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REVIEW

Exploring Nanocellulose-Based Materials for Energy Conversion and Storage Devices

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ABSTRACT

The planet must deal with the two main concerns of the twenty-first century: energy storage and protecting the environment. Energy storage systems urgently require green and sustainable electrode materials due to the rise in worldwide demand for energy and severe environmental damage. The biopolymer-based device reduces e-waste and environmental issues caused by conventional electronic devices. Nanocellulose is a solid choice for green electronics, due to its unique properties, like being eco-friendly, cost effective, biodegradable, having great mechanical strength, and remarkable optical clarity. With its exceptional qualities, sustainability and distinctive structures, nanocellulose has become a hopeful nanomaterial with enormous potential for creating useful energy storage systems. This review aims to offer novel viewpoints on flexible composites made of nanocellulose or nanocellulose-based materials for enhanced energy technologies. Initially, a brief introduction to the special structural features and attributes of nanocellulose is made. To improve these composites' performances, the structure-property-application interactions must be addressed. The most recent uses of nanocellulose-based composites are then thoroughly reviewed. These include flexible solar cells, supercapacitors (SC), lithium-ion batteries and developing energy device innovations. Finally, nanocellulose-based composites for the next generation of energy devices are offered, along with their current difficulties and potential future developments.

KEYWORDS

Nanocellulose; solar cell; supercapacitor; li-ion battery; energy devices

INTRODUCTION

Electronic devices are extensively used in our daily lives. Electronic devices have made a great impact on luxurious living and convenience in recent years. Most electronic devices are non-degradable, creating lots of environmental problems. Due to environmental concerns, biopolymers are becoming more popular due to their cost-effectiveness, degradability, and biocompatibility [**1-3**]. Because of their biocompatibility, environmental friendliness, abundance in nature, higher mechanical strength, flexibility, and low weight, biopolymers provide new possibilities for functional electronics, serving as attractive building blocks for flexible, wearable, implantable, and eco-friendly electronics [**4-8**]. Electronic devices based on biopolymers refer to certain parts of various gadgets that contain biological components and parts that exhibit excellent electrical performance, which is advantageous for minimizing environmental issues. To develop flexible electronics toward green electronics, several fields, including organic electronics, and materials science are currently being merged [**9**]. Comprehensively, biomass has developed into an extreme carbon source for making porous carbon composites. Cellulose, the abundant biopolymer on earth, is also renewable, cost-effective and environmentally friendly. Further, the utilization of cellulose in its nano form makes it more interesting and appealing to researchers due to its remarkable properties, such as high aspect ratio, excellent strength and elasticity, high crystallinity, and so on. Nanocellulose-based materials can be divided into four primary categories: nanocellulose with nanocarbon composites, conducting polymer composites, metal particle composites, and ternary or multiple composites. Flexible electric motors and

batteries are among the primary applications of nanocellulose-based composites in supercapacitors, solar cell and batteries of all kind's electrolytes, electrodes, and separators, shown in **Fig. 1**.

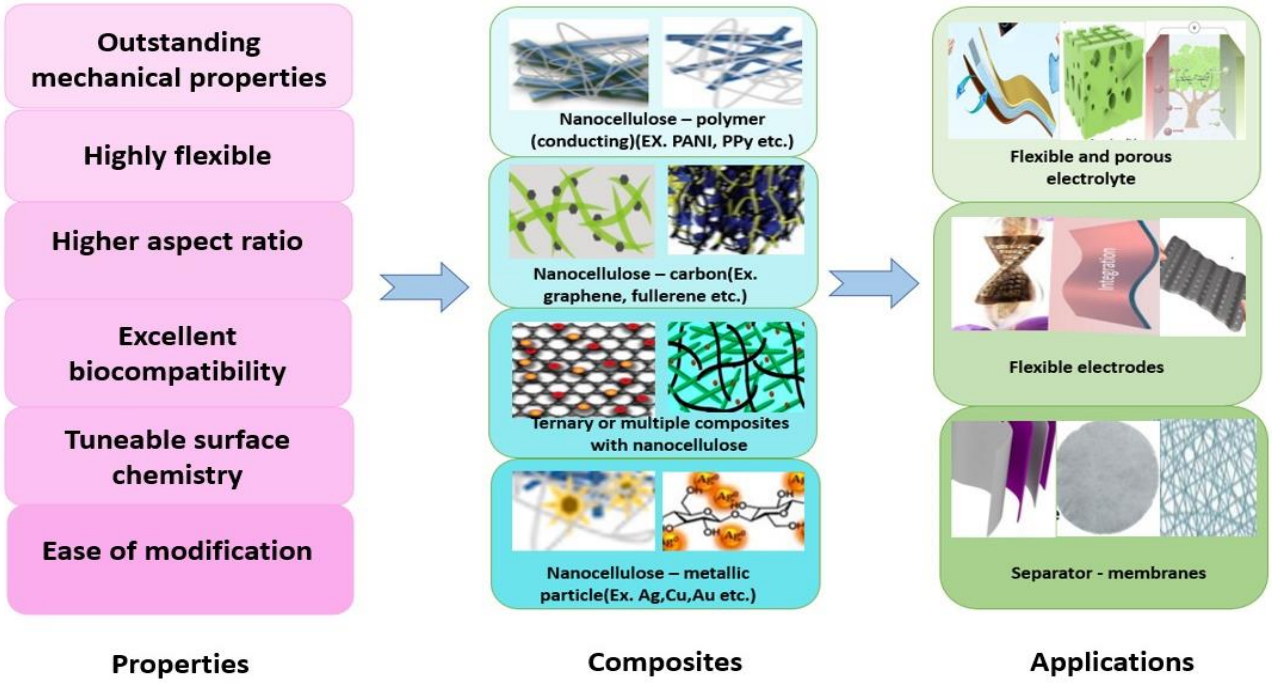

Fig. 1. Nanocellulose-based composites and properties used for applications as energy storage devices, in which it could be utilized as a membrane, electrodes and porous electrolytes and separators.

Due to the rapid development of technology, there is hope for creating energy-efficient conversion and storage devices like solar cells, rechargeable batteries, and supercapacitors (SC) that can be applied to the aerospace, automotive, and other industries. Solar cells, Li-ion batteries capacitors, are the primary types of energy-efficient storage systems in use today. These devices suffer from significant flaws like high operating expenses, rapid self-discharge, as well as low energy density [**10**]. Cathodes, anodes, electrolytes, and separator membranes are components of conventional energy storage systems. These components are ordinarily fabricated from inorganic, or metal compounds, carbonaceous substances, or hydrocarbon chemicals derived from petroleum. However, these conventional materials might struggle to meet the constantly evolving needs of energy storage systems, forcing us to look for novel chemicals and materials that go beyond traditional methods. Nanocellulose has been used in electronic devices like solar cells, OLEDs, supercapacitors (SC), etc. Nanocellulose is used in SC for large-scale production of high-performance energy storage devices in response to the increasingly emerging markets of compact electronic equipment and electric vehicles. Because of the numerous amino and hydroxyl groups, the nanofiber film can improve the adhesion of ions from electrolyte materials as the electrode material and produce a stable electric double-layer capacitor, resulting in exceptional SC energy storage [**11**]. Due to its exceptional qualities, such as lightweight, flexibility and steady cycling performance, cellulosic materials for supercapacitor applications have gained a lot of attention. Lithium-ion batteries (LIBs) are generally mostly utilized in electric vehicles (EVs), portable gadgets and grid-scale energy storage systems (ESSs) [**12-16**]. In addition, replacing toxic or heavy metals in battery electrodes with nanocellulose is safer, more environmentally friendly, and recyclable. Excluding SC, flexible energy systems, such as solar cells or rechargeable lithium-ion batteries (LIBs), have also utilized nanocellulose membranes [**17,18**]. The separators and flexible electrodes for energy devices made of film composites constructed from nanocellulose show tremendous promise. Depending on the traits of obtained film thickness and the nanocellulose-based units, the distinctive characteristics and structure of nanocellulose may offer the film composites admirable hydrophilic properties, excellent flexibility, high mechanical strength and also the optical transparency [**19-21**].

For many years, a material that was optically transparent, strong, and malleable was searched for to replace glass in electronic systems. Cellulose nanofibers can be used to make optically clear paper [**22**] that can be utilized to make substrates for electronic devices like flexible screens, solar cells, organic light-emitting diodes (OLED), antennas and thin film transistors [**23,24**]. OLED produced from recyclable and biodegradable materials are moving forward in attaining sustainable OLED technology.

While compared to glass substrates, organic light-emitting diodes constructed on cellulose nanocrystal substrates had equivalent current efficiencies of 42.79 +/- 8 cd/A at 100 cd/m^2, besides providing the extra benefit of being soluble in water, enabling the product to be recycled [**25**]. The nanocellulose has been employed in the construction of a triboelectric nanogenerator, a type of device that can convert mechanical energy into electric energy by frictional loading [**26-28**]. Due to its inherent insulation, nanocellulose is frequently combined with various electrochemically active materials and pyrolyzed to carbon in order to create novel composites as energy storage materials. This contrasts with the traditional energy device materials (including conducting polymers, carbon-based materials, MXene, metal oxides, etc.). In the ESSs systems, nanocellulose-based composites exhibit extensive micro-pores and mesopores for significant hydrophilic surfaces, charge storage, and rich absorptive sites for improving electrolyte ion absorption and transportation [**29-31**].

Regarding energy devices, the use of existing materials comes with a number of difficulties and restrictions that prevent their general effectiveness. Even though they are essential, traditional materials struggle with problems including performance limitations and environmental effect. One major obstacle to the smooth integration of current materials in energy technologies is the depletion of vital resources combined with rising manufacturing prices. Furthermore, these materials' limits in terms of overall efficacy and efficiency highlight the critical need for substitutes in order to meet the growing need for ecologically friendly, economical, and sustainable energy sources.

This background provides context for analysing materials based on nanocellulose critically. Nanocellulose is a naturally sustainable substitute that has a lower environmental effect and may be more affordable because it is made from renewable plant cellulose. Its composition, which is nanostructured, adds to its improved qualities, which include increased surface area and mechanical strength. Understanding the difficult problems that current energy sources present is essential to realizing nanocellulose's transformative promise. This review paper attempts to carefully analysed these issues, offering a thorough grasp of the complex environment of available energy materials and promoting the use of sustainable nanocellulose-based substitutes as the energy gadgets of the future.

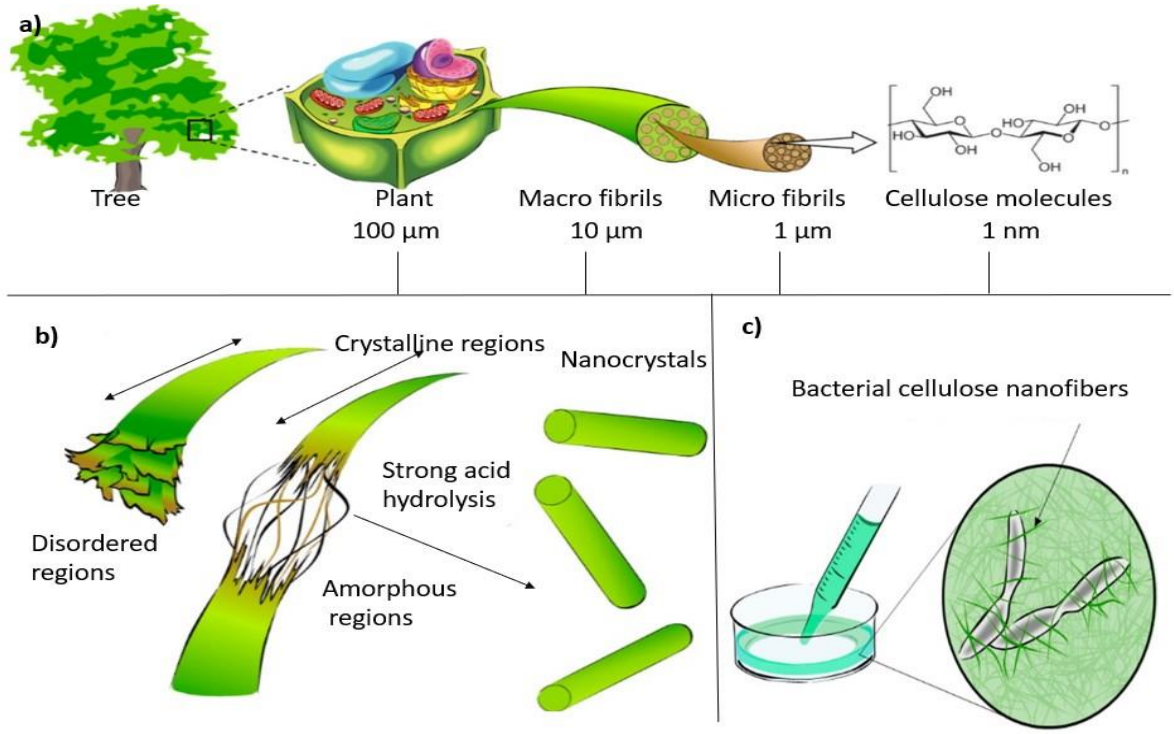

Fig. 2. (a) Hierarchy of cellulose from original to nanoscale molecules, **(b)** Formation of cellulose nanocrystal after acid hydrolysis showing amorphous and disordered regions and **(c)** Synthesis of bacterial nanocellulose [42].

NANOCELLULOSE

The most prevalent carbohydrate on earth is cellulose. A polysaccharide, cellulose is made up of repeated anhydro-D-glucose units that are covalently bonded by ß-1,4-glycosidic linkages [**32**]. Cellulose further extracted at the nanoscale has excellent physical and chemical properties with better environmental capabilities used in electronics, biomedical [**33**] and other industrial applications, illustrated in **Fig. 2(a)**. Nanocellulose (NC) is a highly focused biopolymer. Intermolecular interactions lead to the development of polymer fibers. NC is an entirely natural

nanofiber that can be extracted from biomass resources including wood, plants, herbs and bacteria by defibrillating the cellulose in such materials. NC can be broadly divided into three categories: cellulose nanofiber (CNF), cellulose nanocrystals (CNC) and bacterial nanocellulose (BNC), as shown in **Fig. 2(b) & Fig. 2(c)** [**34,35**]. Amorphous areas with widths of tens to several hundred nanometres and soft, lengthy chains with a length of a few micrometres make up CNF. The flexible, entangled cellulose chains have a huge surface area. Plant-based nanocellulose is an abundant biopolymer in soil with beneficial qualities such as high mechanical strength and slight environmental effect. Plant based nanocellulose offers a wide range of industrial uses, including composites, electronics, electrochemical [**36,37**]. Due to its exceptional thermal qualities, nanocellulose served as a dielectric layer that increased the conductor's heat stability. CNF is an ideal rival as a second option and an eco-friendly substrate for devices thanks to its. advantageous electrical qualities, high flexibility, and transparency [**38**]. Various groups have investigated Si-based digital electronics, gallium arsenide (GaAs) electronics, solar cells, CNF-based transparent paper, on CNF substrates [**39**]. Some applications of nanocellulose based composites are summarized in **Table 1**. In addition, low compactness, simple biodegradability, repeatability, a high volume to surface area ratio, and remarkable supportable mechanical qualities are only a few of the traits that CNFs inherited to gain from recycling garbage into electrical devices that might be useful [**40,41**] Intermolecular interactions are responsible for forming the polymer fibers, which in turn ensure the stiffness of the structure.

CNC, also known as nano whiskers, has a form like an extended crystal rod and is stiffer than NFC. Unlike CNF and CNC, which are made top-down through physical and chemical processes, BNC can be made bottom-up from low molecular weight biomass by improving the circumstances in which cellulose-synthesizing bacteria are cultured. Bacterial cellulose and plant cellulose have same molecular formula, but different chemical and physical properties [**43**]. Plant based cellulose fibers have diameters of around 13–22 μm and crystallinity of nearly 44–65% [**44**]. Bacterial cellulose (BC) fibrils, on the other hand, are generally in the nanoscale, with 10–100 nm diameter and a crystallinity of 90% and have good hydrophilicity naturally due to large number of hydroxyl groups present on their surface. Moreover, BNC has large surface area and high tensile strength (>2 GPa) [**45**], therefore conferring highly desirable characteristics to numerous industries. Furthermore, bacterial cellulose has higher flexibility, purity and hydrophilicity than plant-derived nanocellulose [**46,47**].

Table 1. Comparative Properties of Different Types of Nanocellulose: Production, Dimensions, Durability, and Cost [42].

Nanocellulos	Formation	Size	Sustainability	Cost
CNC	Taken out of cellulose chains by mechanically cleaving the fiber into diameter nanoscale pieces.	5-10 nm	Not good	Low
NFC	only the crystalline region was left after the amprphous region was hydrolyzed by acid to be extracted from cellulose chains.	5-30 nm	Not good	Low
BNC	Bacterial synthesis	10-100 nm	Green approach	High

Table 2. Cellulose based composites for various applications.

No	Cellulose-based composite/aerogel/ film/substrate/paper	Functional materials	Techniques	Applications	Outcomes	Refs.
1.	Cellulose electroactive paper (EAPap) composite	Single wall nanotube (SWNT)/polyaniline with dopants (Cl^- and ClO^{4-})	Spin coating	Smart actuator with ultra-light weight	High stiffness, electrical power consumption and low required power strength	[47]
2.	Cellulose EAPap composites	Multiwall nanotube (MWNT)	In situ pressurized hydrolysis process	Bending EAPap actuator	Increase Young's modulus and resonance frequency	[48]
3.	Cellulose EAPap composite	MWNT	Solvent exchange	Micro-robot, micro-flying objects, sensors	Increase ion mobility, enhance water retention of cellulose, increase output force of actuator	[49]
4.	Cellulose composite	MWNT grafted on cellulose	Mechanical stretching process	Actuator	Increased mechanical and piezoelectric properties, improved actuator performance	[50]
5.	Cellulose nanocomposite	MWNT/TIO2	Hydrothermal process	PH sensors	Enlarged ion adsorption surface area, Improve range for sensing	[51]
6.	Cellulose composite	MWNT	Covalent grafting on cellulose	Flexible paper transistors	Improved mobility, orientation and electrical properties, decrease electrical resistance	[52]
7.	Cellulose composite	Silica and Silica-gold particles	Sol-gel covalent cross- linking process	Solar-cell, optoelectronics, optical coating	Strong-cohesive interaction	[53]

8.	Cellulose EAPap composite	Poly pyrrole-ionic liquid	Dipole rectenna array with modulated microwaves.	Remotely driven actuators, portable electronic, sensing units	Decreased charge accumulation, increased bending displacement of the actuator, highest output voltage and power marginally raised.	[54]
9.	Cellulose composite paper	$BaTiO_3$	Layer by Layer process	Sensing devices	Low cost, eco-friendly, new generation device with excellent value and availability	[55]
10.	Cellulose nanocrystals hybrid thin film	Graphene nanoplatelets	Hot press	Packaging, electrical and heat conducting applications	Increased tensile strength and modulus, young modulus	[56]
11.	Cellulose nanofiber aerogel	MWNT	Supercritical CO2 drying	Flexible supercapacitor	Excellent cycle stability with sustainable capacitance performance, high power density and areal energy density	[57]
12.	Cellulose nanofiber aerogel	MWNT/PANI	In-situ polymerisation, Freeze drying	Supercapacitance	Even after 3000 cycles, capacitance retention greater than 80%, light weight, cycle stability and redox reversibility also good.	[58]
13.	Cellulose nanofiber aerogel	CNT(carbon nanotube)/RGO(Reduced graphene oxide)	Freeze drying	Supercapacitor based electrolyte	Good cyclic stability, high specific capacitance even at after 1000 cycles, light weight	[59]
14.	carboxy methylcellulose aerogel	CNT	Freeze drying	Conductive Aerogel	Highly elastic with approximately 30 percent compressive strain, very low porosity	[60]
15.	Cellulose Nanofiber/ Cellulose Nanocrystal-nanocomposite	CNT	Paper making process	EMI shielding	high electrical conductivity, permittivity, control unwanted reflections, restrict noise in circuits, good flexibility	[61][62]
16.	Cellulose Paper	CNT/SWNT	Paper making process	Smart paper for portable electronics and sensing	High tensile strength and wet strength retention, improved bonding strength	[63]
17.	Cellulose nanofiber film as flexible paper	CNT/LTO (Lithium titanate)	Pressure-controlled aqueous extrusion	Li-ion battery electrode	Higher charge-discharge rate, stable cycle stability, excellent electrochemical performance	[65]
18.	Cellulose nanofiber film	CNT	Aqueous solution process	Li-ion battery anode	Increased ion-mobility, decreased ion-diffusion time	[66]
19.	Bacterial nanocellulose paper	CNT	Vacuum filtration	Flexible supercapacitor	< 0.5% variation, after 5000 charge/discharge cycles indicates outstanding cyclic stability. Performance maintained during the bending cycles, due to excellent interfacial bonding between layers	[67]
20.	Cellulose nanofiber/ nanocomposite	MWNT	Electrospinning	Supercapacitor electrode	Increased the crystallinity and electrical conductivity, reduced activation energy	[68]
21.	2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-CNF /composite fiber	MWNT	3D printing	Wearable electronic devices	Good electrical conductivity, high mechanical strength	[69]
22.	Bacterial nanocellulose fiber	SWNT	Extrusion	Wearable supercapacitor	Out-standing electrochemical stability and properties, reliability.	[70]
23.	Regenerated cellulose fiber film	AgNWs	Free radical polymerization	Flexible electroluminescent device	Highly transparent, partially hydrophilic	[71]
24.	Nano fibrillated cellulose (NFC) film	Cds/quantum dots(qds)	Vacuum filtration and in situ electrostatic adsorption	Green electronics, photocatalytic sensor, photo sensor	Outstanding light transmittance, highly elastic and flexible in nature, excellent optical and physical properties	[72]
25.	Nanocellulose paper	Indium oxide, CNT, silver nanowire	N-type doping	Solar cell, touch screen and LED display	Thermally stable, Excellent optical transparency	[73]
26.	Bacterial cellulose substrate	PANI	In-situ polymerisation	All types of solid-state capacitor	Light weight, very thin, sustainable cycling capacity even after 2500 charge-discharge cycles	[74]

*MWNT-Multiwall nanotube, EAPap- Electro active paper, SWNT- Single-wall nanotube, PANI-polyacrylo nitrile, $BaTiO_3$- Barium titanate, CNT-carbon nanotube, CDS-Cadmium sulphide, LTO- Lithium titanate, AgNws- Silver nanowires

PROPERTIES OF NANOCELLULOSE

The most stiff and resistant natural nanocellulose accessible is cellulose nanocrystals, which have notable features such as high strength, high hardness, and a huge surface area [**75**]. However, one downside of cellulose nanocrystals is their thermal behaviour, which limits their thermo-plasticity when exposed to temperatures between 200 and 300°C [**76**]. Furthermore, because it is a plant-derived polymer, it offers a high-performance, cost-effective method with intrinsic safety, biodegradability and biocompatibility, advantages [**77**]. Plant-based nanocellulose generally has a low degree of crystallization as compared to other celluloses, and at a high degree of polymerization, it shows an increase in mechanical resistance and crystallinity [**78**]. Their size qualities may be affected by a variety of parameters, including the method of preparation, fibre source, nature, duration, acid concentration and temperature utilized in the extraction technique [**79**]. Furthermore, it has lower mechanical characteristics than cellulose produced from other sources due to its relatively low crystallinity and short fiber length [**80**]. One of the drawbacks of deterioration is its poor durability, due to the susceptibility of its fibers to water, carbonation and strong alkalis [**81,82**]. Since many years nanocellulose-based composites are the attraction of a research trend, researchers have reported that plant-based nanocellulose can be reinforced into composites which can resist at high temperatures without degrading [**83**].

Nanocellulose is a highly promising choice for an electrochemical energy device due to its many advantages. 1) Nanocellulose is extremely desirable for producing flexible and durable electrodes and separators due to its excellent mechanical strength (Young's modulus of 130 GPa) [**84**] and thermal stability. 2) Nanocellulose is made possible by its large specific surface area and low density, which makes it an ideal building block for porous electrode materials that are lightweight and have nano-scale pore structures. 3) Since their reactive surfaces are rich in hydroxyl groups, nanocellulose is chemically modified and can be combined with active materials for creating nanocellulose-based novel composites. By reinforcing the active material species in the composites or the composite design, the attributes of these composites can be customized. 4) Nanocellulose is an attractive choice for promoting ionic movement due to its competitive wettability and structural stability in a variety of electrolytes over a broad potential window [**85**]. 5) Nanocellulose is an excellent source material for creating carbon electrodes due to its high aspect ratio, high carbon content, and simple processing, having large surface area, adjustable microstructure, and doping structure [**86**]. The most significant and promising of the aforementioned benefits of nanocellulose is that it can make hybrid materials flexible, allowing for the storage of charge during repeated bending, folding, or compression without significantly affecting the electrochemical performance.

The development of CNC applications in electro-active materials like dielectric materials, microelectronic components, electrically conductive materials, etc. is now being researched. The importance of CNCs in electrical applications is primarily attributable to their adaptability, durability characteristics, and piezoelectric and dielectric capabilities, which are the same as those of other bio-derived materials [**87,88**]. The high degree of crystallinity favoured CNCs to modify polarization densities. It can also be employed as a useful insulating material in a variety of applications thanks to its dielectric characteristics. Due to its role as an electric conductor, moisture plays a significant impact on the ultimate dielectric property.

As for electro-active materials, graphene oxide and CNC both are hydrophilic in nature. Owing to its oxygen-functionalized groups, GO has a high hydrophilicity that makes it ideal for a stable aqueous dispersion in a variety of applications, including composites and inks. Because of its surface hydroxyl groups, CNC is hydrophilic as well. For stable water-based dispersion, surfactants or other modifications may be needed. Their performance in applications is affected by these variations, especially those that rely on formulations that are based on water. Comprehending these hydrophilic differences is essential to maximizing the application of GO and CNC in diverse settings. This specific type of moisture content is derived from CNC sources and is greatly influenced by the degree of crystallinity of cellulose as assessed by cellulose water sorption. The humidity value increases with decreasing cellulose crystallinity and inversely. Higher dielectric characteristics have been found to be closely related to crystallinity, defying assumptions to the opposite. It is obvious that CNCs possess extraordinary dielectric properties which can be employed for electric insulation applications like cable insulation, the value of this feature depends upon the source as well as the morphological aspects of the material [**91**].

Nano fibrillated cellulose (NFC) on the other hand has less thickness than the wavelength of visible light, which results in exceptional optical properties, nanofibril-prepared paper is transparent as an outcome. This can be used in several applications, including those involving solar panels, sensors, and electronics. A team reported the flexible FET (field effect transistors) printed on nano paper for utilization in green electronic transistor applications.[89] Biomimetic robotics, sensors, actuators, and several haptic technologies use electroactive paper that is based on piezoelectric behaviour and ion transport [**28**]. Moreover, bacterial cellulose is rich in hydroxyl groups on the surface compared to plant cellulose, which facilitates doping with carbon-conductive compounds [**89**]. Plant based nanocellulose structure differs from bacterial cellulose, with fewer hydroxyl groups and lower mechanical resistance, which makes it unable to incorporate nano-charges [**89,90**]

Further, the incorporation of carbon nanotubes (CNTs) into cellulose improves the stiffness and tensile strength of paper, resulting in the production of a flexible and durable product. According to research, CNT has long entangled and small components together to improves ductility.

Cellulose nanocrystals serve as a resident of the CNT accumulation and guard the connections of the broad structure of nanotubes [**92**]. Carbon nanotube and cellulose composites are used to create supercapacitor electrodes and biosensors. When it comes to electronic transport and electrocatalytic processes, graphene exhibits unique behaviour. Composites made of cellulose and graphene have excellent shape maintenance and are highly porous. These composites are employed as sensitive and specific solvent sensors, and they work by tracking changes in capacitance during adsorption [**93**]. The processing of cellulose nanofiber-reinforced nanocomposite for various applications and benefits is shown in **Table 2**.

Table 3. Cellulose nanofiber-reinforced nanocomposite processing.

Techniques	Processes	Advantages	Disadvantage	Applications	Ref.
LBL assembly	Immersive/dip-coating, spray-coating, spin-coating	Excellent flexibility, controlled permeability, robustness, easy fabrication, nanoscale thicknesses	Higher assembly time, material incompatibility, limited penetration	Membranes, thermal sensing, biological/chemical sensors (selective diffusion/ adsorption), acoustic microsensors (pressure variation)	[94]
Freeze drying	Removal of water	Pore size, particle distribution, shape and orientation control	Slow process, high energy consumption	Electrolyte for supercapacitor	[59]
Fiber - spinning	1)Wet spinning 2)electrospinning	Low-cost, uniform diameter, defect free surface	Low compatibility, waste generation, degradation, limited scalability	Energy storage, electrode and membranes for batteries	[95]
One pot direct synthesis	1)Cast drying 2)Vacuum assisted filtration	It can develop thick laminating structure, high material compatibility and uniformity	Multiple reaction, waste generation	Flexible energy storage Device and membranes	[96,97]

* LBL- Layer by layer

APPLICATION OF NANOCELLULOSE BASED ENERGY DEVICE

The utilization of nanocellulose in energy device applications in solar cells, supercapacitors and lithium-ion batteries has recently gained interest as it fits the 3E requirements of being eco-friendly, economical and easy to mould into the necessary size and form. Supercapacitors rely heavily on nanocellulose, which is produced using a variety of techniques including pyrolysis, electrospinning, in-situ polymerization, and atomic layer deposition (ALD). As discussed in detail in the supercapacitors section, recent research highlights the better charge storage capacities attained through greater surface area and enhanced electrical conductivity. Nanocellulose-based materials are used in the field of battery technology for electrode design through the use of methods such as wet chemistry and atomic layer deposition. Interestingly, the battery section highlights current developments, emphasizing the significant gains in cycle stability and capacity retention shown in battery systems based on nanocellulose. Because of its outstanding features, such as lightweight, flexibility, and steady cycling performance, biopolymeric materials in supercapacitor applications have received a lot of interest. Furthermore, nanocellulose/other biopolymers are also more sustainable, recyclable, and safer to use as a replacement for heavy metals in battery electrodes.

Solar cell

Biomaterials are abundant, renewable, and sustainable; they are currently being considered for application in solar cells. The devices that convert solar energy into electric energy must have a broad surface area with excellent ion transport properties to do it effectively [**98**]. Paper-based solar cells using CNF (cellulose nanofiber) and silver nanowires have a good conversion efficiency of 3.2%, the same as ITO-glass-based solar cells. The cellulose nanofiber paper retained its high conductivity under and after folding because of its strong attraction and the high degree of entanglement that existed between the cellulose nanofibers and the silver nanowires [**99**]. The solar cells made on CNC substrate have good dark-light rectification and 2.7% power conversion efficiency (PCE). Additionally, they showed that solar cells are divided into their parts and recycled using low-energy methods at room temperature, paving the way for solar cell technology that is truly recyclable [**100**].

Wang developed a unique nanocellulose/epoxy hybrid substrate strengthened by aminated CNFs for providing a remedy for the flexible solar cell (FSC) electrode substrate's poor thermal characteristics while maintaining high optical transparency. Thermal and mechanical characteristics of the developed hybrid substrate are significantly enhanced by A-CNF's high interfacial interaction and compatibility with epoxy matrix. Results showed an improvement in thermal and mechanical properties. The hybrid substrate maintained its transparency even after CNF insertion. Additionally, the PEDOT: PSS conductive layer that was applied to the substrates had consistent conductivity of about 835 S/cm, further there was no electrical degradation even after being exposed to severe temperatures. They anticipate that CNF/Epoxy hybrid substrate open new possibilities for fabricating high-performance and inexpensive flexible solar

cell (FSC) devices through quick and environmentally friendly processes [**101**].

A new beginning in the history of renewable energy has been written, thanks to the rapid advancement of metal halide perovskite solar cells. To achieve high efficiency and long-term stability, the active layers should be made of perovskite materials of superior quality. Here, the perovskite layer is mixed with the incredibly cheap and environmentally friendly polymer ethyl cellulose (EC), to create perovskite films with increased crystal grain size and lower fault density. The devices produced with the EC additive show an improvement in all device characteristics as well as an average power conversion efficiency improved from 17.11 to 19.27% [**102**]. Due to the high-power conversion efficiency (PCE) and lower processing costs, perovskite solar cells (PSCs) have emerged as one of the most promising alternatives to traditional silicon-based solar cells for solving today's urgent energy problems. The petroleum-based polymer substrates used by current flexible PSCs would cause more "white pollution" if they were discarded. Therefore, the development of flexible PSC alternatives that are green, biodegradable, and affordable will be quite beneficial. Here, transparent nanocellulose paper (NCP) covered with an acrylic resin has been developed as a substrate in the fabrication of flexible PSCs that are easily disposable and biodegradable. These NCP-based PSCs have a PCE of 4.25% and a power per weight of 0.56 W/g, which is the ratio of power to device weight. The flexible PSCs demonstrated good stability as well, maintaining >80% of their initial efficiency even after 50 bends. The NCP-based substrates are used in other electronic systems as well, which could lead to the success of the next generation of flexible, green electronics [**103**].

Fig. 3 depicts the creation of NCP-based PSCs. First, NCP was created by combining a viscous solution of natural cotton nanocellulose with acrylic resin to create a waterproof coating on its surface. Cotton was used to make NCP since it is a cost effective, cellulose-rich raw material. It should be highlighted that the produced pure NCP was susceptible to moisture and developed severe wrinkles as a result of nanocellulose swelling, making it unsuitable as a substrate for flexible electronics, notably flexible PSCs made using a solution-processing technique (e.g., poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) is a water-based reagent). Second, flexible PSCs were created on the NCP-based substrate. In order to construct PSCs, they adopted a standard completely solution-processed technique because the NCP-based substrate maintained its stable structure in water. Spin-coating was employed to create a transparent conductive anode on an NCP-based substrate using PEDOT: PSS (PH1000) doped with ethylene glycol and Triton-X 100. The same spin-coating technique was used to create various functional layers, such as the photoelectric active layer (perovskite layer), hole transport layer (PEDOT: PSS 4083) and electron transport layer [**103**].

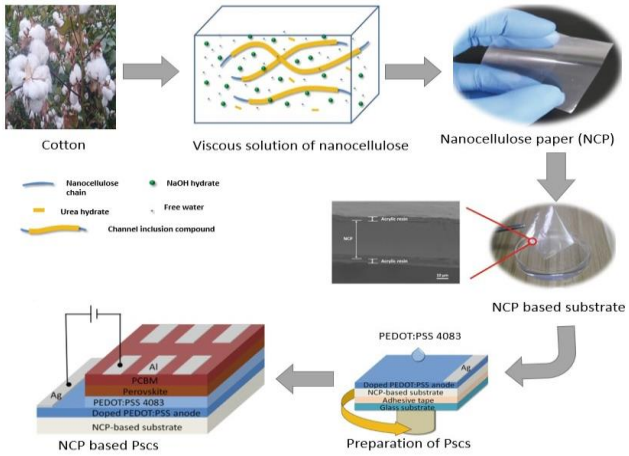

Fig. 3. NCP-based substrate and NCP-based PSCs were prepared. Cotton used to extract the nanocellulose, creating a viscous solution. Nanocellulose was dissolved in a viscous solution to create NCP. Acrylic resin was used to coat the NCP, creating a waterproof layer. The creation of PSCs based on NCP. Ref: [103]

On the NCP-based substrate, we have successfully manufactured flexible PSCs. The top-performing device displayed a PCE of 4.25%, a V_{oc} of 0.69 V, a J_{sc} of 17.46 mA cm^{-2}, and fill factor (FF) of 34.6 %, with no hysteresis shown in **Fig. 4(a), Fig. 4(b)**. over the whole visible spectrum. With an average value of over 3% for more than 80% of the devices, good repeatability was attained in **Fig. 4(c)**. The NCP-based PSCs demonstrated better stability after 50 bends with a 15 mm curvature diameter, keeping more than 80% of their initial efficiency in **Fig. 4(d)** [**103**].

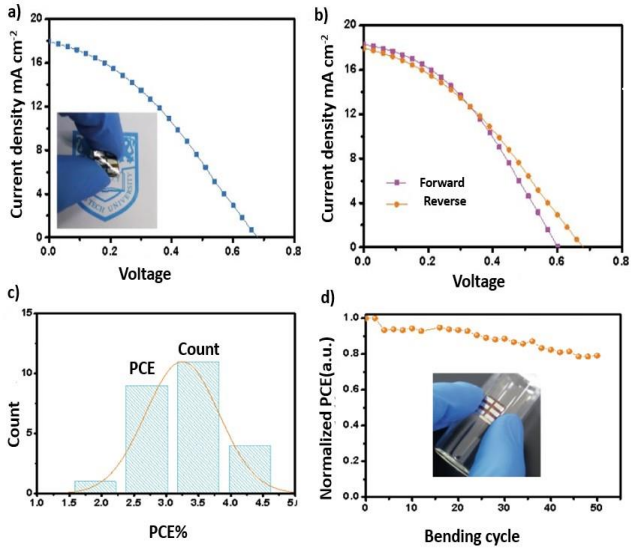

Fig. 4. (a) NCP-based PSCs' devices. Analysis of device based on current density-voltage (J-V). Digital image of bending NCP-based PSCs is shown in the inset. **(b)** J-V curves when scanning in both the forward and backward directions. **(c)** A Gaussian distribution was fitted to a PCE histogram representing 25 devices. **(d)** Characterization of normalized PCE-bending cycles with a digital image of a hand-bent glass bottle [103].

Their comparatively low energy conversion efficiency is one of the main problems with nanocellulose based solar cells. Compared to other materials utilized in solar cells, such CdTe and graphene, nanocellulose materials are not as conductive or optically efficient. The power conversion efficiency suffers as a result. The long-term durability of nanocellulose solar cells may be limited by the material's susceptibility to deterioration when exposed to moisture or ultraviolet light. The cost of producing premium nanocellulose materials may have an effect on how economical nanocellulose solar cells are. Their performance is susceptible to various factors, including humidity, temperature fluctuations, and exposure to pollutants. The transition from lab-scale prototypes to commercial production can also be facilitated by working with industry partners who have large-scale manufacturing experience. Assessing the real-world performance of nanocellulose solar cells can be aided by conducting accelerated aging studies and long-term outdoor testing. More durable and dependable solar cell designs can be developed with the use of these data.

Supercapacitor

Researchers are focusing on energy as a result of the ways in which the world is changing. More work is being put into developing and perfecting energy storage technologies in this area. Supercapacitors (SC) have gained popularity recently as energy storage devices that are designed and manufactured similarly to batteries. The SCs, also known as electrochemical capacitors or ultracapacitors, use high surface area and thin dielectric electrode materials to obtain higher capacitance than traditional capacitors [**104-107**]. Furthermore, currently, the electrode materials in supercapacitors have already received a lot of attention as carbon-based materials due to their accessibility, affordability, outstanding mechanical and electrical properties, high power density, corrosiveness, and resistance properties. Except when functioning or composited with sulphides, metal oxides or carbon nanotubes, graphene, and activated carbon are extensively utilized materials for producing supercapacitors with electric double-layer features [**108-109**]. The cycle endurance of activated carbon is the operating potential that can be achieved at 1.0–3.0 V very high, over 10,000 cycles. MXenes, also known as transition metal nitrides and 2D carbides were regarded to be a promising electrode material for SC due to their hydrophilicity, high electrical conductivity and outstanding electrochemical properties [**110-111**].

Supercapacitors cannot store as much energy as batteries but in the same volume can deliver multiple times more power [**112**]. When high power is needed, they can be utilized alone, or in conjunction with batteries or fuel cells [**113,114**]. While fuel cells offer great energy density, supercapacitors offer high power density. Depending on the materials used for the electrodes, supercapacitors are classified into three categories: i) Supercapacitors with an electrochemical double layer (EDLC) and (ii) hybrid supercapacitor iii) Pseudo-supercapacitors. Comparably, metal oxides and conducting polymers can be used to categorize pseudo capacitors [**115**]. Sheng *et al.* produced a high-performance and flexible fiber-based supercapacitor using wet spinning to create binder-free electrode from synthetic PPy, 2,2,6,6-Tetramethylpiperidine-1-oxyl (TEMPO)-oxidized BC (TOBC), and graphene oxide (GO) [**116**]. They reported to improve the electrode's overall performance, improve electrolyte infiltration, and have enough mesopores to facilitate charge and ion transfer and also get better electrochemical performance even after cycling and bending, better than previously reported graphene flexible supercapacitors. Similarly, Wang *et. al.* made nanocellulose/graphene/MnO2(Manganese dioxide) based aerogel electrode for supercapacitor using hydrothermal process to create excellent SC with high specific capacitance and better capacitance retention after 5000 cycles [**117**]. The creation of high-performance graphene and biomass-based supercapacitors, as well as other increasingly well-liked sustainable energy devices, the electrode can serve as a reference. Researchers fabricated PPy@BC/Mxene ($Ti_3C_2T_x$) composite film via in situ polymerization enhanced electrochemical performance high-performance SCs electrode made of conductive polymer as freestanding supercapacitor electrode [**118**].

Since most conductive materials, including carbon, polymers and metallic particles are brittle, they are typically combined with a supple and flexible substrate to create flexible membrane electrodes for supercapacitors that are light and stand alone. Nanocellulose's very porous structure makes it the perfect substrate for incorporating a lot of conducting materials, and its porous structure also makes it easier for ions to pass through [**124**]. Single- and multi-walled carbon nanotubes, graphene oxide, reduced graphene, and graphite are examples of conductive carbon materials. Great conductivity and higher tensile strength are typical characteristics of conductive carbon materials. Among these, graphene, a two dimensional and atomically thick material has exceptional mechanical qualities like Young's modulus [**72**] While carbon nanotubes are known for their incomparable tensile strength. Carbon compounds could be blended with nanocellulose or deposited onto the nanocellulose's film surface. Due to the possibility of the carbon particles becoming trapped inside the substrate and improving the conductivity of the composite membrane, blending often enables the incorporation of more carbon particles into the nanocellulose substrate than surface coating [**125,126**].

Researcher fabricated MXene/CNFs/porous carbon (PC) porous and flexible hybrid film electrode via simple method of vacuum filtration method. The free-standing hybrid films and the flexible gel electrolyte used to create a quasi-solid-state supercapacitor with a thinner thickness of 0.2 mm and high flexibility [**119**]. A team of researchers designed and fabricated CNF/CNT/RGO based carbon

aerogel electrode via freeze drying and carbonization, obtained with ultralight, superior hydrophilic and compressible properties. The dense honeycomb pore structure of composite aerogel supported faster ion transport while also efficiently transferring stress [**120**].

Past researches have witnessed progress in the fabrication and development of nanocellulose and graphene for energy storage materials toward supercapacitors by improving the electrode's surface area as well as specific capacitance [**121**]. Various nanocellulose-based composites for flexible supercapacitors are compared in terms of their electrochemical performances and production methods, shown is **Table 3**. Porous carbon, another widely explored, electrode material plays a crucial factor in influencing supercapacitor performance. Because of its high specific surface area (SSA), high chemical stability, customizable pore size, and environmental friendliness. porous activated carbon (AC) is extremely attractive for an improving properties of electrode material for supercapacitors [**122**]. Usually, high-quality commercial porous activated carbon is used in electrical double layer capacitors, and it is more expensive compared to ordinary grade of activated carbon. Overall, biomass has evolved into an ideal source of carbon to produce porous carbon composites, because of higher electrical conductivity, consistent chemical and physical properties, and inexpensive cost.

Several other aspects also influence the performance level of supercapacitors, including electrochemical properties of the selection of electrolyte, the electrode, frequency responsiveness, charge transport resistance and the voltage range of electrodes. When compared to graphene-based materials, biopolymers offer steady retention cycling capacity with somewhat lower specific capacitance. The combination of graphene and bio-polymeric materials can give a noticeable result. The outstanding result is closely related to the large specific area and that encourages fast ion adsorption-desorption that can provide an effective transport channel. The process for making 3D porous CNF/MnO_x - m composite for electrodes in supercapacitors is shown in **Fig. 5.** Combination of $Mn(OAc)_2$ solutions with the nanocellulose suspension of various solute concentrations, freeze-dried, and formed within hybrid freestanding aerogels of specific shapes and sizes. When compared to nanocellulose aerogels, the generated hybrid aerogels didn't appear to be significantly different. Hybrid aerogels were converted in the CNF/MnOx-based composite aerogels after being calcined at 850 °C in an argon environment, and the hues became black. The shape of composite was kept, but their volume decreased by roughly 40%. Consequently, the bulk density rose. The composites, meanwhile, showed elastic and flexible characteristics [**123**].

Fig. 5. Schematically diagram of the creation of Carbonized 3D porous (CNF)/MnO_x based composite electrode for supercapacitors [123].

Galvanostatic charge-discharge (GCD) values within 0.5 to 0.5 V were used to evaluate the CNF/MnO_x electrodes. The charge-discharge curves are nearly triangular, including a minor internal resistance drop in **Fig. 6(a)**. Ion intercalation along with de-intercalation, as well as redox processes, occur on the MnOx surface during speedy charge-discharge. The equivalent series resistance phenomena are frequently connected with the occurrence of an internal resistance drop appearing in the start of a discharge. The CNF/MnOx-4 electrode, at the same current density, outperforms the others. The discharge curves based on the CNF/MnOx-4 electrode show a closely linear

response at varying current densities ranging within 0.25 to 2.0 A/g, with discharge times ranging from 300 s to 30 s in **Fig. 6(b)**. May be used to compute specific capacitance from discharge curves, which exhibits a declining trend as current density increase in **Fig. 6(c)**. The Nyquist plot shows general diffusion-controlled Warburg capacitive behaviour with low-frequency diagonal line and a high-frequency tiny, depressed semicircle. The Nyquist plot of the EIS measurements indicates an intercept of 1.1 at the actual impedance (Z′) in **Fig. 6(d)** [**123**].

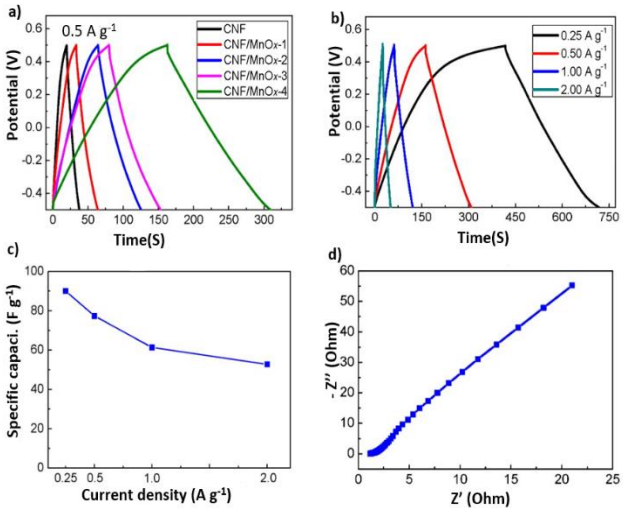

Fig. 6. Galvanostatic electrode fabricated by CNF/MnO_x based electrodes' charge-discharge (GCD) curves. **(a)** Measured at constant current of 0.5 A/g, the charge-discharge curves for three CNF/MnO_x electrodes. **(b)** CNF/MnO_x-4 electrode charge-discharge curves for various current densities. **(c)** The CNF/MnOx-4 electrode's specific capacitance in relation to current density. **(d)** A CNF/MnO_x-4 electrode Nyquist plot [123].

The majority of research on supercapacitors made from nanocellulose has focused on using the material as a substrate or spacer in composites. As was already noted, nanocellulose and other conductive materials are generally combined to form the electrodes of supercapacitors. Supercapacitors with high capacitance and high energy density are currently difficult to manufacture. Researchers must therefore add alternate materials (like sodium lignin thiosulfonate, polypyrrole, etc.) to nanocellulose/graphene and nanocellulose/MXene composites to further enhance the electrochemical performance of the electrode. Additionally, new properties can be obtained by altering the composite structure. Future study will concentrate on number of components, distribution, shape and altering the structure and in nanocellulose and graphene/MXene composites in order to increase the synergy between them [**127-129**].

Supercapacitors based on nanocellulose have enormous potential for energy storage, but they also present a unique set of difficulties. When compared to conventional energy storage, supercapacitors based on nanocellulose frequently have a lower energy density. This restricts their capacity to hold a lot of energy. To improve energy density, scientists can investigate novel materials or hybrid architectures that blend nanocellulose with other high-capacitance materials. This could entail creating nanocomposites or working with materials that are pseudocapacitive. Because nanocellulose materials are often less conductive than metals or other carbon-based materials, supercapacitors may not be able to charge or discharge energy as quickly. The total performance of nanocellulose-based supercapacitors can be increased by increasing the electrical conductivity of nanocellulose by chemical modification or the inclusion of conductive additives. Supercapacitors' compatibility, affordability, scalability, and environmental sensitivity are other key problems. Coatings, economical production techniques, and the creation of hybrid energy devices can all help to solve these problems.

Table 4. Performances, techniques and retention capacities of various nanocellulose based composites for flexible supercapacitor electrode.

No.	Electrode material	Process	Advantages	Dis-advantages	Capacitance	Energy density	Power density	Electrolyte	Capacitance retention	Ref.
1	Poly (3,4-ethylenedioxythipohene) (PEDOT) and nanocrystalline cellulose	Electrochemical polymerisation	High surface area, flexibility and mechanical stability	Limited scalability, degradation	117.02 F/g at 100 mV/s	11.44 Wh/kg	99.85 W/kg	1M KCL	After 1000 cycles - 86%	[130]
2	Al-CNC (Aluminium-cellulose nanocrystal) /PC (porous carbon)	carbonization	Higher conductivity, tunable porosity	Incompatibility issue	804 F/g at 1 A/g	18.2 Wh/kg	(208.33 W/kg	hydrogel Al - CNC	After 6000 cycle - approx 92%	[131]
3	PANI/CNT (carbon nanotube)-CNC (cellulose nanocrystal)/ PVA (polyvinyl alcohol)-PAA (poly acrylic acid)	Directional electrospinning by thermal treatment	Multifunctionality, cycling stability	Complicated synthesis, higher cost	164.6 F g^{-1}	13.8 Wh kg^{-1}	200.3 W kg^{-1}	PVA-PAA–KCl gel electrolyte	After 2000 cycles - 92%	[132]
4	CNCs and multiwalled carbon nanotubes (MWCNTs) aerogel with Ppy	In-situ polymerization/freeze drying	Flexible, light weight	Low scale	2.1 F cm^{-2}	--------		0.5 M Na_2SO_4	Stability over 5000 cycles- 80%	[133]
5	CNC/MnO2	facile one-step in situ-controlled synthesis	Good performance	Finite scalability	306.3 F g^{-1}	-----	42.59 Wh kg^{-1}	PVA/KOH	After 2000 cycles - 64.5%	[134]
6	PVA-GO-MnO_2(Manganese dioxide)/PEDOT	electrospinning and electro-polymerization	Adaptability	Low scalability	144.66 F/g	9.60 Wh/kg	243.72 W/kg	1M KCL	Over 1000 CV cycle – 91.18%	[135]

7	CNC/CNF	Atomic layer deposition (ALD)	Uniformness	Complex processing	152 F g^{-1}	------	----	2M KOH	After 10,000 charge/discharge cycles at 2 A·g^{-1} – 98.5 %	[136]
8	N-CNC	Pyrolysis	High conductivity	Sensitive process	352 F g^{-1}	39.85 Wh kg^{-1}	48.8 Wh kg^{-1}	1M H2SO4	After 2000 cycles – 94.5%	[137]
9	Hierarchically porous N-doped carbon-CNC	Carbonization	Higher conductivity	High cost	172 F/g @ 100 mA/g	23.75 Wh/kg	50 W/kg	6M KOH	After 5000 cycles – 94.5%	[138]
10	nitrogen-doped porous carbon (NPC) and CNC	Carbonization	Tailored porosity	Complex processing	570.6 F/g at 1 A/g	23.75 Wh/kg	50 W/kg	6M KOH	After 1000 cycle – 91.2%	[139]
11	Cellulose nanofiber from BC/PDA (poly dopamine)- Fe+2	Freeze drying and carbonization	Sustainability	Time-consuming	219F g^{-1} at 10 A g^{-1}	10.07 Wh kg^{-1}	1.0 KW/kg	1 M H_2SO_4	cyclic stability within 10000 cycles – to 95%	[140]
12	(cobalt oxide) Co3O4@CN	Sol-gel and carbonization	Chemically stable, higher capacitance	High cost	Approx 214 F/g at 1A/g	10 W h/kg	----	3 M KOH	Even after 5000 cycles – 94%	[141]
13	CNF-N-doped nano porous carbon (NPC)-hybrid zeolite imidazole(HZ)	In situ chemical method via pyrolysis	Highly porous, high capacitance	Sensitive process	146 F/g at 1 A/g	----	----	1 M KOH	After 2000 cycles - 90%	[142]
14	TEMPO-CNF/MnO2/AC	Hydrothermal process via pyrolysis	Collaborative effect	High cost	171.1 F/ g @ 0.5 A/g	8.6 W h/kg	619.2 W /kg	1 M $LiPF_6$	After 5000 cycles at 3 A/g – 98.4%	[143]
15	Mxene ($Ti_3C_2T_x$)/CNF/porous carbon (PC)	vacuum-filtration method	Tunable porosity, higher conductivity	Higher materias cost	143 mF cm^{-2}	2.4 µW h/cm^{-2}	17.5 µW/ cm2	PVA/KOH)	After increasing the power density by 100 folds - ~50%	[119]
16	Nanocellulose derived hierarchical porous carbon HPC/$NiCo_2O_4$	Pump filtration	Highly porous, Synergistic effect	Challenging process	64.83 F g^{-1} @ 0.25 A g^{-1}	23.05 W h/kg	213 W/kg	6 M KOH	After 1000 cycle at 10 A/g – 96.8%	[144]
17	CNF/RGO (reduced graphene oxide)/Tin oxide (SnO_2)	Hydrothermal reduction via free-drying process	Tuneable porosity, enhanced conductivity	High cost material	4.314 F cm^{-2} at 1 mA cm^{-2}	----	----	1 M H_2SO_4	Stability for 2000 cycles with retention up to at 10 mA cm^{-2} - 60.47%	[145]
18	Bio-(Activated carbon)AC/(Reduced graphene oxide) rGO/CNF	self-assembly by one step process	High conductivity	Limited scaling	812.2 mF cm^{-2} at 1 mA cm	0.365 mWh cm^{-2}	18,000 mW cm^{-2}	PVA/Na_2SO_4	After 5000 cycles – 99%	[146]
19	Cellulose nanofibers (CNF), reduced graphene oxide (RGO) and polypyrrole (Ppy)	vacuum-filtration with chemical reduction	Highly porous	Complex processing techniques	625.6 F/g	1.7 Wh/kg	11 kW/kg	1M H_2SO_4	After 5000 cycle – 75.4%	[147]
20	Graphene oxide(GO) and Cellulose nanocrystal(CNC)	Non-liquid-crystal spinning via chemical reduction	Tunable properties	Limited scaling	208.2 F/cm^{-3}	5.1 mW h cm^{-3}	496.4 mW cm^{-3}	PVA/H_2SO_4	After 1000 cycles at 1.0 A/g - 92%	[148]
21	Room temperature ionic liquid(RTIL)-CNT–cellulose	Paper making process and CNT and RTIL incorporate inside cellulose paper	Flexibility and light weight	Low ion diffusion	36 F/g	13 Wh/Kg	1.5 kW/kg^{-1}	6 M KOH and non-aqueous RTIL	very good performance over 100 charge and discharge cycle	[149]
22	CNF/Reduced graphene oxide(RGO)/CNT aerogel electrode	Freeze drying	High surface area	High cost	252 F/g @ 0.5 A/g	8.4 µWh cm^{-2}	9.5 mW cm^{-2}	H_2SO_4/poly(vinyl alcohol) (PVA)	After 1000 charge-discharge cycles at a current density of 1 A/g – 99.5%	[59]
23	Cellulose nanofibers (CNFs)/molybdenum disulphide (MoS2)/reduced graphene oxide (RGO)	Supercritical CO_2drying	High conductivity	Complex processing	916.42 F/g @ 2 mV s^{-1}	22.8 W h/kg	4.3 kW/kg	Polyvinyl alcohol/ H_2SO_4	After 5000 charge–discharge cycles at a current density of 0.5 mA cm^{-2} - 98%	[150]
24	(poly pyrole)Ppy@ TEMPO oxidised bacterial cellulose (TOBC)/rGO	Wet spinning	Enhanced conductivity	High cost	391 F/g	8.8 mWh cm^{-3}	49.2 mW cm^{-3}	1M H_2SO_4	After 5000 cycles – 79%	[116]

*PEDOT Poly 3,4-ethylenedioxythipohene, Al-CNC-Aluminium-cellulose nanocrystal, PC-Porous carbon PANI-Polyaniline, CNT-carbon nanotube-CNC- Cellulose nanocrystal, PVA Polyvinyl alcohol, PAA- Poly acrylic acid, MWCNT- Multiwalled carbon nanotubes, MnO_2 – Manganese dioxide, GO- graphene oxide, Co_3O_4, - cobalt oxide, Ppy- Polypyrrole, RTIL- Room temperature ionic liquid, PDAPoly dopamine, molybdenum disulphide (MoS2), HPC- hierarchical porous carbon, SnO_2- Tin oxide, NiCo2O4- Nickel cobaltite, MoS_2- Molybdenum disulphide.

Lithium-ion battery

Lithium-ion batteries are extensively used in mobile phones, laptops, and electronic vehicles. All batteries depend heavily on separators. Separators, a crucial LIB component, play an important role in the electrochemical performance, safety, lifetime and of LIBs and are essential for the long-term sustainability of energy storage systems. [**151-157**]. Their foremost function is to maintain a distance between the positive and negative electrodes in an electrochemical cell to inhibit electrical short circuits although simultaneously allowing quick movement of ionic charge carriers required for completing the circuit during the flow of current. Consequently, biomaterials are rarely used as separators. Separators, on the other hand, provide routes for lithium-ion migration between two electrodes, and that necessitates high Li-ion conducting efficiency in liquid electrolytes [**158-159**]. Nanocellulose has surfaced as a viable contender to transform Li-ion battery technology among the cutting-edge materials that are garnering interest. Because of its special qualities, nanocellulose is a good choice for acting as both an electrode and an electrolyte in Li-ion batteries. The unique properties of nanocellulose as an electrode material—such as its high surface area, mechanical flexibility, and adjustable porosity—offer potential to enhance Li-ion batteries' electrochemical performance. To solve problems like cycling stability, energy density, and rate capability, high-capacity anodes and cathodes can be designed using nanocellulose in this context. On the other hand, nanocellulose offers a sustainable and environmentally beneficial substitute for traditional liquid and polymer electrolytes as an electrolyte material. Its natural benefits—high ionic conductivity, excellent thermal stability, and nonflammability—offer the chance to improve Li-ion battery longevity and safety. Furthermore, all-solid-state battery architectures, which offer higher energy density and fewer safety concerns, can be made possible by nanocellulose-based solid-state electrolytes.

Nanocellulose fibres have noticeable advantages and have received more attention than traditional synthetic materials because of their unique benefits like abundant availability, high stability, eco-efficient, sustainability and eco-friendly which not only satisfy the requirement of sustainable development, but also enhance the safety and shelf life (lifespan) of LIBs [**29,160-164**]. For the preparation of LIB separators, different types of biopolymer fibres have been used, including cellulose microfibril [**165,166**], cellulose nanofibers [**167**] Because of their beneficial qualities, like great wettability to the electrolyte, great mechanical capabilities, non-toxicity, lightweight, and high thermal stability, biopolymer fibres have recently attracted a great attention as a substitute material for LIB separators [**168**]. Researchers said that the high potential of CNC makes it a desirable choice for manufacturing carbon compounds, which could make for a promising electrode for lithium-ion batteries. However, being electrically non-conductive, CNC cannot be employed directly in the manufacturing of batteries. Therefore, there is a lot of research on using pyrolysis through turn CNC into conducting materials. In one research, nanocrystalline cellulose is transformed into a highly conductive carbon material via pyrolysis, that is employed in lithium-ion batteries by heating it to high temperatures. The results obtained demonstrate an excellent electrochemical performance, discharge capacity for the CNC 800 and CNC 800-SnO_2 electrode in the Lithium-ion battery was 220 mAh/g and 380 mAh/g respectively, after 30 cycles [**169**].

One researcher successfully fabricated CNC-SnO_2NF composites via hydrolysis and CNC becomes extremely conductive as a result based on the pyrolysis this takes place while the annealing of nanoparticles, which enhances the electrochemical performance of the LIB electrode materials. For the development of nanocellulose and metal oxide composites, CNC is a suitable effective protectant, green, non-toxic, and potentially applicable due to its low cost, lightweight, flexibility, and environmental friendliness, ability to standing promising future electrode [**170**]. Another work optimizes novel nanocellulose-based (Gel polymer electrolyte) GPE was created by acetylating CNFs (ACNFs), followed by solvent casting and electrolyte immersion. This GPE has a high electrolyte absorption and stable cycle performance. Report says generated acetylated CNF (GACNF-3) based GPE displayed the good electrolyte, 301% absorption capacity and 0.65 is the ion transference number Li^+ at the ideal degree of substitution (DS) value of 2.37. ACNF precursor films have good thermal stability, high flexibility, and outstanding mechanical strength, Li-ion movement, very low electrolyte resistance, high retention capacity 88% after 100 cycles [**171**].

Researcher fabricated nanocomposite by thermally embedded SnO_2 nanoparticles in cellulose nanofiber. The observed outcomes demonstrate the finished product, significantly enhancing electrochemical performance and inhibiting SnO_2 accumulation and volume expansion. Additionally, the SnO2 nanoparticles stability, during the process of charge-discharge is enhanced by the amorphous carbon structure. This nanocomposite electrode, when used in lithium-ion batteries, could, after 200 operating cycles, reach at higher specific capacity of 619 mAh/g. The noteworthy aspect of studying and creating LIBs electrode materials, performing at higher current densities, enhances the ability of nanocomposite electrodes to work at higher current densities [**172**]. Several research in this field have shown significant promise in ushering in an exciting new generation of high energy and long-lasting batteries for various applications such as windmills, transportation, solar panels, and space applications. **Fig. 7(a)** depicts a theoretical application of nanocellulose in the lithium- sulphur batteries [**173**] and **Fig. 7(b)** shows the porous shape facilitates electron movement within the electrodes and results in a dendritic-free deposition of lithium (Li).

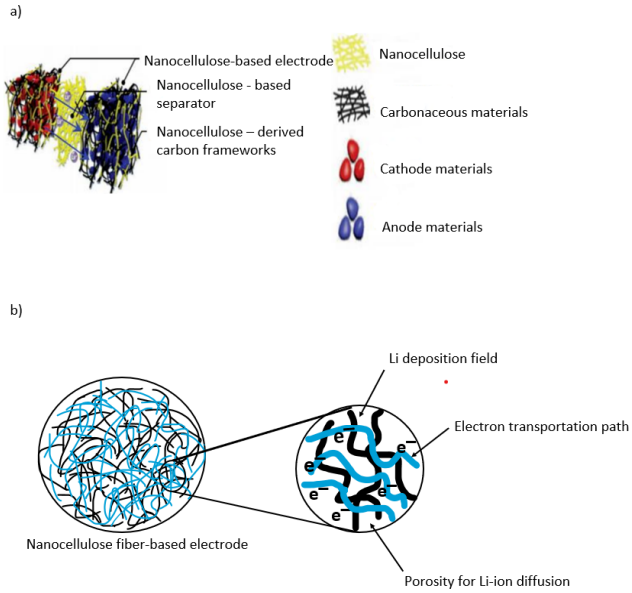

Fig. 7. (a) Illustration of the nanocellulose in a lithium battery [173] (b) Nanocellulose based electrode for Li-ion batteries.

Similarly, **Fig. 8(a-e)** shows a CNC/freestanding Si - based good flexible anode was fabricated using silicon nanoparticles, nanocellulose, carbon nanotubes (CNT) as foundation blocks. The robust attachment of Si particles to the carbon nanotubes and highly porous nanocellulose matrix produced a superior charge-discharge cycle and outstanding specific capacitance performance, which is considerable for LIBs [**174**].

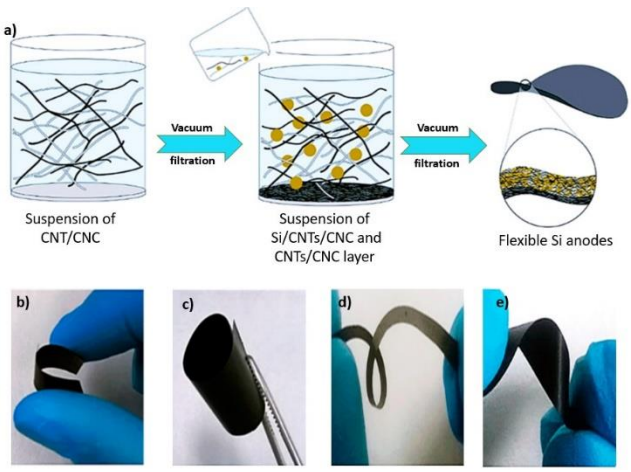

Fig. 8. (a) A schematic depicts the manufacturing technique for creating the flexible CNC-CNT-Si composite electrode by filtering. **(b)** Pictures of electrodes subjected to bending. **(c)** rolling **(d–e)** twisting [174].

Furthermore, the suspension of the nanocellulose and the monodisperse colloidal polystyrene spheres were combined to successfully fabricate a separator based on nanocellulose with a unique microstructure. The findings of the research demonstrate that porous membranes have favourable electrochemical properties, such as a higher ionic conductivity is 1.24 mS cm^{-1}, a basic interfacial resistance with a wide electrochemical stability window of 4.5 V at ambient temperature, thanks to its peculiar microstructure with improved permeability [**175**].

In continuation of membranes, by using a one-step crosslinking technique, a novel polyethylene glycol (PEG) based GPE trapped in a cross-linked cellulose structure is created. During the PEG concentration was increased from 2.5% to 20%, the results showed that composite gel membrane had excellent tensile strength between 33.92 MPa and 211.06 MPa as well as bending resistance. When the PEG concentration was at 5%, it had a significant ionic conductivity $3.31*10^3$ S/cm and an exceptional lithium-ion transfer number was 0.63. By using this gel polymer electrolyte, the built Li/GPE/NCM523 batteries showed an initial discharge capacity of 159.3 mAh/g and a coulomb efficiency of 85.52% at 0.2C. We believe that this high performance and improved mechanical strength GPE membranes used in LIBs might be made affordable and biodegradable [**176**]. The electrochemical performances and processes of several nanocellulose-based composites for lithium-ion batteries, such as electrodes, electrolyte, and separator, are compared and summarized in **Table 5**. For the first time, Lee and colleagues successfully produced cellulose based paper from a suspension of cellulose nanofibers (CNFs) and investigated its potential use as a LIB separator. The produced cellulose membrane resulted in high electrolyte uptake capacity and good electrolyte wettability, which helped to produce separators with good electrochemical performance, high ionic conductivity and low interfacial resistance [**177,178**]. The charge and discharge processes are the two fundamental operations that take place in lithium-ion batteries (LIBs). Because it serves as a physical barrier between anode and cathode, offering mechanical support and working to prevent electrical short-circuits, the separator membrane is a crucial part [**84**]. Contrarily, it must permit ionic flux while blocking electrode electric contact [**154**]. The separator serves as an electrolyte as well during in the charge and discharge cycles as a reservoir for the transportation of ions [**156**].

Nanocellulose-based paper and film materials have been utilized in LIBs to improve energy density, power density, cycle life and safety, by affecting the cell kinetics because of their superior mechanical and thermal properties together with good hydrophilicity. The nanocellulose-based separator/electrolyte must have a high pore structure for encouraging ion migration rate during the electrochemical reaction in order to improve the electrochemical performances of LIB. To increase the mechanical flexibility of the nanocellulose-based electrolyte, a crosslinking network has been developed [**84**]. Biopolymer microfibril and cellulose based membranes possess significant drawbacks to overcome, like weak mechanical strength and wide pores, which are detrimental to the cycle stability and safety of LIBs because dendritic Li causes a short circuit in the cell [**154**]. It's interesting to note that the biopolymer nanofibers nano-meter-scale diameter is anticipated to be vital in giving the separators based on biopolymer

nanofibers well-controlled nano porous architectures. Many researchers were inspired to construct biopolymer nanofiber-based nano porous membranes and investigate the viability of all these LIB separator membranes by the distinctive and advantageous properties of biopolymer nanofibers, as well as their high-cost competitiveness and polarity.

Although they have certain obstacles, Li-ion batteries based on nanocellulose show promise as an energy storage technology. These are the challenges facing current research and potential solutions. When compared to conventional Li-ion batteries, Li-ion batteries based on nanocellulose frequently have a lower energy density. This constraint affects their capacity to deliver and store large amounts of energy. In order to boost energy density, researchers can investigate the creation of hybrid materials and architectures that blend high-capacity electrode materials with nanocellulose molecules. Slower charge and discharge rates may occur from the general poorer conductivity of nanocellulose materials compared to materials based on carbon or metal. Structural breakdown in nanocellulose-based batteries can result from large volume changes that some electrode materials undergo during charge and discharge cycles. Researchers might look into nanocellulose composites that have materials for electrodes that reduce volume expansion. In addition to increasing nanocellulose's electrical conductivity by the inclusion of other materials, batteries can perform better through chemical alteration. The most difficult production is large-scale and economical. To resolve this problem, create scalable production and identify raw resources that are reasonably priced.

Table 5. Performance, techniques and outcomes of nanocellulose based composite for Li-ion batteries.

No	Composite	Processes	Capacitance	Contraction (Thermal)	cycling Stability	Outcomes	Ref.
1	Nanocrystalline Cellulose (CNC)/Tin oxideSnO_2 - electrode	Pyrolysis	380 mAh/g	-----	good stability even after 100 cycles	low cost, lightweight, and good flexibility	[169]
2	3-dimensional MXene-Carbon nanotubes-Cellulose-$LiFePO_4$ - based cathode	vacuum filtration and freeze drying process	19.2 mAh/cm	64.4% coulombic efficiency	stable after 500 cycles	High cycling stability and electrochemical performance	[179]
3	(lithium titanate oxide)LTO/(cellulose nanofiber) CNF/ (carbon nanotube) CNT- based anode	In situ carbonization process	157 mAh/g	100%	96.4% stable after 500 cycles	high conductivity	[181]
4	lithium iron phosphate - nano paper (LFP)/(Carbon black) CB/CNF-based cathode	Freeze drying	8.8 mAh cm^{-2}	-------	90% after 150cycles	Increased ion mobility and enhanced strength of electrode	[181]
5	Cladophora Cellulose nanofiber/Si nanopaper/ carbon nanotube (CNT)	Papermaking	800 mAh/g	More than 95%	Highly stable after 100 cycles	good electrochemical performance, Low cost, flexible and lightweight	[182]
6	polyformaldehyde/cellulose blend separator	Solution casting technique	159.0 mAh/g	-----	70.35% stability after 500 cycles	High tensile strength and young modulus, highly porous, good electrolyte wettability	[183]
7	Cellulose modified by polyvinyl alcohol(PVA)/ styrene-co-acrylate composite (pCSA) - membrane	phase inversion method via wet process	104.1 mAh/g	less than 99%	showed good stability after 80 cycles	good mechanical strength, high porosity and wettability	[184]
8	Lignin-carbohydrate complexes (LCC) and cellulose nanofiber separator	Paper making approach	149.4 mAh/g	------	92.6% stability after 160 cycles	Fast charge transportation, highly porous, environmentally stable	[185]

*$LiFePO_4$- Lithium Iron phosphate, LCC-Lignin-carbohydrate complexes, pCSA – styrene-co-acrylate composite, SnO_2-Tin oxide, PVA, LFP-lithium iron phosphate

PERSPECTIVE AND CHALLENGES

The incorporation of biopolymers into current functional electronics is necessary to reduce electronic waste and expanded use of biopolymers in advanced and intelligent electronic systems, as well as green and renewable energy sources. Molecular engineering holds huge promise for improving the creation of high performance and stable biopolymer-based electronics for use in electronics, energy, packaging and other industries. Nanocellulose has the potential to replace the synthetic, petroleum-based materials used in electronic devices. The industry is eager to absorb and use the most current breakthroughs in order to begin producing batteries, solar cells, supercapacitors, and multi-sensing technologies using eco-friendly and sustainable

materials. Nanocellulose's characteristics can be improved by combining it with carbon compounds such as graphene oxide/reduced graphene oxide/MWNT/SWNT [186]. The primary goal of these studies is to fully integrate nanocellulose with carbon-based composites into everyday goods and products with extended life, biodegradability, eco-friendliness, and reduced e-waste.

Nanocellulose has electrical properties and is highly porous, which can change the physical or chemical properties for molecular modification and allow ion transportation. Although nanocellulose has a low cost, the manufacturing cost remains considerable. Cost-effective large-scale production remains a challenge. Many companies in various nations are concentrating on cost-effective nanocellulose based production. When utilized at low temperatures, nanocellulose has great durability. CNCs and CNFs are highly viscous, even at low concentrations, this limits their application; at higher temperatures, they cannot survive without degradation, necessitating severe base treatment. Furthermore, a high aspect ratio and a high stiffness enhance mechanical reinforcement. A balance between these crucial properties is required when selecting CNC or CNF as reinforcing material because CNFs have a substantially higher aspect ratio while CNCs are stiffer. Although a high aspect ratio is among the most coveted features of nanocellulose, most electrical applications require nanocellulose with homogenous crystallinity.

It is notable that employing nanocellulose as a building block provides composite materials with a high degree of mechanical flexibility, enabling flexible electrical energy systems (EES) for applications such as sophisticated electronics. Although nanocellulose-based materials for EES have made tremendous advances and expanded rapidly in recent years, there are still numerous barriers to overcome before such EES devices can be extensively deployed. So far, only a few ways (like in-situ polymerization and direct blending) have been proposed for mixing nanocellulose with various types of chemically active materials, although some of these have disadvantages such as low efficiency and complex processes. The development of cutting-edge preparation procedures, as well as the manufacture of nanocellulose-based composites with applicable physical and electrochemical properties, will thus continue to receive substantial attention in the coming years. Although nanocellulose gives the composite electrodes a free-standing, electrolyte, separator or membrane structure, the flexibility and mechanical strength of numerous nanocellulose-based composites are insufficient for real-world applications. Low aspect ratio nanocellulose (such as CNC) produces delicate self-assembled structures. Furthermore, because of the increasing need for portable and wearable devices, the expansion of EES systems has focused on scaling integration, efficiency, and intelligence. Currently, the majority of nanocellulose-based composites serve a single purpose with limited specific settings.

Nanocellulose-based materials have many benefits when it comes to energy conversion and storage. These materials are environmentally friendly and sustainable since they are made from plant cellulose, which is renewable. A high surface area resulting from their nanostructured nature allows for increased electrode-electrolyte contact and improved reactant interactions. Consequently, batteries, supercapacitors, and other energy storage devices benefit from increased performance metrics due to this attribute. For applications requiring flexible and wearable energy solutions, nanocellulose's mechanical flexibility makes it an appropriate choice. Its variable porosity also optimizes energy storage capacity by providing precise control over ion diffusion and electrolyte penetration. Not only does nanocellulose have good heat stability, but its remarkable mechanical strength also guarantees structural integrity in high-stress applications. Beyond its functional characteristics, nanocellulose is safe and biocompatible, addressing environmental and safety concerns in energy applications. Through modification, nanocellulose can be made more conductive, overcoming a traditional limitation associated with cellulose-based materials. This versatility enables nanocellulose to be used as both electrode and electrolyte materials, opening up possibilities for hybrid systems, all-solid-state batteries, and multifunctional energy storage devices. Additionally, the lightweight nature of nanocellulose allows for even more applications, particularly in weight-sensitive industries like portable electronics and aerospace. Finally, at the end of its life cycle, nanocellulose-based materials provide a sustainable solution, as they are biodegradable and supportive of recycling efforts.

Despite numerous reports on the compound approaches and structures of various nanocellulose-based composites with enhanced electrochemical performance, owing primarily to the proposed "synergistic effects'' between the nanocellulose and various electrochemical active materials, up till now it is unclear why these composites work so well. As a result, a fundamental knowledge of the molecular interactions between nanocellulose components and active components at the electrode/electrolyte interface is required. Because of the ion trapping in polymer chains and their interaction at different scales, the charge storage mechanisms either in gel form or solid flexible electrolytes have become more complex and difficult to study, necessitating both theoretical advances and the correct experimental instruments. This would enable the development of customized nanocellulose-based composite's usage in various EES devices. Similarly, adding an insulating nanocellulose component to composite electrodes reduces conductivity and prevents electron transportation, which has a negative influence on energy storage device rate properties and electrochemical performance. To optimize the kinetics of electrochemical reactions, active materials that interact with nanocellulose must have interfacial contact modes that maximize the equilibrium between electron mobility and fast ion diffusion. It is critical to better understand the relation between the function, structure and energy storage system of these composites. These nanocellulose-based composites can

be further used in more applications by enhancing their multifunctional dimensionalities and morphologies.

Even though several different nanocellulose-active material combinations have been proposed for the fabrication of separators and composite electrodes with superior electrochemical properties, which primarily improve from their high mechanical strength, high wettability and porous structures. However, there are currently few options for designing and controlling the pore structure of nanocellulose-based composites. It should be mentioned that the porosity and pore size of the composite materials have a considerable impact on the electrical performance of energy storage devices. Electrodes, rational pore structure design, hierarchical micropore-mesopore structure, and wide surface area all provide benefits such as increased electrode-electrolyte contact area, lowered electrolyte ion diffusion resistance, and shortened electrolyte ion diffusion length. As a result, further research into how to manage the pore structure of nanocellulose-based electrodes will be in demand in the future.

Nanocellulose has been tested to see if it can outperform synthetic polymer binders, as a substitute electrode binder for LIBs. The electrode was endowed with electrical conductivity when nanocellulose and nanocarbons were physically and chemically mixed to form a 3D porous conductive network. The use of paper electrodes without the need for binders, metallic current collectors or conductive chemicals was made possible by the nanocellulose/nanocarbon-based conductive network. The generated paper electrodes outperformed commercial electrodes in terms of electrochemical efficacy and were lightweight and flexible. Nanocellulose has also been added to separator membranes and solid-state electrolytes to enhance their mechanical, thermal, and structural strength and durability. Nanocellulose-derived carbon compounds also served well as LIB electrode materials.

However, before these processes can be applied in large manufacturing, two critical issues must be addressed. Shear, for example, reduces the aspect ratio. The second issue is the aggregate. NC re-agglomerates even after drying, making it difficult to distribute evenly in the liquid during polymer manufacturing. Some recent efforts have been made to alleviate these processing issues. However, fixing these challenges still necessitates considerable effort. In humid conditions, it is critical to increasing the durability and shelf life of nanocellulose-based solar cells.

With its outstanding tailoring capabilities, nanocellulose has constantly presented as a difficult substrate in the manufacturing of flexible electrodes, and dividers for supercapacitors and LIBs. LIBs' porosity and its homogeneous surface pore distribution are improving, and research is progressing faster in this direction. Recently, research has concentrated on the development and manufacture of 3D structures and nanocellulose aerogels in order to enhance energy density even more. First and foremost, the production of nanocellulose continues to be plagued by high manufacturing costs and long lead times. It should be able to develop new, low-cost, large-scale nanocellulose production technologies.

The manufacturing sector is eager to accept new technologies, combine them, and begin producing solar panels, batteries, electronics, photonics, and communication equipment. The following research trends may be of future interest. 1) It is critical to investigate various modifying techniques for graphene-based materials, composites and biopolymers in order to adjust their structure, content, and capabilities while considering interfacial interactions and modifications that contribute value throughout the value chain. (From novel materials to individual parts and products). 2) Finding a simple, rapid, eco-friendly, and cost-effective method for extensive preparation is critical for sustainability and marketing. Several laboratory-scale production processes have been recorded but discovering innovative ways to leverage on the versatility and distinctive characteristics of biopolymer/graphene-based composites could drive industry expansion in the future. 3) Based on their features and performance, it is critical to examine biopolymer/graphene-based composites for size management, uniformity, and compatibility. 4) Biopolymer/graphene-based composites may enhance some of their performance and property limitations by introducing an additional building component or functionalizing it with a metal or metal oxide. Metal or metal oxide nanoparticles have unique features that can improve the magnetic, conducting, chemical and electrical capabilities of a particular composite. 5) Life cycle assessment is a critical study subject for the industrialization of new biopolymer/graphene-based composites. Studies need to focus on potential changes in energy usage, human toxicity, blue water footprint and ecotoxicity. A future study on life cycle assessments of commodities in the initial stages of technological development need to be conducted. 6) More research is needed in areas that have not previously been adequately investigated, such as the impact of biopolymer/graphene-based compounds on insulator coating and on anticorrosion materials.

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CONFLICTS OF INTEREST

There are no conflicts to declare.

SUPPORTING INFORMATION

Supporting informations are available online at the journal website.

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GRAPHICAL ABSTRACT

In the context of energy conversion and storage technologies, nanocellulose plays a crucial role, as shown in the graphical abstract. This review focuses mostly on the three applications: solar cells, supercapacitors, and Li-battery. This review will provide a quick overview of the benefits and downsides of nanocellulose, as well as how it may be used in energy devices in the future and how to overcome any obstacles to its development.