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Recent Developments on Nanocellulose-Based Energy Systems

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Abstract
The efficient conversion of lignocellulosic biomass into novel bio-based products can contribute to the transition to a more sustainable, low-carbon economy. However, technological advances are still required in order to better consolidate the biorefinery concept, which involves the processing of biomass into food and feed ingredients, bio-based products (chemicals and materials), and bioenergy. Among these bioproducts, nanocellulose stands as a potential material with remarkable properties and applications in different sectors, such as in the development of energy systems. Recently, the application of nanocellulose as substrate or scaffold in thermoelectric devices has been explored given that these cellulose nanomaterials are degradable, renewable, and biocompatible and present low thermal conductivity (~ 0.03 W/(m·K)) and density (~ 0.04 g/cm^3). This mini-review summarizes some recent technologies being studied to develop energy systems by exploring the remarkable properties of cellulose nanomaterials. The application of nanocellulose to generate energy in thermoelectric devices is discussed in more details aiming to contribute to broaden the use of lignocellulosic biomass into more advanced systems, thus expanding the portfolio of future biorefineries.

Keywords Nanocellulose · Thermoelectric devices · Bioenergy · Biorefinery

1 Introduction

The development of innovative technologies to promote the sustainable exploitation of renewable biomass resources into bioenergy and bioproducts will be required in the transition from a fossil-based to a bio-based, low-carbon economy. Along with that, biomass availability at a competitive price will be necessary to supply the feedstocks that can be derived from a large variety of crops, wood, agricultural and forestry residues, industrial, and food wastes. For instance, the global annual generation of biomass waste is in the order of 140 Gt [1]. Brazil plays an important role in this aspect, leading the production of several crops such as sugarcane biomass, with around 40% of the global production [2]. Currently, most of this sugarcane is used to produce ethanol, sugar, and bioenergy from the burning of sugarcane bagasse [3].

Bioenergy offers a great opportunity to lower greenhouse gas emissions, especially when the process integrates the production of other biomass-derived products. This integration idea is within the biorefinery concept, which involves the sustainable processing of biomass into a spectrum of marketable food and feed ingredients, bio-based products (chemicals and materials), and bioenergy (biofuels, power, and/or heat). Therefore, the whole use of the lignocellulosic biomass to obtain value from the cellulose, hemicellulose, and lignin fractions should be pursued.

Considering that cellulose is the most abundant carbohydrate in nature, formed by crystalline and amorphous domains, its use for nanocellulose production in future biorefineries stands out as a high added-value material with remarkable properties and applications in different industrial sectors [4]. Among these sectors, there are great opportunities for the use of nanocellulose materials in the development of advanced energy systems. In this way, some applications have been proposed for nanocellulose-based materials as a triboelectric nanogenerator for self-powered healthcare products with antibacterial activity [5]; as a gear-like self-powered sensor sensible to human touch and vibration [6]; self-powered paper-based electronics [7–9]; in a self-powered handheld printer system scavenging mouth-blown

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wind energy (that also can be used in mobile devices without an electric power supply) [10]; as a substrate to obtain carbon nanofiber aerogels for supercapacitor applications [11]; to convert solar energy into electricity at near-room temperatures [12, 13], among others [14, 15]. Moreover, the potential use of these nanomaterials is expected to broaden as large-scale nanocellulose plants have already started to operate worldwide, such as CelluForce Inc. in Canada, making these nanomaterials available for evaluation in different applications [16, 17].

This mini-review summarizes some recent technologies being studied to develop energy systems by exploring the remarkable properties of cellulose nanomaterials. A novel and promising application of nanocellulose to generate energy in thermoelectric devices is also discussed in more details aiming to contribute to broadening the use of lignocellulosic biomass in more advanced systems.

2 Nanocellulose

The term nanocellulose is usually applied to describe cellulose materials that present at least one of its dimensions in the nanoscale. These cellulosic nanomaterials can be obtained from a large variety of plant biomass in the form of cellulose nanofibrils (CNF) or cellulose nanocrystals (CNC) (Fig. 1), depending on the extraction procedure used. CNFs are elongated and flexible materials with lengths in the order of micrometers and diameters between 20 and 100 nm, usually obtained by mechanical, chemical, or enzymatic treatments used separately or in association. CNCs consist of stiff nanomaterials with a spindle-like morphology, typical length between 50 and 350 nm, and a diameter between 5 and 20 nm [17, 18]. Sulfuric acid hydrolysis is the most common method to produce CNC, resulting in nanomaterials with a predominant crystalline structure. The morphological characteristic of nanocellulose depends not only on the production process applied, but also on the lignocellulosic source which includes wood and agro-industrial residues as well. In addition to plant source-nanocellulose, bacterial cellulose (BC) is produced extracellularly by microorganisms as pure cellulose with average diameters of 20–100 nm and micrometers lengths.

Due to their nanoscale size, fibril morphology, and large surface area, these cellulose nanomaterials present excellent mechanical properties and interesting characteristics such as hydrophilicity, biodegradability, and biocompatibility [17]. Among the different possible applications of nanocellulose, its use as mechanical reinforcements in polymeric materials stands out, resulting in a significant increase in the mechanical and thermal strength properties of such materials. In the areas involving applied physics, nanocellulose materials find potential use as transparent-flexible electronic devices, batteries, supercapacitors, catalytic supports, templates for electronic components, electroactive polymers, sensors, energy storage systems, nanogenerators based on piezoelectric, triboelectric, and thermoelectric effects, among others [14, 15, 20–22], as exemplified in Table 1.

The piezoelectricity of cellulosic materials is related to the occurrence of electric dipole moments within the CNC particles, due to the anisotropic tri- and monoclinic unit crystalline structure association with unevenly distributed carbon atoms and change of polarization density under electric fields [41]. The piezoelectric modulus changes with the crystallinity of different nanocellulose species. As, e.g., for regenerated nanocrystalline cellulose (II), the piezoelectric constant was found around $35-60$ pC N^{-1}, whereas films of partly aligned CNCs yielded a piezoelectric constant of 0.97 Å V^{-1} [41]. A brief overview about nanocellulose nanogenerators based on piezoelectric and triboelectric effects is presented as follows.

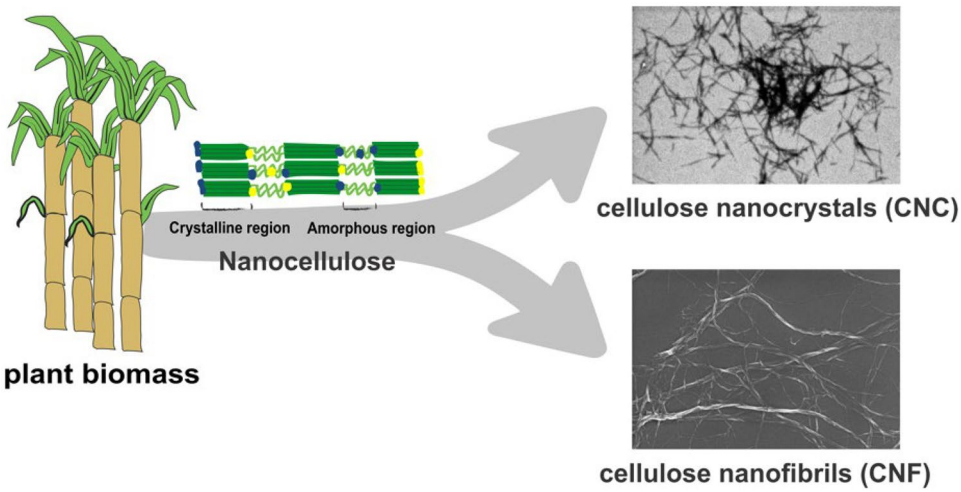

Fig. 1 Schematic illustration and scanning electron microscopy of cellulose nanocrystals (CNC) and cellulose nanofibrils (CNF) (adapted from [19], Copyright 2022, with permission from Elsevier)

Table 1 Recent literature examples on the use of nanocellulose in energy systems

Cellulose type	Application	Designed form	Main results	Refs
CNC	PENG	Electrospun fiber of CNC incorporated in PVDF	Piezovoltage: 60 V (adding 2 wt% CNCs)	[23]
CNC	PENG	BaTiO3 incorporated in CNC films	Piezovoltage: 2.86 V; output current: 262 nA; output power: 373 nW	[24]
BC	PENG	BaTiO3 and MWCNT particles incorporated in BC paper	Outputs: 18 V and 1.6 μA cm^{-2}	[25]
CNC	PENG	CNC incorporated PVDF-HFP polymer	Output voltage: 12 V upon mechanical vibrations; 5.5 V upon elbow movement and 1.1 V upon cloth folding movement	[26]
CNC	PENG	Nanocomposite film with nylon 11	2.6 times higher output voltage as compared to neat nylon 11	[27]
CNC	PENG	Fluorinated CNC-incorporated PVDF	output voltage 8 μV	[28]
CNF	PENG	CNF aerogel film coated with PDMS	Open-circuit voltage: 60.2 V; short-circuit current:10.1 μA; power density: 6.3 mW cm^{-3}	[29]
BC	PENG	BC paper-based matrix assembled with vanadium-doped ZnO mircoflowers	Output voltage of 1.5 V; current density of 80 nA cm^{-1} and power density of 60 nW cm^{-1}	[7]
CNF	PENG	CNF film incorporated acid-oxidized multi-walled carbon nanotubes	Dielectric constant of 73.88; AC conductivity 1.77×10^{-7} Scm^{-1} (at 1 kHz)	[30]
CNF	TENG	Aluminum-doped zinc oxide deposited on CNF paper	Output voltage: 7 V; output current: 0.7 μA; charge transfer: 77 μC m^{-2}	[31]
CNF	TENG	Ag nanowire deposition on CNF paper	Open-circuit voltage: 21 V; short-circuit current: 2.5 μA	[32]
CNF	TENG	CNF/Ag membranes	External load resistance: 20 MΩ; peak power density: 7.68 μW cm^{-2}	[5]
CNF	TENG	CNF film modified with fluorinated ethylene propylene	Output voltage: 30 V; output current: 90 μA	[33]
CNF	TENG	Polyetherimide-grafted CNF aerogel modified with 4 layers of PVDF nanofiber mats	Output voltage: 106.2 V; power density 13.3 W m^{-2} on a 106 Ω external load	[34]
CNC	TENG	PDMS/CNC flake composite	Output power density: 1.65 mW (0.76 mW cm^{-2}) under continuous operation	[35]
CNF	TENG	Polyethyleneimine cross-linked CNF film	Open-circuit voltage: 286 V; power density: 0.43 W m^{-2} with a load resistance of 5×10^7 Ω	[6]
CNF	TENG	Fluorine-modified CNF and polyamide	Output voltage: 28.5 V; current 5.13 μA; power density 1.3 μW cm^{-2}	[36]
BC	TENG	Nanocellulose papers containing silver nanowires and $BaTiO_3$ nanoparticles	Output voltage: 460 V; output current: 23 μA under 5 kgf compressive force	[8]
CNF	TENG	CNF paper with phosphorene	Open-circuit voltage: 5.2 V; current density: 1.8 μA cm^{-2}	[9]
CNF	TENG	CNF aerogel modified with rabbit fur	Power density of 3.4 W m^{-2} achieved on a 4.7 MΩ load under a pressure of 30 kPa	[37]
CNC	TENG	Indium oxide incorporated CNC film	Output voltage: 130 V; short-circuit current: 15 μA; power: 2 mW	[10]
CNF	SC	MXene ($Ti_3C_2T_x$) nanocomposites assembled with colloidal dispersions of CNFs	High capacitance of 298 F g^{-1} and a high conductivity of 295 S cm^{-1}	[38]
BC	SC	N-Self-doped carbon nanofiber aerogels obtained from BC modified with zeolitic imidazolate frameworks nanocrystals	Capacitances of 224 F g^{-1} at 0.5 A g^{-1} and 612 mF cm^{-2} at 1.37 mA cm^{-2}, superior energy density of 31.04 Wh kg^{-1} (13.19 Wh L^{-1}) at a power density of 250 W kg^{-1}	[11]
NC	TE	Copper iodide (CuI) film	S: 228 μV K^{-1} at a temperature range of 290–335 K. PF: 36 μW m^{-1} K^{-2}	[12]
NC	TE	Nanocellulose film modified with PEDOT:PSS, silver nanoparticles (AgNPs), and carbon nanotubes	Thermoelectric voltage output of 1.7 mV for a temperature difference of 125 K	[39]
CNF	TE	CNF-PEDOT:PSS film modified with microparticles of chromium disilicide ($CrSi_2$)	Electrical conductivity: 5.4 ± 0.5 S cm^{-1}; S: 88 ± 9 mV K^{-1}; PF: 4 ± 1 μW m^{-1} K^{-2} at room temperature	[40]

CNF, cellulose nanofibers; *CNC*, cellulose nanocrystals; *BC*, bacterial cellulose; *NC*, nanocellulose; *PENG*, piezoelectric nanogenerator; *TENG*, triboelectric nanogenerator; *SC*, supercapacitors; *TE*, thermoelectric; *PVDF*, polyvinylidene fluoride; *MWCNT*, multi-walled carbon nanotube; *PVDF-HFP*, poly(vinylidene fluoride-co-hexafluoropropylene); *PDMS*, polydimethylsiloxane; *S*, Seebeck coefficient; *PF*, thermoelectric power factor; *PEDOT:PSS*, poly(3,4-ethyelenedioxythiophene)-polystyrene sulfonate

3 Nanocellulose in Piezo- and Triboelectric Generators

In view of the development of a new class of electrical and electronic devices, the cellulose (either on a nano-scale) is inherently dielectric, presenting an electrical response to mechanical changes [20]. When the CNCs are manufactured as ultrathin films, a piezoelectric response of 2.1 Å V^{-1} can be reached, being comparable to a film of a piezoelectric metal oxide, convenient for energy harvesting and power generation [41]. The intrinsic piezoelectric property of nanocellulose is attributed to hydrogen bonding between the linear cellulose chains, predicted through modeling [42]. In the same direction, the triboelectric effect (based on mechanical deformation and electrostatic transduction) can be reached by tailoring charges on the cellulose surface and manipulating the morphologies of the active layers that can be composed by a combination of conducting polymers or inorganic piezoelectric particles [20].

For instance, a triboelectric nanogenerator composed of a thin film of CNC with indium trioxide (ITO) nanoparticles (Fig. 2) are able to harness mild wind-blown energy and generate an output voltage up to 130 V, a short-circuit current of 15 μA and power of 2 mW, capable to power supply a handheld printer [10]. Yang et al. [31] presented degradable, flexible, and transparent conductive substrates composed of aluminum-doped zinc oxide atomically layer deposited on transparent cellulose nanofibril papers to serve as triboelectric nanogenerator in wearable and disposable electronic devices. They obtained a sheet resistance in the range from 130 to 180 Ω sq^{-1}, which was at the same level as ITO- and graphene-coated PET substrates. In another work, Nie et al. [36] functionalized cellulose nanofibrils with triethoxy-1H,1H,2H,2H-tridecafluoro-n-octyl-silane (PFOTES) to control the surface polarizability and hydrophobicity. The triboelectric charge density and hydrophobicity were improved by grafting fluorine-bearing silane chains to the surface of CNFs, reaching currents of 9.3 μA upon the short circuit, which is about twice the previous obtained. Zhang et al. [7] presented a hybrid flexible piezoelectric nanogenerator assembling vanadium-doped ZnO microflowers in bacterial cellulose matrix to be applied in self-powered motion sensors. They reached an output voltage of 1.5 V, current density around 80 nA cm^{-2}, power density of 60 nW cm^{-2}, and a response time of 0.1 s when over motion. Li et al. [25] presented a hybrid tribo/piezoelectric nanogenerator using nitrocellulose nanofibril paper and BaTiO3/MWCNT@bacterial cellulose paper as the triboelectric and piezoelectric layer, respectively. They obtained outputs of 18 V and 1.6 μA cm^{-2} and a response for dynamic pressure from 0.5 to 3 N cm^{-2} with a sensitivity of 8.276 V cm^2 N^{-1} and a detection limit of 0.2 N cm^{-2}.

Other examples of nanocellulose application in piezoelectric and triboelectric generators are presented in Table 1. A novel and promising application of nanocellulose to generate energy is in thermoelectric devices, which will be discussed in more details below.

4 Nanocellulose for Thermoelectric Applications

To move toward a more carbon–neutral and sustainable society, research on the development of active materials derived from renewable supplies to generate clean and sustainable energy alternatives are needed. In addition to the environmental friendliness of energy generator devices, making them safer, efficient, affordable, flexible, and lightweight is also the key challenge [43, 44]. Thermoelectric devices composed of electrically active macromolecules are emerging as promising platforms for the next generation of environmentally friendly energy conversion technologies that can provide efficient conversion of thermal gradients into electricity, with silent operation, readily synthesized, air-stable, good flexibility, ease of manufacture, and comparatively long lifetimes [45, 46].

In thermoelectric devices, when a thermal gradient is applied (ΔT), charge carriers diffuse from a higher temperature to a lower temperature zone generating potential difference (electric voltage ΔV). The direct conversion of ΔT into ΔV is called the Seebeck coefficient or thermopower, $S = -\Delta V/\Delta T$. The thermoelectric efficiency of energy conversion is evaluated by a non-dimensional figure of merit (ZT), determined by their ability to allow the flow of charge while resisting the flow of heat, defined as:

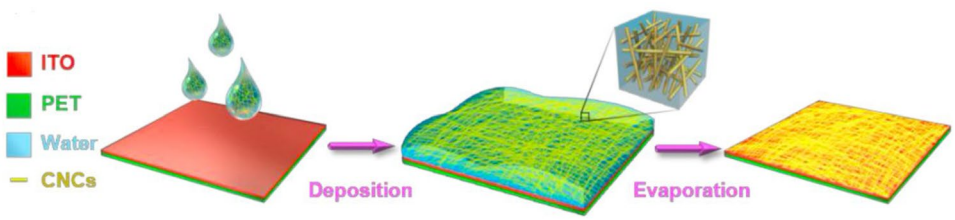

Fig. 2 Schematic production of the CNC/ITO composite electrodes (reprinted from [10] Copyright 2021, with permission from Elsevier)

$$ZT = \frac{S^2\sigma}{\kappa}T$$

where σ is the overall electrical conductivity and κ is the thermal conductivity [45, 46].

Chalcogenide compound semiconductors (such as the families of Pb, Sn, S, Se, Te, Bi, and S) are among the most relevant for thermoelectric energy conversion, but these include complicated chemistry, a compatible surfactant removal process, large ZT (> 1) at very high temperatures (500–600 °C) [47] and a lack of compatible precursors for S, Se, and Te [48].

Conducting polymers including polyaniline (PANI), polypyrrole (PPy), and poly(3,4-ethylenedioxythiophene) (PEDOT) have been displayed attractive thermoelectric properties, showing unique advantages with respect to typical thermoelectric materials such as intrinsic low thermal conductivities ($\kappa < 1$ W m^{-1} K^{-1}), relatively high Seebeck coefficients (S ~ 20–100 μV K^{-1}) and high electrical conductivities upon doping ($\sigma \sim 10^4$ S cm^{-1}) [43, 49]. Together with others, carbon-based materials, such as carbon nanotubes and graphene, present good processability and flexibility, low cost, and scalability [50].

A high potential material to be used in thermoelectric devices as substrate or scaffold is the nanocellulose, because they are degradable, renewable, biocompatible, presents low thermal conductivity (~ 0.03 W m^{-1} K^{-1}) and density (~ 0.04 g cm^{-3}) [50–53]. It is possible to prepare nanocellulose films and aerogels, with a rich three-dimensional network, which are mechanically robust, flexible, but electrically insulating. In particular, nanocellulose (either in the form of CNCs or CNFs) has been exploited as an emerging material for new electrochemical energy devices due to its superior physical and chemical properties (biocompatibility, low cost, renewability, excellent mechanical and optical properties, flexibility, easy modifications, unique geometry, and hydrophilicity) [54]. As an aerogel, nanocellulose presents inter-connected networks with low thermal conductivity [55], and when prepared with nanocomposite materials, its pore sizes can reach values lower than 100 nm, becoming able to present thermal conductivities values lower than 18 mW/m/K (thermal super-insulators) [56].

Nanocellulose-based films/aerogels have been considered a fascinating substrate to fabricate functional materials for energy devices [43, 44]. Its integration with conducting materials opens avenues for the development of thermoelectric devices with flexibility and mechanical strength [57]. Some conducting polymers with highly p-conjugated polymeric chains, as presented above, and their derivatives have been deposited on the surface of nanocellulose by in situ polymerization, chemical vapor treatment, or layer-by-layer technique [45, 57–59], resulting in devices with relatively high electrical conductivity, charge-storage capacities, rapid charge/discharge rates, and stable cycling performance [60]. All the presented electrical/electrochemical performances suffer a direct influence by the films/aerogel pore size [54, 55].

In this context, Li et al. [50] presented a free-standing paper-based thermoelectric generator (Fig. 3 a–c) composed of multi-walled carbon nanotube (MWCNT)/carboxylated nanocellulose (C-CNF) with flexible properties and electrical output as 3.7 nW for a ΔT of 70 °C. The use of C-CNF reduced significantly the thermal conductivity and improved the MWCNT dispersibility and thermoelectric performance. Jia et al. [51] obtained a thermoelectric and flexible PEDOT/single-walled carbon nanotube (SWCNT)/bacterial cellulose (BC) nanoporous films after pressing the precursor aerogel. The schematic of electron transport and heat transport along and through the pores are represented in Fig. 3d–f. The uniformity of PEDOT surface coating form multiple paths to perform the electron transport, achieving an electrical conductivity of 290.6 S cm^{-1} and a maximum output power of 169 nW at ΔT = 65.6 °C. The porous structure and abundant junctions resulted in reduced thermal conductivity and increased phonon scattering, improving the thermoelectric properties. Mehés et al. [13] proposed a biohybrid energy-harvesting device composed of nanocellulose and poly(3,4-ethylen edioxythiophene):polystyrene sulfonate (PEDOT:PSS) to convert solar infra-red radiation into energy via thermoelectric mechanisms, reaching sixfold enhancement in the harvested bio-photovoltaic power. Klochko et al. [12] converted solar energy into chemical energy using a nanocellulose-based biodegradable flexible thin film with copper iodide (CuI) deposited on both sides of the substrate, generating a maximum output power of 1.5 nW for a ΔT of 50 °C. Han et al. [59] obtained thermoelectric polymer aerogels mixing nanofibrillated cellulose with PEDOT:PSS and glycidoxypropyl trimethoxysilane (GOPS) before freeze-drying. The resulting nanocellulose-based conducting aerogel was exposed to a secondary dopant, in this case, the dimethylsulfoxide (DMSO) vapor, reaching semiconductor properties, where it was successfully applied as a dual-parameter sensor, with decoupled pressure and temperature sensing through an electrical readout. They obtained aerogels with different shapes and good flexibility, as shown in Fig. 3g–i. Another similar temperature–pressure dual-parameter sensor was presented by Khan et al. [57] using a nanocellulose-based aerogel and the conductive polymer PEDOT:PSS, to be future applied as electronic skin. Other important studies using nanocellulose in thermoelectric devices are presented in Table 1.

All those studies show the suitability of using low-cost plant-derived nanocellulose substrates to create environmentally friendly biodegradable thermoelectric materials, generating

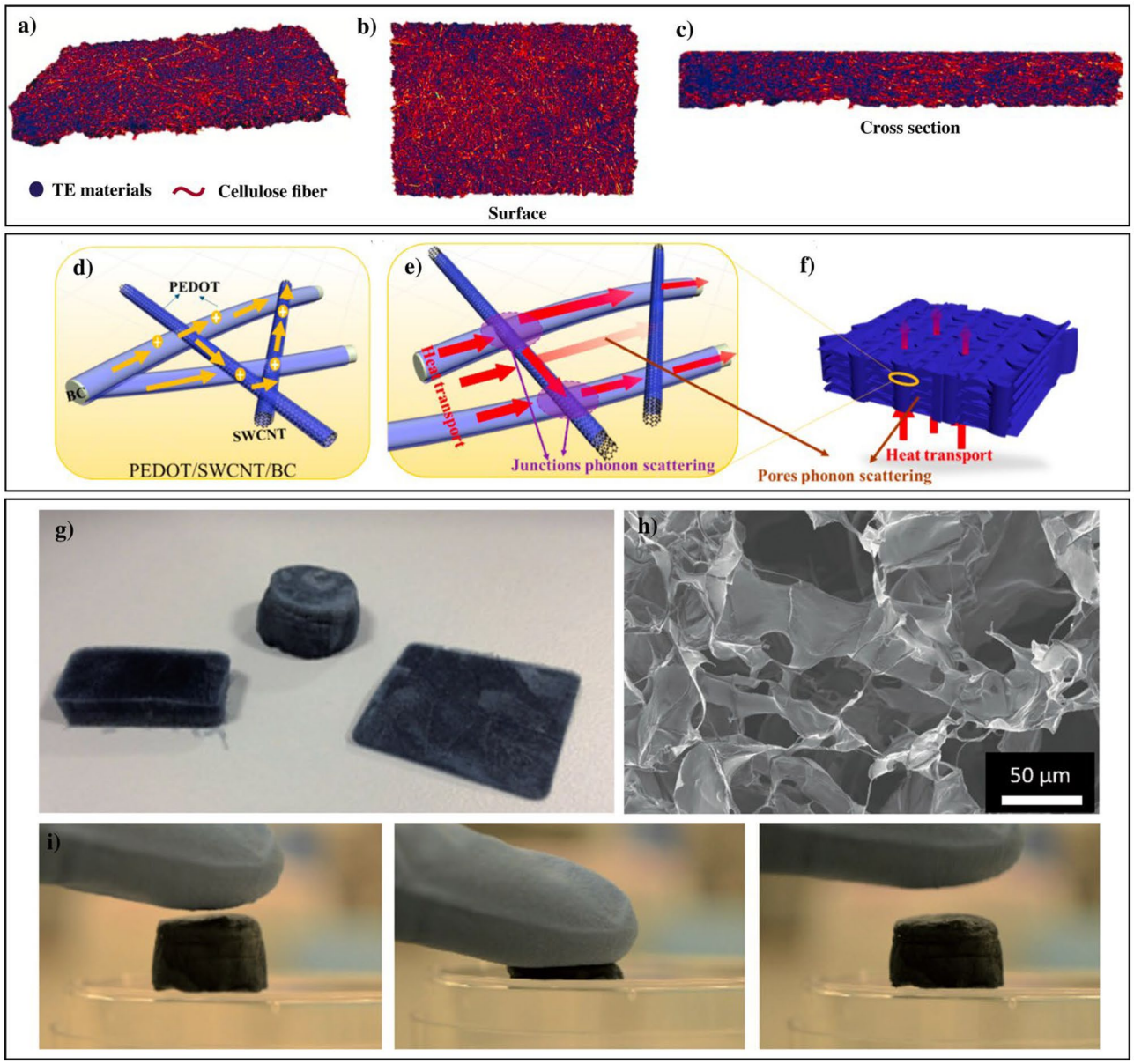

Fig. 3 3D structure of MWCNT/C-CNF paper-based TE materials (**a**), its surface (**b**), and cross-sectional (**c**) views (the brown part represents the TE materials and the blue part the cellulose fiber) [50]. Schematic of electron transport (**d**), heat transport along (**e**), and through (**f**) the pores in PEDOT/SWCNT/BC films [51]. NC-PEDOT:PSS aerogels with different shapes (**g**), its SEM typical images (**h**), and the aerogel flexibility before, under, and after pressing (**i**) [59] (**a–c** reprinted from [50] Copyright 2021, with permission from Elsevier; **d–f** adapted with permission from [51], Copyright 2021 American Chemical Society; **g–i** reprinted from [59], Copyright 2021, with permission from John Wiley and Sons

electricity by harvesting residual environmental heat at near-room temperatures. The nanocellulose acts as reinforcement, increasing the strength, flexibility, and thermal stability of the films/scaffolds. The design of high-performance thermoelectric materials has a sizeable impact on the commercial industry as a sustainable energy recycler for the constant losses, besides being a key for several applications such as energy storage devices, autonomous microsystems, sensors, tissue engineering, wearable electronics, and polymer electrolyte fuel cells.

5 Concluding Remarks

Nanocellulose materials find potential use in different areas involving applied physics, showing great promise for application in energy harness due to its advantageous chemical, mechanical, and biocompatible properties, being also capable of supporting self-powered electronic components. The variety of examples on the use of nanocellulose as a substrate to fabricate functional materials for energy

devices opens avenues for the development of thermoelectric devices with flexibility and mechanical strength. Such applications of nanocellulose-based energy systems offer potential opportunities for expanding the use of lignocellulosic biomass as high-value bio-based products in future biorefineries.

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Author Contribution All authors contributed equally to this work.

Declarations

Conflict of Interest The authors declare no competing interests.

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