kapok, pineapple leaf, coir	CNF	–	10–25	[70]
Oil Palm trunk	CNF	170–800 nm	4–10	[71]
Bamboo	CNC	–	50–100	[72]
Waste paper	CNC	100–300	3–10	[73]
Banana Peels	CNF	455 nm	10.9	[74]
Palm Residue	Coir NC	–	108	[50]
	Fronds NC	–	90	
Coir Fiber	CNF	–	37.8	[75]
Jute Fiber	CNF	–	50	[76]
Beer industrial residues	NC	–	73–145	[77]
Coconut residues	CNF	–	70–120	[78]
Silkworm silk fibers	CNF	–	0.1–0.4	[79]
Flax	CNC	57 nm	6	[80]
Cotton	CNC	68 nm	8	

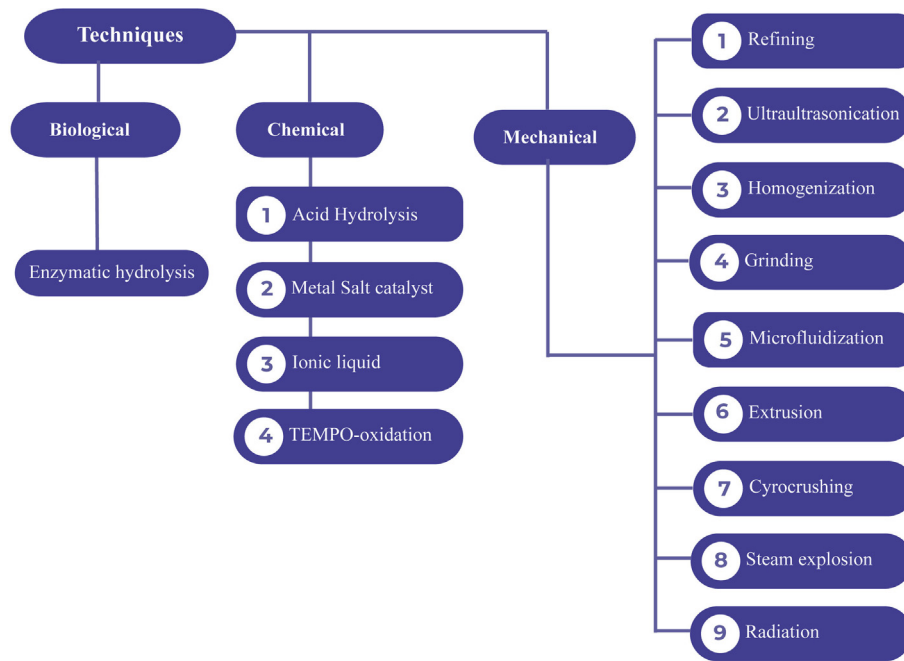


Fig. 6 – The different processes adopted in the manufacturing of nanocellulose.

these nanocomposites in the industries is due to the low density and cost, nobler mechanical properties and thermal insulation, less environmental impacts combined with sustainability trends. Furthermore, the mechanical and physical characteristics of the nanocellulose-reinforced composites are comparable with the properties of the synthetic fibers. Traditionally glass, carbon, and aramid fibers are utilized in fiber-reinforced composites. For example, glass fiber possesses elastic modulus between 70 and 110 GPa and a density of 2.5 g/cm³, and aramid fiber possesses modulus between 63 and 67 GPa with a density of 1.4 g/cm³ [28]. Therefore, when materials are required to be chosen based on the modulus of elasticity and weight, nanocellulose is an excellent choice for this scenario having roughly 65 J/g for NFC and 85 J/g for CNC. In comparison, traditional materials like steel provide only 25 J/g. Nowadays, these materials have also been utilizing in many structural applications as a substitute for steel owing to their excellent mechanical properties [131].

Nanocellulose-based composites exhibit excellent mechanical properties due to these nanofillers' stiffening influence, especially at a lower concentration. It results in a large specific surface area that provides excellent interaction between the polymer matrix and nanocellulose. The distribution and dispersion of the nanomaterials in the hydrophobic matrix must be uniform to ensure significant force transfer in the nanocomposites. Furthermore, the homogeneous dispersion and metallurgical interaction between the polymer matrix and nanocellulose produce nanocellulose composites of the properties that lie between the properties of polymer matrix and cellulose. Ghasemi et al. [132] developed CNF-based PLA composites, and microscopic analysis depicted excellent dispersion of the CNF in the polymer

matrix. As a result, significant mechanical and thermal properties were achieved. Therefore, an optimum concentration of nanocellulose promotes dispersion and distribution in the polymer matrix, providing better mechanical interlocking and interfacial strength. In another study, Carrillo et al. [133] obtained dispersion in the non-polar matrix using the proposed double emulsion technique. The authors balanced the nanocellulose with the organic polymer matrix in a double emulsion (water-in-oil-in-water) solution. It avoided the drying of the dispersion of nanocellulose before the processing. In this way, the microstructure and mechanical properties of the nanocomposites can be tuned by managing the ionic strength of nanocellulose in the dispersion.

Recently, the attention of the manufacturers and consumers has been diverted to develop green composite by employing cellulosic materials as reinforcing agents [10]. Numerous researchers reported that the addition of NFs, NFC, and CNC into the polymer matrix significantly improved the mechanical properties of the composites. To replace the glass fiber sheet of the vehicles, Wu et al. [134] fabricated kenaf fiber-reinforced composites using the vacuum bag resin transfer molding technique. The results revealed that these composites exhibited excellent mechanical characteristics and minimized energy consumption, contributing to promoting a sustainable environment. Morelli et al. [135] found that the CNC incorporation increased tensile strength and elastic modulus of the polybutylene adipate-co-terephthalate (PBAT). In another study, Tan et al. [136] investigated the influence of the addition of CNC in the polyethylene-co-vinyl acetate polymer matrix (PEVA), and the results indicated significantly improved mechanical properties of the nanocomposites. Likewise, Spinella et al. [137] tried

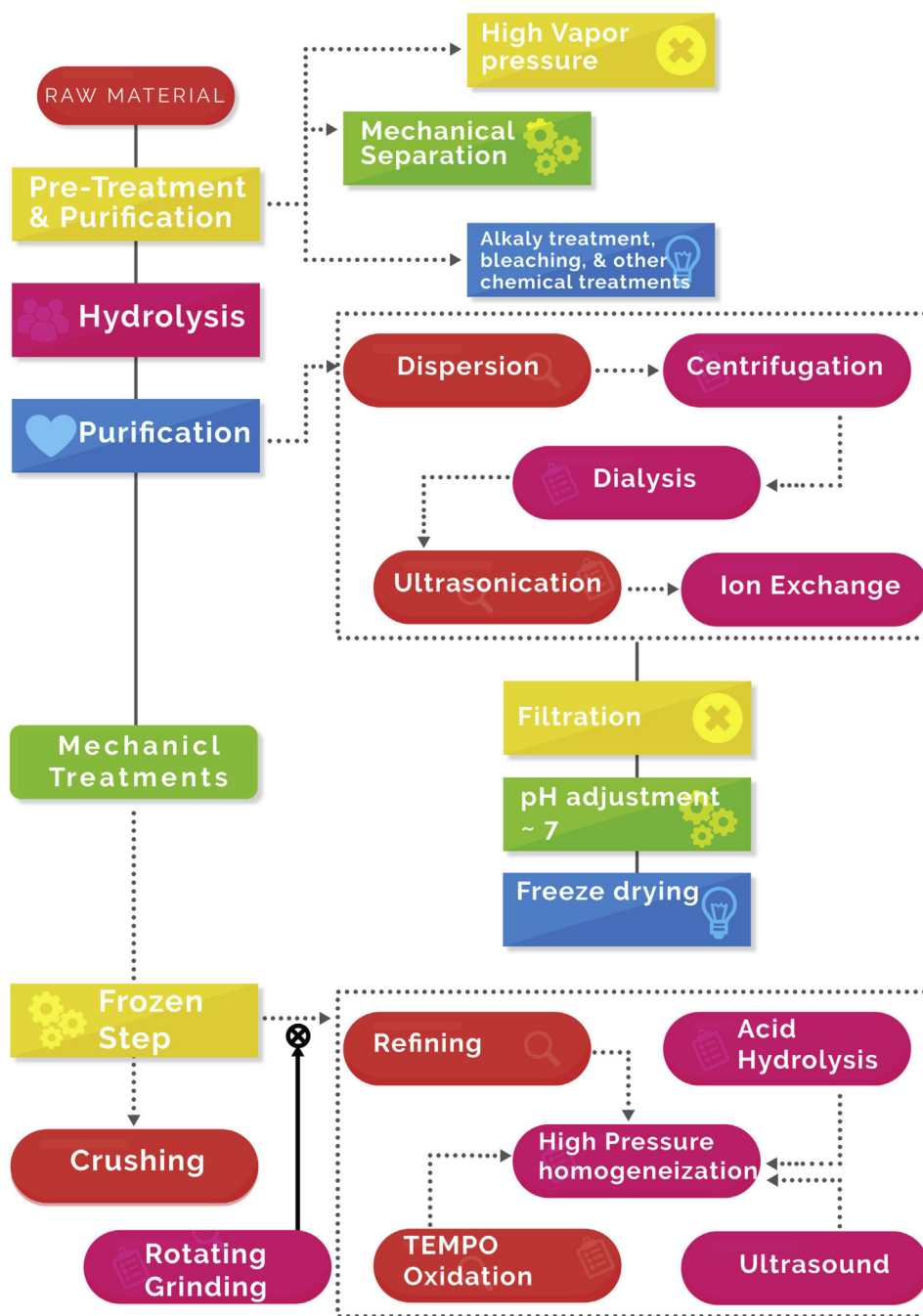


Fig. 7 – Generic flow charts of obtaining nanocellulose through different steps.

to improve the mechanical characteristics of the PLA by adding CNC and found that both loss modulus and storage modulus of the nanocellulose-based composites were enhanced in comparison to pure polymer. The method of obtaining nobler mechanical properties with the incorporation of nano cellulosic materials depicted that these nanocomposites can be employed as potential replacements of synthetic fiber-reinforced composites in the engineering and material science fields.

To summarize, the type of fibers, dimensions, microfibril orientation, the volume contents of the nanocellulose materials in the fibers, chemistry with matrix, surface, and chemical treatments are critical parameters that influence the overall mechanical properties of nanocellulose materials [138–140]. Some recently reported works on the mechanical characterization of different nano cellulosic composite materials treated with different chemicals are also summarized in Table 4.



Fig. 8 – Different processes involved in NaOH/urea-based treatment.

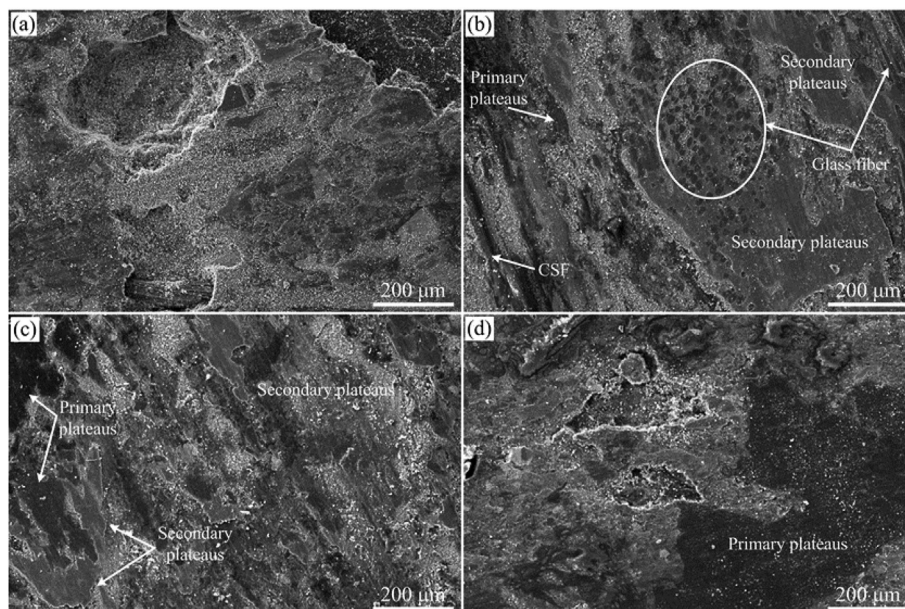


Fig. 9 – Micrographs of the corn stalk fibers composite with different percentage of silane treatments (a) 1% (b) 5% (c) 9% (d) 13% [95] (Images reproduced with permission from Elsevier).

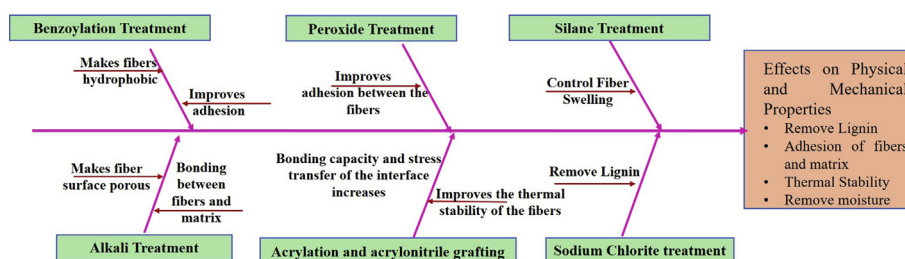


Fig. 10 – Cause and effect diagram of different chemical treatment procedures on NFs.

Table 2 – Advantages and drawbacks of some treatments used on NFC.

Surface Treatment	Improvements	Drawbacks	Ref.
Polymer Grafting	Inherit the NFC along with some mechanical properties	The organic solvents use in this method and have negative effects on the environment.	[48]
Plasma Discharge	Enhance the carboxyl and carbonyl groups, and the quantity of oxygen increases at nanocellulose surface.	Expensive experimental setup is required (like arc discharge) and parameters like high frequency and low power are difficult to achieve.	[111]
Physical adsorption	Improves the bonding process between nanocellulose and polymers.	Large water is consumed, and wastewater contains many hazardous materials.	[112]
Grafting (UV based)	Environment-friendly technique as no organic solvent is consumed.	Highly labor skilled is required. Color fading and chromatic homogeneity are largely influenced.	[113]
Silane based Treatment	Improves the hydrophilic nature of fibers.	Sometimes different mechanical properties like elongation are not improved through this technique.	[114]

Table 3 – Recent work on BNC production through different microorganism.

Methods	Microorganism	Chemical Process Adopted	Time	Temperature	Final BNC amount (g/L)	Ref.
Static Culture	<i>G.xylinus</i> CH001	–	5 days	28 °C	0.66	[119]
Static Culture	<i>Komagataeibacter saccharivorans</i>	d-H ₂ O & NaOH	7 days	28 °C	3.9	[120]
Static Culture	<i>K. medellinensis</i>	Citric Acid	12 days	–	5	[121]
Static Culture	Yeast (SCOBY)	NaOH treated	20 days	30 °C	29.2	[122]
Static Culture	<i>K. medellinensis</i>	Cross couplings Reactions	8 days	28 °C	6	[123]
Static Culture	<i>K. xylinus</i> BPR 2001	–	9 days	30 °C	6.4	[124]
Static Culture	yeasts (SCOBY)	NaOH treated	20 days	30 °C	60	[125]
Stirred Culture	<i>G. hansenii</i> CGMCC 3917	–	–	30 °C	7.02	[126]

5. Applications of nanocellulose

The nanocellulosic materials are used as packages on different food products due to their bio-friendly properties [164,165]. For example, low absorbency of cellulose and the ability to produce a thick percolating network of hydrogen bonds are attractive characteristics for the filtration process [166]. Two important areas are primarily focused on this particular application. One is oxygen (gas) permeability [167,168], and the second is water transmission in vapor form [169]. The low porosity and surface harshness of NFCs make them suitable materials for the printing of conductive tracks.

5.1. Flexible electronics

In this modern world, engineering and extensively used different equipments are going through a widespread change

from rigid counterparts to flexible electronics, owing to their numerous benefits over rigid electronics, for example, mechanical flexibility, durability, and lightweight components [170–172]. A lot of research has been conducted in the past that demonstrates flexible electronics as a high-performance competitive device as compared to rigid electronics (Fig. 11). The equipments with mechanical flexibility, lower creation cost, and feasible manufacturing ultimately determine their efficiency and durability [174–176]. Nanocellulose-based films [177] are highly emerging because of their several merits, making the current electronics move forward towards high performance and multi-functional applications. Moreover, the features like low cost, portability, biodegradability, and disposability are attracting other applications such as biomedical, aeronautical, and energy conversion devices [169,178]. Nanocellulose-based materials are applied to develop the devices such as solar cells, transistors, touchscreens, supercapacitor applications [179].

Table 4 – Some recent work (2015–20) on the Cellulose-based natural fibers/nanocellulose reinforced composites.

Cellulose containing natural fibers reinforced composites	Matrix Source	Adopted Chemical Treatments	Mechanical characteristics			Important Finding	Ref.
			Tensile Strength (MPa)	Flexural Strength (MPa)	Impact Strength		
MFC	PPG	Silane	42.7	—	—	The tensile strength is increased up to 70%.	[141]
Wood fiber	PLA	—	65	93	24 J/m	Multi-functional bio-additive improved the mechanical properties.	[142]
Cotton linters	—	NaOH/urea (Solvent)	122	—	—	The addition of embedded CNTs generates high electrical conductivity.	[143]
Wood pulp-based ACC	—	NaOH/urea (Solvent)	45.9	—	—	Tensile strength improved more than ten times with a short solvent treatment.	[144]
CNF	Epoxy/DGEBA	—	The addition of CNFs improves thermal as well as storage and loss modulus of these nanocomposites drastically.				[145]
Salix psammop il powder	—	—	The tensile strength improved to 304.6 MPa with 10 wt.% MWCNTs, which was nearly 106.8% higher than that of MWCNT/cellulose fibers with a similar quantity of MWCNTs.				[146]
Pulp softwood craft fibers	—	NaOH solution	The produced ACC cryogels and aerogels shows improved compressive modulus and absorbed energy up to 40%. Strain increased.				[147]
Cellulose nanofibrils	—	—	The cellulose nanofibrils lessens the syneresis of the alginate gels and enhances its compression against deformations.				[148]
MCC	rPP	MAPP	28	—	7.5 kJ/m ²	Mechanical properties were improved with MAPP.	[36]
MCC	—	Citric acid	2.25	7.8	—	The maximum mechanical properties were attained at 5% citric acid concentration.	[149]
MC	—	PEI	10	A good match between theoretical and experimental results shows uniform dispersion of PEI-RGO in MC.			[150]
MFC	—	NaOH	The uniform distribution of NFC improves its mechanical properties.				[151]
SCB	HDPE	Zirconium oxide	15.7	18.6	38.9 kJ/m ²	Influential effects of mentioned treatment on SCB mechanical properties.	[152]
Cotton linters	—	NaOH/urea	At 5% wt. these redacted GO composite behaves as multi-functional sensor material.				[153]
L-CNCs	PLA	—	A uniform dispersion of L-CNCs and an equal crystallinity of PLA improves thermo-mechanical properties.				[154]
CNF	—	Acetylation	14.7	The ACNFs lowers the degradation rate of CNF as a result of fungal degradation.			[155]
BC, CNF	PVA	Acetylation	43	These CNFs exhibit good visible light transmittance along with improved mechanical properties.			[156]
MCC	PVA	NaOH/Urea	The PVA based MCCs has improved fracture toughness of 44.3 MJ/m ³ .				[157]
MCC	Epoxy	Alkali	50	70	12 kJ/m ²	The alkali treatment increases the mechanical properties of MCC.	[158]
CLNP	—	—	The manufactured composites films possess a high UV resistance at 5% wt. of CLNP.				[159]
SCB, CNC	k-carrageenan	Alkali	85	The inclusion of CNC reduced the light transmittance capacity but improved the mechanical properties.			[160]
CNF	—	—	72.3	Addition of graphene in CNF matrix improves its tensile strength while maintaining good flexibility.			[161]
Jute, kenaf, hemp fibers	Epoxy Amite	—	The hybridization of NFs improves the mechanical properties.				[162]
CDA	—	Alkaline	45	The ACC films produced from cellulose II as a result of alkali treatment on CDA and NFCs conversion.			[163]

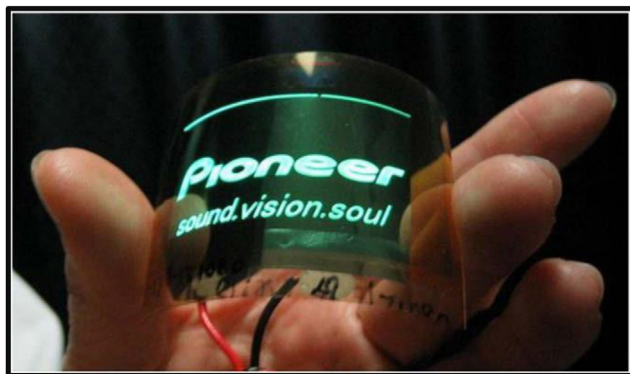


Fig. 11 – Flexible display on CNF substrate [173] (Adopted with permission from Elsevier).

5.2. Skin electronics

One of the essential applications of nanocellulose-based materials is skin electronics that exhibits flexible, and self-healing characteristics, and can be biocompatible with human skin tissues. It has enormous applications in the biomedical sector, such as medical diagnostics and biomimetic prosthetics [180]. Daniele et al. [181] proposed a conformal electronic decal that was based on a polysaccharide circuit board (PCB) incorporated with thin nanocellulose-based film and water-soluble pullulan layer that showed not only the good mechanical and thermal properties but also exhibited high water vapor transmission rates, which is a remarkable property for skin electronics. Nanocellulose-based films [182] have also revealed their applications in diagnosing neurological disorders because of their excellent biocompatibility, wettability, and good surface roughness at the nanoscale [122,178].

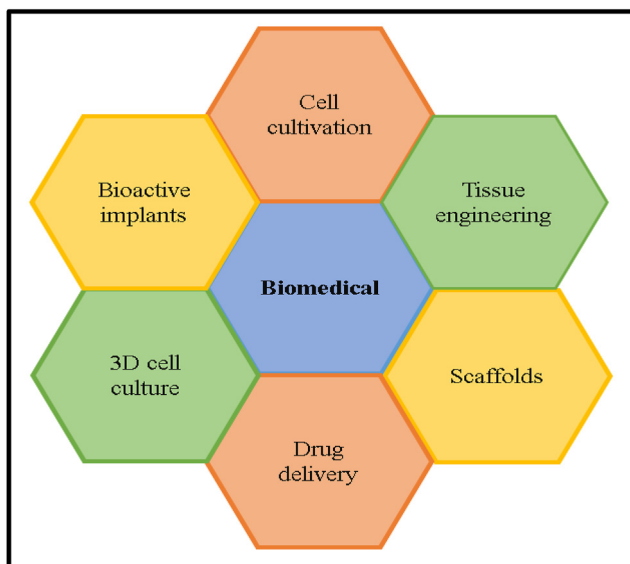


Fig. 12 – Pictorial representation of Biomedical Applications of Nanocellulose.

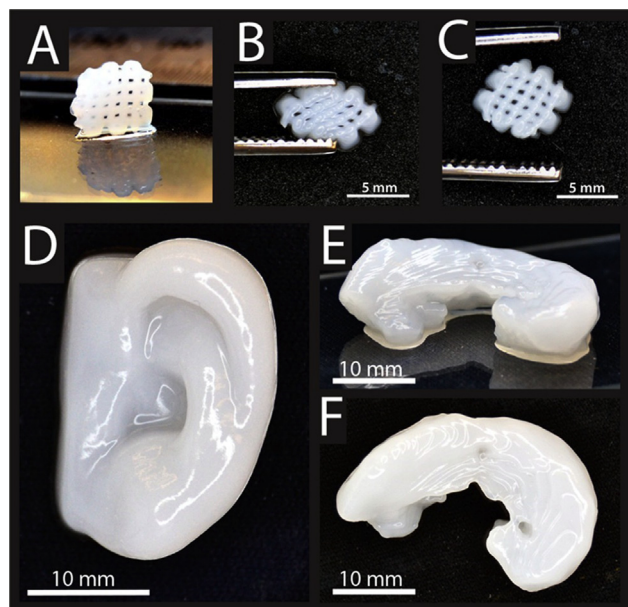


Fig. 13 – Different 3D printed nanocellulose–alginate scaffolds. “(Reprinted with permission from [208]. Copyright 2021 American Chemical Society)”.

5.3. Photodetectors

Photodetectors are the most used elements for sensing and monitoring the different motions and environmental changes. However, it remained challenging to produce a photodetector of non-toxic, transparent, and biodegradability features [183]. Park et al. [184] proposed pure nanocellulose-based transparent, non-toxic, and high-performance films, which displayed good electric properties, including the saturation mobility of $1.4 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. It also exhibited excellent electrical features under the mechanical bending test. In this way, nanocellulose-based films can be applied as substrates for environment-friendly sensor systems.

5.4. Water treatment

Water pollution is causing severe threats to human lives and other living organisms because of the rapidly growing population, industrial and agricultural activities. Several engineering tools/equipments have been introduced to purify this polluted water, including reverse osmosis (RO) membranes, solar evaporation, and filtration through the membrane [185]. From all of these, one of the most used filtration techniques is membrane filtration which is used to remove the pollutants from the contaminated water owing to its low environmental concerns, high efficiency, chemical stability, and durability [186]. Furthermore, nanocellulose-based films are the most suitable and feasible source of purification due to their inherent properties such as sustainability, less costly, wider availability, hydrophilicity, eco-friendly, and can be redeveloped [187]. Moreover, these nanocellulose-based membranes are attractive materials for water treatment because of their capacity to modify pore

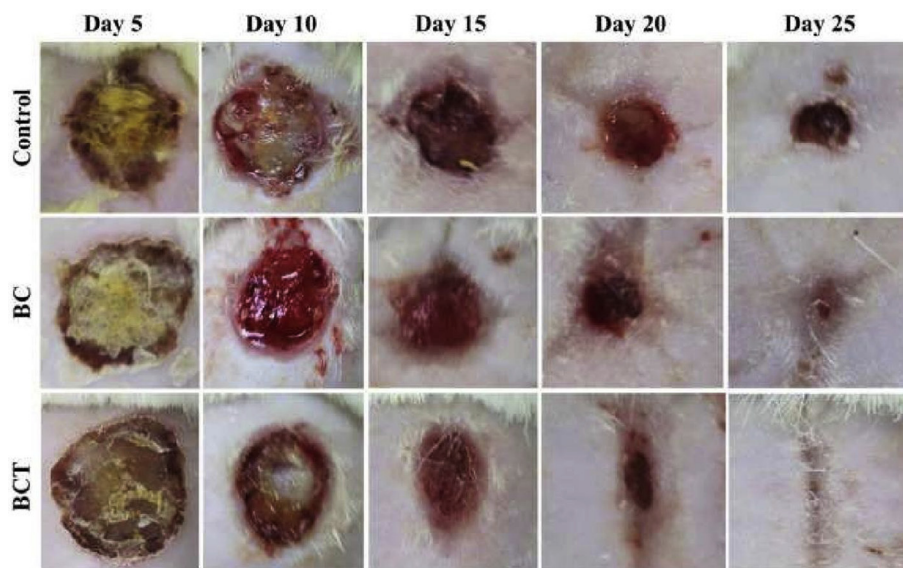


Fig. 14 – Effect of BNC and BCT hydrogels on wound healing on different day 5, 10, 15, 20, and 25 [209]. (Adopted with permission from Elsevier).

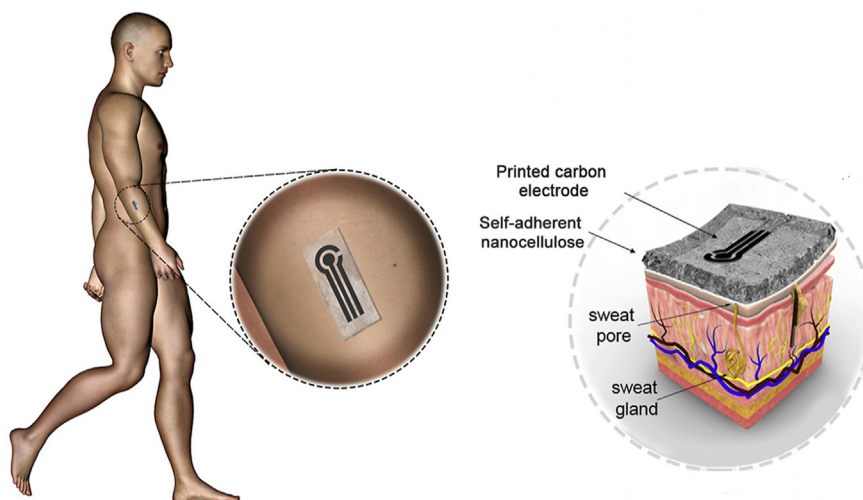


Fig. 15 – Pictorial depiction and schematic layered diagram of SPCEs on BNC substrate for electrochemical biosensing application [210]. (Adopted with permission from Elsevier).

size, mechanical properties, water flux, adsorption capacity, and operating pressure [188,189].

Different types of nanocellulose-based membranes are available with distinct pore sizes. Here, only two types of membranes for water treatment are discussed: pure nanocellulose made membranes and composite membranes containing a significant portion as nanocellulose and minor portion of other relevant materials. Mihranyan et al. [190] developed a membrane using CNFs extracted from *Cladophora* algae to eliminate the virus from contaminated water. These CNFs can filter 80–120 nm-sized viruses (swine

influenza and xenotropic murine leukemia) with high efficiency. High-performance composite membranes have also been introduced with reinforcement of inorganic and metallic particles. For dye degradation, highly efficient nanocellulose-based composite membranes were developed by introducing graphene oxide (GO) into the BNC matrix, followed by in situ syntheses of palladium nanoparticles on the GO flakes. Pd/GO/BNC-based layered membranes enhanced the contact and interaction between dye contaminants and catalytic Pd NPs, thus, maximized the dye degradation performance while filtering the water [191].



Fig. 16 – BNC effectively utilized in dental tissue regeneration (a sample pic of 39-year-old female patient [211]).

5.5. Biomedical applications

Nanocellulose is a promising biomaterial for different medical applications, owing to its biocompatibility [192], low cytotoxicity, distinct shape, surface science, rheology, and crystallinity [193–195]. The cytotoxicity of these materials relies upon molecule size, surface science, and impeccability of the manufacturing process. Furthermore, drug delivery [196], tablet coatings [197,198], and protein immobilizations [126] are also the other research hotspots in which these nanocellulose-based materials can be effectively utilized [199].

Furthermore, these materials also gain importance in health-care products due to their significant mechanical properties, nano-scaled fibrous interlinked structure, and natural availability. These features have attracted researchers to develop their applications in different biomedical material synthesis, such as nanocellulose in hydrogels, scaffolds, threads, bioactive implants, drug delivery, cell cultivation, and tissue engineering (Fig. 12).

Efforts have been made to synthesize biomaterials with osteoconductive and osteoinductive properties, similar matrix structure as natural extracellular to build stronger cell–material interaction for bone engineering. In this scenario, bacterial cellulose contributes as an effective biopolymer and is used for tissue engineering applications due to its excellent mechanical strength. Among all the types of nanocellulose, BNCs are considered the most important in biomedical applications [138] due to their excellent biocompatibility, thus suited for many applications like scaffold [200], tissue engineering [201], and membranes. Antimicrobial treatment, auricular cartilage regeneration [202,203], and blood vessel replacements [204] are by far the essential areas of BNCs applications [205–207]. For example, Markstedt et al. [208] produced human chondrocytes within an ear-like shape, as depicted in Fig. 13.

Wounds can cause a risk of infection and need to be treated for proper healing to avoid serious health problems. These

Table 5 – Applications of different nanocellulose in various biomedical and other engineering fields.

Applications	Nanocellulose Forms				Ref.
Electronics Devices	CNF	0D, 1D, 2D	–	–	[180,217]
Tissue Engineering	–	–	–	–	[218]
Medical/Pharmaceutical Industries	–	CNC	–	–	[219]
Energy-based devices	CNF	–	CNC	–	[180,220]
Biosensors	CNF	–	CNC	–	[183,221]
Self-Heating Polymer films	CNF	–	–	–	[222]
Water Treatment	–	–	CNF	–	[180]
Rubbery Materials	CNF	CNC	–	–	[223]
Optical Devices	CNF	–	–	–	[224]
Food Industries	–	–	CNC	–	[225]
Agriculture Industries	–	CNF	CNC	–	[57]
Package Industries	NC	–	CNC	–	[226–228]
Hydrogels	–	BNC	CNC	–	[129,229]
Automotive Windows	–	–	CNC	–	[230]
Wound Healing/Diagnosis	CNF	–	–	–	[183,231]
Biomedical Devices	CNF	–	–	–	[232]
Barrier Films	–	CNF	–	–	[233]
Continuous Papermaking	CNF	–	–	–	[234]
Coatings Materials	CNF	–	CNC	–	[235]
Aerogels	ACC	–	CNC	–	[147,236]
Ballistic Protection	BC	CNFs	CNC	–	[178]
Cryogels	NFC	–	–	–	[237]
Biocomposites	BC	–	–	–	[238]
Textile industries	Plant Stem (giant reed and Dutch rush) with different polymer materials	–	–	–	[239]
Aerospace structure	BC	–	–	–	[240]
Self-cleaning Materials	Superhydrophobic Cellulose	–	–	–	[241]
Viscosity modifiers	CNF	–	–	–	[242]
Antibacterial Materials	MFC	–	–	–	[243]
Rheology modifiers	CNF	CNC	–	–	[244]
Porous Materials	–	NFC	–	–	[245]
As catalysts	Ethylenediamine-functionalized nanocellulose	–	–	–	[246]
Hygiene products	OPEFB Nanocellulose	–	–	–	[247]

different nanocellulose-based materials are effective for wound healing applications. For example, Jiji et al. [209] proposed thymol enriched bacterial cellulose hydrogel for a third-degree burn. This study revealed that these bacterial cellulose hydrogels (BCT) have an exceptional biocidal activity for burn-related pathogens and improve this type of wound (Fig. 14). Furthermore, these hydrogels enabled the development of fibroblast cells bearing low toxicity and enhanced cell viability than traditional bacteria cellulose (BC) technique.

BNC is an excellent skin substitute, usually utilized for wound dressing, and offers extraordinary substrate for wearable sensors. Silva et al. [210] proposed a comprehensive study on skin-adherent biosensors to detect metal ions in human sweat and urine. The sensing unit is composed of screen-printed carbon terminals (SPCEs) on BNC. These prepared SPCEs were utilized to identify the poisonous metals cadmium (Cd^{2+}) and lead (Pb^{2+}) with an accuracy of detecting 1.01 and 0.43 μM , individually, which is adequate for detecting these metals ions in the human body (Fig. 15). In the form of the *Gluconacetobacter xylinus* strain, BNC is effectively used to regenerate dental tissue [211] (Fig. 16). Furthermore, NC such as Gore-Tex® and Gengiflex® have been observed as promising materials within the dental industry [212].

New nanocellulose-based threads have been synthesized that exhibited biocompatibility with human stem cells to accelerate the healing procedure [213]. These threads are generally prepared with the patient's cells one week before human surgery to avoid the risk of human immune reaction [214]. Nanocelluloses have also been used in polyvinyl alcohol-based hydrogels for ophthalmic applications as these composite materials are mechanically strong regardless of their softness and flexibility [215,216].

To summarize this, nanocellulose is extensively utilized in many applications of engineering and biomedical fields. The nanocellulose is produced from waste fruit and organic products. Researchers concentrate further on the extraction, surface modifications and explored further applications of nanocellulose. A summary of previous works on exploring different applications of nanocellulose is summarized in Table 5.

6. Conclusion

In this study, nanocellulose-based materials are discussed based on the recently published studies, mostly from 2015 to 2021. The major three types are CNCs, CNFs, and BNCs; in terms of synthesis, different surface characterization techniques and emerging applications in various fields are summarized. The fundamental mechanisms behind these three types are also highlighted. The different intrinsic characteristics of nanocellulose-based composites can be improved considerably by different manufacturing methods and by incorporating efficient materials with nanocellulose. For instance, by adding different nanoparticles like MWCNTs, the mechanical characteristics of nanocellulose based composites have been improved. Furthermore, chemical treatments also provide the improved mechanical properties of nanocellulose. It is worth mentioning that there is ever-growing interest for researchers in developing a strong relationship between the different properties like fibril morphology, crystallinity,

hydrophilic capacity, and mechanical properties (elasticity, elongation, thermal stability, and surface roughness). Among all the types of nanocellulose, BNCs and NFCs have strong potential to reinforce nanocellulose-based composite materials. To our best knowledge, a maximum young's modulus of 21 GPa and the maximum strength of 320 MPa were found in tensile property characterization of these nanocellulose composites, which have been achieved so far. One of the major issues that need to be sorted in nanocellulose composite is the poor dispersion of the nanofillers in the polymer matrices. It is concluded that cellulose should be considered the main component in polymers. Industrial-scale production of nanocellulose can be possible from cellulose and subsequently their utilization in engineering and medical applications.

7. Future challenges and opportunities

Although considerable improvements in nanocellulose-based materials properties are reported. However, removing different substances to improve their performance and recycling issues are still the major concerns associated with nanocellulosic materials. Future investigations on nanocellulose-based materials must consider various aspects like biomimetics, numerical models, artificial intelligence (AI), and machine learning-based algorithms to address the current difficulties. The novel manufacturing routes like additive manufacturing or 3D printing should also be considered for uniform and on-demand utilization of nanocellulose. 3D printing based novel processes must be explored to produce nanocellulose, which should be technically feasible for large-scale production. These considerations will further empower the researchers to get better-functionalized nanocellulose materials and their composites in the future. With the significant advancements in multi-disciplinary areas, nanocellulose-based composites will show improved characteristics. Furthermore, these materials should have considerably low environmental impacts. These research gaps will lead towards a new research area of sustainable nanocellulose for high-performance and advanced applications.

Data availability

Data sharing does not apply to this article as no datasets were generated or analyzed during the current study.

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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