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# Nanocellulose and carbon nanotube composites: A universal solution for environmental and energy challenges

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## ABSTRACT

In recent years, the interest of researchers in developing materials that can solve the problems of energy scarcity and environmental pollution has led to a special focus on nanocellulose and carbon nanotube-based composites. Nanocellulose has outstanding characteristics that promote highly ordered structures that are renewable and biodegradable. Carbon nanotubes, in turn, have high surface area, strength and electrical functionality. Their combined use holds promise for the development of composites with unique properties applicable in a variety of applications. However, despite the research conducted, questions remain unresolved regarding the technological aspects of production and specific applications of nanocellulose and carbon nanotubes. This review presents recent advances in the field of multifunctional hybrid nanocellulose and carbon nanotube-based nanocomposites, with a focus on their role in addressing environmental and energy challenges. The paper discusses the characteristics and advantages of the components of the composites, methods for their synthesis, as well as potential problems and possible directions for further research. Particular emphasis is placed on improving the interactions between the components, reducing the cost of production, and extending the scope of applications by integrating additional nanomaterials.

## 1. Introduction

Current trends in industrial and technological development towards high demand for energy resources have stimulated active research on nanomaterials derived from natural resources [1,2]. Unlike conventional alternatives, these materials offer multifaceted properties including ease of processing, enhanced performance, and sustainability, which has attracted increased attention from the research community. Nanomaterials offer significant advantages such as high surface-to-volume ratio and customizability, making them promising for various applications [3]. Their diverse range of nanoscale sizes provides

high surface area that favors adsorption and modification of contaminants. This makes nanocomposites attractive for the design and development of sustainable materials in energy and environmental applications, emphasizing their potential in the context of creating innovative solutions for future needs [4].

Nanocellulose (NC) is currently attracting considerable interest as a promising, non-toxic, and environmentally friendly material due to its high mechanical strength, excellent biocompatibility, and good degradation and regeneration abilities [5]. Its nanoscale rod-like or fibrillar structures exhibit outstanding characteristics, modifying the traditional properties of NC building blocks and facilitating the development of

*Abbreviations:* APS, Ammonium persulfate; BNC, bacterial nanocellulose; CMWCNTs, carboxylated multi-walled carbon nanotubes; CNC, cellulose nanocrystals; CNF, cellulose nanofiber; CNTs, carbon nanotubes; CPR, ciprofloxacin; DCF, diclofenac sodium; EC, ethyl cellulose; GO, graphene oxide; HNS, heterofibrous network scaffold; LbL, layer-by-layer; LIBs, lithium-ion batteries; LiPS, lithium polysulfide batteries; MWCNTs, multi-walled carbon nanotubes; NC, nanocellulose; PANI, polyaniline; PCM, paracetamol; PS, pigskin; SWCNTs, single-walled carbon nanotubes; TEMPO, 2,2,6,6-tetramethyl-1-piperidine-N-oxyl; UDF, unidirectional freeze drying; UDP-Glc, uridine diphosphate glucose; WPF, wood pulp fibers; ZAB, zinc-air batteries.

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highly ordered hybrid structures. NCs can effectively act as a stable carrier and matrix for the eco-friendly synthesis of multifunctional nanocomposites in the form of hybrid filaments, films, and aerogels with strong and rigid chained monomolecular structures [6]. The electrostatic stabilization by hydroxyl centres (-OH) on the NC surface and hydrogen bonds between hydroxyl and oxygen-containing groups is critical to achieve a stable colloidal suspension in polar solvents [6]. In addition, the critical concentration of a colloidal suspension of NCs can lead to unique ordering of chiral nematic self-assembly, providing a variety of template routes with unique electrochemical, adsorption and mechanical properties [7]. Thus, NCs functionalized according to targeted precursors offer alternative perspectives for the development of nanocomposites with unique architectures and tunable properties for a variety of applications.

On the other hand, carbon nanotubes (CNTs), which are derivatives of carbon nanomaterial, possess strong covalent carbon-carbon bonds along the axis of their regular 1D tubular structure, which provides ultrahigh stiffness (up to 1 TPa) and tensile strength approaching 100 GPa [8]. Whereas the sp<sup>2</sup> hybridization ability gives them exciting electrical properties. The combination of low density and outstanding mechanical, electrical and thermal properties provides CNTs with the opportunity and functionality to develop innovative hybrid nanocomposites in combination with NC to improve performance over individual components. This hybridization of CNTs can serve as a further direction to improve the functionality of renewable nanocellulose. However, the aggregation of CNTs due to the large specific surface area and strong Van der Waals forces between nanotubes creates difficulties in the uniform dispersion process, resulting in deterioration of the excellent mechanical and electrochemical performance [9]. Methods of improving dispersion, such as the use of surfactants or chemical modification through covalent bonding, using traditional strong acids like H<sub>2</sub>SO<sub>4</sub>/HNO<sub>3</sub> [10], may disrupt the surface structure and integrity of CNTs, hindering the full realization of their superior properties [11]. Recent studies suggest that NC can effectively serve as a stable dispersant for CNTs with high concentration, circumventing the need for additional surfactants and avoiding modification by strong acids [12–15]. In addition, the uniform mixing of CNTs with NCs used as matrix favors channel dilation, allowing unimpeded ion transport [12]. This enables the fabrication of lightweight, flexible and highly porous composite structures of 1D filaments, 2D films, nanopapers and 3D hydrogels and aerogels directly from dispersions of NC/CNT derivatives, without the use of additional structural materials. Thus, important aspects such as affinity, stability, dispersibility, and strong covalent and non-covalent interactions between the molecular chains of NC and CNTs represent fundamental characteristics that form the basis for the development of multifunctional hybrid nanocomposites with preferred properties.

In this review, we have considered the current state of the art in functional materials comprising hybrid structures derived from NC and CNT derivatives. Initially, the characteristics and advantages of nanocellulose and carbon nanotubes individually, as well as the potential offered by composites based on them, were summarized. Then, the strengths and possible challenges of different methods in forming various 1D, 2D and 3D structures based on NC/CNT nanocomposites were analyzed. Additional attention was given to the consideration of innovative hybrids applied in various applications including environmental remediation and energy storage and conversion systems. Finally, the current challenges and future prospects of NC and CNT hybrids were briefly discussed.

Due to the characteristics described above, in recent years, there has been an increased research interest in hybrid multifunctional nanocomposites incorporating nanocellulose and carbon nanotubes. However, it is worth noting that no recent reviews have been published that fully address nanocellulose and CNT-based nanocomposites, their technological aspects of fabrication, and specific applications in the fields of environmental clean-up and energy. The present review aims to fill this specific scientific vacuum and to rethink aspects of the utilization

of environmentally friendly functional materials.

## 2. Features of nanocellulose and carbon nanotubes

### 2.1. Overview of nanocellulose

Cellulose (C<sub>6</sub>H<sub>10</sub>O<sub>5</sub>)<sub>n</sub> is a linear semi-rigid biopolymer consisting of repeating D-glucose units linked together in a <sup>4</sup>C<sub>1</sub> chain configuration and twisted 180° relative to each other [16,17]. These glucose units combine to form crystalline structures known as elementary fibrils. These fibrils combine to form microfibrils, which in turn form macrofibrils or cellulose fibers. The structure of cellulose includes three hydroxyl groups: a primary (-OH) group at the C-6 position and secondary (-OH) groups at C-2 and C-3, which have hydrophilic properties [18]. The chemical groups within and between cellulose molecules give it unique properties such as ease of functionalization, chirality, hydrophilicity, and insolubility in most aqueous solvents [19]. The chain length and degree of polymerization of cellulose vary depending on its source and extraction process. The main sources of cellulose are plants, animals, bacteria and others. Cellulose has both ordered crystalline regions and more chaotic amorphous regions.

Different extraction and processing methods can produce different polymorphic forms of cellulose, making it a versatile starting material for a variety of applications based on its origin and structure. For example, the crystal structure of cellulose I includes two different forms: cellulose I $\alpha$  (triclinic) and cellulose I $\beta$  (monoclinic), where the latter is considered the most stable from a thermodynamic point of view [20]. The type of cellulose allomorph depends on the source [21]. Algae, bacteria and other aquatic organisms usually produce cellulose I $\alpha$ , while trees, plants and some animals such as shellfish provide cellulose I $\beta$  [22].

On the other hand, nanocellulose is an ultrafine cellulose particle, at least one of its dimensions (length, width or diameter) is between 1 and 100 nanometers. It is obtained from conventional cellulose using mechanical methods (micro-annealing, high-pressure homogenization, micro-milling and high intensity ultrasound treatment) [23] or chemical methods (acid hydrolysis, 2,2,6,6,6-tetramethyl-1-piperidine-N-oxyl (TEMPO) oxidation, ammonium persulfate (APS)) [24]. Nanocellulose has unique properties such as high Young's modulus and high tensile strength, flexibility, hydrogen bonding ability, biodegradability, low coefficient of thermal expansion and large surface area, which makes it a sought-after material in various fields including energy, environmental, biomedical, food processing and many more [17,22,25]. Nanostructured cellulose has outstanding characteristics, notable for the extensive presence of hydroxyl function groups that can be further modified to integrate binding sites. The above properties predispose NC as an attractive building block for synergistic interactions with various functional materials such as polymers, graphene and CNTs. The applications of NC in science and technology reveal promising opportunities for the creation of novel nanocomposite materials [26]. Nanocellulosic materials can be classified into three main types depending on how they are obtained [27].

#### 2.1.1. Cellulose nanocrystals

The first type is cellulose nanocrystals (CNC), also called nanocrystalline cellulose (NCC) (Fig. 1(a,d,g and j)). This type of nanocellulose is mainly obtained by controlled chemical treatment of cellulose fibers. The process involves alkaline, bleaching and acid hydrolysis of cellulose fibers. As a result of this treatment, disordered regions of cellulose like chain-like dislocations along the elementary fibril undergo hydrolytic decomposition due to the reduction of spatial interferences and kinetic factors [28]. While the ordered domains, which are more resistant to hydrolysis, remain intact [25]. Thus, amorphous domains are removed from the original cellulose, leading to the formation of needle-like cellulose nanocrystals with excellent crystallinity, with a cross section on the order of 5–20 nm and a length of 100–200 nm, i.e. high aspect ratio, and colloidal stability in aqueous

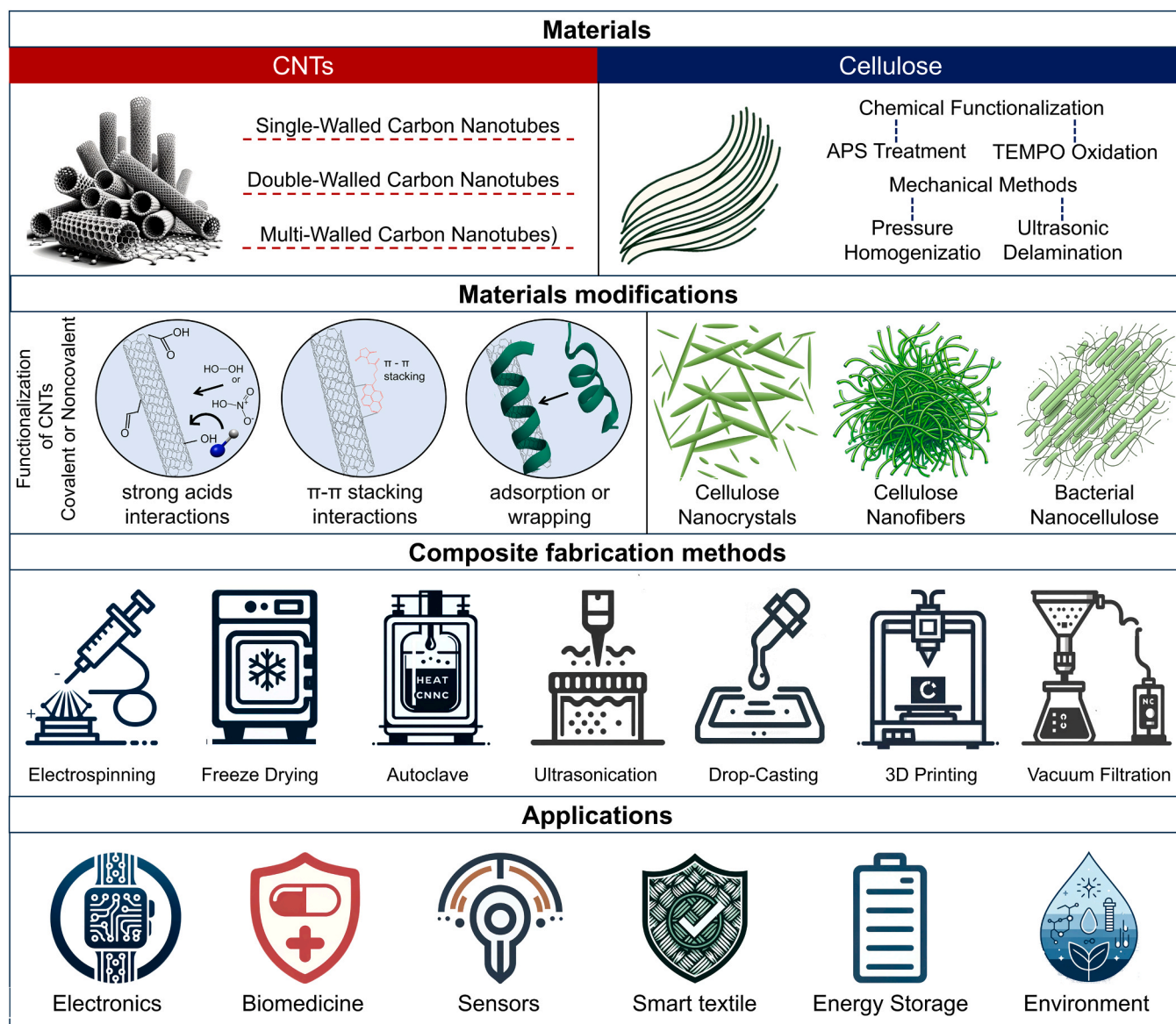


Fig. 1. Schematic overview of this review.

suspensions [25,29]. The latter characteristic is determined by the electrostatic repulsion between neighboring CNC particles, while the first two stem from the destruction of disordered regions of cellulose during hydrolysis. In many cases, these properties play an important role in applications requiring high productivity.

In addition to their high length versus diameter and significant surface area, CNCs have remarkable physical characteristics. They are capable of self-organizing into chiral nematic liquid crystal phases [29] and also respond to magnetic and electric fields [30]. Furthermore, the authors confirm that they exhibit piezoelectric properties [31]. When CNC is embedded in polymeric materials, hydrogen bonding networks are formed that facilitate the transmission of mechanical loads [32]. This is particularly interesting from the point of view of additive manufacturing. However, CNC-derived materials such as films and aerogels exhibit brittleness and break easily when deformed due to their low size ratio. This limits their application in various fields including the development of flexible supercapacitors [33]. To successfully integrate active materials with CNC and develop nanocomposites with enhanced mechanical strength, thermal stability and flexibility, it is necessary to rationally design and control the surface properties and interactions between CNC and active materials. This will achieve homogeneous

integration with high active material loading without reducing the specific surface area of the bulk materials.

### 2.1.2. Cellulose nanofibers

The second type is cellulose nanofibers (CNFs), which are preferably produced by mechanical methods (sometimes with enzymatic or chemical pretreatment) including high-pressure homogenization, ultrasonic delamination or ball milling, which help to break down the cellulose into nanometric fibers (Fig. 1(b,e,h and k)) [28]. These production methods allow CNFs to retain an amorphous-crystalline structure. This blended structure and processing methods allow CNFs to have more flexible properties relative to CNC and bacterial nanocellulose (BNC) with thicknesses in the range of 20–100 nm and lengths >10,000 nm, depending on the isolation process and the source of the nanocellulose [28].

Such nanofibers have a large surface area, which makes them attractive for electrochemical energy storage devices. In addition, the large surface area allows successful surface modification of nanocellulose due to the presence of hydroxyl group on the nanofiber surface [34]. Although the Young's modulus of CNF is approximately  $10.2 \pm 1.2$  GPa [35], which is found to be lower than that of bacterial and

crystalline celluloses, successful integration with carbon nanomaterials allows for improved mechanical performance [36,37]. It is important to note that these nanofibers exhibit thixotropic properties and can become less viscous under shear stress [38]. However, their stability in solution is often limited due to fiber aggregation, driven by a high affinity for forming interparticle bonds, which can reduce their effectiveness in various applications such as composite materials [39]. Several solutions to the aggregation problem of CNF have been proposed by researchers in this field [40–42]. The most common approach involves the introduction of surfactants or polymers to stabilize the CNF dispersion and prevent aggregation. Specifically, increasing the electrostatic charge on the surface of nanofibers promotes like-charge repulsion, thereby preventing aggregation and flocculation [43,44]. It is important, however, to exercise caution and not exceed the critical charge density, as an excessively high charge density can lead to counterion condensation. Additionally, mechanical disaggregation methods, such as ultrasonic treatment or high-speed mixing, can break interparticle bonds and ensure uniform distribution of CNF in colloidal solutions [40]. Furthermore, chemical modification of cellulose nanofibers can improve their dispersion, making them more compatible with various matrices and surfaces.

### 2.1.3. Bacterial nanocellulose

The third type of nanocellulose, BNC, or bacterial nanocellulose, has attracted attention for its unique properties (Fig. 1(c,f,I and l)). This material is considered pure and devoid of plant or animal components [16]. While CNC and CNF are produced by top-down approach, BNC is obtained by bottom-up technology. The bacteria used for BNC synthesis belong to known genera such as *Alcaligenes*, *Agrobacterium*, *Sarcina*, *Acetobacter*, *Rhizobium* or *Pseudomonas* [34]. BNC production is a complex and multistep process controlled by multiple genes, enzymes and protein complexes. Cellulose synthase plays a major role in BNC synthesis, acting in concert with other enzymes and regulatory proteins. The process involves several metabolic pathways such as the phosphogluconate pathway and the Krebs cycle. These pathways involve four key steps including the conversion of glucose to glucose-6-phosphate, conversion of glucose-6-phosphate to glucose-1-phosphate, creation of uridine diphosphate glucose (UDP-Glc) and the final polymerisation of UDP-Glc into cellulose by cellulose synthase [45]. Although the process of UDP-Glc production has been studied, the mechanism of glucose polymerisation into long chains remains poorly understood.

BNC has outstanding mechanical characteristics, including high crystallinity (75–96 %) and significant Young's modulus (between 78 GPa and 114 GPa), which makes it stronger than CNC and CNF [46]. The structure of BNC is formed due to internal and external hydrogen bonds, which gives the fibers density and insolubility in water, but the ability to retain moisture. BNC nanofibers have a small diameter (20–100 nm) and a large surface area, which provides high water absorption capacity and good adhesion [47,48]. These unique characteristics, combined with high biodegradability, make BNC an excellent material for creating valuable products such as temporary artificial skin for medical applications. BNC also finds applications in drug delivery systems, energy storage devices, solar cells, energy harvesting mechanisms and other fields [34,49].

## 2.2. Carbon nanotubes

CNTs have attracted considerable scientific interest since their discovery in 1991. Carbon nanotubes are one of the modifications of carbon possessing a hexagonal lattice of carbon atoms in the  $sp^2$  hybridization state [58]. These nanotubes are tubular nanostructures produced by a variety of methods. To better understand their structure, carbon nanotubes can be viewed as coiled graphene nanosheets, where the ends of the nanosheets are connected to form cylindrical nanotubes. CNTs are a significant class of nanomaterials that differ in the number of graphene layers in their structure. Among them, SWCNTs (single-walled carbon

nanotubes), DWCNTs (double-walled carbon nanotubes), and MWCNTs (multi-walled carbon nanotubes) are the best known (Fig. 2) [59]. SWCNTs consist of a single layer of graphene and possess various structural shapes such as zigzag, crescent and chiral. DWCNTs, on the other hand, exhibit higher structural stability and interesting electronic and optical properties due to their unique internal coaxial structure, which allows functionalization of the outer wall without compromising the integrity of the inner wall [60]. MWCNTs are structures with several graphene layers interacting with each other due to Van der Waals forces [61].

Conventional methods such as catalytic chemical vapor deposition (CCVD), arc discharge and peapod methods are used to synthesize CNTs. These methods yield different types of CNTs (SWCNTs, DWCNTs and MWCNTs), depending on the process parameters such as pressure, temperature, carbon precursor and catalyst. Illustrations of the main CNT synthesis methods are shown in Fig. 2. The qualitative characteristics of CNTs, such as defects, diameter and length, are determined by the synthesis method and affect their thermal and mechanical properties. The diameter of nanotubes varies on the nanometer scale (from 0.4 to 2.5 nm for SWCNTs), while their length can reach several micrometers [62]. This high diameter-to-length ratio favors an increase in specific surface area.

In addition to their cylindrical nanostructure, carbon nanotubes exhibit unique physicochemical characteristics. These properties include outstanding mechanical strength, mainly due to  $sp^2$  bonds (e.g., the tensile strength of SWCNTs is almost 100 times that of steel, while the specific gravity of the latter is about six times higher) [63]. Carbon nanotubes also have excellent electrical properties and excellent chemical and thermal stability, maintained at temperatures of about 700°C in air and up to 2800°C in vacuum [61]. Due to their extensive surface area and the presence of delocalized  $\pi$ -electrons on their surface, carbon nanotubes also possess outstanding adsorption activity and present interesting prospects for energy storage and in the field of environmental cleaning. However, pure CNTs are insoluble in aqueous media, which is a major disadvantage in practical applications. To overcome this problem, functionalization is applied, which is achieved using a variety of molecules including absorption, electrostatic interaction or covalent bond formation [58]. These modifications lead to efficient removal of impurities in CNTs and improved water solubility, as well as enhanced electrochemical performance [26].

Despite the wide range of CNT applications, the main obstacle that restrains its large-scale application is toxicity, which negatively affects human health and the environment [64]. However, the actual source of CNT toxicity is still unclear. The toxicity of carbon nanotubes is believed to be related to various factors including surface modification, degree of aggregation, dose, morphology, size and length [65,66]. Despite the complexity of the intracellular processes that are affected by CNTs, some studies have suggested strategies to overcome their toxicity. These strategies include modifying the surface of the material using functionalized groups, coating with metal oxides or attaching proteins [67]. Modification of their hydrophobic characteristics, making them incompatible with their biological environment, can be addressed using chemical functionalization and stabilization of the nanofluid [68]. The essence is that stabilization through mechanical action prevents degradation of the chemical structure and optoelectrical characteristics of nanomaterials. Nevertheless, increased attention should be paid to the potential toxicity of surfactants at the required fraction of CNTs in the mixture [69,70].

In this situation, innovative nanostructures based on biopolymers such as nanocellulose represent a promising approach to effectively stabilize carbon nanotubes in aqueous media within the framework of biocompatibility and environmental sustainability [71]. Moreover, functionalization with hydroxyl, epoxy, carboxyl and carbonyl surface functional groups helps to strengthen the bond with nanocellulose, which facilitates the development of NC/CNT-based composite [26]. Nanocellulose exhibits interaction with carbon nanotubes through

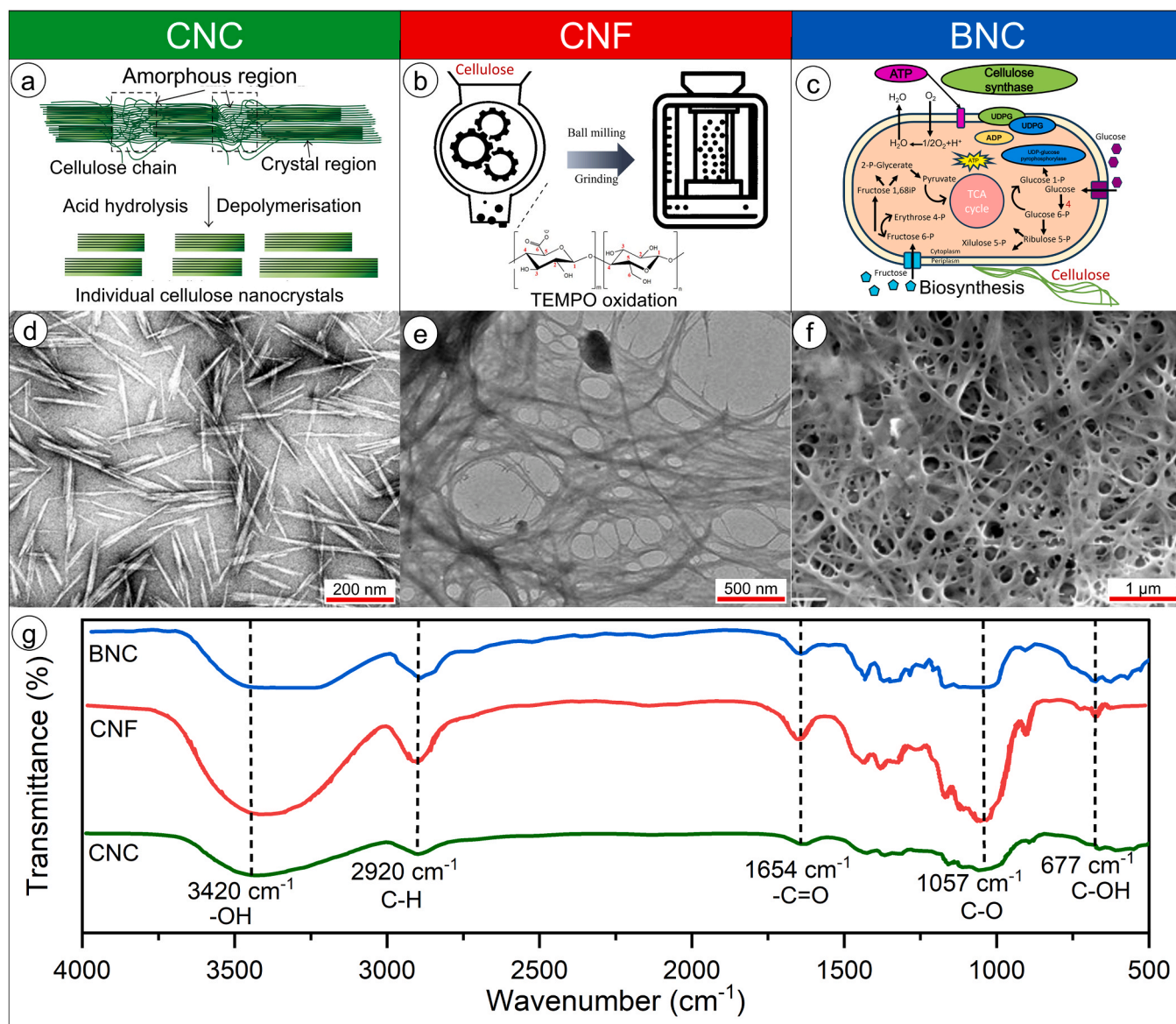


Fig. 2. Schematic illustrations of preparation processes of CNC (a), CNF (b) and BNC (c) [50–52]; TEM images of CNC (d) [53], CNF (e) [54] and FESEM image of BNC (f) [55]; FTIR spectra (g) for CNC [56], CNF [57] and BNC [55].

intermolecular physical forces, forming hybrid nanostructures that exhibit high stability when dispersed in water [72]. This collective approach provides nanocellulose with outstanding properties as a stabilizer for CNTs, surpassing the effectiveness of inorganic surfactants, biological molecules and other traditional polysaccharides.

### 3. Nanocomposites based on NC/CNTs

In today's world of materials science for solving environmental and energy problems, where emphasis is placed on environmental friendliness, low cost and high performance at the same time, nanocellulose derivatives (CNC, CNF and BNC) with large surface area, high porosity content and excellent mechanical strength are unique materials that fulfil all these requirements [81]. However, the main feature of nanocellulose is the possibility of synergy with other materials in the form of composites, due to the abundance of surface functional groups, facilitating even more improvement of initially existing properties and imparting additional characteristics by using another material involved in composite formation and the possibility of expanding and improving the arsenal of environmental and energy applications. Recently,

nanocellulose-based composites and carbon-based nanomaterials, especially carbon nanotubes, have attracted particular interest due to a number of advantages provided by these nanomaterials, in particular: (1) carbon nanotubes are known for their outstanding mechanical strength and their addition to nanocellulose-based composites can significantly improve the mechanical properties of the composites, making them stronger and more resistant to deformation; (2) CNTs have outstanding electrical and thermal conductivity, making nanocomposites based on them attractive for electronic and thermal conductive applications; (3) CNTs have low density and high stiffness and their incorporation into nanocomposites can improve the stiffness of the material without increasing its mass; (4) the large specific surface area of CNTs promotes good adhesion and interaction between the phases of the composite, which in turn improves the mechanical properties.

Thus, the advantages of NC/CNT composite attract the attention of researchers from all over the world as a versatile material for a wide range of applications. Therefore, there is a need to develop and optimize synthesis methods to create NC/CNT-based materials with desired architecture and properties.

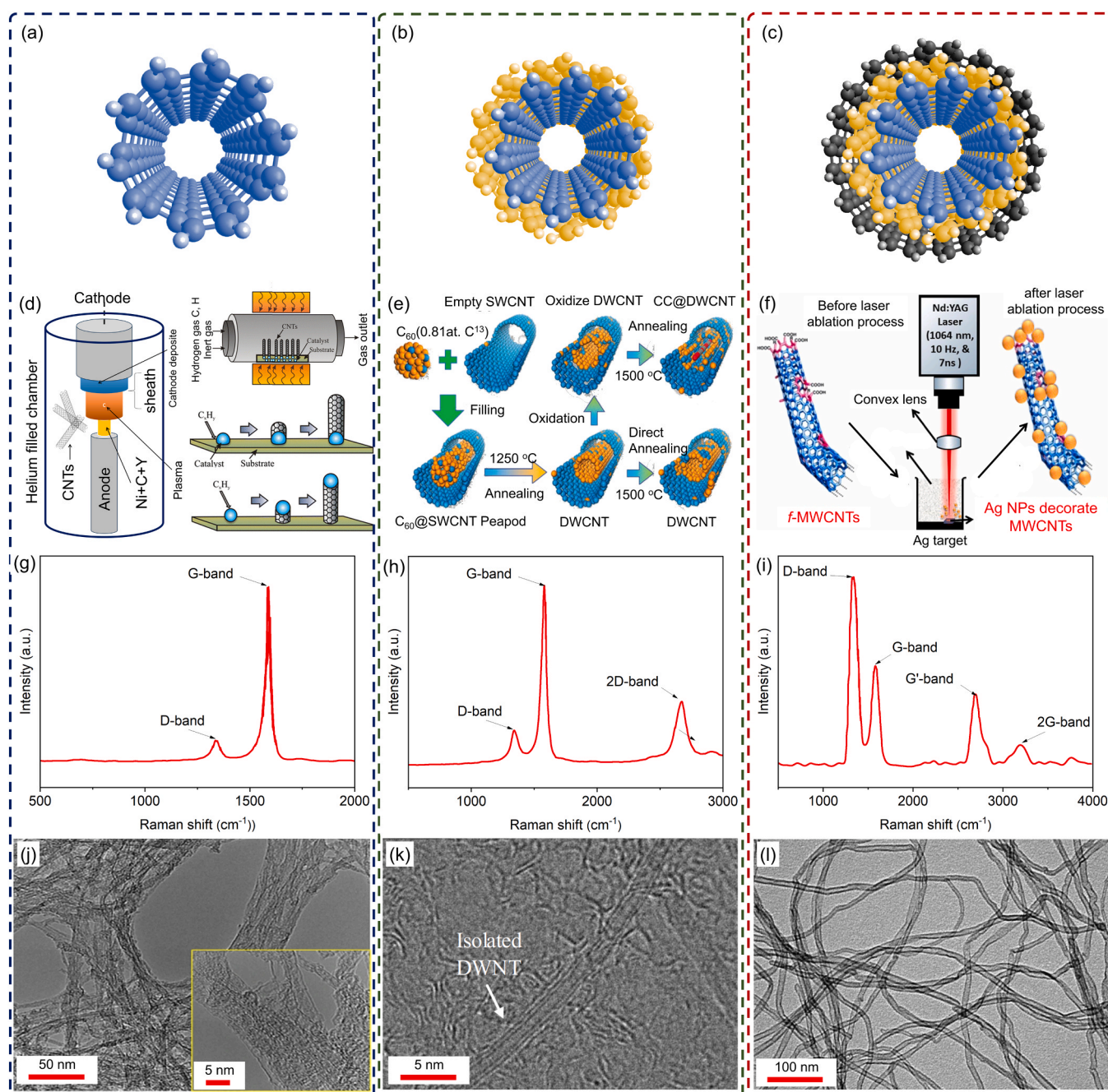
### 3.1. Methods of fabrication of NC/CNT-based nanocomposites

#### 3.1.1. Thin films based on NC/CNTs

Despite the fact that CNTs have excellent electrochemical properties, the main challenge in working with CNTs is their hydrophobicity, which increases the probability of aggregation in aqueous solutions, thus preventing these advantages from being fully utilized [82]. Existing methods to improve dispersion, such as the use of surfactants or treatment with strong acids on the CNT surface to provide solvation [10], have their limitations. For example, the use of surfactants can cause adhesion problems to polymers in composites due to weak interfacial layers. Whereas, strong acids like  $H_2SO_4/HNO_3$  during processing can disrupt the electronic structure of CNTs, which reduces the conductivity of the composites, thereby jeopardizing the superior properties of CNTs

[11]. Instead, the uniform mixing of CNTs with NCs used as matrix favors channel expansion and allows unimpeded ion transport [12,83].

As mentioned in Section 2.2, modification of CNTs using a variety of functional groups on their surface promotes improved interaction with the matrix of nanocellulose derivatives. The presence of hydrogen bonds between nanotubes and various NC surface functional groups can contribute to the electrostatic stabilization and efficient bonding of NC and CNT chains using strong Van der Waals and hydrogen bonds [84–86]. Dispersion techniques involve dissolving NC and CNTs in a solvent or dispersion medium with the possibility of use in downstream processes such as drop-casting [13], vacuum filtration [87] or electroforming [88,89]. These methods require the creation of a stable colloidal dispersion, which in turn allows the development of composite materials with high electrical conductivity and strength, bypassing the need for



**Fig. 3.** Representation of SWCNT (a), DWCNT (b) and MWCNT (c); Schematic of preparation processes of CNTs (d, e, f) [60,73–75]; Raman spectra for SWCNT (g) [76], DWCNT (h) [77], MWCNT (i) [78]; TEM images of SWCNT (j) [76], DWCNT (k) [79], MWCNT (l) [80].

chemical functionalization with harmful acids or the addition of undesirable surfactants. Ultrasonic processing is a simple method to disperse CNTs in aqueous nanocellulose solutions without the use of surfactants [87].

However, in order to achieve optimal dispersion and functionalization of CNTs, it is important to choose the right energy level of ultrasound treatment, taking into account the effect of the medium, in order to overcome only non-covalent forces without damaging the CNT structure [90]. It should be noted that colloidal stable aqueous dispersions of nanocellulosic materials can be successfully obtained, especially if these materials possess certain surface functional groups with sufficient ionic charge [91]. The interfacial and intermolecular interactions between NC and CNTs play a key role in stabilizing the composite and contribute to increasing the dispersion limit of CNTs, which represents a key aspect for the ideal design and assembly of high-performance composite nanomaterials [9,92]. In addition, functionalized nanocellulose-based materials can provide ionic conductivity in composite matrices, making them attractive for a wide range of electrochemical and other applications [13].

Of particular importance is the high stability of aqueous NC/CNT dispersions, which can be deposited by drop casting onto tetrahedral amorphous carbon substrates over a wide potential range (-0.6 to +1 V) and in a variety of electrolytes (Fig. 3(a)) [13]. The use of a simple and environmentally friendly vacuum filtration method allows the creation of thin and strong structures of NC/CNT nanopapers with different thicknesses (Fig. 3(b)) [93]. Moreover, buckypaper (BP) obtained by horizontal vacuum filtration from MWCNTs after addition and chemical treatment using CNC exhibits super hydrophilic properties with wetting angle (CA) ranging from 46.6° to 81.3° compared to untreated MWCNT BP, which has a CA value of 132.2° [94].

### 3.1.2. 1D structures based on NC/CNTs

Other promising materials that have attracted great attention of scientists include mechanically strong and conductive yarns due to qualities such as excellent flexibility, knitting ability and light weight. The development of NC/CNT based composite yarns contributes to the improvement of mechanical strength and electrical conductivity [95]. For example, in the case of CNF/SWCNT yarns (SWCNT content 43 wt %), the strength reached a value of about 220 MPa and electrical conductivity of ~207 S/cm [96], whereas yarns spun from pure SWCNT had a mechanical strength of ~124 MPa and electrical conductivity of ~100 S/cm [97]. Moreover, the one-dimensional (1D) structure provides significant property enhancement due to the dual alignment of both CNFs and SWCNTs [84]. Conventional methods to create NC and CNF-based filaments include wet spinning [84,98], flow-focusing [96] and 3D printing [99]. In particular, the work conducted by Zhangmin Wan et al. [84] presented a simple and scalable method to produce unidirectional CNF/SWCNT composite filaments for use as a high-performance structural material. This method utilized a three-roll mill and wet spinning technology to provide a strong bond between NC and CNTs. This leads to effective component alignment, efficient load redistribution and enhanced electrical conductivity of CNF/CNT composite filaments. The resulting filaments exhibit moderate electrical conductivity while maintaining outstanding mechanical properties superior to those of the individual components.

In another study, a sustainable strategy for interfacial nanoparticle complexation (INC) was developed to create conductive composite nanofibers made of anionic (TEMPO)-oxidized cellulose nanofibers (TO-CNF) with the addition of carboxylated multi-walled carbon nanotubes (CMWCNTs) [100]. The study demonstrated that increasing the CMWCNT content in the composite leads to a decrease in the tensile strength and Young's modulus, but the electrical conductivity increases significantly, reaching 392 S/m at a concentration of 0.4 % CMWCNT in the INC filament. Thus, the obtained conductive and green filaments based on the INC method have high potential for applications in wearable electronics and biomedicine [101]. An additional method for

dispersing NC and CNTs is electrospinning, which enables the formation of composites in the form of fibers with diameters as small as a few tens of nanometers (Fig. 3(c)). This method differs from other methods such as wet spinning, dry spinning, melt spinning and spinning with the formation of interfacial complexes, which are limited to micrometer dimensions [98,102]. Using electrospinning, it is possible to produce continuous one-dimensional CNT-based fibers that have increased stability, significant specific surface area and high porosity. In addition, the introduction of NC into the electrospinning process not only promotes the stabilization of aqueous CNT suspensions, but also improves the mechanical flexibility and electrical conductivity of the resulting electrospun nanofibers [88]. These advantages greatly enhance the performance of NC/CNT composites, especially in terms of their permeability and ability to remove contaminants and store energy [103].

### 3.1.3. Hybrid aerogels based on NC/CNTs

In addition to one-dimensional structures and thin films, NC and CNT-based composites are widely used in the form of hybrid aerogels. These aerogels retain a three-dimensional porous mesh structure, which improves the transport and diffusion of charge carriers (electrons and ions) in electrochemical reactions and mass transfer processes in water purification. This configuration is achieved through the efficient utilization of branching pores and channels [104]. The main difficulty in the development of porous aerogels using nanocellulose is the preservation of the porous structure during solvent removal, as this can induce capillary stress that can change the shape of the pores and cause them to collapse or deform. One common method for producing low density, high porosity aerogels based on NC/CNT composite is freezing followed by sublimation. For example, the TOCN/SCNT/PPy aerogel obtained by freeze drying has a specific surface area of 334 m<sup>2</sup>/g and a pore volume of 0.76 cm<sup>3</sup>/g, indicating high porosity with preservation of the structure of the original dispersion [12]. This method helps to prevent the formation of capillary pressure, which can damage the porous structure. In addition, freeze drying followed by carbonization allows the cellulose nanofibers, which have essential oxygen-containing functional groups, to successfully form hydrogen bonds with CNFs in the process of creating hybrid nanostructured aerogels (Fig. 3(d,e)). The FTIR curves clearly demonstrate the formation of hydrogen bonds between CNF and CNT chains, which is explained by the obvious shift of -OH, -CH, -COOH and C-O-C peaks towards lower wave numbers [105].

However, conventional methods for the preparation of aerogels (by chemical vapor deposition [106], hydrothermal methods [107], freeze drying [108]) mainly focus on optimizing impedance matching of carbon materials and have isotropic porous structures, which limits their other practical applications where directional porous structures are required. Whereas, the anisotropy of pores gives aerogel remarkable properties by controlling the morphology and size distribution of pores and pore channels as well as wall thickness [109]. In particular, Cong Wang et al. [110] by unidirectional freeze drying (UDF) method developed an anisotropic aerogel based on microcellular CNF-CNTs for use in "directional" strain gauges. Typically, the UDF technique involves slowly freezing a suspension of a two-component precursor by immersion in liquid nitrogen at a constant rate, leading to the separation of the solvent and dispersed phase, followed by freeze drying to remove the solidified dispersant, thereby creating an ordered structure. It is observed that aerogels obtained by UDF technology have morphology directly dependent on the precursor material. Also [110] reported that CNF cells transform into xylem-like microcellular aerogels after the UDF process, while the presence of CNTs imparts electrically conductive properties and also allows further blending with polymers due to the hydrophobic nature of carbon nanotubes [111].

Despite the above advantages of the lyophilization method, aerogel production is difficult to implement on a large scale as it requires additional energy and time and specialized equipment for the drying process. However, Zheng Huang et al. [112] propose a novel method to produce high-performance anisotropic aerogels on a large scale based on



CNFs and CNTs at room temperature and without chemical crosslinking. This strategy involves unidirectional cyclic repeated freezing and thawing to crosslink individual CNFs and CNTs to form a hybrid composite based on hydrophobic/hydrophilic nanofibers. It is observed that repeated freeze-thawing can strengthen the pore walls and align the structure of the anisotropic porous aerogel. The aerogels obtained by this method have increased porosity at the level of 98.1–98.6 %, ultra-low density, which is 0.0262–0.0296 g/cm<sup>3</sup> and sufficient mechanical strength, where at a strain of 75 % compressive stress was equal to 0.19–0.33 MPa.

Thus, the creation of NC/CNT-based composite materials is of considerable interest due to the expansion of practical possibilities and improvement of properties of the starting materials compared to their individual use. The efficiency of combining NC and CNT derivatives depends primarily on the chosen synthesis method. The choice of the optimal method affects the final properties of the composite material by considering various characteristics such as mechanical strength, electrical conductivity, porosity, and others. In addition to the possibility of controlling the desired material properties, the availability of raw materials, minimization of harmful waste during synthesis and the purpose of the end product application also plays an equally important role in the selection of a particular method for the production of NC/CNT nanocomposites.

## 3.2. Application of NC/CNT nanocomposites

### 3.2.1. Environmental applications of NC/CNT nanocomposites

**3.2.1.1. Water treatment applications of NC/CNTs composites.** Recently, the use of carbon nanomaterials, especially carbon nanotubes, in the processes of flocculation [113], adsorption [65] and catalysis of pollutants in aqueous media [114] has attracted interest. CNTs have outstanding mechanical properties, high aspect ratio and large specific surface area, which make them effective water purification agents. Nevertheless, their high cost and difficulty in regeneration after use can be a problem. Comparatively, NCs derived from an extensively distributed natural polymer also represent promising materials for water purification [115,116]. Unmodified NCs, depending on the production technology, may contain a variety of surface hydroxyl groups (-OH), sulfate or carboxylate groups (-COO-), making them effective for adsorption of cationic pollutants. While chemical modification by introducing positively charged amino groups allows the development of nanocellulose-based materials capable of adsorbing anionic dyes [117]. However, such modified nanocellulose adsorbents can no longer be used for cationic dyes. Process optimization can be achieved by combining carbon nanomaterials and nanocellulose, considering their different interactions with different molecules.

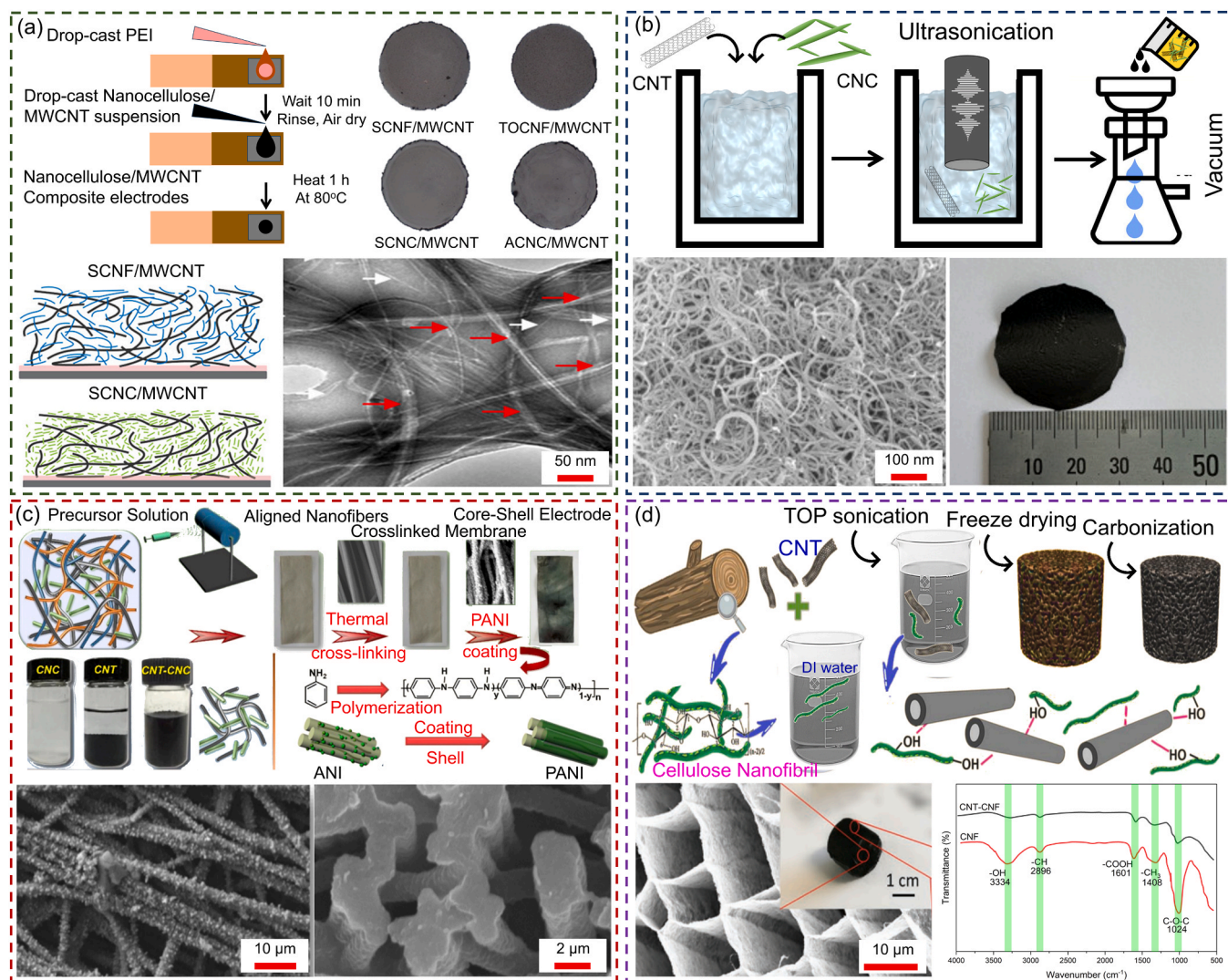


Fig. 4. Schematic illustration of preparation processes of NC/CNT composite and their characteristics and morphological images: drop-casting (a) [13], vacuum filtration (b) [93], electrospinning (c) [88] and freeze drying method (d,e) [105,110].

Earlier studies [90,105] also indicate a positive effect of CNFs on the dispersion of carbon nanotubes in aqueous solutions, which opens perspectives for the combined use of these nanomaterials to improve their adsorption characteristics and optimize costs. In particular, hybrid aerogels based on CNF and carbon nanomaterial (carbon nanotubes and graphene nanoplates) have adsorption capacity and adsorption rate close to CNF aerogels in cationic solutions of methylene blue and more efficient than CNF aerogels in anionic solution of Congo red [104]. It is also observed that these composites have high mechanical strength and the ability to recover their shape when exposed to water. The stabilizing effect of NC in the fabrication of highly conductive carbon nanofluids can produce paper films with excellent mechanical, crystalline and adsorption properties, which can be used for water filtration. For example, CNT/CNC-based nanocomposite buckypaper showed improved filtration and adsorption capacity for salt removal, which is 48 % more efficient than without CNC involvement [118] (Fig. 4). In addition, composite BPs have excellent potential for adsorption of dye molecules and  $Mn^{7+}$  ions from aqueous solution thus proving to be a potential competitor to conventional water treatment analogs. Furthermore, the possibility of multiple reuse by distillation and combustion of CNF/MWCNT-based ultralight and super porous aerogels as an effective material for separating various oil compounds from water has been reported [119].

Apart from adsorption and filtration, photocatalytic oxidation is the most promising method for water treatment of organic pollutants due to

its environmental friendliness and efficiency [120–123]. Despite the limitations of pristine nanocellulose such as low electrical conductivity, photocatalytic inertness and poor light permeability, other properties such as large surface area, ability to form frameworks, low cost and environmental friendliness are very attractive for photocatalysis applications in water treatment. Recently, Gallegos-Cerda et al. [124] developed a hybrid aerogel based on cellulose fibers functionalized with CNTs and  $TiO_2$ , which demonstrated photocatalytic removal of >90 % for rhodamine B and methylene blue under UV irradiation. In this composite structure, ultralight and highly porous aerogels with high mass transfer are used as carrier matrices, whereas the introduction of CNTs not only improves the physical and mechanical properties of the aerogel but also enhances the catalytic activity of the photocatalyst due to its electrical and optical properties.

**3.2.1.2. Pollutant sensing and antibacterial applications of NC/CNTs composites.** It is known that cellulose not only reduces the cost and makes electrochemical sensors environmentally friendly, but also increases their sensitivity and selectivity in the detection of analyzed substances [126,127]. The adhesion properties prevent the nanoparticles from leaching from the surface of the modified electrode, providing long-term stability. The large number of OH groups in the structure of nanocellulose can be modified for selective adsorption of different types of analytes [26]. To increase the electrical conductivity, biocompatibility, sensitivity, rapid electrochemical response and

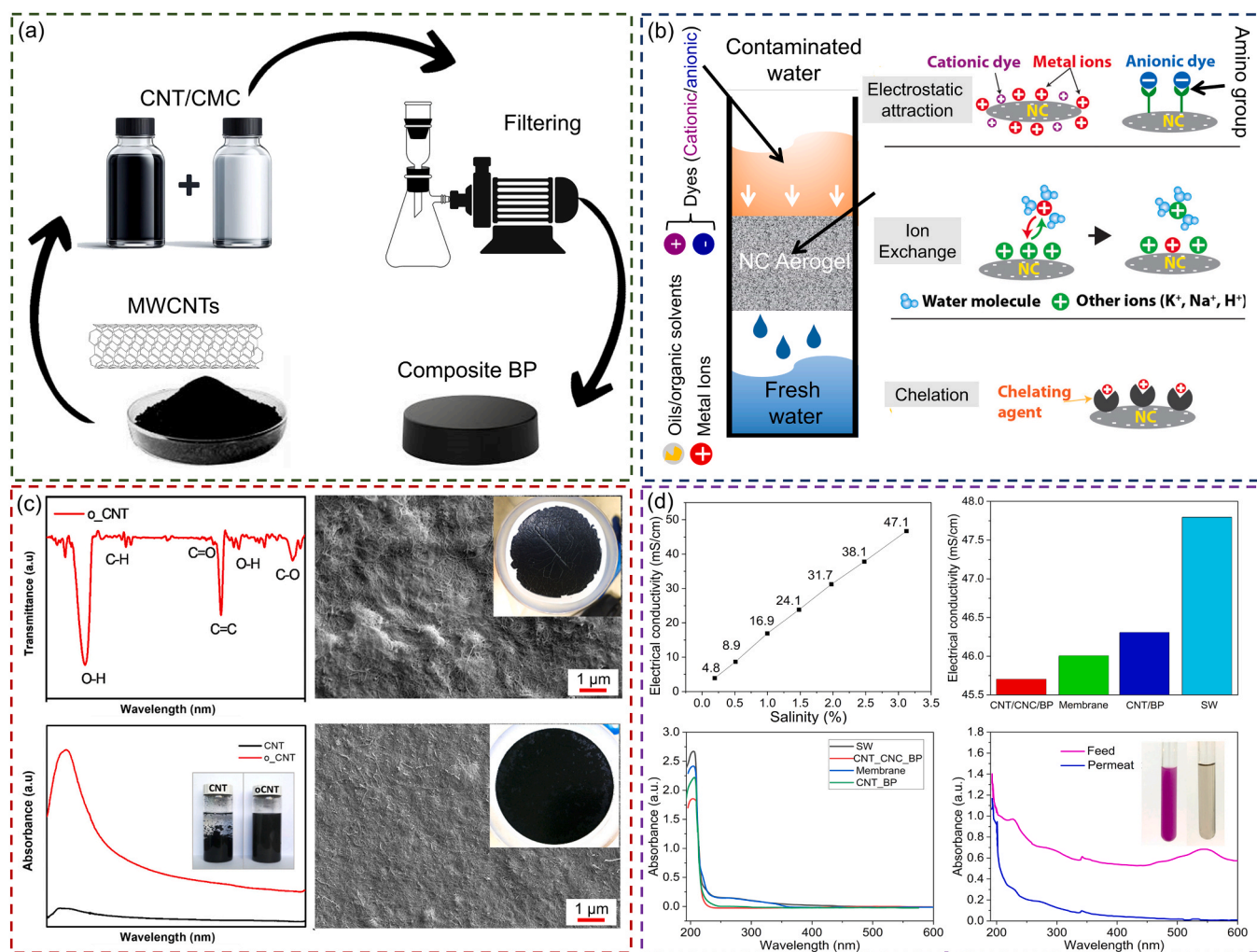


Fig. 5. Schematic illustration of preparation processes of NC/CNT composite (a) [118]; representation of adsorption mechanism of metal ions, dyes and oils (b) [125]; characteristics and morphological images (c) [118]; water filtration performance (d) [118].

expand the electroactive surface area, NCs are combined with various carbon nanomaterial compounds, including carbon nanotubes [128–131]. Moreover, various modifications of CNTs improve the ionic conductivity, providing efficient ion transport in electrochemical applications [132]. Recent studies confirm the effective use of NC/CNT composites for the detection of pharmaceuticals (paracetamol (PCM) [133], ciprofloxacin (CPR) [133], acetaminophen [132], diclofenac sodium (DFC) [26]) causing serious problems to human health and the environment (Fig. 5). In particular, Md Shalauddin et al. [133] have developed an electrochemical sensor based on NNC-PPY/SWCNTs nanocomposite with outstanding analytical response to simultaneously measure PCM and CPR concentrations in biological media, pharmaceutical drugs, and water samples. The sensor has a wide concentration range from 0.05 to 40  $\mu\text{M}$  for PCM and 1–50  $\mu\text{M}$  for CPR, and sensitivity in the range of 50.341  $\mu\text{A } \mu\text{M}^{-1} \text{ cm}^{-2}$  and 18.610  $\mu\text{A } \mu\text{M}^{-1} \text{ cm}^{-2}$ , respectively. In another study, Khan AAP et al. [126] showed that a nanocomposite sensor based on polypyrrole (PPy) combined with carboxymethylcellulose (CMC) and multi-walled carbon nanotubes (PPy/CMC/MWCNTs) is effective for monitoring heavy metal ions such as antimony cation  $\text{Sb}^{3+}$  in real conditions (tap water, well water, seawater and mineral water).

In addition, studies showed significant antibacterial activity of the synthesized PPy/CMC/MWCNTs sample against Gram-positive bacteria *B. subtilis* together with *S. aureus* (zone of inhibition  $22 \pm 0.50$  and  $19 \pm 0.25$  mm) and Gram-negative bacteria *P. aeruginosa* together with *Escherichia coli* (zone of inhibition  $18 \pm 0.25$  and  $17 \pm 0.5$  mm). The antibacterial properties of the composite can be explained by the presence of anionic compounds in the structure of the biopolymer during chemical modification with sodium carboxymethyl groups and combination with MWCNTs, which contain a high concentration of negatively charged sulfate functional groups [100]. Thus, the negatively charged surface of the composite provides electrostatic interaction with Gram-positive bacteria *B. subtilis* together with *S. aureus*, which explains the higher inhibition than against *P. aeruginosa* together with

*Escherichia coli*, which have negatively charged cell membranes [134]. In addition, cellulose partially substituted with ethyl cellulose (EC) in the structure of PUM/PDA@Ag-CNT based nanofiber membrane promoted shape recovery and imparted stability to the membrane during long-term use of antibacterial and conductive strain gauge [135]. At the same time, the uniform distribution of CNT combined with Ag on the EC-PUM surface provided excellent electro-sensitive and antibacterial properties.

Based on the above study, it can be concluded that nanocellulose and carbon nanotube-based composites are complementary with outstanding mechanical strength, low cost and extensive surface area. These characteristics make them promising for various applications such as filtration and adsorption of aqueous pollutants, photocatalysis, pollutant sensors and antibacterial effects. High stability due to strong ionic and hydrogen bonds has been observed. Nevertheless, to achieve more efficient environmental cleaning processes, more attention should be paid to fundamental research, especially in the field of the behavior of functional groups on the surface of NC/CNT composites. Recent performances and fabrication strategies of various NC/CNT based composites for water purification and pharmaceutical detection applications are collected in Table 1 and 2.

### 3.2.2. Energy applications of NC/CNT-based composites

The progress of modern society has stimulated an increasing demand for energy. In the last three decades, electrochemistry-based energy systems such as supercapacitors and rechargeable batteries have been widely recognized as power sources for portable gadgets and electric vehicles [142–144]. These technologies are characterized by high energy density, long lifetime and enhanced safety standards [145]. Supercapacitors, or electrochemical capacitors, represent a promising solution for electrochemical energy storage due to their high capacitance, long lifetime, and wide range of operating temperatures. Despite advances in thin, flexible and lightweight supercapacitors, their practical applications are limited by high raw material costs and complex

**Table 1**

Recent performances and fabrication strategies of various NC/CNT based composites for water purification applications.

N <sup>o</sup>	Nanocellulose	CNT	Other Additives	Composite fabrication technique	Performance	Application	Ref.
1	CNC	MWCNTs	-	Vacuum-assisted filtration	CNT/CNC buckypaper exhibited 48 % of salt rejection and excellent filtration and adsorption ability	Filtration and adsorption of dye molecules and $\text{Mn}^{7+}$ ion	[118]
2	TEMPO oxidized CNFs	CNTs	-	Sonication followed by freeze-drying	Adsorption capacities of CNF-CNT aerogels for MB and CR reached 1130.1 $\text{mg g}^{-1}$ and 381.4 $\text{mg g}^{-1}$ at initial dye concentrations of 500 $\text{mg L}^{-1}$ and 2000 $\text{mg L}^{-1}$ , respectively	Dye adsorption	[104]
3	CNFs	CNTs	Graphene	Hydrothermal method followed by freeze-drying	Adsorption capacities of aerogels for MB and rhodamine 6 G (R6G) reached 96.1 $\text{mg g}^{-1}$ and 79.2 $\text{mg g}^{-1}$ at initial dyes concentration of 15 $\text{mg/L}$ , respectively	Dye adsorption	[107]
4	NC	MWCNTs-NH <sub>2</sub>	Reduced graphene oxide (RGO)	Hydrothermal with freeze-drying and high-temperature annealing method	Adsorption performance for ethyl acetate (161.76 $\pm$ 7.69 $\text{g g}^{-1}$ ), dichloromethane (156.14 $\pm$ 8.24 $\text{g g}^{-1}$ ), cyclohexane (92.73 $\pm$ 5.22 $\text{g g}^{-1}$ ), acetone (103.87 $\pm$ 7.83 $\text{g g}^{-1}$ ), and sesame oil (124.09 $\pm$ 6.64 $\text{g g}^{-1}$ ), and an oil/water separation within 20 s	Oil and organic solvent adsorption	[136]
5	Cellulose aerogel	CNTs	TiO <sub>2</sub>	Freeze-drying	Photocatalytic removal of RB and MB (97 % and 93 %) occurred after 110 min of UV irradiation	Photocatalytic degradation of organic dyes	[124]
6	CNFs	MWCNTs	Chitosan	Impregnation and freeze-drying	The maximum adsorption capacity for copper ions was 46.05 $\text{mg g}^{-1}$ . No toxic substances were used in the cooking process	Adsorption of heavy metal ions	[137]
7	CMC	MWCNTs	PPy	Ultrasonication and in situ polymerization	Zone inhibition range $22 \pm 0.50$ and $19 \pm 0.25$ mm appeared against <i>S. aureus</i> and <i>B. subtilis</i> separately; $18 \pm 0.25$ and $17 \pm 0.5$ mm zone inhibition against <i>E. coli</i> and <i>P. aeruginosa</i> at 100 $\mu\text{g/ml}$ concentration	Antibacterial performance	[126]
8	Cellulose acetate	MWCNTs	Ag	Electrospinning	100 % bactericidal efficacy (0 % viable cells) against <i>E. coli</i> (b) <i>S. aureus</i> . within 48 hours.	Antibacterial performance	[138]

**Table 2**

Recent performances and fabrication strategies of various NC/CNT based composites for pharmaceutical detection applications.

N <sup>o</sup>	Nanocellulose	CNT	Other Additives	Composite fabrication technique	Performance	Application	Ref.
1	CNF	SWCNTs	Ppy	Drop-casting	Dynamic linear range of 0.05–40.0 $\mu\text{M}$ and 1–50 $\mu\text{M}$ for PCM and CPR; a limit of detection (LOD) of 0.072 nM and 0.196 nM; sensitivity of 50.341 $\mu\text{A } \mu\text{M}^{-1} \text{cm}^{-2}$ and 18.610 $\mu\text{A } \mu\text{M}^{-1} \text{cm}^{-2}$ , for PCM and CPR	PCM and CPR sensor	[133]
2	NC	f-MWCNTs	–	Drop-casting	Two linear dynamic ranges from 0.05 to 1.00 $\mu\text{M}$ and 2–250 $\mu\text{M}$ DCF with LOD of 0.012 $\mu\text{M}$	Diclofenac sodium (DCF) sensor	[26]
3	CMC	MWCNTs	Ppy	Ultrasonication and in situ polymerization	$\text{Sb}^{3+}$ ions sensor's sensitivity (22.4304 $\mu\text{A } \mu\text{M}^{-1} \text{cm}^{-2}$ )	Metal ion sensor	
4	NC	f-MWCNTs	Cu	Ultrasonication and drop-casting	A broad linear range of 0.01–80 $\mu\text{M}$ for ACOP and 0.05–80 $\mu\text{M}$ for DCF, and a LOD of 0.00014 $\mu\text{M}$ for ACOP and 0.00048 $\mu\text{M}$ for DCF. Sensitivity of 13.85 $\mu\text{A } \mu\text{M}^{-1} \text{cm}^{-2}$ and 4.142 2 $\mu\text{A } \mu\text{M}^{-1} \text{cm}^{-2}$ for the detection of ACOP and DCF, respectively	ACOP and DCF sensor	[132]
5	Sulfated CNFs	MWCNTs	Nafion	Sonication and drop-casting	A linear dopamine (DA) detection range of 0.05–100 $\mu\text{M}$ with LOD of 65 nM in PBS and 107 nM in interferent solution	DA detection	[139]
6	Lygands/NC	MWCNTs	$\text{Co}_3\text{O}_4$	Drop-casting	A wide linear range was achieved over the concentrations ( $10^{-13}$ M to $10^{-2}$ M), with LOD of $1.5 \times 10^{-13}$ M	Cadmium ion sensor	[140]
7	Sulfated CNCs	MWCNTs	–	Sonication and drop-casting	The linear range of 0.1–100 $\mu\text{M}$ DA detection concentrations; LOD of 176 nM	DA, PCM and uric acid detection	[141]
8	Sulfated CNFs	MWCNTs	–	Sonication and drop-casting	The linear range of 0.1–100 $\mu\text{M}$ DA detection concentrations; LOD of 250 nM	DA, PCM and uric acid detection	[141]
9	TEMPO oxidized CNFs	MWCNTs	–	Sonication and drop-casting	The linear range of 0.1–100 $\mu\text{M}$ DA detection concentrations; LOD of 250 nM	DA, PCM and uric acid detection	[141]

manufacturing processes. Thus, there is a need to develop high-performance supercapacitors using new, stable, renewable and more affordable materials. Among many materials, nanocellulose is the best candidate for use in the fabrication of modern supercapacitors because of its significant advantages.

In the development of NC-based electrochemical energy storage sources, the following features of this material are highlighted: (1) excellent mechanical properties with the possibility of being used as a high-strength material for electrodes and separators [146,147]; (2) the high content of hydroxyl groups on the NC surface makes it possible to chemically modify and combine with active agents to improve electrochemical properties [23]; (3) the high aspect ratio of CNFs, presenting a spiderweb-like structure, suitable for the fabrication of nanocellulose nanocapacitors; (4) the high specific surface area, porosity control and ease of processing enables efficient electrode separation and at the same time easy ion transport for further use in thermostable nanoporous separators [147]; (5) the relatively high presence of carbon in NC components allows efficient combination with carbon nanomaterials (graphene, CNTs, nanodiamonds) for use in creating high-performance electrodes in electrochemical energy storage devices [148]; and lastly, (6) nanocellulose is an abundant and green material for hybrid building blocks of energy storage devices, which can obtain the desired structure and chemical properties by applying multiple fabrication techniques [149]. Moreover, the NC exhibits impressive electrolyte absorption capacity across a broad potential range, making it a strong contender for energy storage purposes [27].

Considering the above advantages, the NC structure is mainly considered as a matrix for binary nanocomposites. However, NC itself is not a conductive material. Generally, to achieve superior conductivity for supercapacitor electrodes, NC is used as a carbon precursor or NC is combined with conductive materials such as conductive polymers and/or carbon nanomaterials [150]. Despite significant advances in the development of NC-based carbon electrodes, the low bulk density, which is less than 0.8 g/cm<sup>3</sup>, causes problems due to inefficient utilization of the porosity of the structure and reduced energy density [27]. In addition, the complex trade-offs between heteroatom doping and electrical conductivity and the mismatch between electrode and electrolyte are further challenges to be overcome to improve the

performance of NC-based carbon electrodes. Whereas the synergy of the best properties of NCs and conductive carbon nanomaterials can solve the conductivity problems and impart additional mechanical strength, flexibility and high active center area. Among carbon nanomaterials, CNTs with outstanding specific surface area, electrical conductivity and electrochemical properties represent an attractive electrode material for supercapacitors [12,151,152]. Studies emphasize the outstanding characteristics of one-dimensional CNTs such as high tensile strength, Young's modulus and electrical conductivity [48,150,152]. Compared to two-dimensional carbon nanomaterials, the presence of a one-dimensional structure with a high length-to-diameter ratio at the nanometer scale provides high flexibility of the CNT layer on substrates, creating long continuous conducting pathways [153,154]. In addition, the geometry of CNTs allows conformal coatings on various surfaces, providing high integrity [48].

**3.2.2.1. High-performance supercapacitors based on NC/CNTs composites.** In recent years, interest in NC and CNT composites has increased due to their potential for energy applications. The unique properties of CNTs make them an excellent choice for imparting electrical conductivity to non-electrically conductive nanocellulose templates through coating or blending processes. While simple coating with carbon nanomaterials provides NC films with higher conductivity than polymer films, increasing conductivity by increasing the thickness or number of coatings is often ineffective due to poor adhesion of carbon particles. This problem leads to the delamination phenomenon [150]. Unlike surface coatings, blending allows more carbon particles to penetrate the substrate because the NC substrate physically holds numerous particles. By filling nanocellulose templates with a continuous network of carbon materials, conductive composites with high electrical conductivity and tensile and bending stability can be created. The uniform mixing of CNTs or graphene with nanocellulose fibers used as a matrix promotes channel expansion and allows unimpeded ion transport, thereby solving the problem of low dispersibility of carbon nanomaterials arising from hydrophobic properties [12,83]. This, in turn, gives the supercapacitor an excellent charging and discharging rate and helps to reduce the volume change of the electrode materials in various pseudocapacitive reactions.

In addition, the combination of CNTs and NCs can significantly enhance not only the conductive properties of the composite, but also show excellent mechanical properties. In particular, the mechanical properties of uniformly dispersed TEMPO oxidized NC and CNTs in pigskin (PS) hydrogel were improved five times compared to pure PS hydrogel, while the specific capacitance was  $\sim 65 \text{ F g}^{-1}$  and the capacitance retention was nearly 60 % after 2000 cycles [155].

To increase the specific capacitance in energy storage systems based on modified CNTs and NCs, in most cases, blends with conducting polymers, which have a specific capacitance 3–7 times higher compared to carbon materials, are used [12]. In such hybrid electrodes, CNTs, which have innate high conductivity and mechanical stability, are considered as a material that contributes to electrode stability, since conducting polymers are limited by poor cyclic stability during the charge-discharge process. On the other hand, nanocellulose possesses carboxyl and hydroxyl links that can directly interact with the functional groups of the conductive polymer and, in addition, nanocellulose has the required components with relatively large amounts of carbon, which provides a close affinity between the two materials and serves as an excellent binding material. For example, a solid-state supercapacitor assembled from PANI@CNT-CNC/PVA-PAA nanofiber-based electrodes exhibits a capacitance of  $155.5 \text{ F g}^{-1}$  at  $0.5 \text{ A g}^{-1}$  [88]. In addition, it exhibits remarkable capacitance retention characteristics of 92 %, 90 %, and 89 % of its original value after 2000 charge/discharge cycles under plane strain, bending, and twisting, respectively (Fig. 6a). Such excellent results are attributed to the excellent synergy of polymer material,

nanocellulose and CNTs combined to form a composite with unique architecture. In another study [156], a CNF/CNT composite based on PPy polymer exhibited a higher capacitance of  $200.8 \text{ F g}^{-1}$  at  $0.5 \text{ A g}^{-1}$  with a remarkable capacitance retention of 90 % after 5000 cycles at  $1 \text{ A g}^{-1}$ . Furthermore, layer-by-layer (LbL) assembly of various carbon nanomaterials and conducting polymers represents an innovation in the field of electrochemistry. In such systems, the use of porous nanocellulose-based aerogels as a matrix allows the electroactive material to penetrate inside its porous structure greatly enhancing the electrochemical performance of the electroactive material, which is almost impossible to achieve in the case of membrane-like substrate materials.

A study by Shaoyi Lyu et al. [157] presents CNC and wood pulp fibers (WPF) aerogels for LbL assembly of polyaniline (PANI) and CMWCNTs or graphene oxide (GO) in clean energy storage devices. The efficient nanoarchitecture with extensively interconnected channels and electrode hydrophilicity in WPF/CNC-(PANI/CMCNT)10 and WPF/CNC-(PANI/GO)10 composites provided high specific capacitance ( $716.62$  and  $636.63 \text{ F g}^{-1}$ , respectively), and the energy densities of the assembled supercapacitors were  $168.64$  and  $113.57 \text{ mWh}\cdot\text{cm}^{-2}$ . In a similar study, Shaoyi Lyu [154] used CNF aerogels with a network of interwoven nanofibers to assemble electroactive materials, which resulted in a larger specific surface area of the aerogel and outstanding electrochemical performance, significantly improving the flexibility of the electrode. The specific capacitance of CNF-[PANI/CMWCNT]10 and CNF-[PANI/RGO]10-based electrodes were  $965.80$  and  $780.64 \text{ F g}^{-1}$ ,

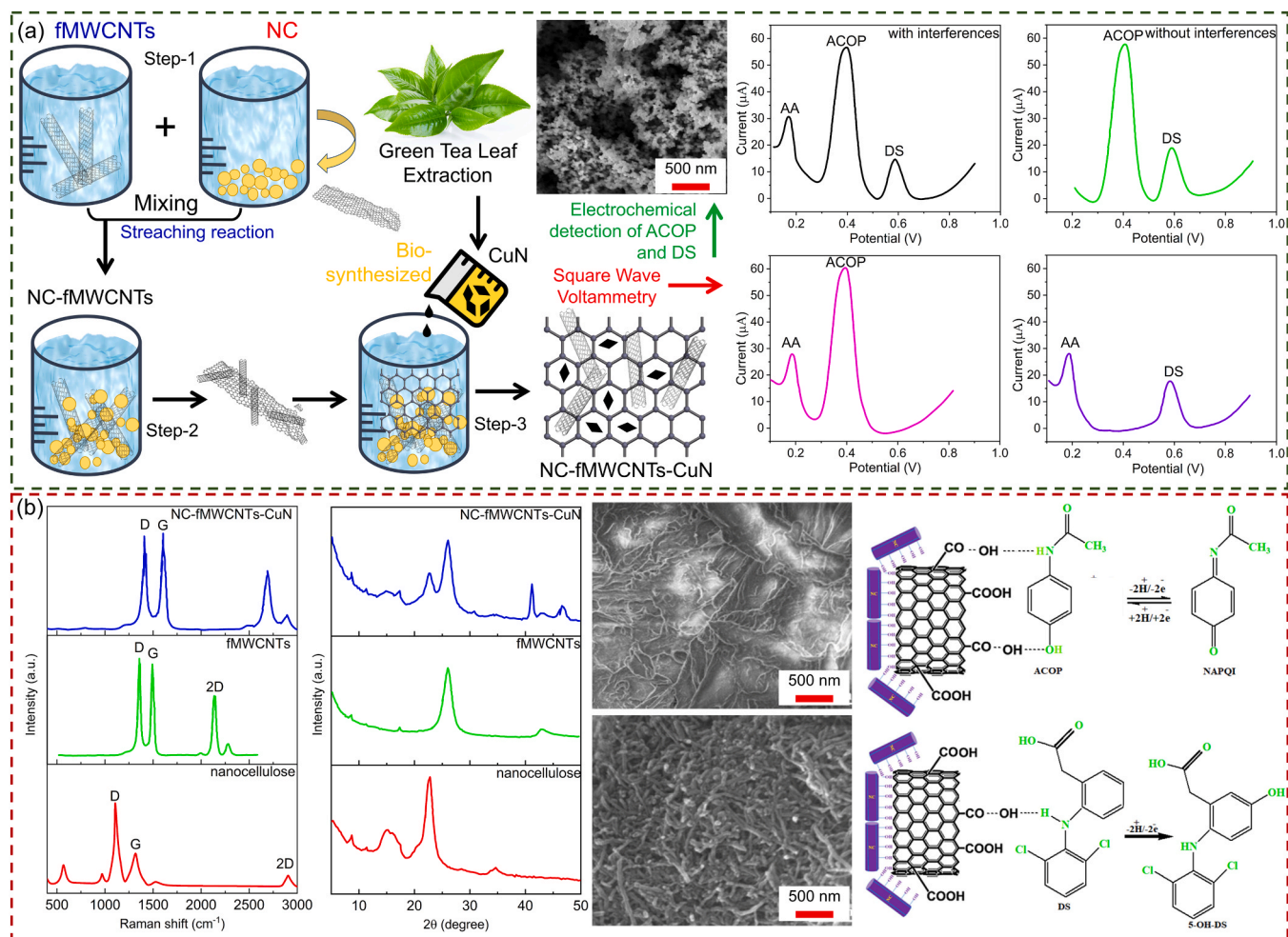


Fig. 6. Schematic illustration of preparation processes (a); SWV response to acetaminophen (ACOP) and DS (b) characteristics (c); and proposed sensing mechanism of ACOP and DS of NC-fMWCNTs-CuN/GCE electrode [132].

respectively, and the area specific energy for the symmetrically assembled supercapacitors were 147.23 and 112.32 mWh·cm<sup>-2</sup> (Fig. 6b). By analyzing the results of microstructure and electrochemical curves in both studies, it can be concluded that the electrode composite using CMWCNTs has a more developed specific surface area and better electrolyte accessibility, due to its unique homogeneous structure, compared to the GO-based electrode. This is a more favorable condition for ion diffusion and electron transfer, which affects the electrochemical properties of the aerogel electrode.

There are also known cases of combining CNTs with graphene oxide and nanocellulose to create highly porous electrode materials with excellent electrical and mechanical properties desirable for the production of flexible solid-state supercapacitors [158,159]. Since, graphene oxide nanosheets possess a rich arsenal of oxygen atoms this helps them to easily form a homogeneous solution when mixed with CNFs (Fig. 6c). In the CNF-CNT/GO system, graphene oxide acts as a "surfactant" to directly disperse the primary CNTs, while CNFs improve the conductivity by providing continuous conductive pathways between the layers of GO nanosheets and CNFs prevent agglomeration of both hydrophobic carbon nanomaterials in aqueous suspension. In addition, CNF further improves the mechanical properties of the whole composite [160].

**3.2.2.2. Rechargeable batteries based on NC/CNTs composites.** In addition to supercapacitors, composites with the presence of NC and CNTs can also be used in rechargeable batteries such as lithium-ion batteries (LIBs) [161], zinc-ion batteries [162], zinc-air batteries (ZAB) [163], and lithium polysulfide batteries (LiPS) [164] (Fig. 8). In recent study [165], NC/CNT-based materials with porous structure, have been found to have robust mechanical properties and high electrochemical stability. These materials have been successfully applied in various types of rechargeable batteries where they are used as key components of electrodes, solid electrolytes and separation membranes and show significant potential for the development of batteries with improved electrochemical properties and durability. In particular, Shaomei Cao et al. [161] presented a hierarchical MoS<sub>2</sub>/CNF/CNTs-based hybrid film for use as a paper electrode in LIBs, while eliminating the traditional use of a metal current collector. This property makes the presented material promising for possible future applications in flexible and wearable electronics. In this study, TEMPO-oxidized CNFs were used in a liquid-phase peeling process in aqueous medium to exfoliate the MoS<sub>2</sub> monolayer. This method shows high efficiency in preventing the aggregation of two-dimensional MoS<sub>2</sub> nanosheets into volume during cycling and subsequent drying of the electrodes. In addition, the TEMPO-oxidized CNFs act as a biological binder and act as a fiber-like framework in the formation of the hierarchical MoS<sub>2</sub>-based hybrid film. The presence of CNTs in the composite provides a dual function of preventing the aggregation of MoS<sub>2</sub> nanosheets and serving as an electrical channel to accelerate the transport of charge carriers, thus improving the overall electrical conductivity. The next step, namely carbonization, further increases the electrical conductivity and improves the porosity of the mesh structure, providing the paper electrode with high specific capacitance, stability and efficiency over long LIBs use cycles.

CNF/CNT-based composites are also used in rechargeable zinc-ion batteries as an alternative to current LIBs. For example, the cathode electrode presented by SWCNT/CNF/MnO<sub>2</sub> composite with a unique architecture of a three-dimensional two-continuous heterofibrous network scaffold (HNS) exhibited enhanced performance and stability in cycling [162]. It should be emphasized that the structure and aggregation of the cathode-active material remain inaccessible to destructive effects. The gravimetric energy and power performance based on the electrode sheet is also significantly higher than that traditionally achieved with standard electrodes (up to 91 Wh kg<sub>electrode</sub><sup>-1</sup> and 1848 W kg<sub>electrode</sub><sup>-1</sup>, respectively). The new HNS strategy successfully

disperses MnO<sub>2</sub> particles in the composite with CNFs, allowing electrolyte permeability as well as high electrical conductivity of SWCNTs, thus accelerating redox kinetics without the need for a metal foil current collector.

Flexible and thin composite papers based on renewable CNFs and CNTs also find wide applications in flexible zinc-air battery electrodes, which are promising candidates for integration into wearable electronic devices considering their outstanding theoretical energy density estimated at 1086 Wh·kg<sup>-1</sup>. The entanglement between CNFs and CNTs effectively improves the mechanical performance of the material, including tensile strength (7.47 ± 0.74 MPa) and fracture strain (1.67 ± 0.12 %), providing outstanding bending stability [163]. The integration of silver (Ag) nanoparticles with high electrical conductivity and significant active surface area-to-volume ratio significantly enhances the activity of the oxygen reduction reaction (ORR) on the ZAB air cathode and further improves the electrical conductivity (Fig. 7b). Based on this approach, researchers have developed CNT/Ag/CNF composite paper as an air cathode for ZAB, exhibiting an excellent power density of 70 mW·cm<sup>-2</sup>. This value is significantly higher than that for ZAB using Pt/C (59 mW·cm<sup>-2</sup>) [163].

Moreover, researchers [164] have observed that highly conductive MWCNT and CNF-based papers for the cathode of lithium polysulfide batteries suffer a rapid loss of columbic efficiency and capacity due to a phenomenon known as the "shuttle effect". This effect leads to unexpected damage to lithium anodes and causes the deposition of solid insulating blocks such as Li<sub>2</sub>S<sub>2</sub>/Li<sub>2</sub>S or S<sub>8</sub>, which blocks the electrode and affects the inefficient utilization of sulfur during cycling. However, the introduction of a LiPS-like material such as NiCo<sub>2</sub>S<sub>4</sub> into the MWCNT/CNF composite maximizes the utilization efficiency and achieves excellent capacitance retention at both low and high currents.

Supercapacitors and rechargeable batteries based on NC/CNT composites are characterized by high conductivity, mechanical properties, stability, and capacitance. NC is an unrivaled material for creating aerogels with a unique porous hierarchical architecture, allowing the electroactive material to penetrate inside its porous structure significantly enhancing the electrochemical performance of the electroactive material, both for supercapacitors and rechargeable batteries. Whereas the presence of CNTs in the composite, in addition to improving the mechanical properties, serves as an electrical channel for accelerating the transport of charge carriers, which helps to improve the overall electrical conductivity. At the same time, the addition of polymers to such systems further improves capacitive properties, while the combination with graphene oxide improves strength and conductivity. Electrochemical performances and fabrication strategies of various NC/CNT based composites for energy storage devices are collected in Table 3 and 4

### 3.2.3. Other applications of NC/CNT nanocomposites

The additional areas include electromagnetic protection, temperature control, and biomedical applications. The extraordinary properties of nanocellulose, such as its biocompatibility, biodegradability, and impressive mechanical strength, combined with the excellent electrical conductivity and thermal stability of carbon nanotubes, open new possibilities for multifunctional materials. In electromagnetic shielding, for instance, hybrid nanocomposites provide efficient absorption and reflection of electromagnetic waves, which is crucial for protecting sensitive electronic equipment and reducing electromagnetic interference in various devices. For temperature control, these composites can be engineered to achieve precise thermal management, essential in electronic devices, energy storage systems, and wearable technology. In biomedical applications, the synergy between nanocellulose's compatibility with biological tissues and CNT's superior electrical properties can lead to innovative solutions for biosensors, drug delivery systems, and tissue engineering scaffolds. The integration of these materials creates a platform for developing advanced technologies that address current challenges in multiple high-impact fields.

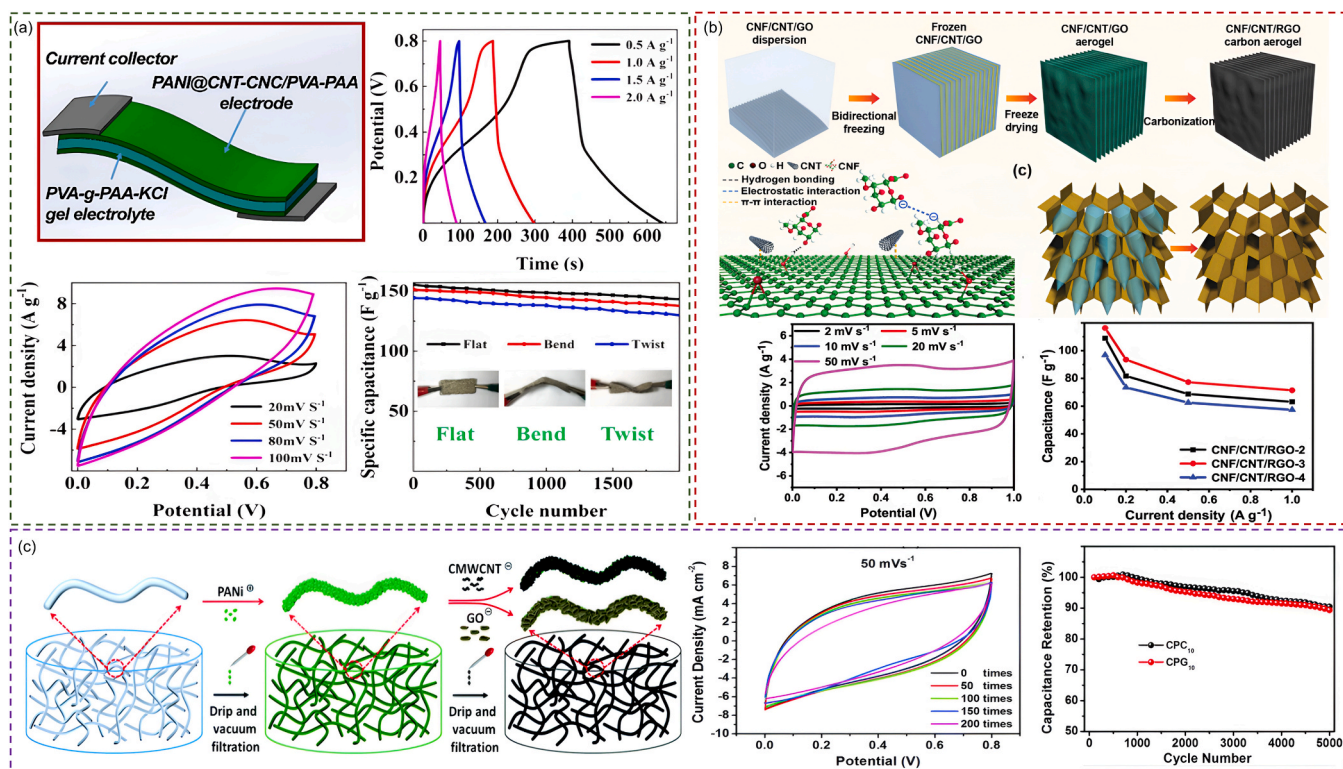


Fig. 7. Electrochemical performance of (a) PANI@CNT–CNC/PVA–PAA nanofibrous electrodes. Reproduced with permission [88]. Copyright 2019, American Chemical Society; (b) CNF/CNT/RGO Carbon aerogels electrode Reproduced with permission [154] Copyright 2019, The Royal Society of Chemistry; (c) LbL assembled CNF-[PANI/CMWCNT]10 electrode Reproduced with permission [158]. Copyright 2022, John Wiley and Sons.

**3.2.3.1. Electromagnetic shielding.** With the increasing number of electronic devices, there is a growing need for effective electromagnetic interference (EMI) shielding to prevent signal disruption and protect sensitive components. Nanocellulose and carbon nanotube (CNT)-based hybrid nanocomposites exhibit excellent electrical conductivity and high aspect ratios, making them ideal for EMI shielding. These composites leverage the conductive properties of CNTs and the structural integrity of nanocellulose to effectively attenuate electromagnetic waves (Fig. 9a). Research has demonstrated that such composites can provide protection comparable to traditional materials while offering the advantages of flexibility and lightness [15,169–171]. Potential applications include the shielding of electronic devices, communication systems, and protective clothing for workers exposed to electromagnetic fields. The integration of nanocellulose and CNTs creates a synergistic effect, enhancing the overall performance of the material and making it a promising candidate for next-generation EMI shielding.

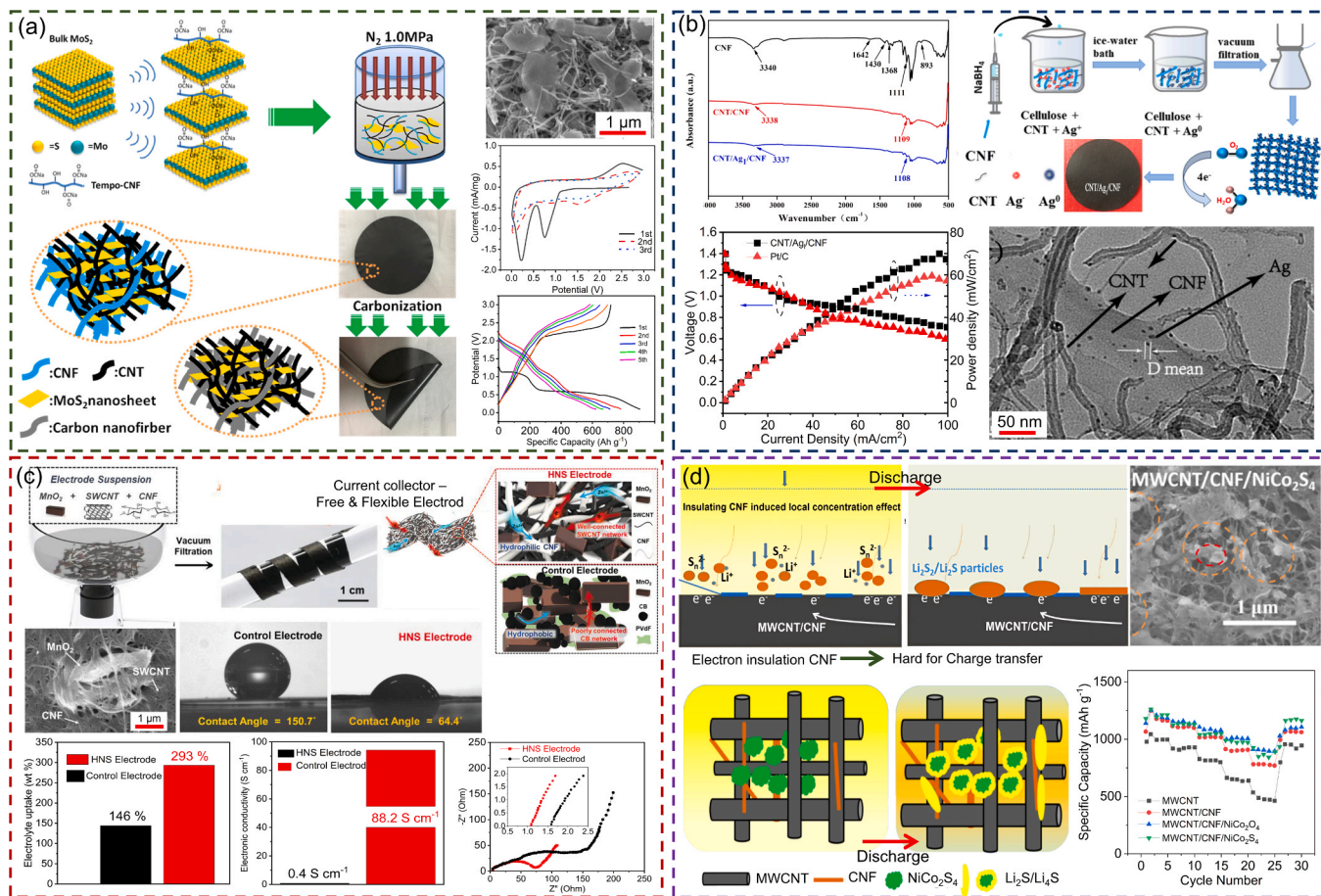
CNF/MWCNT hybrid composites fabricated by vacuum filtration and hot-pressing exhibit an EMI shielding efficiency (SE) of up to 45.8 dB at X-band with a film thickness of only 0.15 mm. These composites also demonstrate high mechanical strength, reaching up to 48 MPa, which is superior to many other MWCNT-based materials [172]. Achieving high EMI SE values and good mechanical properties necessitates a uniform distribution of MWCNTs within the CNF matrix. The use of TEMPO-oxidized cellulose nanofibrils (OCNFs) enables a stable MWCNT dispersion without the need for chemical functionalization or surfactants, promoting the formation of strong and flexible composite films with high electrical conductivity up to 3024 S/m and low thermal conductivity. These properties make the composites highly promising for flexible electronics and wearable device applications [172].

Another promising area is the use of 3D printing to create lightweight and high-performance EMI shielding materials. MWCNT and OCNF-based inks for direct printing (DIW) enable the creation of porous structures with arbitrary architectures. These porous frameworks

exhibit ultralight structure ( $0.11 \text{ g/cm}^3$ ) and high EMI SE up to 43.5 dB at X-band. 3D printed frameworks with hierarchical pores exhibit perfect EMI, which makes them promising for application in advanced electronic devices [173]. Morphological analysis shows that CNF and MWCNT-based composites possess a dense and consistent layered structure, which contributes to high electrical conductivity and improves mechanical properties. These composites have a specific surface area up to  $106.2 \text{ m}^2/\text{g}$  and a porous structure, which provides high EMI shielding efficiency. In addition, they exhibit high flexibility and strength, making them suitable for use in a variety of applications requiring lightness and high performance [174]. Thus, hybrid nanocellulose and carbon nanotube-based nanocomposites offer new opportunities for the development of innovative solutions in electronics and EMI shielding, providing high performance and robustness in real-world applications.

**3.2.3.2. Biomedical applications.** Nanocellulose (NC) and carbon nanotube (CNT) hybrid nanocomposites show significant potential for biomedical applications due to their unique structural and morphological features [141,175–177]. For example, biopatches made with carbon nanotubes and bacterial nanocellulose (BNC) can restore electrical conductivity in areas of the myocardium disrupted by damage or scarring. Studies in dogs have shown that the use of biopatches can restore conduction after surgical incisions. Electroanatomical mapping demonstrated that conduction velocity at the center of the incision increased from  $1.1 \pm 0.6 \text{ m/s}$  to  $2.7 \pm 0.4 \text{ m/s}$  after patch application, which was almost the same as the initial velocity before the incision ( $2.5 \pm 0.6 \text{ m/s}$ ) [178]. The flexibility and electrical conductivity of these biopatches allow them to function effectively in a dynamically changing heart shape.

These properties are important for other applications such as the development of tactile sensors. Sensors utilizing the principle of the piezoresistive effect to measure pressure and the thermoelectric effect to



**Fig. 8.** Recent applications of NC/CNT composite in different types of rechargeable batteries: LIBs (a) [161]; ZAB [163] (b); zinc-ion batteries [162] (c); and LiPS (d) [164].

**Table 3**

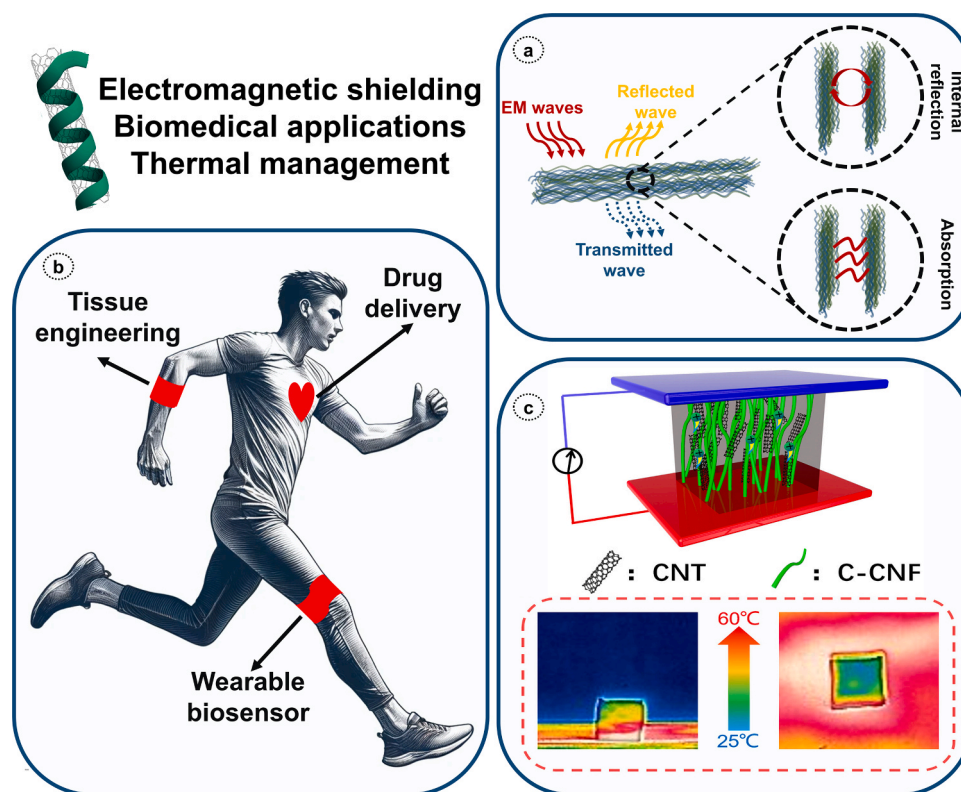
Electrochemical performances and fabrication strategies of various NC/CNT based composites for supercapacitors and energy storage devices.

N <sup>o</sup>	Nanocellulose	CNT	Other Additives	Composite fabrication technique	Electrochemical performance	Application	Ref.
1	CNF	MWCNTs	Ppy	Ultrasonication, followed by in situ polymerization	200.8 F g <sup>-1</sup> with 90 % after 5000 cycles at 1 A g <sup>-1</sup>	Supercapacitor electrode	[150]
2	CNC	CMWCNTs	Wood pulp fiber and PANI	LbL assembly technique	716.62 F g <sup>-1</sup> ; high areal specific energies 168.64 mW h cm <sup>-2</sup> ,	Supercapacitor electrode	[157]
3	CNF	CMWCNTs	PANI	LbL assembly technique	965.80 F g <sup>-1</sup> ; 1.59 F cm <sup>-2</sup> ; 147.23 mW h cm <sup>-2</sup>	Supercapacitor electrode	[154]
4	TEMPO-oxidized CNF	Sulfonated CNT	PPy	Fe <sup>3+</sup> in-situ oxidation, followed by solvent-exchanging and freeze-drying	5299 mF/cm <sup>2</sup> at 1 mA/cm <sup>2</sup> ; 163.2 % capacitance retention after 2500 cycles (at 1 mA/cm <sup>2</sup> )	Solid-state supercapacitors	[12]
5	CNC	CNTs	PVA, PAA and PANI	Directional electrospinning, followed by a thermal treatment and in situ polymerization of PANI	Tensile strength of ~54.8 MPa, and a favorable electroconductivity of ~0.44 S m <sup>-1</sup> ; 164.6 F g <sup>-1</sup> with 91 % after 2000 cycles	Solid-state supercapacitor	[88]
6	CNF	CNTs	rGO	Bidirectional freezing and subsequent annealing	Ultralow density (2.64 mg cm <sup>-3</sup> ) and superhydrophilicity (water contact angle ≈0° at 106 ms); 109.4 mF cm <sup>-2</sup> at 0.4 mA cm <sup>-2</sup> ; 88 % after 5000 cycles	Solid-state supercapacitors, wearable electronics, electronic skin	[158]
7	Cladophora cellulose	CNT	V <sub>2</sub> O <sub>5</sub> · H <sub>2</sub> O nanosheets	Sonication, followed by vacuum filtration	250 C g <sup>-1</sup> at 1 A g <sup>-1</sup> ; 85 % after 8000 cycles	Energy storage device	[87]
8	Cationic CNF	MWCNTs	-	Sonochemical treatment	Films with adjustable anisotropic thermal and electrical conductivity	Biomedical and energy device	[90]
9	TEMPO-oxidized CNF	MWCNTs	Pigskin hydrogel	Sonication, followed by freeze-drying	~65 F/g with 60 % after 2000 cycles	Flexible sensor and supercapacitor electrode, electronic skin	[155]



**Table 4**  
Electrochemical performances and fabrication techniques of various NC/CNT composites for rechargeable batteries.

N <sup>o</sup>	Nanocellulose	CNT	Other Additives	Composite fabrication technique	Electrochemical performance	Application	Ref.
1	TEMPO-oxidized CNF	CNTs	MoS <sub>2</sub> nanosheets	In situ carbonization	Specific capacity (930.0 mAh·g <sup>-1</sup> ), nice rate capability, and longtime cycling stability	Lithium-ion batteries	[161]
2	CNC	SWCNTs	–	Ultrasonication, vacuum-drying, followed by carbonization	Specific capacity of 443 mAh·g <sup>-1</sup> at a current density of 232 mA·g <sup>-1</sup> after 120 cycles with the capacity retention of 89 % and high-rate capability	Lithium-ion batteries	[166]
3	CNF	SWCNTs	MnO <sub>2</sub> particles	Ultrasonication, vacuum-assisted filtration, followed by freeze-drying	((129 mAh g <sup>-1</sup> <sub>MnO2</sub> at 20 C (vs 78 mAh <sub>MnO2</sub> <sup>-1</sup> for the control electrode)) and cyclability (capacity retention after 450 cycles at charge/discharge current density of 2 C/2 C = 77 %	Aqueous Zn-Ion Batteries	[162]
4	CNF	CNTs	Zn	Ultrasonication, vacuum-assisted filtration, followed by freeze-drying	Specific capacity of anode at 0.2 A g <sup>-1</sup> is 208.4 mAh g <sup>-1</sup> . Anode delivers 89.5 mAh g <sup>-1</sup> at the 1000th cycle, corresponding to 95.4 % and 90.1 % of the capacity at the first and second cycles, respectively	Aqueous Zn-Ion Batteries	[167]
5	CNF	CNTs	Ag nanoparticles	Liquid-phase reduction method	Excellent conductivity up to 293.17 ± 32.8 S·m <sup>-1</sup> ; high tensile strength of 7.47 ± 0.74 MPa, a fracture strain of 1.67 ± 0.12 %; an excellent power density of 70 mW·cm <sup>-2</sup>	Zinc-Air Batteries	[157]
6	CNF	Dual-Phase MWCNT	NiCo <sub>2</sub> S <sub>4</sub>	Ultrasonication, vacuum-assisted filtration, followed by freeze-drying	Specific capacity of 1154 mAh g <sup>-1</sup> at 0.1 C and a high-rate performance of 963 mAh g <sup>-1</sup> at 1 C; high electronic conductivity (above 50 S cm <sup>-1</sup> )	Li-S batteries	[164]
7	TEMPO-oxidized CNF	CNTs	Sulfur	Ultrasonication, vacuum-drying, followed by freeze-drying	The initial specific capacity could be up to 1168 mAh g <sup>-1</sup> at 0.1 C and the capacity remained 86 % after 120 cycles at 0.5 C.	Li-S batteries	[168]



**Fig. 9.** Recent applications of NC/CNT composite in other fields: electromagnetic shielding (a); biomedical applications (b); and thermal management (c) [14].

measure temperature show high sensitivity and stability. The use of nanocellulose as a substrate provides flexibility and environmental friendliness to the sensors, making them promising for use in wearable electronic devices [179].

In addition, hybrid nanocomposites based on cellulose nanofibrils (CNFs) and multi-walled carbon nanotubes (MWNTs) exhibit high biocompatibility and mechanical strength, making them promising for use in bone tissue regeneration (Fig. 9b). These composites support the viability, adhesion, and proliferation of osteoblasts as evidenced by a

25 % increase in proliferation rate compared to control samples [180]. High mechanical properties are also important for the development of strain sensors that demonstrate high sensitivity and stability under repeated strain cycles. Such sensors have been successfully used to monitor human body movements such as finger and hand flexion, accurately recording cyclic changes in resistance in response to movements [181].

An important application of nanocellulose is the development of hydrogels for wound healing. Bacterial nanocellulose (BNC)-based

hydrogels are highly adhesive, biocompatible, and able to retain a significant amount of fluid, which promotes rapid wound healing and prevents infections. Studies have shown that these hydrogels can effectively stop bleeding and promote tissue regeneration, as evidenced by a reduction in clotting time of up to 25 minutes in vivo experiments [84].

Hybrid nanocomposites based on carbon nanotubes and nanocellulose have high mechanical, electrically conductive, and functional properties, making them promising for use in shape memory smart materials and strain sensors. For example, strain sensors based on TPU/CNF-C/CNT composites exhibit a change in relative resistance ( $\Delta R/R_0$ ) under strain of up to 40 % of 1.93, remaining stable for more than 1000 cycles [181]. Thus, hybrid nanocellulose and carbon nanotube-based nanocomposites exhibit high performance and multifunctionality in various biomedical applications. These materials possess unique structural and morphological features that provide improved physicochemical and biological properties, including high electrical conductivity, mechanical strength, flexibility, and biocompatibility. These properties open new perspectives for the development of innovative medical solutions such as biopatches for myocardial conduction restoration, tactile sensors, bone regeneration materials, and hydrogels for wound healing.

**3.2.3.3. Thermal management.** Efficient temperature management is crucial in the electronics, automotive, and aerospace industries to prevent overheating and ensure optimal performance. Due to their high thermal conductivity and stability, hybrid composites such as CN/CNTs are promising components for use in thermal interface materials, heat sinks, and thermal barriers. Research indicates that these composites can improve the performance and durability of electronic components by providing efficient heat transfer pathways [182–185]. The ability to tune the thermal properties of these composites through controlled synthesis and functionalization makes them adaptable to various temperature control tasks.

In particular, aerogels based on carbon nanotubes and cellulose nanofibers (CNFs) have shown compressive strengths of up to 152 kPa at 90 % strain and low thermal conductivity ranging from 0.03 to 0.08 W/mK, making them promising for thermal management and heat harvesting [14]. Paraffin-stabilized composites showed thermal conductivities in the range of 0.1–0.35 W/(m·K), which is significantly higher compared to pure paraffin, while retaining their shape and properties even after 100 melting/freezing cycles, confirming their high thermal stability and reliability for solar thermal energy harvesting and storage applications [186]. In another study, composites based on sulfated nanocellulose (SNC) and single-walled carbon nanotubes (SWCNTs) achieved ultra-high conductivity of 1140 S/cm and excellent thermal conductivity, reaching a temperature of 172 °C in 10 seconds when a voltage of 2 V was applied. These composites also exhibited excellent mechanical properties with a tensile strength of 222 MPa and excellent flexibility, maintaining resistance stability under 1000 bending cycles [9]. These results indicate that the modification of carbon nanotubes and nanocellulose, as well as their proper ratio and structure, play a key role in improving the properties of composites (Fig. 9 c). For example, fluorination of multi-walled carbon nanotubes (MWCNTs) and their combination with cellulose nanofibers (CNFs) helped to improve the thermal conductivity and mechanical strength of composites due to enhanced interfacial interaction and efficient phonon transport [187]. The membrane made of F-MWCNTs/CNFs reached a temperature of 88.9 °C in 4 seconds, whereas the unmodified membranes only heated up to 67.6 °C. The temperature rise rate of F-MWCNTs/CNFs is 32.02 °C/s, which is significantly higher than 9.82 °C/s for unmodified membranes. The tensile strength of F-MWCNTs/CNFs reaches 141.97 MPa, which is 32 % higher compared to unmodified composites.

These studies highlight the importance of proper functionalization and structural organization of components to achieve optimal performance of composites, opening new perspectives for their applications in

heat transfer control, printed electronics, and energy harvesting and storage systems.

#### 4. Economic analysis of nanocellulose and carbon nanotube (NC/CNT) Composites

The global nanocomposites market is forecasted to grow significantly in the coming years, driven by demand in the environmental and energy sectors. According to MarketsandMarkets, the nanocomposites market is expected to grow from \$4.1 billion in 2019 to \$8.5 billion by 2024, with a compound annual growth rate (CAGR) of 16 %. This market is growing due to the high demand from packaging, aerospace, defence, electronics and semiconductor applications (Nanocomposites Market Global Forecast to 2024 | MarketsandMarkets). Composites based on cellulose nanocrystals (NC) and carbon nanotubes (CNT) are well positioned to capitalize on this trend due to their superior properties. The primary materials, cellulose nanocrystals, and carbon nanotubes, vary in cost depending on their source and purity. The cost of cellulose nanocrystals can range from \$5 to \$80 per gram, while carbon nanotubes range from \$30 to \$300 per gram. Today, several successful companies in the global market produce cellulose nanocrystals and carbon nanotubes. These companies include:

CelluForce produces crystalline nanocellulose under the CelluRods™ brand. The company works with cellulose nanocrystals and has a production capacity of 1 ton per day. CelluForce has the world's largest research network for cellulose nanocrystals, allowing it to develop and modify products to meet specific customer requirements.

Blue Goose Biorefineries Inc. produces crystalline nanocellulose under the BGB Ultra™ brand. Pilot production is located in Saskatoon, Canada, with a production capacity of up to 100 tons per day. The company has successfully scaled up the production of cellulose nanocrystals and plans to further expand its capacity.

Carbon Solutions Inc. (CSI) produces carbon nanotubes. The company is a leader in the development of nanotube purity and quality control technologies. CSI has strong business relationships with leading companies in the development of nanotubes for sensors, flexible electronics and other applications.

OCSiAl produces graphene nanotubes under the TUBALL™ brand. The company is the largest manufacturer of graphene nanotubes with a production capacity of 1 ton per year and has a presence in several countries, allowing it to serve the global market.

A comparative economic analysis of the viability of various NC/CNT composite production methods is essential to identify the most economically efficient approaches (Table 5 and 6). Ultrasonic treatment and vacuum filtration are the most economically efficient methods for producing stable and strong composites, while wet spinning and electrospinning methods, though requiring higher initial costs, may be justified for obtaining composites with unique characteristics for specialized applications. It is recommended to choose the production method based on the specific industry needs and the required properties of the final product.

#### 5. Future prospects

Nanocellulose as a green and inexpensive biopolymer has good high specific surface area, good mechanical strength and biodegradability. CNTs have excellent electrical conductivity, excellent mechanical properties and high specific surface area. Compared with two-dimensional graphene, the one-dimensional structure of CNTs provides flexibility by creating long conductive pathways. Combining NC and CNTs gives a synergistic effect, making the composite better. NC effectively overcomes the limitations of CNTs, improving the dispersion, stabilization and overall performance of the composite while ensuring environmental friendliness and biocompatibility. Thus, NC/CNT composites have outstanding mechanical, electrical, adsorption properties and high surface area, but solving certain problems to optimize their

**Table 5**  
Comparative economic analysis of various production methods.

Method	Advantages	Disadvantages	Equipment Cost	Operating Costs	Economic Evaluation
Strong Acid Treatment	High dispersion degree	High cost, hazard	High	High	High costs and safety issues
Ultrasonic Treatment	Eco-friendly, simple	Requires precise tuning	Medium	Low	Economically efficient method
Vacuum Filtration	Eco-friendly, stable	Long process time, specialized equipment	Medium	Medium	Economically viable for high-quality composites
Electrospinning	Nanostructured fibers, high porosity	High equipment cost	High	Medium	High costs justified for high-tech applications
Freeze-drying with Sublimation	High porosity, stable structure	Long process time, high equipment cost	High	Medium	High costs justified for specialized applications

**Table 6**  
SWOT Analysis of NC/CNT Composites.

Strengths	Weaknesses	Opportunities	Threats
Exceptional mechanical, electrical, and thermal properties	High cost of carbon nanotubes	Increased demand for eco-friendly and high-performance materials	Stiff competition from alternative materials
Renewable and biodegradable	Challenges in achieving uniform dispersion	Breakthroughs in production technologies	Impact of economic fluctuations on production costs and demand
High surface area	Advanced production methods require significant investment	Support for sustainable development through regulatory measures	Challenges in regulating the use of nanomaterials

fabrication and application requires further research, which include the following points:

- (1) Improving the effective interaction between NCs and CNTs as well as simplifying the synthesis process of composites represent key areas for future research. The ability to control the structure, morphology, and intrinsic properties in detail with a high level of quality remains an important challenge.
- (2) The high cost and toxicity of CNTs remains a major limitation for large-scale applications of NC/CNT composites. Consequently, more efficient and cost-effective and green CNT fabrication methods are needed, taking into account the scale-up of production to reduce the overall cost of composites.
- (3) Few studies address the economic aspects of research. Incorporating the cost of materials or production expenses could assist researchers in analyzing the market viability and determining the feasibility of using specific materials or synthesis methods, as well as comparing economic aspects when evaluating different approaches.
- (4) Basic research on NC/CNT composites is needed to better understand water purification processes. This will allow for a more detailed study of their potential to create effective systems for purifying water resources from heavy ions, dyes, harmful medical drugs, bacteria, and other hazardous substances.
- (5) Continue to investigate the influence of structure and structure parameters on the mechanical, capacitive and electrochemical properties of composites to optimize their use in advanced supercapacitors. In addition, innovative technological solutions should be developed to introduce composites into flexible supercapacitors with high energy capacity and fast charge.
- (6) To further improve the properties of NC and CNT-based composites, it is recommended that additional nanomaterials such as conducting polymers, carbon nanomaterials, and active nanoparticles should be investigated and introduced. This will enhance their functionality and applications, especially in the fields of electrochemical energy storage and water treatment.

## 6. Conclusion

This review presents a comprehensive study of nanocomposites based on nanocellulose and carbon nanotubes, highlighting key aspects of their development and application. The review identifies the main advantages of these composites compared to their individual components and traditional materials, particularly in the contexts of energy storage and conversion, as well as environmental remediation. The primary benefits of the composite include its eco-friendliness and low cost, attributed to the use of nanocellulose, which reduces the overall cost and toxicity of carbon nanotubes. Carbon nanotubes, due to their unique properties such as excellent electrical conductivity, strength, and high surface area, significantly enhance the overall efficiency of the composite depending on the specific application.

The review also provides a detailed examination of effective methods for mixing these two nanomaterials and proposes research approaches for creating 1D, 2D, and 3D structures based on this hybrid composite. Special attention is given to current research on the application of this composite in environmental water purification, pollutant detection, antibacterial technologies, and energy solutions such as high-performance supercapacitors and rechargeable batteries. Furthermore, the hybridization of these materials with various functional groups improves interfacial and intermolecular interactions, promoting the efficient use of both nanocellulose and carbon nanotubes, thus expanding the application range and overall effectiveness of the composite. The addition of conducting polymers, graphene, and metal nanoparticles can further enhance the composite's capacitive, mechanical, adsorptive, and catalytic properties. In addition, detailed market prospects of the investigated samples were clarified, demonstrating the economic feasibility of these materials, which could be valuable for researchers.

Based on a thorough analysis, we have proposed recommendations for further research on this composite, including improving its characteristics, refining effective production methods, large-scale application, hybridization, and studying the impact of incorporating additional materials.

### CRediT authorship contribution statement

**Azamat Taurbekov:** Conceptualization. **Meiram Atamanov:** Writing – original draft, Supervision, Funding acquisition, Conceptualization. **Zulhair Mansurov:** Supervision. **Bayan Kaidar:** Writing – original draft, Project administration, Conceptualization. **Zhengisbek Kusanov:** Formal analysis, Data curation. **Vanessa Fierro:** Supervision. **Tolganay Atamanova:** Visualization, Project administration. **Alisher Abdisattar:** Visualization, Conceptualization.

### Declaration of Competing Interest

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

## Data availability

Data will be made available on request.

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