

REVIEW ARTICLE

Cellulose-based smart materials: Novel synthesis techniques, properties, and applications in energy storage and conversion devices

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Abstract

There has been a significant scope toward the cutting-edge investigations in hierarchical carbon nanostructured electrodes originating from cellulosic materials, such as cellulose nanofibers, available from natural cellulose and bacterial cellulose. Elements of energy storage systems (ESSs) are typically established upon inorganic/metal mixtures, carbonaceous implications, and petroleum-derived hydrocarbon chemicals. However, these conventional substances may need help fulfilling the ever-increasing needs of ESSs. Nanocellulose has grown significantly as an impressive 1D element due to its natural availability, eco-friendliness, recyclability, structural identity, simple transformation, and dimensional durability. Here, in this review article, we have discussed the role and overview of cellulose-based hydrogels in ESSs. Additionally, the extraction sources and solvents used for dissolution have been discussed in detail. Finally, the properties (such as self-healing, transparency, strength and swelling behavior), and applications (such as flexible batteries, fuel cells, solar cells, flexible supercapacitors and carbon-based derived from cellulose) in energy storage devices and conclusion with existing challenges have been updated with recent findings.

KEYWORDS

batteries, cellulose-based materials, energy storage devices, fuel cells, hydrogels, supercapacitors

1 | INTRODUCTION

Cellulose is a naturally occurring substance found in plants that humans have used for thousands of years. Today, it plays a significant role as a raw resource for producing textiles, paper, and forest goods. Additionally, because it is renewable, sustainable,

biodegradable, and not made from petroleum, it has a substantial environmental appeal.^{1,2} A second generation of cellulosic products based on the nanometer scale extraction of cellulose has evolved in response to the increasing need for high-performance materials. By doing this, shortcomings brought on by its hierarchical structure can be eliminated, leading

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to the production of high-quality products.^{3,4} Cellulose does not exist as a single molecule in the environment but exists in the form of fibers formed by multiple (18–36) cellulose chains. These elementary fibrils are then bundled into larger microfibril bundles (macro fibrils). Additionally, hemicellulose, lignin, and micro- and macro-fibrils build the cell walls, which combine with other substances to make plants.^{5,6} Recently, there has been an increased interest in using lignocellulosic materials in developing smart bio-based hydrogels.⁷ Biopolymer-aided hydrogels, or cellulose-based (CB) hydrogels, are primarily derived from plants, animal extracts, and cellulose. Bacterial cellulose (BC) or chemically softened pristine and innate cellulose using LiCl/dimethylacetamide (DMAc), *N*-methylmorpholine-*N*-oxide (NMMO), ionic liquids (ILs), and alkali/urea (or thiourea) could be used to make these hydrogels.^{8,9} Most cellulose derivatives can be classified as ethers or esters, including methylcellulose (MC), ethyl cellulose, hydroxyethyl cellulose (HEC), carboxymethyl cellulose (CMC), cellulose acetate butyrate, cellulose acetate trimellitate, and hydroxypropyl MC phthalate.^{10–12} These materials are also being explored as an electrolyte for energy storage/electronic devices. Indeed, the continuously increasing demand for wearable electronic gadgets has driven the progress of flexible energy storage systems (ESSs) with protection, low cost, superior wearability, and high energy density (ED). Aqueous rechargeable Zn-ion batteries (AZIBs) have also been recognized as secure energy storage technologies because of their superior ion conductivity and nonflammability. As a result, choosing the suitable electrolyte is increasingly essential for wearable and flexible electronic devices that must remain stable irrespective of external shocks or even deformation. In supercapacitors, ZIBs, and Zn-air batteries, hydrogel electrolytes have received much attention because of their unique characteristics, including flexibility and H-bonding.^{13,14} Electroactive conductive polymers (CPs) have attracted considerable attention due to their promising applications, such as rechargeable batteries, electrochromic devices, supercapacitors, chemical sensors, and biosensors. Polyaniline is a promising cathode material used in rechargeable batteries, such as Zn batteries.¹⁵ This active material was applied to fabricate an aqueous rechargeable Zn-conducting polymer battery with a high specific energy.¹⁶ The article explains how to extract cellulose and cross-link it using physical, chemical, and polymerization techniques to turn liquefied cellulose and its byproducts into hydrogels.

Here, in this review article, we have discussed the role and overview of CBMs in ESSs. Additionally, the extraction sources and solvents (such as ILs, NMMO system, NaOH/urea and deep eutectic solvents

[DESSs]) used for dissolution have been discussed in detail.¹⁷ Finally, the properties, applications (e.g., lithium-ion battery [LIB], Zn-ion battery [ZIB], fuel cell [FC], solar cell and flexible supercapacitors [FSCs]) and conclusion with existing challenges have been updated with recent findings.

2 | GENERAL OVERVIEW OF CELLULOSE

The rapid commercialization and enormous developments in the human population have seriously polluted the environment. A broad range of pollutants, such as toxic heavy metal ions, inorganic anions, micro-pollutants, and organic compounds, such as dyes, phenols, pesticides, humic substances, detergents, and other persistent organic pollutants, have been found to have elevated concentrations in water in several articles published in the past few decades. The discharge of these dangerous chemicals into natural water bodies has severely upset the balance of the ecosystem, harming both plants and animals.^{18–21} Over the past few decades, several different water purification methods have been proposed and used.²² Various low-cost and effective processes, including adsorption, absorption, flocculation, and filtration, can remove pollutants from wastewater. Recently, there has been a massive increase in interest in using naturally occurring materials for the techniques mentioned above.^{23,24} The structural component of plants and some species of algae cell walls and in bacterial biofilms makes cellulose a promising candidate to become the most profuse renewable biopolymer upon the ground.²⁵ Due to its widespread availability, reusability, sustainability, and potential for surface modification, lignocellulosic material is one of the most promising uses for water or wastewater treatment. We have recently extensively summarized the use of cellulose in water remediation, pesticide removal, and other advanced applications in our previous works.^{26–29} So, here we have briefly discussed the potential of cellulose for such applications. The removal of organic and inorganic contaminants using lignocellulosic adsorbents has been the subject of numerous studies.^{30,31} When obtaining cellulose from lignocellulosic materials, the cellulose-hemicellulose-lignin complex must typically be broken down over several steps using physical, chemical, or biological means. Lignin, hemicellulose, and pectin can be effectively removed from these materials using alkaline treatments, which are inexpensive and environmentally friendly.³² A carbohydrate polymeric substance, that is, cellulose, contains carbon, hydrogen, and oxygen. It also falls under the larger category of polysaccharides, which are naturally

occurring polymeric materials. Some molecules share structural similarities with cellulose but contain additional atomic groups, such as nitrogen. Chemically, cellulose comprises repeated $-1, 4$ glycosidic linkages in linear chains. Microfibrils and macro fibrils are constantly present in cellulose, allowing it to create 3D hierarchical structures in the form of powder, film, and hydrogel.^{33,34} It is the most widely used material in the world, which has been used for many millennia as wood for construction and fire, as fibers to make paper, and as textile materials for clothes, ropes, and other uses.³⁵ The three most important ingredients in the creation of hydrogels are the monomer, initiator, and cross-linker. Diluents (such as water or other aqueous solutions) can be used to regulate the temperatures during polymerization as well as the final hydrogels' characteristics. The hydrogel mass needs to be cleaned to get rid of any contaminants from the preparation procedure. These consist of the unreacted monomers, such as initiators, cross-linkers, and undesired byproducts of side reactions, as shown in Figure 1.³⁵

One of the most notable green materials of the present day has been identified as nanocellulose (NC), which is cellulose in the form of nanostructures. NC materials are becoming more and more popular due to their excellent and appealing potential, which includes their high aspect ratio, biocompatibility, renewability, superior mechanical capabilities, and abundance. Numerous functional groups found in hydroxyls allow for a range of functionalization's through chemical processes, leading to the creation of various materials with adjustable properties. Generally, there are two primary types of NC: (1) nanostructured materials, such as cellulose microfibrils and microcrystals, and (2) nanofibers, which include BC, cellulose nanofibers (CNFs), and cellulose nanocrystals (CNCs). Natural cellulose can be used to make NC, which has a size range of a few to tens of nanometers in at least one dimension.³⁷ BC

and nanofibers are two forms of NC derived from bacterial origins.³⁸ Smart materials with a base in NC exhibit responses to external stimuli such as light, temperature, electrical input, pH, and magnetic force. The research's focus has significantly expanded to create nanostructured materials such as CNC, CNF, and BC for various applications.³⁹ It can be acquired through diverse approaches, including mechanical and chemical procedures.⁴⁰ These distinctions underscore the impact of diverse origins and extraction methods on the final product. The material commonly appears as short rod-like structures or whisker shapes, with diameters falling within the 2–20 nm range and lengths spanning 100–500 nm. It possesses a cellulose chemical composition, primarily concentrated in crystalline regions where crystallinity levels typically vary from 54% to 88%.⁴¹ NC is susceptible to surface modification due to its high surface area and abundant hydroxyl groups. Essential methods include direct chemical modification and covalent bonding of functional groups to the hydroxyl groups on the NC surface.^{42,43}

When cellulosic materials are hydrolyzed in concentrated sulfuric acid, the outcome is sulphonated nanocrystalline cellulose (NCC), with sulfate groups attached through an esterification process with surface hydroxyl groups. Aqueous NCC suspensions' physical and structural properties are influenced by two important colloidal phase behaviors: the liquid–solid transition and the isotropic–anisotropic transition. The development of colloidal glass or gel is typically favored by increasing the colloidal volume fraction or decreasing the stability of NCC particles in water. These usually display rheological characteristics that resemble those of solids, such as elasticity and yield stress. The study of colloidal suspensions includes rheology as a critical component. An increasing number of investigations on the rheology of colloidal NCC scatterings have been published in recent years due to the increased demand for solutions to formulation and processing problems in academic and industrial settings.⁴⁴ While CNF is normally produced using a variety of methods, such as mechanical, physical, and physiochemical approaches, CNC can only be created by acid pretreatment, which necessitates acid–solid hydrolysis. CNF is composed of long, thread-like bundles of cellulose molecules. Hydrogen bonds, which are created between neighboring molecules' hydroxyl groups and oxygen atoms, hold these bundles together laterally. Meanwhile, CNC has a rectangular-shaped, remarkably crystalline, short-structured NC. Their size is influenced by the temperature, time used for hydrolysis, and native cellulose raw material.⁴⁵ However, bacterial NC (BNC) is a type of NC that is produced by microorganisms.⁴⁶ BNC differs from many kinds of plant cellulose in that it has high chemical purity, high levels of polymerization,

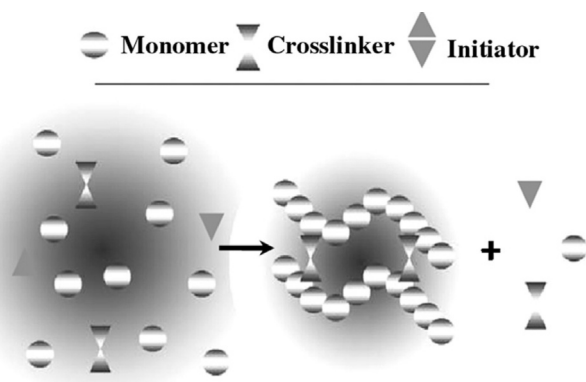


FIGURE 1 An illustration showing the preparation of hydrogel. Reproduced with permission.³⁶ Copyright 2013, Elsevier B.V.

and high levels of crystallinity. It is important to note that hemicellulose and lignin are frequently present in plant NC.⁴⁷

3 | CELLULOSE EXTRACTION SOURCES

The adsorption of heavy metals involves the utilization of many cellulose-containing compounds. One such substance that has 50% cellulose is bagasse. It can be utilized to adsorb Cu(III) and Cu(VI) ions from tannery wastewater in native and immobilized forms.^{48,49} The possibility of obtaining cellulose-enrich compounds from fruit wastes, such as orange bagasse (OB), has also been investigated. The complicated composition of OB changes depending on the species and the season. Insoluble fibers such as cellulose, hemicellulose, lignin, and soluble fibers like pectin and gums make up the fibrous structure.⁵⁰

Under mild chemical sequential extraction conditions, OB from nature and OB from industry were examined as starting materials for the manufacture of NC. Mariño et al.⁵¹ investigated the OB in nature and industrial OB was used as a starting material for the production of NC, which accounted for acid (5% v/v and 100°C) and/or alkaline conditions (NaOH 1.6%–4.0% m/v, 120°C), and bleaching with NaClO₂ (1%–3% m/v, 80°C). Comparable CNFs with 60%–70% crystallinity and great purity (above 98%) were produced by ultrasound treatment. These kinds of bagasse are utilized as dietary fiber sources in animal feed, baked goods, jam or fruit juice, meats, dairy products, sauces and dressings, and fermentation substrates.³²

A one-pot two-step homogeneous transesterification reaction in a 1-ethyl-3-methylimidazolium acetate (EmimOAc) DMSO mixed system was used to directly create a CB biomass plastic from untreated sugarcane bagasse. The lignin component was then removed by precipitating it in methanol.⁵² Cellulose can be

removed from several sources and unified into various neat bulk materials, including membranes, films, fibers, hydrogels, and aerogels. It can also be mixed with other materials to make composites. Wood that grows naturally comprises cellulose, lignin, and hemicellulose. The primary use of cellulose can be as a wood component for buildings, furniture, paper, and containerboard. Textiles, other value-added materials, and emerging uses of cellulose to compete with plastics for primary packaging all confront the issues of variable characteristics, long-term strength, and high construction prices.⁵³ Figure 2 shows a few examples of sources of cellulose extraction.

4 | CB HYDROGELS AND SOLVENTS USED FOR DISSOLUTION

4.1 | Hydrogels

The hydrogel is composed of a three-dimensional (3D) network of hydrophilic polymers. The hydrogel can hold large amounts of water or nutrients for an extended period. Hydrogels in aqueous solutions are formed by hydrated polymer networks or hydrophilic groups such as -OH, -COOH, -NH₂, -CONH₂, and -SO₃H. Since biomass resources are biodegradable, biocompatible, and biological, they have been extensively used in hydrogel fabrications. Hydrogels are first produced chemically, using either a single step or a series of steps. One cross-linking agent, for instance, has been utilized to create CB hydrogels from cotton linter pulp as a hydrogel material.⁵⁴ Zhao et al.⁵⁵ found that hydrogels are made of chitosan, which is made from chitin. Cross-linked hydrogels have been treated with dextran via the grafting procedure. Dextran and chitosan are highly hydrophilic polysaccharides, biodegradable, and capable of being pendant-functionalized. The hydrogels show

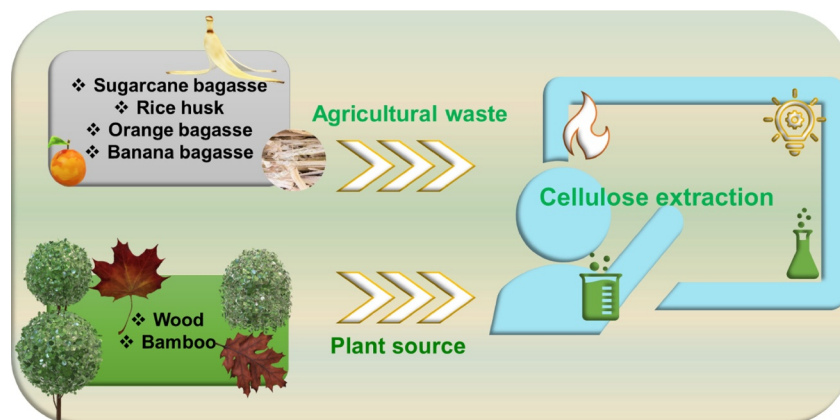


FIGURE 2 Sources of cellulose extractions.

cytocompatibility with mouse fibroblast NIH3T3 cells, exhibiting good swelling properties. As a promising carrier, biomimetic polysaccharide hydrogels have been used to treat wounds. Physical cross-linking can be used to create cellulose hydrogels from cellulose solutions. Due to cellulose's extended hydrogen-linked structure, which makes it difficult to dissolve in standard solvents, the main barrier to forming cellulose hydrogel is the lack of suitable solvents. Various solvents, such as ILs, alkali/urea (or thiourea), NMMO, etc. are used to produce hydrogel from native cellulose. By grafting copolymerization of acrylic acid, acrylamide, and 2-acrylamido-2-methyl-1-propane-sulfonic acid onto Na-CMC with a cross-linking agent and powdered montmorillonite, the superabsorbent composite was produced. A free radical initiator, potassium persulfate, was utilized. The superabsorbent composite's water absorption was 680.2 g/g, a 107% improvement over its counterpart polymer (328.2 g/g). Figure 3 depicts the predicted chemical cross-linking and grafting reaction mechanism.⁵⁶

Table 1 shows the CB hydrogels and their characteristics for energy storage devices (ESDs).

4.2 | Solvent used for dissolution of cellulose

4.2.1 | ILs system

An organic/inorganic anion and a sizable organic cation comprise molten salts or ILs. The large dimensions of their ions cause charge dispersion, which hinders the formation of a regular crystalline structure. Because of their special characteristics, ILs can be used to emphasize their strong solvation ability for a wide range of compounds and their high thermal and chemical stability, provided they are designed properly.⁶² As shown in Figure 4, cellulose is found in lignocellulosic biomass as microfibrillar three-dimensional structures with a broad range of lengths (0.1–100 μm) and widths (2–20 nm). These structures comprise films of parallelly aligned chains that are connected together. Cellulose is broken down into three main categories: CNCs, which have a diameter of 5–30 nm and a length of 100–500 nm; nano fibrillated cellulose, which has a diameter of 5–70 nm and a size of several micrometers, and BNC, which has a typical

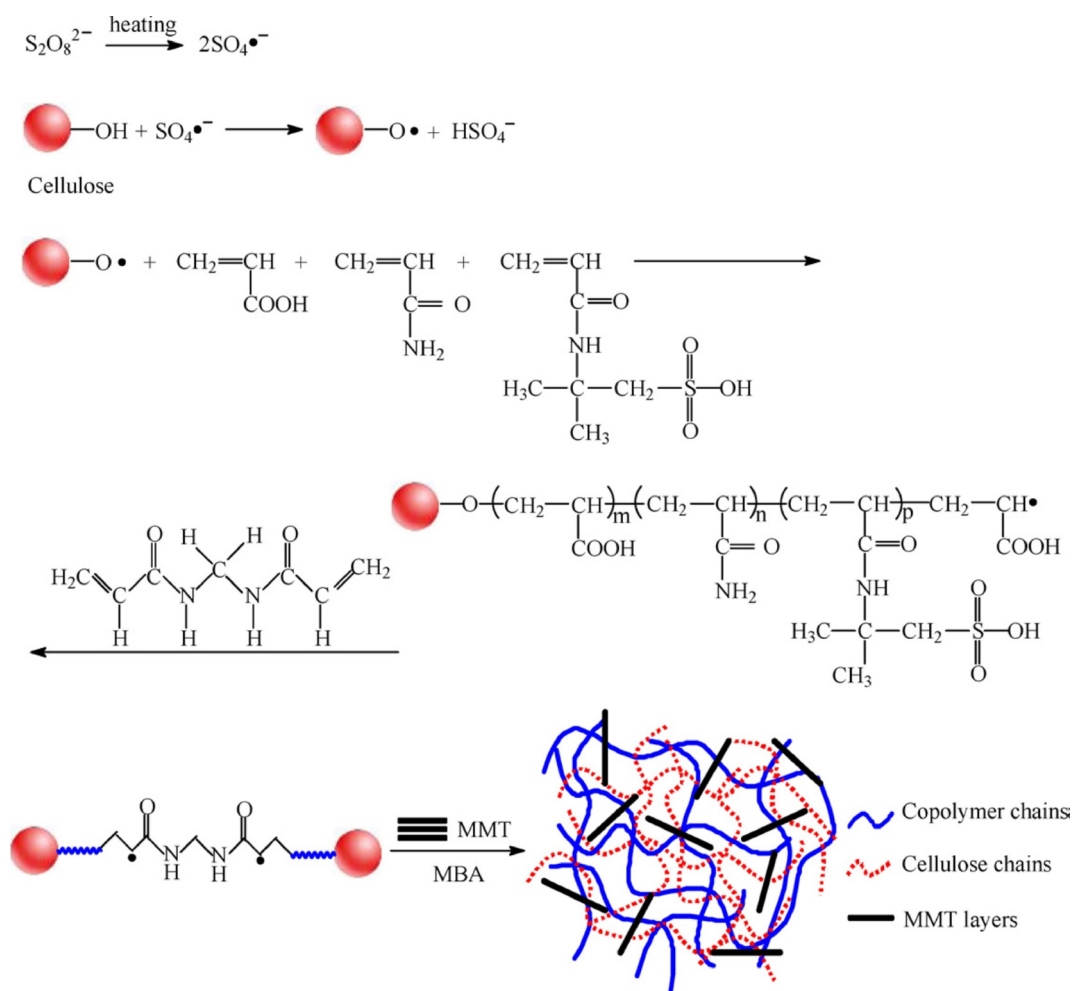


FIGURE 3 The predicted chemical mechanism for manufacturing the superabsorbent hydrogel composed of cellulose-g-P(AA-co-AM-co-AMPS). Reproduced with permission.⁵⁶ Copyright 2010, Elsevier Ltd. MBA, *N,N'*-methylene bisacrylamide; MMT, montmorillonite.

TABLE 1 CB hydrogels and their characteristics for ESDs.

Cellulose	Fabrication process	Bio nano composite	Battery type	Electrolyte	Energy density	Electrochemical capacitance	References
Cellulose fiber	Hydrothermal with vacuum percolation	CF/CNT/rGO/ δ -Na _x V ₂ O ₅ ·nH ₂ O	ZIB	2.0 M ZnSO ₄	295.4 Wh kg ⁻¹ at 162.3 W kg ⁻¹	1.87 mAh cm ⁻²	57
Carboxymethyl cellulose	Hydrothermal, calcination, and covering	CMC/Mn/Co/styrene butadiene rubber/Cu	LIB	1.0 M LiPF ₆ liquefied within a combination of EC, DEC, and fluoroethylene carbonate	—	—	58
CF	Hydrothermal and calcination	CF/NiO	LIB	1.0 M LiPF ₆ liquefied within a mix of EC and DEC	—	476 mAh g ⁻¹	59
CNF	Vacuum percolation	CNF/ZIF8	LIB	1.0 M LiPF ₆ liquefied within the mix of EC and dimethyl carbonate	—	—	60
CNF/polyaniline (PANI) foam	Freeze drying	—	—	—	51.45 mg/cm ³	59.26 mF/cm ² (at 10 mV/s)	61

Abbreviations: CB, cellulose-based; CF, carbon fiber; CMC, carboxymethyl cellulose; CNF, cellulose nanofiber; CNT, carbon nanotube; DEC, diethyl carbonate; EC, ethyl cellulose; ESDs, energy storage devices; LIB, lithium-ion battery; rGO, reduced graphene oxide; ZIB, Zn-ion battery.

diameter of 6–70 nm and a length of several micrometers. Even though the chemical composition of all NCs is similar, different aspects such as crystallinity, particle size, and morphology vary depending on the sources and extraction methods used.⁶³

Cellulose dissolves in the ILs like 1-butyl-3-methylimidazolium chloride (BMIMCl) and 1-allyl-3-methylimidazolium chloride (AMIMCl). After dissolution, regenerated cellulose is produced by adding water, ethanol, or acetone, exhibiting the same level of polymerization and lack of degradation as the original cellulose. The form of regenerated cellulose materials, such as film, beads, and gels, can be changed by altering the regeneration instrument and procedure.⁶⁴ Xu et al.⁶⁵ suggested making cellulose/graphene composite hydrogel and aerogel with high mechanical strength and thermal stability. Highly tough cellulose/graphene composite hydrogels were successfully produced by merely regenerating the mixture of wood pulp and reduced graphene oxide (rGO) from their IL solution using deionized water as a coagulant instead. Vitamin C, a safe and affordable reducing agent, prepared rGO in IL without additional steps. This cellulose/graphene composite hydrogel, which is extremely firm, has a broad range of potential implementations in the biomedical, environmental, and energetic materials sectors. After dissolving, structural integrity must be provided by physical or chemical cross-links to create hydrogels. Physical cross-links can result from various interactions, including ionic contacts, hydrogen bonds,

chain entanglements, strong van der Waals forces, and crystallite connections. Chemical cross-links are covalent connections made between polymeric chains or the polymers and a cross-linker used to gel the solution. Physical cross-linking in hydrogels can be formed from solutions by coagulation with an anti-solvent in a certain way or by curing at a given temperature (e.g., molding, casting, or dropping). Chemical cross-linkers or irradiation can cross-link materials to create chemical hydrogels.⁶⁶

4.2.2 | *N*-methyilmorpholine *N*-oxide system

Without the formation of an intermediary component, cellulose dissolves directly in an organic solvent during the direct dissolving process. This is a key distinction between the derivatizing process and direct dissolution. The cuprammonium, LiCl/DMAc, and Lyocell processes are examples of direct dissolving processes. Of these, the NMMO route and the IL approach are the two different ways that the Lyocell process can be executed. Lyocell is a relatively new type of regenerated cellulosic fiber made by a direct solution spinning method based on the solvent's ability to dissolve cellulose.⁶⁷ Initially, cellulose pulp and an aqueous NMMO solution are used to create the slurry in the Lyocell process. This homogeneous slurry is fed into the dissolution process to create a dope, a Lyocell polymer. This extremely viscous solution is then

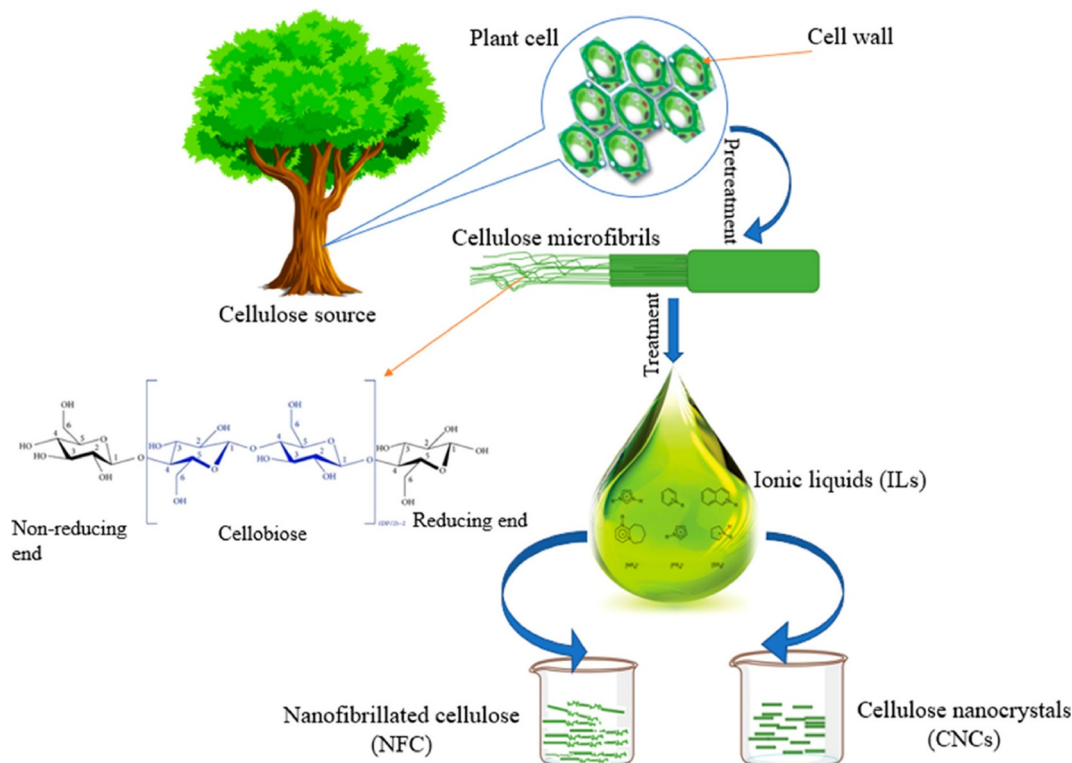


FIGURE 4 Schematic illustrating the processing of cellulose microfibrils using ILs to create NC. Reproduced with permission.⁶³ Copyright 2021, American Chemical Society. ILs, ionic liquids; NC, nanocellulose.

extruded through an air gap into a regeneration bath containing aqueous NMMO to create Lyocell fiber.⁶⁸ Typical slurry compositions in a commercial Lyocell process are 50%–60% NMMO, 10%–15% pulp, and 20%–30% water. In comparison to conventional heating, the use of microwave heating (105–490 W and 2450 MHz microwave energy until the dissolution was completed) to dissolve cellulose in NMMO has the advantage of reducing reaction times, according to a research study by Dogan and his co-worker.⁶⁹ The microwave-heated cellulose dissolving time was calculated according to the nominal value of water in the solution. It was demonstrated that 210 W microwave heating is a viable alternative heating technology for cellulose dissolution in NMMO.⁶⁹ Because of the presence of metallic and hemicellulose impurities that degrade cellulose and NMMO and lower the dope's onset temperature, which is necessary to initiate the reaction, Jadhav et al.'s⁷⁰ use of inexpensive paper-grade pulp is not practical for the Lyocell process. For safety concerns, the conventional Lyocell process can only use high-grade dissolving grade pulp. The effectiveness of a modified Lyocell process for faster dissolution and impurities removal for pulps/pulp blends is demonstrated by pretreating pulps to remove metallic and hemicellulose impurities in order to get around this limitation. Compared to the traditional method, the modified Lyocell process is safer, more affordable, and has a faster rate of dissolution. 99.6%

NMMO recovery and no yield loss of regenerated fiber are seen, which is similar to the conventional Lyocell process. The modified process would also be cost-effective if it used low-cost paper-grade pulp, which could be used to reduce raw material costs by 7%–8% and increase processability to save operating costs.⁷⁰

4.2.3 | Sodium hydroxide/urea

In an aqueous alkali solution, urea can increase cellulose's solubility and stability. To stop the dissolved cellulose molecules from re-aggregating, it may build up on the hydrophobic area of the cellulose molecule. Hydrophobic interaction may be the primary mechanism behind the self-assembly of cellulose and urea molecules. In the dissolution of cellulose, the hydrophilic hydroxyl groups are stabilized by Na^+ hydrations, the hydrophobic portion is stabilized by urea, and OH^- breaks the hydrogen bonds between the molecules. Van der Waal interactions are the main link in cellulose inter-sheets, whereas hydrogen bond interactions between hydroxyl groups are the main interaction in cellulose intra-sheets. When cellulose is dissolved in a NaOH and urea solution, the urea affects the hydrophobic components of the cellulose. In contrast, the hydrophilic hydroxyl groups of the cellulose interact with the NaOH. By lessening the hydrophobic effect of the cellulose

backbone and preventing the hydrophobic sections from reuniting, urea improves the dissolution of cellulose in alkali solutions. This can also make the solution more stable.⁷¹ A solution of suitable alkali hydroxide, urea, and distilled water was pre-cooled to -5 to -20°C . A specified quantity of dried cellulose was then instantaneously dissolved in the solvent and agitated for about 5 min at room temperature. The cellulose was assumed to have disintegrated when an optical microscope revealed a translucent cellulose solution free of any native fibers. To assess the effects of alkali hydroxide concentration, urea concentration, and dissolution temperature, cellulose sample Cell-3 was used, and its concentration was maintained at 4%.⁷²

4.2.4 | Deep eutectic solvents

DESs are a rapidly expanding substitute for traditional and IL-based solvents to manufacture functional materials. The excellent available qualities, flexibility, and biocompatibility of the related materials, when incorporated into inorganic or polymeric matrices, give the materials exciting qualities that have expanded their applicability in numerous fields of material chemistry and polymer research. Recent years have seen significant advancements in creating stimuli-responsive smart hydrogels employing DES.⁷³ Several polymeric hydrogels have been prepared using DES systems in various applications. These include frontal polymerization, bulk use of polymerizable eutectics, or alternate solvent systems, where one or more eutectic components are polymerizable compounds.⁷⁴ Ren et al.⁷⁵ reported that cholinium

acetate ($[\text{Ch}][\text{CH}_3\text{COO}]$), being an IL, causes cellulose fibers to expand. However, when chloride salts are added to $[\text{Ch}][\text{CH}_3\text{COO}]$, such as tributyl methylammonium chloride ($[\text{N}_{4441}]\text{Cl}$), a eutectic mixture is created that can dissolve cellulose up to 6 wt%. The values found are comparable to those found with some ILs. This eutectic combination is a better initial point for manufacturing new DESs that dissolve cellulose. DESs composed of $[\text{Ch}]\text{Cl}:\text{im}$ (3:7), $[\text{Ch}]\text{Cl}:\text{U}$ (1:2), and $[\text{Na}_{111}]\text{Cl}:\text{oxalic acid}$ (1:1) can dissolve cotton linter pulp.⁷⁶ The DES $[\text{Ch}]\text{Cl}:\text{ZnCl}_2$ was employed by Lynam et al.⁷⁷ for the dissolution of cellulose; however, following microwave heating, only 3 wt% microcrystalline cellulose was dissolved. The O-acetylation of cellulose was then successfully continued in this medium with good yields. Several methods such as physical cross-linking, chemical cross-linking, and polymerization techniques can be used to prepare the hydrogels. Figure 5 shows the general overview of hydrogel sources and preparation methods.

5 | PREPARATION METHODS OF HYDROGELS

5.1 | Physical cross-linking

Physically cross-linked hydrogels have demonstrated tremendous potential in recent decades.⁷⁸ Physically cross-linked hydrogels are typically produced through reversible intermolecular interactions, such as metal coordination, H-bonding, π - π stacking, polymerized predicaments, hydrophobic/hydrophilic connections, crystallization, and stereo-complex formation.⁷⁹

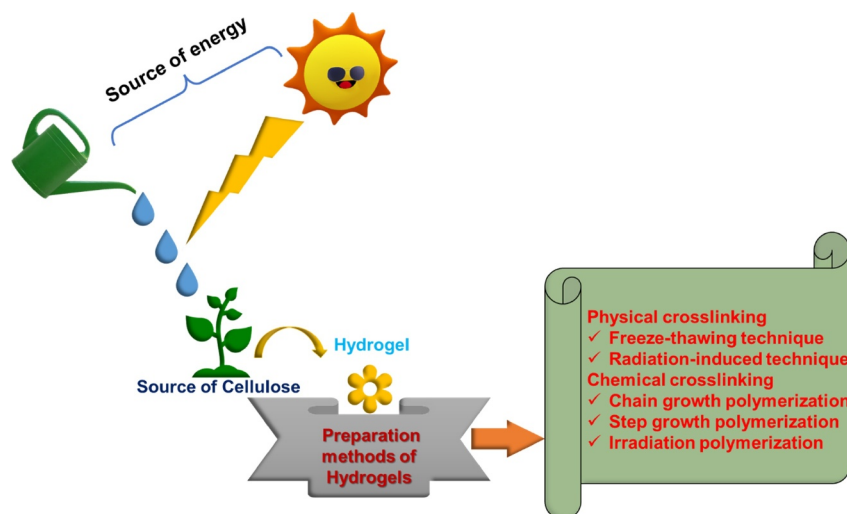


FIGURE 5 General overview of the sources and preparation methods for hydrogels.

5.1.1 | Freeze–thawing technique

The freeze–thaw method is used when bulk solvents, such as low molecular solutes, crystallize when frozen. This causes the concentration of polymer to rise, the spacing between polymer chains to contract, and the chains to align and associate the hydrogel's joining network structure. This method usually joins a network held together through covalent interactions and hydrogen bonds. Along with cellulose, other biopolymers that have been processed using the freeze–thaw method include hyaluronic acid (HA), carboxymethylated curdlan, β -glucan, locust bean gum, and mixtures of amylopectin and amylose.⁵⁴ Using hemicelluloses, polyvinyl alcohol (PVA), and chitin nano whiskers, novel hydrogels were produced by freezing and thawing them in 0, 1, 3, 5, 7, and 9 cycles. Acid hydrolysis was used to create chitin nano whiskers with an average length of 200 nm and width of 40 nm. Without using any chemicals, hemicelluloses, PVA, and chitin nano whiskers were blended using a freeze–thaw method to create hydrogels.^{80,81} The chitin nano whiskers were incorporated consistently into the PVA/hemicelluloses matrix, minimizing the packing and syneresis process and improving the hydrogels' thermal stability, compressive strength, and crystallinity. Using scanning electron microscopy (SEM) images, Figure 6A–F displays the hydrogels with porous network structures made by hemicelluloses/PVA/chitin nano whiskers through freeze/thaw cycles for 0, 1, 3, 5, 7, and 9 times. After one freeze/thaw cycle, the spider-web pattern was more visible in the cross-section of the hydrogel (Figure 6B). However, as the number of freeze/thaw cycles increased, the design gradually faded away by five cycles (Figure 6D). The study found that hemicelluloses, PVA, and chitin nano whiskers all form hydrogen-bonded networks easily because these polymers have many hydroxyl bonds and the network assemblies are stable at low temperatures. Gel-5 discovered an intriguing pattern with a pore structure akin to a honeycomb. Because of the inherent pressure that ice crystals experience after freezing, the bridges connecting the ice crystal structure's lamellae looked broken and uneven. During the freezing process, chain packing was formed by the combination of hemicelluloses, PVA, and chitin nano whiskers. Water can readily escape from the dense networks during thawing, which leads to the development of syneresis. With a compressive stress of 10.5 MPa, the hydrogels that underwent nine freezing and thawing cycles demonstrated the highest compressive strength. These freeze–thaw hydrogel designs are made of a permeable polymer system in which the crystals function as knots, the polymer

components connect the visible hydrogel specimen throughout, and the pores are supplied by free water. Since water forms hydrogen bonds with the hydroxyl classes of all three polymers, it also acts as a swelling representative in the disordered zones of the polymer matrix. The crystalline knots improve the hydrogel's mechanical properties and thermal durability. This hydrogel construction procedure is shown in Figure 6G. Thermogravimetric analysis (TGA) is a useful method for resolving polymer form and taking into account the intermolecular interactions amongst the polymers in the hydrogel. Figure 6H shows the TGA and derivative thermo-gravimetric thermogram of the two hydrogels with 0 and 3 freeze/thaw cycle periods. Due to various factors, such as water evaporation and hydrogel degradation, the two pieces with differing weight-loss stages had different arcs. The primary cause of the weight loss below 100°C was moisture evaporating from the specimens.⁸²

PVA is a synthetic polymer that is water-soluble, non-toxic, biodegradable, and biocompatible. Because each repeating unit of PVA contains hydroxyl groups, PVA has a solid hydrophilic and hydrogen bonding property that enables it to form cross-linked hydrogels.⁸³ Physical hydrogels can be produced by repeatedly freezing and thawing PVA aqueous solutions, forming crystallite. Peppas first reported the freeze–thaw process for producing ultrapure PVA hydrogels in 1975. It was found that increasing the concentration of PVA and adding CNC can improve the compressive properties of as-prepared hydrogels. The compressive properties of hydrogels were negatively impacted by the increase in the number of freeze–thaw cycles from three to five. According to microstructure analysis, the hydrogels' porosity significantly decreased as PVA concentrations rose. The microstructure of the hydrogels did not change noticeably either with the addition of CNC or when the number of freeze–thaw cycles was increased. PVA/CNC hydrogels had a slightly lower crystallinity than PVA hydrogels with the same PVA concentration.⁸⁴

5.1.2 | Radiation-induced technique

Radiation assisted synthesis is a novel preparation technology that is environmentally friendly. Microwave irradiation is often used to prepare hydrogels because the solution is heated to high temperatures in a short span of time, which makes it react faster than conventional thermal conditions.⁸⁵ Four cellulose derivatives, MC, CMC-Na salts, HEC, and hydroxypropyl cellulose (HPC), were cross-linked in aqueous solutions by ionizing radiation to create hydrogels with significant water absorption. Gamma

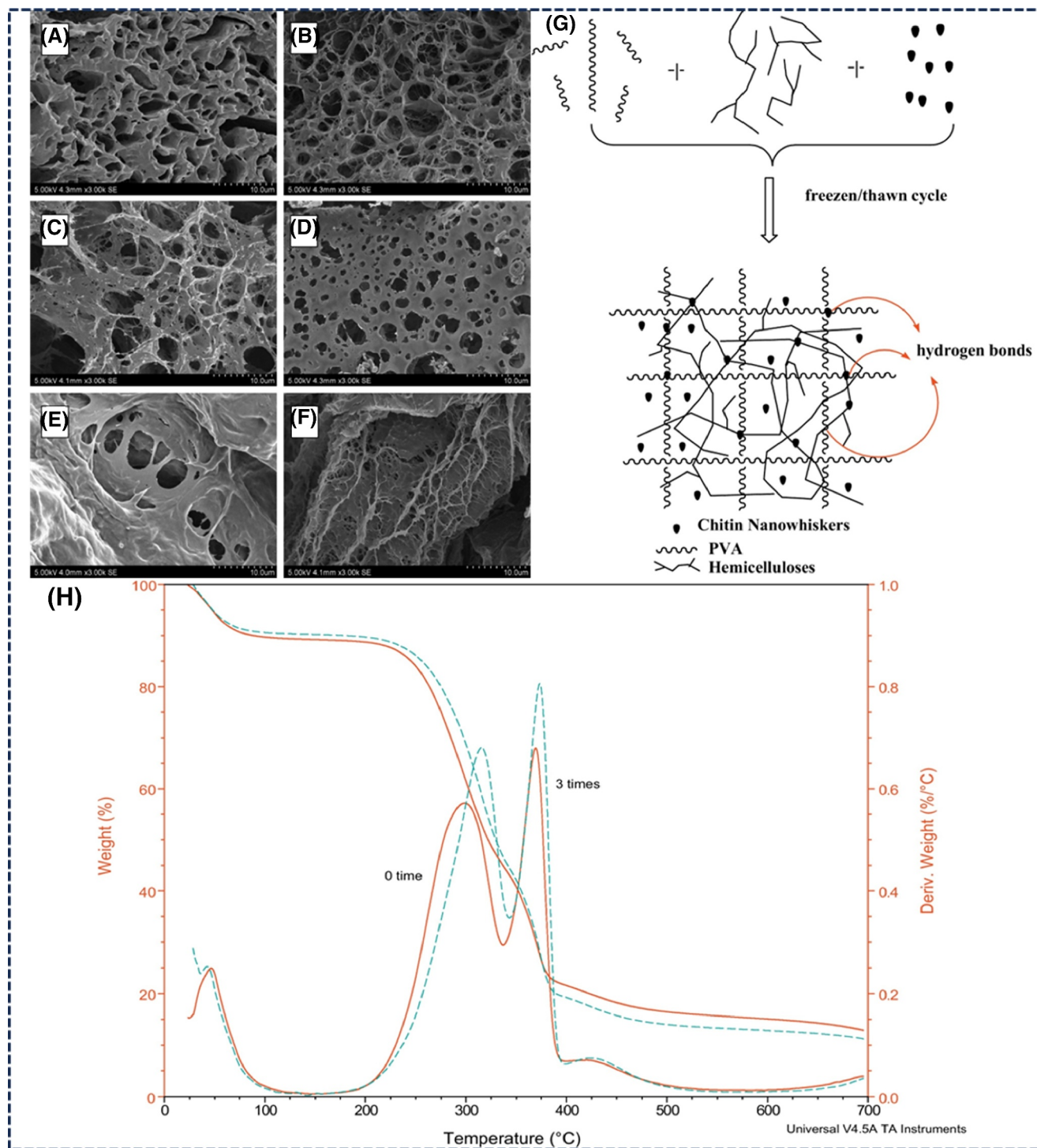


FIGURE 6 (A–F) Gel-0, Gel-1, Gel-3, Gel-5, Gel-7, and Gel-9 SEM pictures. (G) Hemicelluloses, PVA, and chitin nanowhiskers gelation tool in six freeze/warming cycles. (H) TGA and DTG graph of Gel-0 and Gel-3. Reproduced with permission.⁸² Copyright 2013, Elsevier Ltd. DTG, derivative thermo-gravimetric; PVA, polyvinyl alcohol; SEM, scanning electron microscopy; TGA, thermogravimetric analysis.

radiation was used to create superabsorbent hydrogels based on aqueous solutions of cellulose derivatives (CMC, MC, HEC, and HPC). Second-order swelling kinetics provided a good explanation for the swelling behavior of all gels in pure water. It was discovered that the solvent diffusion

process into the polymer network was unusual. Less hydrophilic derivatives, such as HPC and MC, can absorb much less water than the hydrophilic CMC-based hydrogel, which exhibits the greatest swelling ability in pure water. Nevertheless, a high electrolyte sensitivity goes hand in hand with the

high-water absorption. HEC has the greatest absorption capacity among the derivatives in solutions that mimic the real system of potential hydrogel applications, while the performance of CMC is poor.^{86,87} Because of their affordability, plentiful availability, and environmentally beneficial qualities, polysaccharides, such as starch, sodium CMC, chitin/chitosan, carrageenan, sodium alginate, and HA have garnered increased interest in recent times.⁸⁸ An electron beam (EB) accelerator and γ -ray irradiator are familiar commercial radiation sources. γ -rays and EB can be used to effectively induce the generation of radicals in polysaccharide chains. These radicals can then initiate the grafting and cross-linking of functional vinyl monomers onto polysaccharide polymer chains. One of the cellulose derivatives, CMC, has been extensively studied and used because of its high-water solubility. Similar to carboxymethyl starch, CMC can also be directly cross-linked by EB or gamma-ray irradiation. An alternative way to functional acrylate vinyl monomer is attached upon CMC using radiation to produce cross-links. CMC can also be produced by radiation cross-links.⁸⁹ Fekete et al.⁸⁶ synthesized CMC-g-poly (acrylic acid) hydrogels by gamma-ray irradiation with montmorillonite clay and *N,N'*-methylene bisacrylamide. The water swelling ratio of the hydrogels was greater than that of the sodium chloride solution.

5.2 | Chemical cross-linking

A covalent bonding must be present in the chemical cross-linking of the polymer chain. There are three primary categories of chemical cross-linking methods.⁹⁰ A different method called bulk polymerization is also used to create gels. The high density of free hydroxyl groups in the cellulose structure allows for their chemical and functional modification, which makes it possible to produce cellulose derivatives.⁹¹

5.2.1 | Chain growth polymerization

This process has three stages: initiation, propagation, and termination, just like free radical polymerization. A monomer should have unsaturation (a double bond) to follow the chain growth polymerization process and create a polymer chain. Three types of polymerizations can occur as the reaction mixture begins to gel: solution polymerization, suspension polymerization, and photopolymerization.⁹⁰

- (i) Solution polymerization: In solution polymerization, neutral or ionic monomers are

cross-linked with a multifunctional cross-linking agent using a thermal, redox, or UV initiator. The main advantage of solution polymerization over bulk polymerization is the solvent's capacity as a heat sink.⁹² Water, ethanol, a combination of water and ethanol, and benzyl alcohol are common solvents used in this procedure. The free-radical initiated polymerization of AAc and its salts by a cross-linker called methylene bisacrylamide is widely employed and is a simple procedure to manufacture nano-hydrogels by solution polymerization.⁹³

- (ii) Suspension polymerization: Many significant polymers, such as poly(vinyl chloride), poly(methyl methacrylate), expandable polystyrene, styrene-acrylonitrile copolymers, and various ion exchange resins, are commercially prepared via suspension polymerization. Suspension polymerization involves the production of polymer inside drops of a monomer-containing phase dispersed throughout a continuous liquid phase.⁹⁴ Surfactant is often scattered throughout the ongoing phase, while monomer is typically dissolved in the dispersed phase. This process can produce particles of the desired size, but removing organic solvents such as *n*-hexane and toluene is difficult. The random migration of water droplets into the oil phase under vigorous stirring produced spherical particles. A steady and controllable size emulsion of an oil phase enclosed in water droplets develops under these circumstances. Hydrophilic reactants cause the reactions to take place inside the tiny water droplets. It benefits producing high molecular weight particles simply, quickly, and efficiently. Amphiphilic PVA, an emulsifying agent that reduces the interfacial tension through adsorption, can stabilize the water/oil emulsion. Additionally, quickly synthesizing hydrogel particles with a predetermined shape using ultrasound-assisted polymerization has proven an excellent method for preventing thermal degradation.⁹⁵
- (iii) Photopolymerization: Photopolymerization uses light to start a polymerization reaction to obtain a linear or cross-linked polymer structure. The photopolymerization method has frequently been used to create hydrogels, typically in situ.⁹⁶ Recent advancements have made the photopolymerization technique a potent tool for producing multi-layered hydrogels with complex designs commonly used for tissue engineering.⁹⁷ The use of photo-initiated polymerization has attracted the attention of numerous researchers; for instance, Reeves et al.⁹⁸ synthesized CMC hydrogels with UV light as the initiator, while Mann et al.⁹⁹ produced a tissue-engineered

hydrogel scaffold by photopolymerizing poly(ethylene glycol) (PEG) derivatives with proteolytically degradable peptides. The gelation process can be more efficiently controlled using photopolymerized hydrogel systems. The alginate-based hydrogels were cross-linked; physiological factors and UV light exposure regulated the rate of cross-linking using photoirradiation.¹⁰⁰

To achieve intercalated monomer polymerization in the lattice layers of montmorillonite, Wang et al.¹⁰¹ proposed two types of photopolymerization reactions. Benzoin ether, a common photo-initiator, was utilized in the first system to produce the radicals that caused the polymerization. In the other system, the pre-polymer cross-linking reaction is initiated by acid generation through a photo-induced electron-transfer reaction.

5.2.2 | Irradiation polymerization

A new method for synthesizing CB hydrogels is radiation polymerization. Unsaturated substances can be polymerized using high-energy radiations such as γ rays and EB.⁹¹ High-energy radiation could be used to create hydrogels from water-soluble polymers. Ionizing high-energy radiations are required to ionize simple particles within air or water, such as EBs and γ -rays. Alongside the polymer strands, many reactive sites are generated during the radiation of a polymer solution. The subsequent combining of these radicals results in a significant number of cross-links.¹⁰²

5.2.3 | Step-growth polymerization

Step-growth polymerization is a slow process that involves covalent bonding between poly-functional monomers. The average molecular weight increases slightly as a result of step growth polymerization, but a high-rate reaction is no longer required for a polymer chain.¹⁰³ Furthermore, at least two multifunctional monomers are reacted with mutually reactive groups to form homogeneous hydrogels; each monomer serves as a cross-linking point. Different hydrogel structures for networks are produced by the two cross-linking techniques, and variations in the concentration or molecular weight of the monomers may have distinct effects on the hydrogel's development. Azide-functionalized polypeptides and four-arm PEG functionalized with cyclooctene were reacted via the strain-promoted alkyne-azide cycloaddition reaction to form a hydrogel network utilizing a step-growth technique.¹⁰⁴

5.2.4 | Sonochemistry technique using ultrasound

There are multiple applications for ultrasonic radiation. Numerous applications involving polymers have also considered using ultrasound. These include the production of free radicals for the synthesis of polymers, the activation of initiators of free radicals, the disintegration of polymers in solution at high shear rates, and the physical mixing of heterogeneous emulsion/suspension polymerization systems. Water-soluble monomers and macromonomers have been polymerized using ultrasonic technology to produce a variety of acrylic hydrogels. Utilizing glycerol, sorbitol, or glucose additions in viscous aqueous monomer solutions, ultrasound was used to create initiating radicals in an open system at 37°C. Glycerol was the most effective of the water-soluble additives used in the hydrogel synthesis. Hydrogels were prepared using the monomers 2-hydroxyethyl methacrylate, poly(ethylene glycol) dimethacrylate, dextran methacrylate, acrylic acid/ethylene glycol dimethacrylate, and acrylamide/bis-acrylamide (Figure 7).¹⁰⁵

5.2.5 | Solution polymerization in water

This method is mainly used to prepare superabsorbent hydrogels based on cellulose. Compared to other methods, this one has more substantial advantages. Because aqueous polymerization has a high degree of polymerization and is safe and harmless, it has been used extensively. To put it simply, it has better control over the polymerization heat and is both affordable and practical. An initiator produced by radiation or temperature initiates the polymerization process. The final product can be powdered and dried after some time for various uses. By polymerizing in pure water, CB superabsorbent hydrogels may be produced; however, the interaction of the mixture in aqueous media is considered another method of producing such hydrogels. For example, a CB superabsorbent hydrogel was made via the exchange of cellulose and Na-CMC in an alkaline/urea aqueous media instead of unpolluted water.¹⁰⁶

Zeng et al.¹⁰⁷ employed freeze-thaw-assisted hydrothermal separation to extract hemicellulose from poplar, achieving a notable 32.25% increase in separation efficiency. This demonstrates the effectiveness of freeze-thawing as a supplementary method for enhancing biomass component isolation.¹⁰⁷ Freezing and thawing significantly impacted gel formation, particularly over short time intervals. Following a single cycle, gel formation percentage was observed at 10%, 15%, and 19% for 50/50, 80/20, 100% and 100% PVA blends, respectively. Conversely, at a low

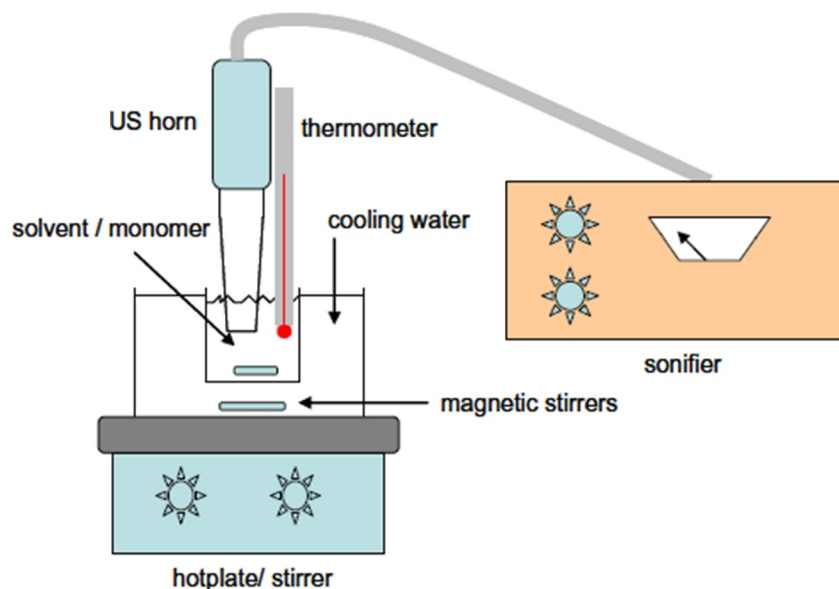


FIGURE 7 A schematic representation of the ultrasound equipment and setup used to polymerize vinyl monomers and create hydrogels is provided. Reproduced with permission.¹⁰⁵ Copyright 2009, Elsevier B.V.

irradiation dose of 10 kGy, all blends with all its composition exhibited complete solubility, indicating insufficient cross-linking. This suggests that freezing and thawing proved more effective than low-dose irradiation within shorter time frames.¹⁰⁸ PVA hydrogels, produced through the freezing and thawing method, were studied by Urushizaki et al. Their research revealed a strong correlation between tack and viscoelasticity in these gels, providing a valuable predictive tool for material properties in transdermal drug delivery. Additionally, the rate of gel swelling exhibited a linear increase with the square root of the immersion time in water.¹⁰⁹

Therefore, the choice of method depends on factors such as the source material, desired properties of the extracted cellulose (purity, crystalline, etc.), and the intended application. The feasibility of these methods for large-scale energy storage applications depends on factors, such as scalability, cost-effectiveness, performance, environmental impact, and technological advancements. As research in CB materials continues to evolve, optimizing these methods for commercial viability and large-scale production is crucial for their widespread adoption in energy storage technologies.

6 | PROPERTIES OF HYDROGELS

Our group has extensively explored the unique properties of hydrogels, emphasizing their remarkable water-absorbing capacity, biocompatibility, and

tunable mechanical characteristics. These have been thoroughly summarized in previously published review articles.^{110–112} So here, we have precisely summarized the hydrogel properties. We scrutinized their ability to swell and deswell in response to environmental stimuli, as well as their permeability and controlled diffusion behavior. Additionally, we delved into the chemical functionalization of hydrogels to tailor their properties for targeted applications. Understanding these properties has laid a solid foundation for our research, enabling us to harness these features for diverse applications in fields, such as biomedicine, environmental sciences, and materials engineering.^{111,113,114} However, we have tried to compile more literature on the properties of hydrogels, which will pave the path for beginners in this field.

6.1 | Self-healing

Self-healing is a characteristic of all living things. It allows tissue to be fully or partially restored following physical, chemical, or mechanical damage, and has recently been used in materials design. Hydrogels that can self-heal, can repair fractures and return their mechanical attributes. Even if the hydrogels are destroyed while being administered, this can stop the flow of loaded chemicals. Making the cross-links between polymer chains capable of reforming within a suitable period after breakage is a standard method for making a hydrogel self-healable. This method diminishes the safety worry about the toxicity produced

by releasing the healing agent and avoids the requirement to integrate a healing agent within the hydrogel.

However, cellulose does not have intrinsic self-healing effects like some manufactured materials explicitly designed for self-repair. Nevertheless, CB materials can be self-healing when integrated with specific additives or procedures. Cellulose molecules include multiple hydroxyl (-OH) groups, which can initiate hydrogen bonds with neighboring molecules. This property permits CB materials to create a cohesive network that can heal from inevitable deterioration. Further, cellulose materials can swell and reorganize in water or certain solvents. When damage happens, such as microcracks or minor fractures, water or solvent can penetrate the material, pushing it to swell and close the gaps, thus facilitating self-healing.

Despite the encouraging potential outlined overhead, hydrogels typically exhibit insufficient mechanical strength partly due to their high-water scope and delinquency to resist wear and tear. This makes hydrogels sensitive to structural breakdown and uncontrolled agent discharge, as shown in Figure 8A. While Figure 8B has also reported the use of dynamic covalent bonds involving imine groups, disulfide groups, and acylhydrazone groups, cross-links in self-healing hydrogels are usually noncovalent, ranging

from hydrophobic or host-guest interactions to reversible ionic bonds and hydrogen bonds.¹¹⁵ Digital images presently proved the self-healing capability of hydrogel, and the empty hydrogel in the absence of 5-Fu/MGMs was preferably shown within Figure 8C–E. Also, they designed two hydrogels; one was empty, and the difference was 30 mg/mL 5-Fu/MGMs/Gel, as illustrated in Figure 8F–I. With the identical development mentioned above, the cured hydrogel was robust enough to sustain its weight and extend to separate the whole. Figure 8J–L directed to the inoculation, self-healing and magnetism of the 30 mg/mL 5-Fu/MGMs/Gel, consequently.¹¹⁶

By means of magnesium-bisphosphonate (BP) ligand interactions, a pH-responsive and self-healing hydrogel has been produced from BP-functionalized HA-BP and magnesium silicate NPs. NPs serve as cross-linkers and bioactive agent carriers during the creation of hydrogels.¹¹⁷ Recent research has developed a self-healing hydrogel as a transporter of bioactive substances by using hydrazone bonds to cross-linked oxidized xanthan and 8-arm PEG hydrazine. Every time the strain reduces during the cycle of tension and relaxation, the carrier's storage modulus can be restored. Additionally, the carrier displays swelling and degrading behavior that responds to pH.¹¹⁸ This enables the controlled release of bioactive agents in response to environmental

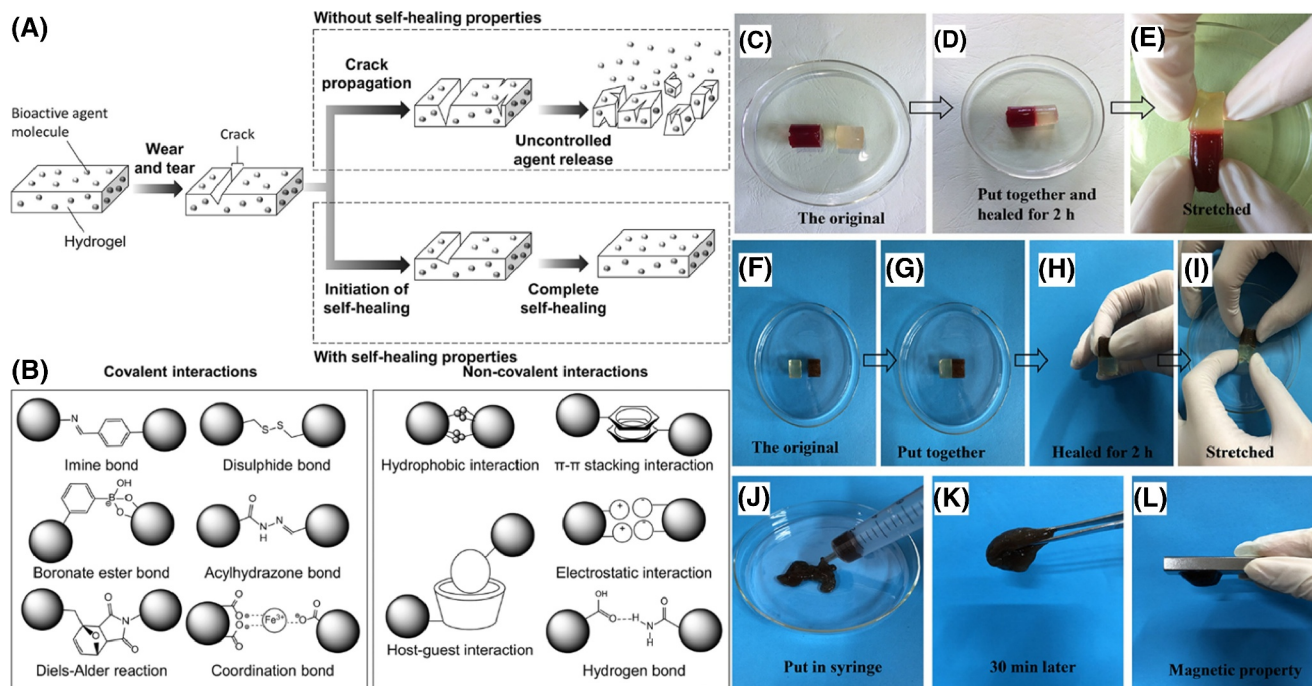


FIGURE 8 (A) A graphic illustration shows self-healing's role in controlling the unrestrained liberation of the loaded agent from a hydrogel. (B) Significant covalent and noncovalent interactions are used to develop self-healing hydrogels. Reproduced with permission.¹¹⁵ Copyright 2021, American Chemical Society. (C–E) Self-healing method of blank Gel in the absence of 5-Fu/MGMs. (F–I) Self-healing procedure of hybrid platforms. (J–L) Efficiency of hybrid supports on 37°C. Reproduced with permission.¹¹⁶ Copyright 2019, Elsevier B.V.

modifications. Proteins and peptides are frequently employed to create self-healing hydrogels in addition to polysaccharides.¹¹⁹

Phadke et al.¹²⁰ proposed achieving self-healing in hydrogels by incorporating polar-functionalized hydrocarbon side chains into the polymer network. These chains contain polar functional groups facilitating hydrogen bonding across separate hydrogel pieces or ruptures. They noted that when two lightly cross-linked A6ACA hydrogels were introduced to each other in a low-pH aqueous solution ($\text{pH} \leq 3$), they swiftly bonded together within 2 s, thus validating their hypothesis. The recovered hydrogels display a robust interface, able to endure their weight, repeated stretching, and exposure to boiling water. Once the stress is released, the healed samples can withstand significant deformations and return to their original size and shape. This pH-mediated healing is reversible: Two healed hydrogels separate when exposed to high pH. The healing, separation, and rehealing process is repeated over 12 times without hysteresis. The healing process occurs on the same timescale and exhibits comparable weld-line strength to the original hydrogels. It has been observed that the healing properties of hydrogels decrease over time when subjected to a low-pH environment before healing. However, researchers have found a solution: immersing these hydrogels in a high-pH environment can revive their healing ability when reintroduced to a low-pH environment. This decline in healing ability under prolonged exposure to low pH is attributed to the formation of intramolecular hydrogen bonds within the hydrogels. These bonds reduce the availability of the hydrogels to form intermolecular hydrogen bonds across interfaces, thereby impeding their healing efficacy.

6.2 | Transparency

Hydrogels' "clarity" or "transparency" is a quality determined by how well they transmit light and is quantified as light transmittance. The degree of phase separation that occurs during the formation of hydrogels—also referred to as spinodal decomposition or spinodal phase separation, wherein one thermodynamic phase forms two coexisting phases—means that the transparency of these materials varies after they are created and is dependent on the coagulation bath or curing temperature used. For hydrogels made in NaOH/urea aqueous systems, an increase in healing temperature causes a significant loss of transparency (e.g., 85% transmittance at -10°C , 66% transmittance at 20°C , and nontransparent at and beyond 30°C). It is brought about by a greater degree of phase separation,

which increases the heterogeneities of the hydrogel.¹²¹ Acetone is added to water as a coagulation solution to increase the hydrogels' transparency. This may be due to a "specific polymer structure" that develops due to the gel's equilibrium between swelling from water and shrinking from acetone.¹²² In contrast to "water-coagulated gel," gel beads made using a novel technique, deionization of the cellulose/LiCl/DMAc solution with ion exchange resins, appear colorless and transparent without a fibrous texture. This is likely because, during ion exchange, crystallite formation occurs before phase separation.¹²³

6.3 | Strength

Guan et al.⁸² reported that hemicellulose can be effectively transformed into hydrogel through a freeze-thaw process, improving its crystallinity, compressive strength, and thermal stability. It resulted in a structure with a high degree of crystallinity, increased rigidity, and stability. As the concentration of biopolymers increases, the compressive stress of hydrogels rises.⁶⁴ The tensile capabilities of high-strength hydrogels derived from CNFs are demonstrated by micrographs that display the top (upper) and side (lower) views of the freeze-dried hydrogels produced in 9 wt% and 15 wt% NaOH cellulose (alkaline treatment). Due to the crystalline network in the gels, the two types of hydrogels (cellulose I and II) with differing crystal shapes displayed high Young's modulus and high tensile strength. The nanofiber hydrogel with a cellulose II crystal structure showed a continuous network made by the engagement of the surrounding CNFs, in contrast to the hydrogel with a cellulose I crystal structure. It also displayed more excellent tensile properties. The hydrogels made from CNFs have low compressive characteristics because CNFs are relatively weak when compressed or bent but quite strong when subjected to tensile stress. The double-network approach is feasible and helpful for enhancing poor mechanical qualities such as compressive modulus and strength since it combines CNF gels with natural polymers and polysaccharides.¹²⁴

6.4 | Swelling behavior

The ability of hydrogels to expand in the presence of a solvent that is suitable thermodynamically is one of their advantages. When an initial state hydrogel comes into contact with it, solvent molecules move toward the hydrogel surface and penetrate the polymeric network. In this instance, a moving border divides the rubbery hydrogel region from the undissolved glassy phase.¹²⁵ The elastic phase of the

network's meshes will periodically expand, allowing other solvent molecules to enter the hydrogel network. The rate of swelling, which is influenced by several physicochemical factors, most notably the degree of porosity and the type of porous structure, is one of the key characteristics of hydrogel swelling. Four categories of hydrogels can be distinguished: non-porous (very slow swelling ratio, dependent on sample size), micro-porous (slow swelling ratio, dependent on size), macro-porous (rapid swelling ratio, dependent on size), and super-porous (swift swelling ratio and size-independent) hydrogels.¹²⁶ Althans et al.¹²⁷ concluded that the degree of swelling at low acid concentrations depends on the kind of acids at constant temperature below the phase transition point in pure water. No matter what type of acid is present in the shrunken condition, swelling will still occur. Swollen gels contract when the acid concentration rises; the type of acid also affects the concentration at which the contraction occurs. The hydrogels swell again as the acid concentration is increased further, and the type of acid being employed significantly impacts the acid concentration at which the swelling takes place. The extremely high levels of swelling seen for the swelling in formic acid may be a sign that the cross-linker or the polymer itself will eventually deteriorate, which will cause a sharp rise in the network's flexibility.

7 | APPLICATION OF THE HYDROGELS IN ESDs

Because of their great elasticity, transparency, adjustable mechanical properties, and consistent electrical conductivity, flexible ionic conductors have drawn a lot of attention in light of the Internet of Things' rapid development and the growing need for human-machine interfaces. Conduction hydrogels, with their distinct properties of flexibility, durability, and versatility, have shown remarkable promise recently for wide-ranging and important applications in sustainable energy, including flexible electronic devices, batteries, and sensors.^{128,129} Novel flexible storage technologies such as supercapacitors and rechargeable batteries are exciting because of their broad range of potential applications in flexible electronics and implants. Hydrogels are among the most promising electrolyte substitutes since they are soft and have high water absorption rates, which fulfills the necessary requirements for flexible ESDs. Additionally, modifying the chemical structures can produce a variety of properties, such as self-healing capacity, ultra-stretchability, electrochromic, and extreme-temperature resistance, paving the path toward the formation of novel flexible ESDs with extended working lives and scintillating functions.¹³⁰

7.1 | CB electrically conductive hydrogels for flexible batteries

CB conductive hydrogels are a perfect candidate for flexible electronic appliances owing to their exceptional characteristics, better electronic effects, tunable mechanical flexibility, and easy preparation method. Additionally, some hydrogel materials exhibit unique biological features (such as self-healing, self-adhesive, anti-microbial performance, and biocompatibility) to achieve special needs.^{131–133} Thus, in recent years, conductive hydrogels have been a vital area of study; flexible electronics show advantageous opportunities for flexible electrodes, mechanical sensors, and other appliances with the human body.^{134,135} Batteries typically comprise of two electrochemically active electrodes divided by an electrically insulating electrolyte that is ionically conducting. Batteries are categorized as primary or secondary, depending on whether they can be recharged.

7.1.1 | Lithium-ion batteries

The most popular batteries for a wide range of products, including consumer electronics and electric vehicles, are LIBs. Flexible high-performance LIBs offer much potential as power sources for wearable devices. Despite the enormous efforts, a well-developed adaptable LIB for commercial use has yet to be shown. The following are the current technological barriers for Li-ion electrodes: (1) rigid batteries cannot be designed with conventional materials; (2) ionic and electronic conductivity is inefficient during operation; and (3) electrochemical performance degrades during deformation. More specifically, hydrogels provide compact loading of electrochemically active materials, enough space for volume expansion and contraction caused by Li insertion, continuous electronic and ionic transport pathways, and effective resistance to mechanical deformation when used in flexible LIB electrodes.¹³⁶

Anode materials can be supported by flexible conductive scaffolding made of carbon materials derived from cellulose. Inorganic alkali-assisted carbonization has the potential to significantly increase the porosity and specific surface area of biomass carbon, which is beneficial for improving LIB cycle stability and rate performance. For instance, BC-based carbon compounds having a specific surface area of $1235 \text{ m}^2 \text{ g}^{-1}$ are frequently prepared by a KOH-activated pyrolysis method. The BC-based carbon electrode demonstrated a specific capacity of more than 857 mAh g^{-1} as an anode material for LIBs after 100 cycles at 100 mAh g^{-1} . In order to create a flexible LiFePO_4 -carbon paper that

could function as a free-standing cathode, the precursors were solution-based and impregnated in a microcrystalline cellulose network. This was followed by a carbothermal reduction process.¹³⁷

Carbon-based materials, including graphene, carbon nanotube (CNT), and TiO₂-based compounds, are the most widely used anode materials.¹³⁸ Scientists have dedicated significant efforts to address capacity loss in battery systems. For example, they employed an electrospinning method to create flexible carbon fibers (CFs) derived from cellulose acetate (CA). These fibers were then utilized in the negative electrode materials for LIBs.¹³⁹ The cycling performance of batteries utilizing SnO₂ has been significantly improved by incorporating nano-SnO₂ particles and various carbon-based materials known for their low expansion capabilities.^{140–142}

The intramolecular hydrogen bonds are mainly seen between the hydrogen held with the hydroxyl group of the C3 carbon and the ring oxygen of the adjacent O5 unit, as shown in Figure 9A. By appropriately replacing the hydroxyl groups in the cellulose to varying degrees, other water-soluble cellulose by-products have been obtained. Cereal by-products are mainly classified as cellulose esters and cellulose ethers due to the function of the linked oxygen upon the –OH groups of individual anhydroglucose unit. The most typical water-soluble cellulose by-products are shown in Figure 9B, which also shows them separated into cellulose esters and cellulose ethers. Mixed ethers, such as hydroxyethyl carboxymethyl cellulose and methyl carboxymethyl cellulose, would also be found among other ethers. The charge and discharge processes in sodium-ion batteries (NIBs) and LIBs are shown in Figure 9C. It is evident that the separator

membrane is a crucial component since it acts as a physical barrier between the anode and the cathode, providing mechanical support and averting an electrical short circuit. The separator also serves as an electrolyte reservoir for the movement of ions during the charge and discharge cycles. The separator membrane must have the following qualities, among others: thickness, porosity, pore size, permeability, cost, mechanical strength, and chemical and electrochemical stability. Numerous cellulose derivatives have been created after Kuribayashi's pioneering study to serve as effective LIB separators. For instance, it has been suggested that BC is a viable substitute for conventional polyolefin LIB separators.¹⁴³

LIBs have become the backbone of modern moveable electronics and electric vehicles owing to their high ED, long cycle life, and excellent performance. However, the ever-growing demand for these batteries raises concerns about the sustainability of their components, especially the electrode materials. To address these concerns and reduce the environmental impact of LIBs, researchers have been exploring alternative materials. In this regards, Figure 10A–E schematic illustration of the recent elastic and permeable division established upon hydroxyapatite nanowires (HAP-NW) systems with outstanding thermal durability, fire resistance, and exceptional electrolyte wettability. Figure 10F–K shows the electrochemical activity at the cells' ambient conditions designed by various separators. Figure 10F,G shows the cycling activity and rate capacity of the LiFePO₄/separator/Li half cells manufactured with the HAP/CF partition and retail polypropylene (PP) separator. Additionally, using graphite as a functional electrode, the cell with the HAP/CF separator exhibits a more useful cycling activity and

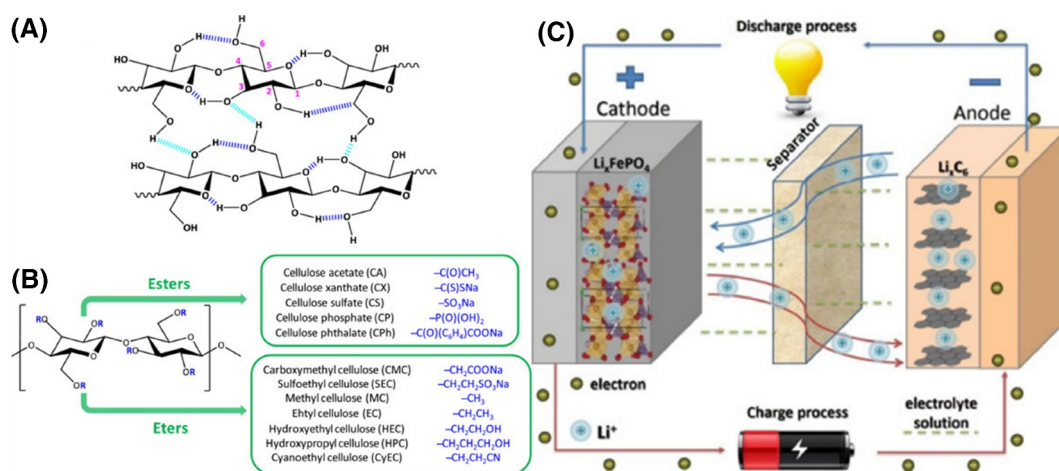


FIGURE 9 (A) An example of the original cellulose's chemical composition in graphic form. While the intermolecular hydrogen bonds between adjacent chains are shown in cyan, the intramolecular hydrogen bonds between identical chains are displayed in blue. The role of carbons in the cellulose chain is represented by the magenta digits; (B) configuration of water-soluble cellulose by-products highlights the –R replacement of the most appropriate cellulose ester/ethers. (C) A LIB's components and electrochemical processes are shown schematically. Reproduced with permission.¹⁴³ Copyright 2020, Elsevier Ltd. LIB, lithium-ion battery.

rate capacity (Figure 10H,I). To verify the effective application of this novel HAP NW-based separator toward LIBs with improved activity, pouch-kind LiFePO_4 /partition/graphite whole cells with elevated mass loading cathodes (18.5 mg cm^{-2}) have been built and analyzed (Figure 10J,K). An abrupt decrease in the open-circuit voltage (OCV) can happen if a partition experiences significant shrinkage because both the cathode and the anode may become damaged. The interior short circuit caused by the PP separator's thermal shrinkage causes the OCV of the cell with the PP separator to rapidly decline to 0 in <20 min, as shown in Figure 10L. The electrochemical activity of

the batteries designed with the retail PP separator and the HAP/CF separator at 150°C is shown in Figure 10N. Prior to being exposed to high-heat dimensions, five scans were conducted under ambient conditions to verify the standard battery requirements. Additionally, a battery with an approximate HAP/CF separator at ambient conditions (138 mAh g^{-1}) is tested and shown to have a higher discharge capability of 157.8 mAh g^{-1} . It is explained by a lower interfacial resistance at higher temperatures and an enhanced ion scattering rate (Figure 10O). The HAP/CF separator's remarkable thermal endurance and its excellent potential for use in batteries related to elevated temperatures are

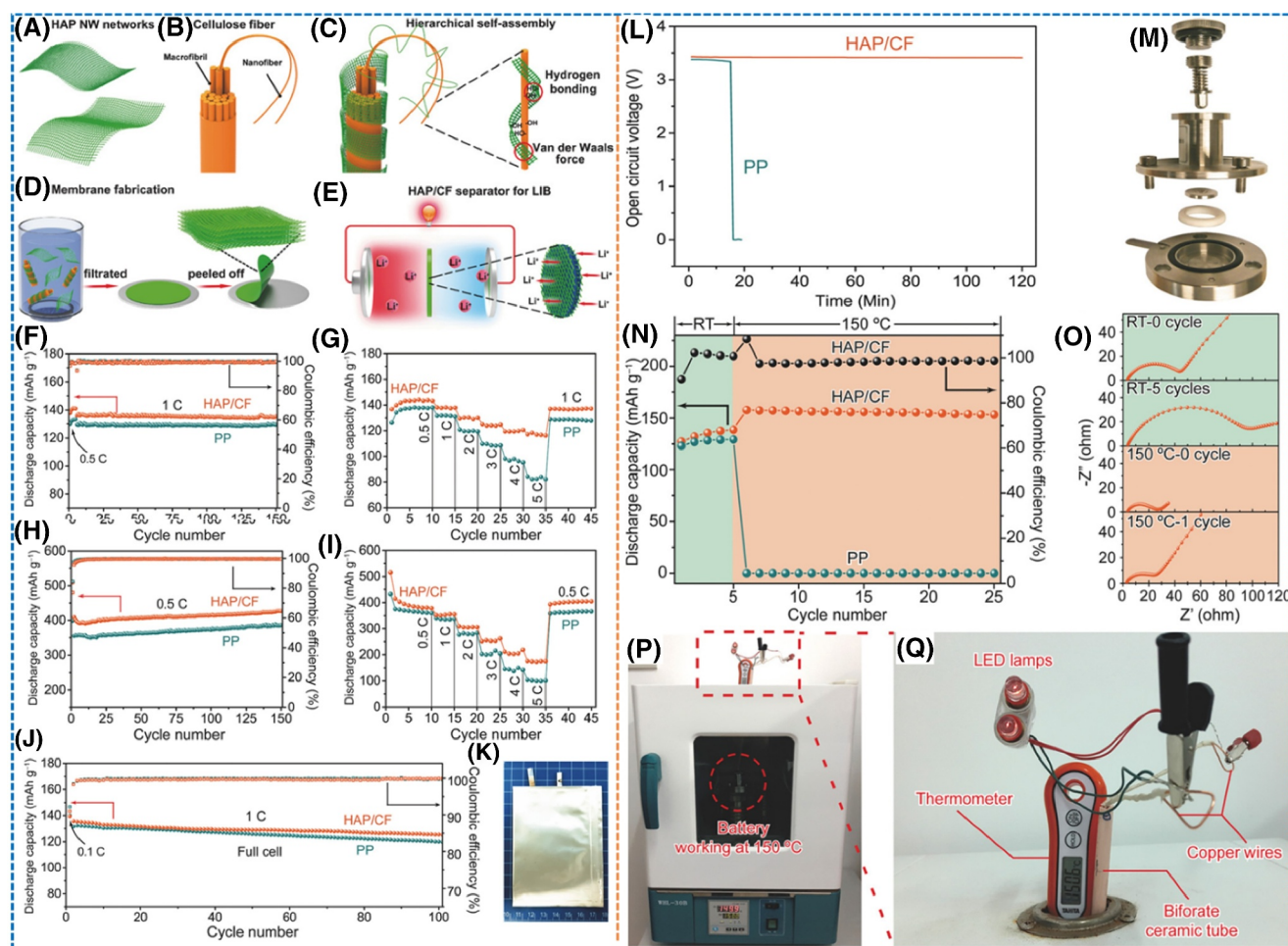


FIGURE 10 Graphic description of the pristine, approvingly flexible and permeable separator established upon HAP NW systems with superior thermal durability, flame resistance, and outstanding electrolyte wettability. (A) HAP NW chains. (B) CFs. (C) Group of CFs and HAP NW systems arranged hierarchically. (D) The process of suction percolation used to create the layered, highly permeable HAP/CF separator. (E) The LIB's installation of the HAP/CF separator. The room temperature electrochemical performances of the cells. (F) The LiFePO_4 /separator/Li half cells' cycling activity and (G) their rate capacity using the HAP/CF and PP divisions ($1 \text{ C} = 170 \text{ mAh g}^{-1}$). (H) Cycling activity and (I) half-cell rate capacity of graphite/partition/Li using the PP divider and HAP/CF ($1 \text{ C} = 375 \text{ mAh g}^{-1}$). (J) LiFePO_4 /divider/graphite whole cells' cyclability using the PP and HAP dividers ($1 \text{ C} = 140 \text{ mAh g}^{-1}$). (K) An image of the fully synthesized pouch-type cell taken with the HAP/CF divider. The LiFePO_4 /divider/Li batteries' electrochemical activity at 150°C . (L) At 150°C , the LiFePO_4 /separator/Li batteries' OCV arcs with the PP divider and HAP/CF. (M) An image of the battery-operated device used for the high-temperature test. (N) The PP divider and the HAP/CF are cycling at 2 C. (O) Battery EIS using the HAP/CF divider. (P) Description of the battery designed utilizing the HAP/CF divider operating at 150°C . (Q) Augmentation of the labeled parts within (P). Reproduced with permission.¹⁴⁴ Copyright 2017, Wiley-VCH Verlag GmbH & Co. CFs, carbon fibers; EIS, electrochemical impedance spectrum; HAP NW, hydroxyapatite nanowires; OCV, open current voltage; PP, polypropylene.

demonstrated by Figure 10P,Q, which shows that the battery included with the partition can safely ignite up to two 3.0 V light emitting diode (LED) lights at a temperature as high as 150°C.¹⁴⁴

This might result from cellulose's higher porosity, which swells in the electrolyte and facilitates Li ions to pass through the pores and contact the separator, electrolyte, and positive and negative electrodes. One factor that impacts the battery's ability to perform safely is the thermal shrinkage of the wall.^{145,146} Integrating CBMs into LIBs holds great promise for achieving more sustainable and environmentally friendly energy storage solutions.

7.1.2 | Zinc-ion battery

The rapid growth in the demand for energy storage solutions has intensified the search for advanced battery technologies that are both sustainable and high-performing. In this pursuit, ZIBs have emerged as a capable substitute for conventional LIBs. Moreover, integrating CBMs into ZIBs presents an opportunity to create eco-friendly and efficient ESSs. Metallic Zn is the most widely used stable metal in aqueous electrolytes. High electrical conductivity, ease of manufacturing, non-flammability, low toxicity, and comparatively low polarizability in comparison to metallic materials, such as magnesium and aluminum are just a few of its advantages. AZIBs have thus been widely regarded as a promising electrochemical energy system due to their high theoretical capacity (gravimetric capacity of 820 mAh g⁻¹ and volumetric capacity of 5855 mAh cm⁻³), low redox potential (0.762 V vs. the standard hydrogen electrode), availability of resources, and environmental friendliness. Because it is cheap, renewable, and ecologically friendly, biomass materials offer much promise for use in AZIBs as the two maximum plentiful land-based renewable bioresources, cellulose and lignin, have received the most significant consideration. The use of lignin, a naturally occurring polymer molecule with a comparatively high degree of polymerization, is constrained by its complex structure, high manufacturing costs, and labor-intensive preparation method. Additionally, lignin still faces significant application hurdles because of its large molecular weight dispersal and limited mechanical strength while having demonstrated excellent potential as binders and polymer electrolyte. Cellulosic materials have greater appeal and are more practical for use with AZIBs than lignin.¹⁴⁷ The critical component for building a flexible electrochemical energy storage device is a solid-state electrolyte, which provides the charge carriers and can be used in place of a separator to prevent short circuits between two electrodes. Water-in-salt electrolytes were suggested to achieve electrochemical

stability and a large potential window. A Zn-based rechargeable battery's performance was enhanced using a water-in-salt ZnCl₂. In water-in-salt ZnCl₂, the potential of the oxygen evolution process was increased, and Zn²⁺ ions displayed nearly ideal deposition and striping on the Zn metal anode. ZnCl₂ can potentially enhance zinc-ion hybrid supercapacitor (ZHS) performance in a water-in-salt electrolyte because it is inexpensive and readily soluble. Yang et al.¹⁴⁸ devised an intriguing technique to extract highly concentrated ZnCl₂ from a cellulose matrix and use it as a gel polymer electrolyte. Cotton cellulose was dissolved in a concentrated ZnCl₂ solution to produce cellulose hydrogel, a gel polymer electrolyte. After adding a small amount of calcium ions, the metal-cellulose interactions would enclose Zn²⁺ and Ca²⁺ in the cellulose hydrogel. Then, a flexible ZHS was made by using the cellulose hydrogel as the electrolyte. Due to its high ionic conductivity and stable electrochemical potential window of 2.0 V, the ZHS has an ED of 192 Wh kg⁻¹ at a power density (PD) of 499 W kg⁻¹ and a capacity of 193 mAh g⁻¹ at a current density of 0.5 A g⁻¹. The ZHS exhibits stable cycle performance with 94.7% capacity retention after 5000 cycles and a low temperature tolerance of -20°C.¹⁴⁸

7.1.3 | Sodium-ion battery and capacitors

The rising focus on renewable energy technologies has sparked interest in Na-related ESSs. NIBs (SIBs) and sodium ion capacitors (SICs) have emerged as promising alternatives to Li-related ESSs due to their abundant sources and low cost. Researchers are increasingly drawn to these systems because of their potential to address energy storage needs sustainably.³³ Amatucci et al.¹⁴⁹ introduced metal ion hybrid capacitors as a novel ESS type, garnering significant research attention. Like SIBs, SICs comprise a capacitor-type electrode, a battery-type electrode, a Na⁺ conducting electrolyte, and a separator. The battery-type electrode stores energy through redox or reversible intercalation/de-intercalation reactions with a metal ion on the surface. Meanwhile, the capacitor-type electrode, boasting a high specific surface area, stores charge through ion absorption-desorption on its surface. With the growing focus on SIBs, SICs have emerged as promising contenders for large-scale energy storage due to their high ED, rapid energy delivery, and prolonged lifespan. Casas et al.¹⁵⁰ devised a pioneering separator utilizing renewable water-soluble cellulose derivatives. Cross-linking CMC and HEC generated membranes with substantial specific surface areas through non-solvent-induced phase separation. Extended galvanostatic cycling in a symmetric Na/Na cell configuration demonstrated a notable reversible voltage

response, maintaining a square wave polarization shape even after 250 h of cycling. This indicated robust Na plating and stripping with suppressed dendrite growth. Evaluating the novel membrane as a separator in $\text{Na}_3\text{V}_2(\text{PO}_4)_3/\text{Na}$ half cells revealed promising results. After 10 cycles at C/10, the cellulosic separator exhibited a capacity of 74 mAh g^{-1} with a 100% Coulombic efficiency, surpassing Whatman GF/D, a commercially available separator, which achieved 61 mAh g^{-1} and 96% efficiency. The study suggests avenues for developing biomass-derived porous membranes as battery separators, outperforming fossil resource-based commercial separators in capacity retention, Coulombic efficiency, and dendrite growth suppression. These separators, potentially adaptable to other battery systems, promise to advance sustainable energy storage technologies. Mittal et al.¹⁵¹ proposed advancements in energy storage technologies, aiming to rival mature lithium-ion batteries by addressing ED and operational lifespan concerns. Employing a circular economy approach, the authors utilize biodegradable cellulose nanoparticles to formulate a gel polymer electrolyte with notable characteristics. This electrolyte exhibits a high liquid electrolyte uptake of 2985%, an ionic conductivity of 2.32 mS cm^{-1} , and a Na^+ transference number of 0.637. A mesoporous hierarchical structure is achieved through a balanced combination of mechanically rigid CNCs and flexible CNFs, facilitating close interaction with metallic Na. This architecture enables stable Na plating/stripping at current densities of up to $\pm 500 \mu\text{A cm}^{-2}$, surpassing conventional fossil-based NIBs with separator-liquid electrolytes. Coupled with an environmentally sustainable and economically viable $\text{Na}_2\text{Fe}_2(\text{SO}_4)_3$ cathode, the battery attains an ED of 240 Wh kg^{-1} , delivering 69.7 mAh g^{-1} after 50 cycles at a rate of 1 C. In contrast, Celgard in liquid electrolyte yields only 0.6 mAh g^{-1} at C/4.

7.1.4 | Magnesium-ion batteries

Rechargeable magnesium-ion batteries (MIBs) show promise as electrochemical ESS due to their low cost and high safety. However, the need for more suitable anodes has hindered the advancement of high-performance MIBs. Cheng et al.¹⁵² introduced a novel anode for MIBs, comprising Bi nanospheres embedded in a carbon aerogel derived from CNCs (CNC-CA@Bi-NS). By employing ion-induced gelation and in situ thermal reduction processes, they achieved homogeneous incorporation of near-monodisperse Bi nanospheres (4–9 nm diameter) into the interconnected CA matrix. This hybrid structure effectively mitigated volume changes during magnetization/demagnetization, prevented nanosphere agglomeration,

and provided ample space for electrolyte storage and penetration. The electrode exhibited impressive performance, delivering a reversible specific capacity of 346 mAh g^{-1} at 0.5 C after 100 cycles, excellent rate capability, and unprecedented long-term cycling stability with a high Coulombic efficiency of ~100% at 2.0 C after 5000 cycles. Han et al.¹⁵³ developed flame-retardant cellulose aerogels from waste cotton fabrics by synthesizing magnesium hydroxide nanoparticles within cellulose gel nanostructures, followed by freeze-drying. The nanoporous cellulose gel acted as a scaffold for the non-agglomerated growth of magnesium hydroxide nanoparticles, resulting in excellent flame retardancy with extinguishing within 40 s. Despite a slight decrease in specific surface area from 38.8 to $37.6 \text{ cm}^2 \text{ g}^{-1}$, thermal conductivity moderately increased from 0.056 to $0.081 \text{ W m}^{-1} \text{ K}^{-1}$, maintaining excellent heat insulating performance. The simple process and availability of biomass waste make this approach suitable for industrial-scale production, offering potential for future green building materials. Pandey et al.¹⁵⁴ showcased a nano-composite polymer electrolyte in a prototype magnesium cell, utilizing a composite of magnesium and multi-walled carbon nanotubes as the negative electrode and vanadium pentoxide as the positive electrode. They substituted magnesium with an Mg-MWCNT composite as the negative electrode to enhance rechargeability. However, the discharge capacity diminished after 10 cycles due to the passivation of the negative electrode. Nonetheless, the electrolyte exhibited flexibility and mechanical strength, remaining free-standing.

7.2 | Fuel cell

The need for alternative renewable energy sources has grown due to the depletion of fossil fuels and natural gas, as well as their detrimental effects on the environment. The polymer electrolyte FC is an appealing kind of FC that has shown to be an interesting field for further investigation. This is because of a number of advantages, such as high PD, short warm-up times (because of low operating temperatures), and low risk of electrolyte leakage.¹⁵⁵ Both chitosan and cellulose have demonstrated exceptional potential because of their superior mechanical, thermal, and water-retention properties. A high-performance membrane needs to have a few specific qualities. Among the cellulose and its derivatives that are frequently used as a base electrolyte are BC, CNC, and CNF. Furthermore, after blending with other polymers, this BC's well-developed 3D network structure with pores allows for ion transfer and good biocompatibility.¹⁵⁶ The production of organically produced ion-exchange membranes using bio-based materials is speeding up the development of sustainable dividers for PEFCs.

This study combined lignosulfonates (LS), a derivative of the sulfite pulping method, and BNC, a bacterial polysaccharide to create fully bio-based ion-exchange membranes. The natural cross-linker tannic acid and the lignin derivative were dissolved into an aqueous solution and infused into the wet BNC nanofibrous 3D structure. The resulting free-standing membranes showed improved mechanical properties with a maximum Young's modulus of 8.2 GPa, good moisture-uptake capacity with a maximum value of approximately 78% after 48 h, and suitable thermal-oxidative stability up to 200°C in either N₂ (inert) or O₂ (oxidative) atmospheres. Furthermore, by combining the conducting LS with the mechanically robust BNC, the membrane with the highest lignosulfonate concentration was given a maximum ionic conductivity of 23 mS cm⁻¹ at 94°C and 98% relative humidity. These BNC/LS water-mediated ion conductors have good mechanical performance, thermal-oxidative stability, and water-uptake capacity, making them environmentally friendly substitutes for ion-exchange membranes in PEFCs. Furthermore,

humidification and their conductivity were positively correlated (Figure 11).¹⁵⁷

Carbon-based substances have significant potential to replace noble metal composites owing to their excellent starting material, design controllability, and high stability.¹⁵⁸ The major stoppages for carbon-based substances within commercial-scale implementations are their low electrocatalytic activity and working durability.¹⁵⁹ Doping heteroatoms within the carbon substrate have been regarded as a sufficient technique to improve oxygen reduction reaction (ORR) performances, even though nitrogen-based CNTs have been shown to exhibit effective electrocatalytic performance.¹⁶⁰ This has demonstrated that heteroatoms may disrupt the electroneutrality of carbon particle connections and create more charged sites to adsorb and reduce oxygen, thereby improving ORR performances.¹⁶¹ The electrocatalytic performance may be enhanced by controlling the material network to facilitate electron and mass transfer.¹⁶² Because of the inherent properties of self-doping heteroatoms and distinct channels, biomass resources have attracted

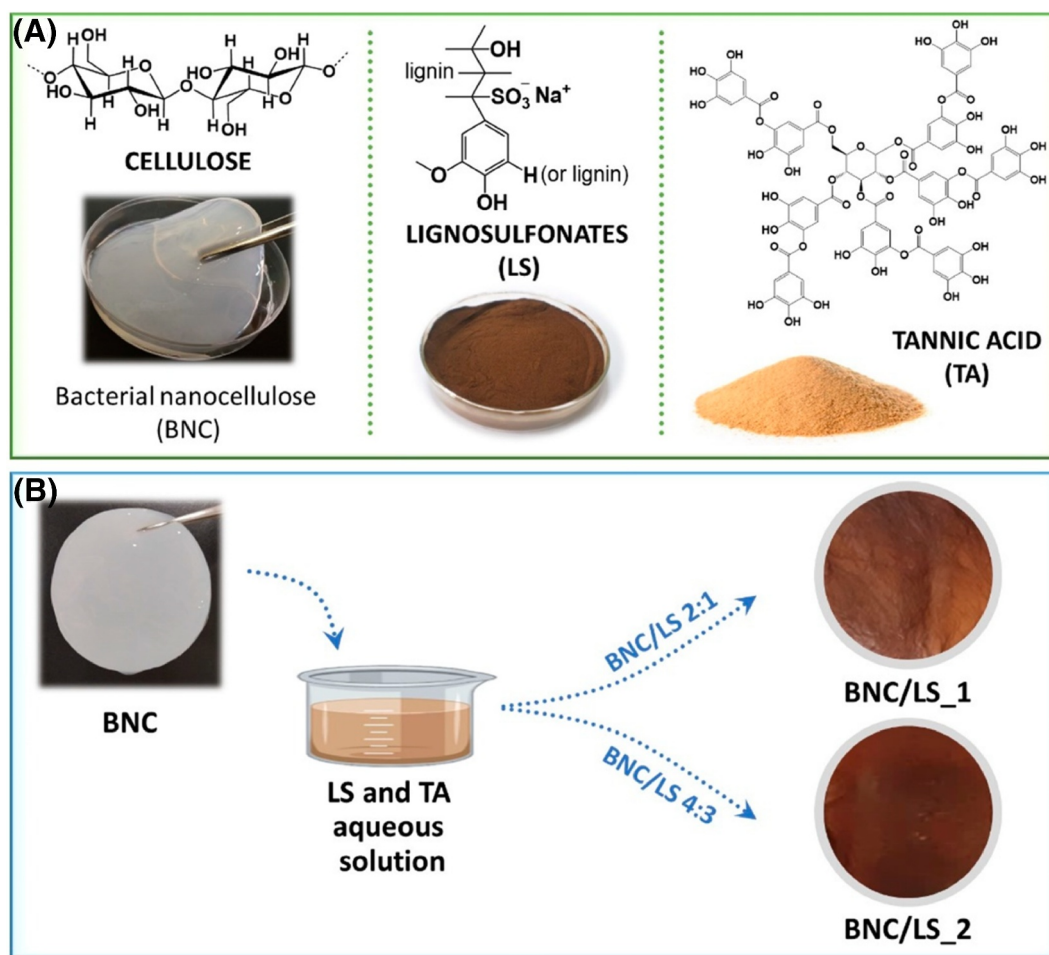


FIGURE 11 (A) Chemical composition and images of the precursors, which are BNC (wet membrane), LS (powder), and TA (powder), as well as (B) the production process for the BNC/LS-based membranes and the corresponding dry membrane photos. Reproduced with permission.¹⁵⁷ Copyright 2020, MDPI. BNC, bacterial nanocellulose; LS, lignosulfonates; TA, tannic acid.

increased attention. Figure 12A shows the adequate and stable materials originated from cellulose for microbial FCs (MFCs). The edges of the particle network, the increased specific surface area, and the additional functional zones toward CON-900 for MFCs were demonstrated by the highest PD and polarization measures that were documented (Figure 12B). After culturing all MFCs for 21 days, an enhancing result voltage may be maintained, and all CBMs gradually reach a steady voltage (Figure 12C). The ORR on the CON-900 air cathode is more efficient than that of the OC-900 and CN-900 air cathodes, as demonstrated by the elevated potential scaffold. As illustrated in Figure 12D, the CN-900-MFC produces the maximum PD of $(412 \pm 5) \text{ mW m}^{-2}$, which is 1.45 times greater than that of the OC-900-MFC ($283 \pm 4) \text{ mW m}^{-2}$ but significantly less than that of the Pt/C-MFC ($1062 \pm 14) \text{ mW m}^{-2}$). The polarization arcs of every MFC are displayed in Figure 12E. The OCV of CON-900-MFC is 704 mV, which is significantly higher than that of C-900-MFC (552 mV) and CN-900-MFC (665 mV) and achieves 95.1% of Pt/C-MFC (740 mV). As seen in Figure 12F, the anode and cathode voltages were computed to determine which electrode was more

common in MFCs. Chronoamperometric (CA) analysis showed the working durability of CON-900 and 20% Pt/C. As schemed within Figure 12G, CON-900 only shows a current reduction of 9.0% after steady use for 12,000 s, which is higher compared to 20.0% current reduction of its primary current appears in 20% Pt/C. Additionally, linear scanning voltammetry (LSV) profiles were documented after and before the i - t measurement, as illustrated within Figure 12H, where the little lowering half-wave voltage (0.01 V) and determining current density (0.2 mA cm^{-2}) may be followed within CON-900. By using Figure 12I, it is possible to obtain the reducing values of half-wave voltage and determining current density in 20% Pt/C. For methanol, these values reach 154 mV and 0.193 mA cm^{-2} , while for potassium thiocyanate, they reach 310 mV and 0.349 mA cm^{-2} . On the other hand, CON-900 experiences a slight 10 mV drop in half-wave voltage (Figure 12J). Subsequently calcining the combination of NH_4Cl and oxidized cellulose on 900°C , the surface of CON-900 was distinguished using SEM and transmission electron microscopy (TEM) investigation. Corresponded with the enormous carbon block within OC-900 (Figure 12K), CN-900 only shows surface

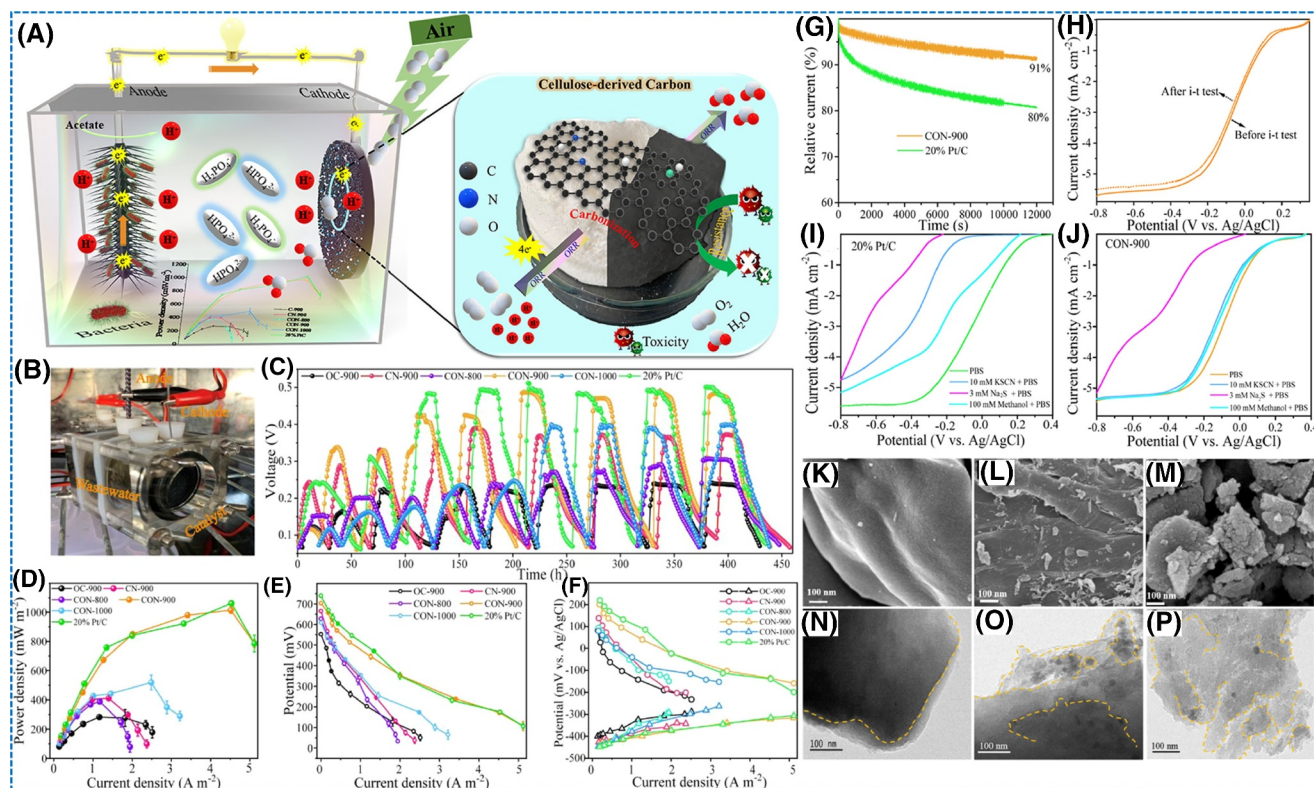


FIGURE 12 (A) Adequate and stable materials emanated from cellulose for MFCs. (B) Graphic expression for operating MFCs. (C) Cell voltage profile, (D) PD curves, (E) broad MFC polarization arcs, and (F) cathode and anode polarization profiles toward MFCs. (G) CA analysis and (H) comparing the LSV arcs around 12,000 s before and after the i - t test. The effects of different toxic implications on the LSV curves for (I) 20% Pt/C and (J) CON-900. SEM images of the (K) OC-900, (L) CN-900, and (M) CON-900. TEM images of the (N) OC-900, (O) CN-900, and (P) CON-900. Reproduced with permission.¹⁶³ Copyright 2022, American Chemical Society. CA, cellulose acetate; LSV, linear scanning voltammetry; MFCs, microbial fuel cells; PD, power density; SEM, scanning electron microscopy; TEM, transmission electron microscopy.

deterioration with a diminutive portion of carbon filaments connecting to the carbon block (Figure 12L), while CON-900 ultimately delivers multiple carbon particles by a coarse texture (Figure 12M). Their surfaces were additionally determined by TEM photos exhibited in Figure 12N–P.¹⁶³

Here, Kim et al.¹⁶⁴ showed the efficacy of BNC as a reinforcement toward proton-exchange membrane fuel cell composites, creating it with a graphene-incorporated Pt nano sandwich configuration prepared using a facile chemical vapor deposition (CVD) process (Figure 13A). TEM imaging of Pt nanoparticles on BNC substrates made with Pt-CVD shows that NPs with a diameter of about 2 nm were uniformly sustained on BNC's exterior (Figure 13B,C). TEM images of the graphene-incorporated Pt composite are shown in Figure 13D–F, showing that the particles have one to two inferior graphitic coatings. Because the Pt NPs

matured under the somewhat elevated heat of CVD, up to about 2.6–3.0 nm, their dimensions did not significantly improve. Cyclic voltammetry (CV) was used to estimate the electrochemical activity of the Pt-based BNC (Pt/BNC) and graphene-incorporated Pt/BNC (G-Pt/BNC) composites, as well as a conventional retail Pt/C composite for comparison (Figure 13H–J). The G-Pt/BNC, Pt/BNC, and Pt/C composites' electrochemical surface areas were approximated and estimated (Figure 13K,L). Subsequently, in 2000 scans, the Pt/BNC catalyst showed an activity decline (65%).

7.3 | Solar cell

The fundamental parts of a solar cell are an aluminum frame, light-transmissive glass, ethylene vinyl acetate glue, a battery sheet, and a backplane.

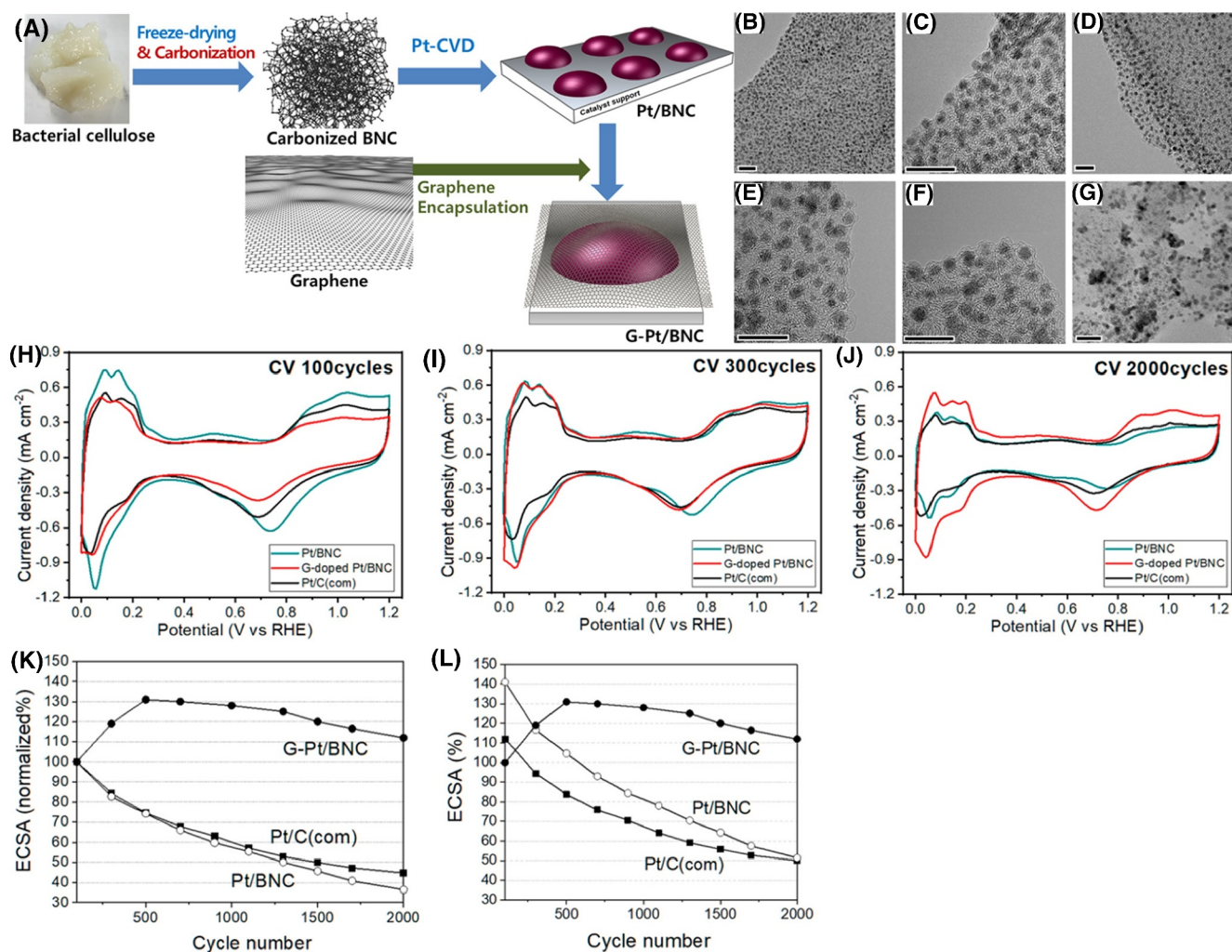


FIGURE 13 (A) Graphic description of the synthesis of our graphene (G)-incorporated G-Pt-BNC roll composite. (B, C) TEM pictures depicting Pt/BNC composites (Pt 20 wt%) prepared using Pt-CVD at 200°C for 2 h. (D–F) TEM photos of fresh G-Pt/BNC composite (Pt 20 wt%) shielded with G incorporation and (G) Pt/C (HiSPEC3000, retail) as a point of reference. G-Pt/BNC, Pt/BNC, and Pt/C (H), (I) after 300, (J) after 2000, and (K) after 100 CV scans; (L, K) after 2000 CV scans; and (K) standardized ECSA (m²/g) plot. Reproduced with permission.¹⁶⁴ Copyright 2021, American Chemical Society. BNC, bacterial nanocellulose; CV, cyclic voltammetry; CVD, chemical vapor deposition; ECSA, electrochemical surface area; TEM, transmission electron microscopy.

Solar cells use the photovoltaic effect to convert light into electrical energy. Its primary functioning component is the battery piece.¹⁶⁵ The development of heat-shielding materials that autonomously control sunlight without electricity is necessary to suppress temperature increase in an environmentally friendly way because reducing the amount of electricity used by cooling systems is crucial to lowering CO₂ emissions. HPC has attracted attention as a temperature-responsive polymer with a lower critical solution temperature. A thermochromic substance called HPC adapts its optical characteristics to variations in the ambient temperature.¹⁶⁶ Evaporative and radiative cooling, standard methods of heat dissipation in nature, are viable alternatives to more traditional methods for delivering large cooling capacities with minimal energy use and waste discharge. Here, hydrophobization, freeze drying, hydrogelization, and dip-coating were used to create a hydrophobized BC aerogel/PVA hydrogel bilayer gel that combined radiative and evaporative cooling. The resulting bilayer gel has high visual reflectance (98.8%) and emissivity in the atmospheric window band (0.86) due to the intrinsic fiber architectures of BC aerogel. This significantly reduces heat input and increases thermal aggregation dissipation.¹⁶⁷ The performance of SC devices is optimized by increasing the transparency and optical haze of substrates, which improves light scattering and increases the rate at which active species use light.¹⁶⁸ Hydrophilic gels, also known as hydrogels commercially, are promising for applications requiring long-term hydration or heat regulation. Hydrogels are a unique material for this use because of their ability to contain water, intelligence, and thermal properties. Hydrogels act as a heat-removal agent from the back surfaces of solar

panels, lowering their temperature and boosting electricity output. The system primarily uses the three-step water desorption cooling process.¹⁶⁹ The solar panel's hydrogel bed acts as a heat conductor; the first process is heat diffusion. The second phase is water desorption, in which water vapors remove waste heat from the system. The solar panel must be cooled down in the final step using further heat removal and water absorption, as shown in Figure 14. The size of the hydrogel beads reduced in the final phase as the water evaporated, as can be seen below in Figure 14. Four different setups were evaluated and compared with an uncooled system. One-row beds, two-row beds with fins, and three-row beds with fins were the four configurations that were looked at. In the studies, three radiations were used: the highest (1000 W/m²), average (800 W/m²), and lowest (600 W/m²). The hydrogel bed effectively lowered the panel's temperature at different radiation intensities, according to the results. At radiation intensities of 600 and 1000 W/m², the temperature was lowered to 9.6 and 9°C, respectively. At 1000 W/m², the fin-equipped three-row bed improved estimated efficiency by 7.2%.¹⁷⁰

7.4 | Flexible supercapacitor

A supercapacitor cannot function properly without separators because they prevent short circuits from happening in the first place and guarantee that electrolyte ions can pass through, facilitating efficient charging and discharging. CNF with a tunable thickness and high porosity make good separators for FSCs.¹⁷¹ Recently, polymeric materials have attracted much attention for supercapacitor applications due to

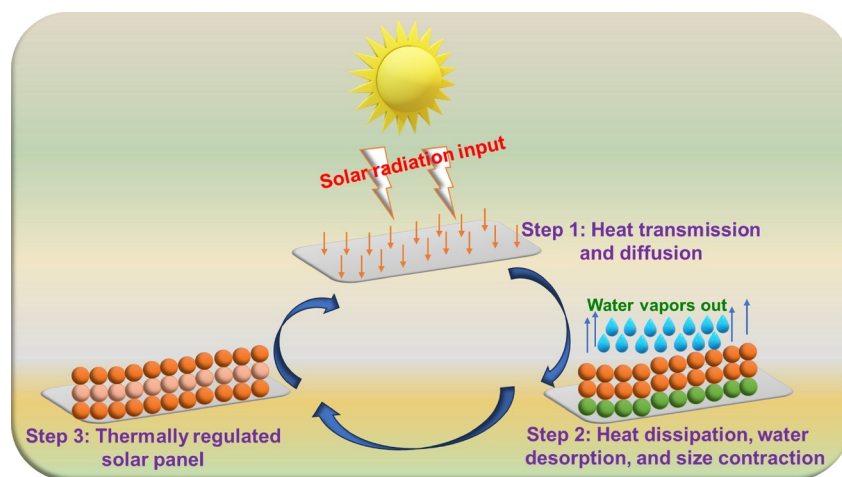


FIGURE 14 The thermal control process for the hydrogel bed-cooled solar panel. Adapted from Ref. 170.

their advantageous qualities, such as flexibility, lightweight, and steady cycling performance.^{172,173} Organic polymeric materials have been expanded to include biopolymers to create ESDs for the future. Supercapacitors need an electrolyte that links the two electrodes ionically in addition to having the two electrodes separated by an ion-permeable separator.^{174–176} The four steps of the vapor phase polymerized (VPP) approach are illustrated in Figure 15A: (1) inking the CP matrix with Fe^{3+} oxidant media; (2) water-assisted vacuum vapor step polymerization; (3) flushing the sample to release excess oxidant; and (4) drying of the polymerized CP model and CP matrix. Two VPP operations using the aforementioned phases are experienced by one VPP cycle, which is examined in this paper. To demonstrate the electronic performance, the material's resistance and film resistance were both displayed. The resistance of

1×1 cm specimens was estimated, as shown in Figure 15B. Resistances of 109.4, 63.1, and 24.3Ω were the main resistances obtained by poly(3,4-ethylenedioxythiophene) (PEDOT)/CP-1, PEDOT/CP-5, and PEDOT/CP-10. Consequently, the resistance of the PEDOT/CP decreased as the VPP processes improved. A four-point investigation process was used to register the specimen's film resistances, as shown in Figure 15C. As a result, the resistances of the electrode were recorded after each of the 50 bending processes to complete the bending analysis of PEDOT/CP electrodes. Since conductivity and resistance have an inverse relationship, the conductivity retention against bending processes was calculated, as shown in Figure 15D. For every bending process, a bending curve of $\sim 90^\circ$ was achieved (inset of Figure 15D). The high conductivity of the PEDOT/CP composite material allowed for a double character job as an

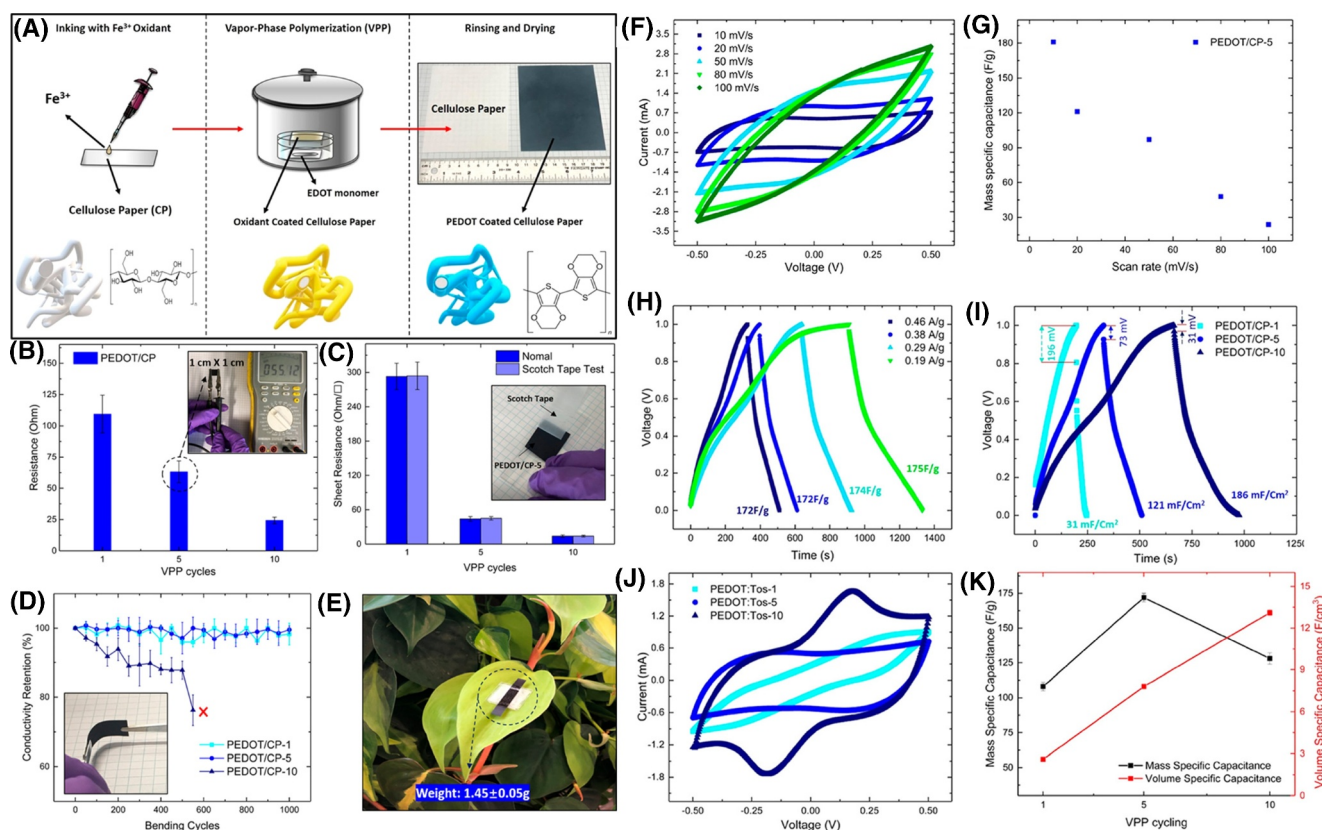


FIGURE 15 (A) Using a photograph of the finished product, the VPP technique consists of inking the substrate with an oxidant, polymerization, rinsing, and drying. Their matching three-dimensional representations can be seen in the bottom row. (B) Resistance measurements upon PEDOT/CP composites with varying VPP scans using the two-probe technique. (C) PEDOT/CP composite sheet resistances for different VPP cycles both before and after the Scotch tape test. (D) All of the PEDOT/CP materials undergo bending cycling testing. (E) An constructed supercapacitor that uses PEDOT/CP composite as the electrode and current collector is demonstrated in a lightweight manner. (F) PEDOT/CP-5 supercapacitor CV profiles at several sweep rates. (G) PEDOT/CP-5 supercapacitor mass specific capacitances at various scan speeds. (H) PEDOT/CP-5 supercapacitor CD profile at various current densities: 0.19, 0.29, 0.38, and 0.46 A/g . (I) CD profiles of 1, 5, and 10 VPP cycled composite-based PEDOT/CP supercapacitors with a constant current density of 0.32 mA/cm^2 . (J) CV profiles at a constant sweep rate of 10 mV/s for assembled PEDOT/CP supercapacitors with various VPP cycles. (K) Calculated mass specific capacitances and volume specific capacitances for PEDOT/CP electrodes with varying VPP cycles using CD profiles as a reference. Reproduced with permission.¹⁷⁷ Copyright 2020, American Chemical Society. CD, charge-discharge; CP, conductive polymer; CV, cyclic voltammetry; PEDOT, poly(3,4-ethylenedioxythiophene); VPP, vapor phase polymerized.

electrode and a current collector. The typical built simple layered assembly PEDOT/CP-based supercapacitor weighed 1.45 g. It consisted of two electrodes measuring 2.3 cm by 0.6 cm, a KimWipe separator measuring 2.0 cm by 1.2 cm, and two tapes measuring 1.9 cm by 2.5 cm. Its lightweight characteristic was further illustrated by positioning the constructed capacitor on a leaf without any discernible strain (Figure 15E). The mass-specific capacitance of the supercapacitors was calculated by plotting their CV curves, as shown in Figure 15F. Capacitance decreased with increasing scan rate, and 24 F/g was found at 100 mV/s (Figure 15G). The charging-discharging (CD) curves were plotted to determine the PEDOT-specific capacitance for device performance. There were no discernible changes in mass-specific capacitances within the chosen current densities range of 0.19–0.48 mA/g (Figure 15H). Compared to CV profiles, the specific capacitance results computed using CD profiles were 2% lower. As seen in Figure 15I, CV curves for various VPP scans were evaluated at the same current density of 0.32 mA/cm². As seen in Figure 15J, CV profiles were performed at a constant sweep rate of 10 mV/s, confirming the capacitance shift as the number of VPP scans increased. Higher capacitance was shown by the rise in integrated areas seen after extra cycles of polymerization from PEDOT/CP-1 to PEDOT/CP-10. The volume-specific capacitances (C_v) of the entire electrode, including the PEDOT and CP matrix, increased dramatically as the VPP cycles increased. They were 2.6, 7.9, and 13.7 F/cm³ for PEDOT/CP-1, PEDOT/CP-5, and PEDOT/CP-10 supercapacitors, respectively (Figure 15K). The increasing concentration of PEDOT elements in the CP matrix was suggested as the cause of the notable improvement in volume specific capacitance.¹⁷⁷

In the flexible/wearable electronics market, FSCs are competitive options for ESDs due to their high PD, incredibly long cycle life, and remarkable mechanical flexibility to withstand bending, twisting, or folding.¹⁷⁸ The development of high-performance flexible electrode materials has attracted tremendous effort. Supercapacitor performance may be enhanced by a combination of factors, including high porosity, high electroactive material loading (66.67 wt% SupCP1/2 total electrode mass), and the unique supramolecular structures of the SupCP aerogels. All three samples had a boxed or box-like CV shape, as indicated by the CV curves in Figure 16A–C, which suggests good capacitive behavior. The excellent capacitance behavior at high scan rates was attributed to the excellent transport of electrolyte ions through the highly porous networks. Electrochemical impedance spectrum and Nyquist plots were used to analyze the internal impedance and ion transport performance of the SupCP aerogels (Figure 16D–F). From the CV curves,

the area-normalized capacitance (C_s) was computed. SupCP1/2 had the biggest C_s in Figure 16G, whereas SupCP1/1 and SupCP2/1 had smaller C_s values. As can be seen in Figure 16H, the capacitor retention rate of SupCP1/2 maintained a good degree of symmetry at 75.6% following 2000 cycles of charge and discharge. The ohmic (IR) drop of only about 0.1 V showed that the SupCP1/2 electrode had good cycle stability, a small equivalent series resistance, and fast CD performance. To create an FSC, they chose a PP nonwoven composite PVA/H₂SO₄ solid electrolyte, and we evaluated the SupCP electrode's performance in the H₂SO₄ electrolyte first. On the CV curve in Figure 16I, three pairs of redox peaks in the SupCP1/2 electrode are visible. We designed a sandwich-type FSC by compressing a SupCP1/2 aerogel on both sides of the ultrathin PP/H₂SO₄/PVA flexible electrolyte (Figure 16J). This innovative structure gave the supercapacitor a high level of mechanical strength and stability and guaranteed its flexibility. The PP nonwoven fabric's pores were filled with H₂SO₄/PVA in Figure 16K,L, and the three parts were subsequently combined to form a single, flexible electrolyte interlayer with fiber diameters ranging from 1 to 5 μ m. This arrangement aided ion and electron diffusion. As shown in Figure 16M, the ultrathin flexible electrolyte was closely bonded to the SupCP1/2 electrode. This improved the electrochemical performance of the FSC and significantly reduced the electrical resistance between the interfaces. To test the performance of the FSC, we connected two flexible all-solid supercapacitors to a blue LED light (Figure 16N,O). The LED could be turned on in both its bent and unbent states thanks to the FSCs.

7.5 | Carbon materials derived from cellulose

Carbon materials are the most promising electrode materials for industrial applications. Using 1D CNT, CNF, and 2D rGO signifies the most promisingly explored carbon materials for flexible devices. The development of 1D structured CNT has led to the creation of flexible electrodes, leveraging their remarkable attributes such as high electrical conductivity, excellent chemical stability, elevated mechanical strength, and the ability to control regular pore structures. Wet spinning of a mixture solution containing BC and CNT can yield 1D nanofibers. The entangled system guarantees extended and continuous conductive paths along the nanofiber lengths and imparts a high degree of flexibility.¹⁷⁹ Incorporating NC with diverse electroactive materials is a substitute electrode binder, replacing traditional synthetic polymer-based binders. Most carbonaceous materials employed in ESSs are derived from

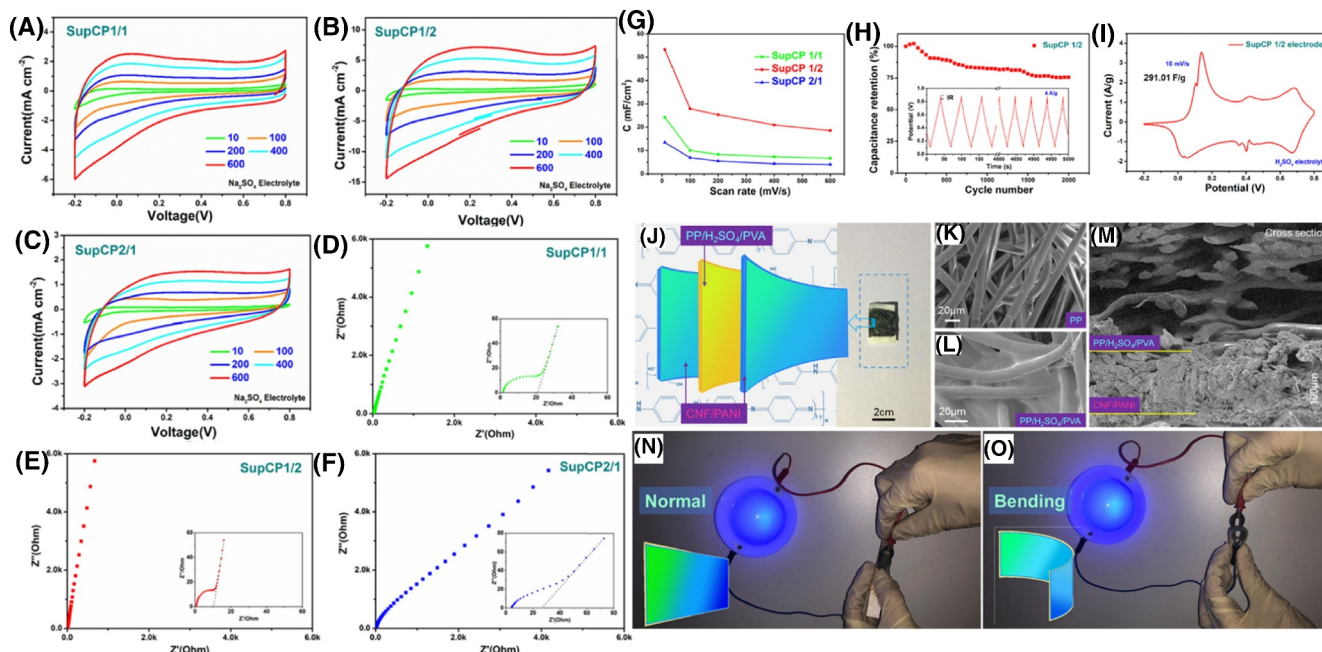


FIGURE 16 (A–C) CV data recorded in a 1 M Na_2SO_4 electrolyte at scan rates ranging from 10 to 600 mV/s. (D–F) Displays the Nyquist plots of (E) SupCP1/2, (D) SupCP1/1, and (F) SupCP2/1. (G) Abilities at different scan rates. (H) SupCP1/2 capacitance retention after 2000 charge and discharge cycles (the charge and discharge curve is inset). (I) SupCP1/2 electrode CV curve in H_2SO_4 electrolyte. (J) The FSCs structural diagram and digital image. (K, L) Field-emission scanning electron microscopy pictures of the composite $\text{H}_2\text{SO}_4/\text{PVA}$ solid electrolyte and CNF/PANI aerogels made of PP nonwoven fabric. (M) The flexible solid electrolyte and electrode cross sections. (N, O) Images of a FSCs that, when bent and tiled, illuminates a blue LED light with an increased brightness thanks to its outer casing. Reproduced with permission.⁶¹ Copyright 2019, American Chemical Society. CNF, cellulose nanofibers; CV, cyclic voltammetry; FSCs, flexible supercapacitors; PANI, polyaniline; PP, polypropylene; PVA, polyvinyl alcohol; SEM, scanning electron microscopy.

precursors sourced from fossil fuels. Recently, there has been considerable interest in NC as an eco-friendly precursor for producing carbonaceous materials. High-temperature pyrolysis on NC in an inert atmosphere transforms it into conductive carbon materials.^{180,181} Vocht et al.¹⁸² presented the novel stabilization process using a low-pressure furnace combined with an ammonium tosylate carbonization agent, which significantly enhances the mechanical properties of cellulose-derived CFs. Stabilization dwell time influences properties, with increased time leading to higher elongation at break. Tire cord fiber-based CFs exhibit higher tensile strength, while air gap-spun cellulosic fibers-based CFs show superior Young's moduli. The influence of low-pressure during stabilization requires further understanding, emphasizing the importance of controlled pyrolysis for optimal CB CF mechanical properties.

A straightforward, environmentally friendly, universal, and efficient method for creating large-scale 3D heteroatom-doped carbon nanofiber networks is described by Chen et al.¹⁸³ This technique allows for the successful pyrolysis of BC dissolved in H_3PO_4 , $\text{NH}_4\text{H}_2\text{PO}_4$, and $\text{H}_3\text{BO}_3/\text{H}_3\text{PO}_4$ aqueous solution to create 3D P-doped, N, P-co-doped, and B, P-co-doped carbon nanofiber networks. Figure 17A shows the general approach for creating free-standing heteroatom-doped carbon nanofibers. As illustrated

in Figure 17B, Long et al.¹⁸⁴ synthesized N-doped activated carbon (a-CBP) and carbon- MnO_2 hybrid material (c-BP/ MnO_2) as the negative and positive electrode materials for the asymmetric supercapacitor device, respectively, using BC as both a template and precursor. The CV curves at scanning rates from 20 to 500 mV s^{-1} generally retain rectangular shapes due to the material's excellent conductivity and porous structure (Figure 17C), even at high rates of 500 mV s^{-1} , indicating excellent capacitive behaviors. Additionally, the a-CBP electrode's galvanostatic CD curves (Figure 17D) are remarkably symmetrical and linear, suggesting excellent electrochemical reversibility, CD characteristics, and an ideal double layer capacitive characteristic.

Furthermore, even at a high current of 20 A g^{-1} , there is no noticeable iR drop for the charge/discharge curve, indicating low overall resistance. Figure 17E illustrates that a-CBP exhibits a significantly higher specific capacitance of 296 F g^{-1} at 2 mV s^{-1} in comparison to c-BC (161 F g^{-1}) and c-BP (208 F g^{-1}). Evidently, a high scan rate of 200 mV s^{-1} was also used to evaluate the cycle life of a-CBP (Figure 17F). It exhibits outstanding cycling stability after 10,000 cycles, maintaining a capacitance retention of 99% of the initial capacitance with no discernible change for the CV curves before and after 10,000 cycles (inset in Figure 17F).

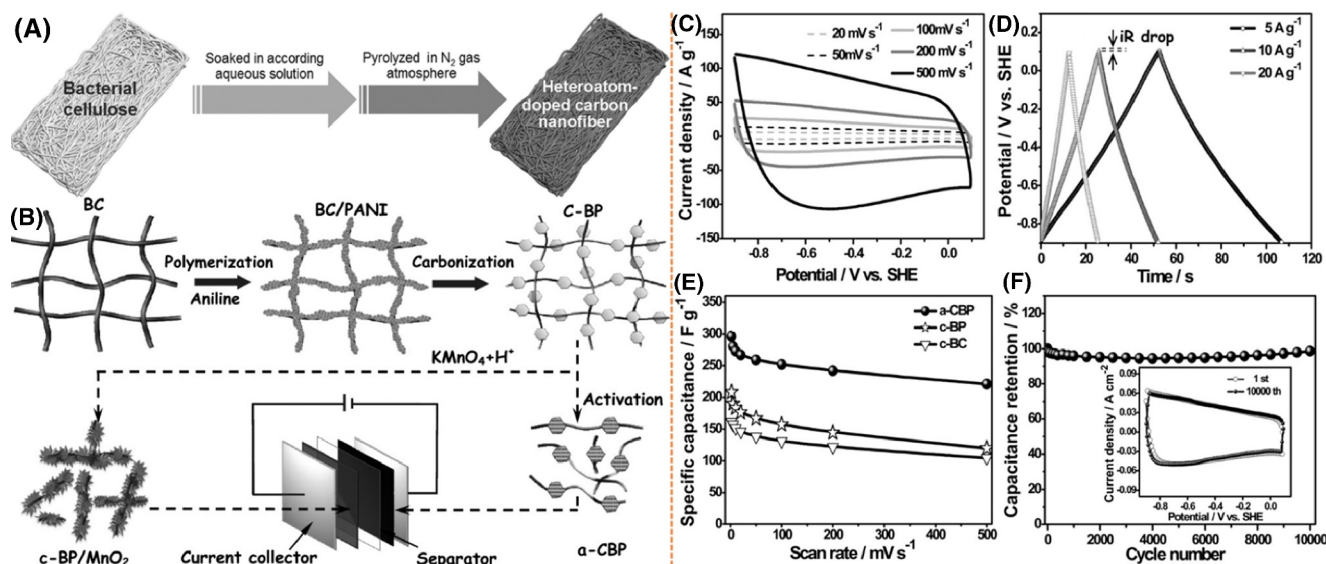


FIGURE 17 (A) Diagram showing the steps involved in creating the free-standing, heteroatom-doped carbon nanofibers. Reproduced with permission.¹⁸³ Copyright 2014, Wiley-VCH Verlag GmbH & Co. (B) A schematic diagram showing how BC served as a model and precursor to create the electrode materials used in the asymmetric supercapacitor device. (C) a-CBP CV curves in 6.0 M KOH electrolyte at various scan rates of 20, 50, 100, 200, and 500 $mV s^{-1}$. (D) a-CBP GCD curves at various current densities of 5, 10, and 20 $A g^{-1}$. (E) The specific capacitance of the electrodes a-CBP, c-BP, and c-BC at various scan rates ranging from 2 to 500 $mV s^{-1}$. (F) a-CBP cycle performance at 200 $mV s^{-1}$ scan rate (the 1st and 10,000th cycle CV curves at 200 $mV s^{-1}$ are displayed in the inset). Reproduced with permission.¹⁸⁴ Copyright 2014, Wiley-VCH Verlag GmbH & Co. a-CBP, activated carbon; BC, bacterial cellulose; CV, cyclic voltammetry; GCD, galvanostatic charge-discharge.

These days, there is a sharp increase in interest in batteries and other CB electrochemical ESDs such as supercapacitors. This interest is sparked by the fact that cellulose can be used to create functional separators, lightweight current collectors, and electrodes with high energy and power densities.^{185,186} As a result, CB supercapacitors and batteries are now possible. Given that cellulose can be produced inexpensively and in large quantities through processes such as papermaking, it makes sense to believe that cellulose will play a significant role in the development of sustainable electrochemical ESDs. These devices have the potential to serve as affordable substitutes for batteries based on lithium, for example, and to enable the creation of novel electrochemical energy storage applications.¹⁸⁷

In order to tackle this difficult problem, Cho and his coworkers¹⁸⁸ employed a naturally occurring/ecofriendly cellulose-mediated (CM) cell architecture strategy. They were able to demonstrate a new class of hetero-nanonet (HN) paper batteries based on 1D CNF (CNFs)/multiwall carbon nanotubes (MWNTs). The HN paper batteries are made of microporous CNF separator membranes and electrode-active powders (CM electrodes) that are interwoven with MWNT. Electrochemical reaction kinetics are facilitated by the construction of 3D bicontinuous electron/ion transport pathways in the CM electrodes, which is made possible by the CNF/MWNT heteronet-mediated material/structural uniqueness. A schematic illustration

was provided (Figure 18A), which showed the overall fabrication procedure as well as the photographs that represented each step. Without compromising its dimensional stability, the self-contained, ultrathick (thickness $\approx 1400 \mu m$) CM $LiFePO_4$ cathode was successfully fabricated (Figure 18B).

The ultrathick CM $LiFePO_4$ cathode's SEM images (Figure 18C) show that the $LiFePO_4$ powders and CNF/MWNT heteronets are evenly distributed in the through-thickness direction. Measuring the discharge capacity per cathode area as a function of areal mass loading allowed researchers to better understand the electrochemical performance of thickness-tunable CM $LiFePO_4$ cathodes. This was achieved by simply adjusting the suspension amount of the electrode mixture during the vacuum-assisted infiltration. With areal mass loading, the areal capacity tends to increase linearly (Figure 18D,E). During the cycling test, the HN paper cells displayed stable charge/discharge profiles (Figure 18F). The long-term structural and electrochemical durability of the ultrathick CM electrodes in the HN paper cell was confirmed by the morphological analysis (Figure 18G), which shows that the CNF/MWNT HN structure of the electrodes was well preserved following the cycling test.

As shown in Figure 19, NC-based composites are used as flexible electrodes, separators, and electrolytes in supercapacitors and batteries. The separator is essential in preventing short circuits between the electrodes of supercapacitors and batteries. It must

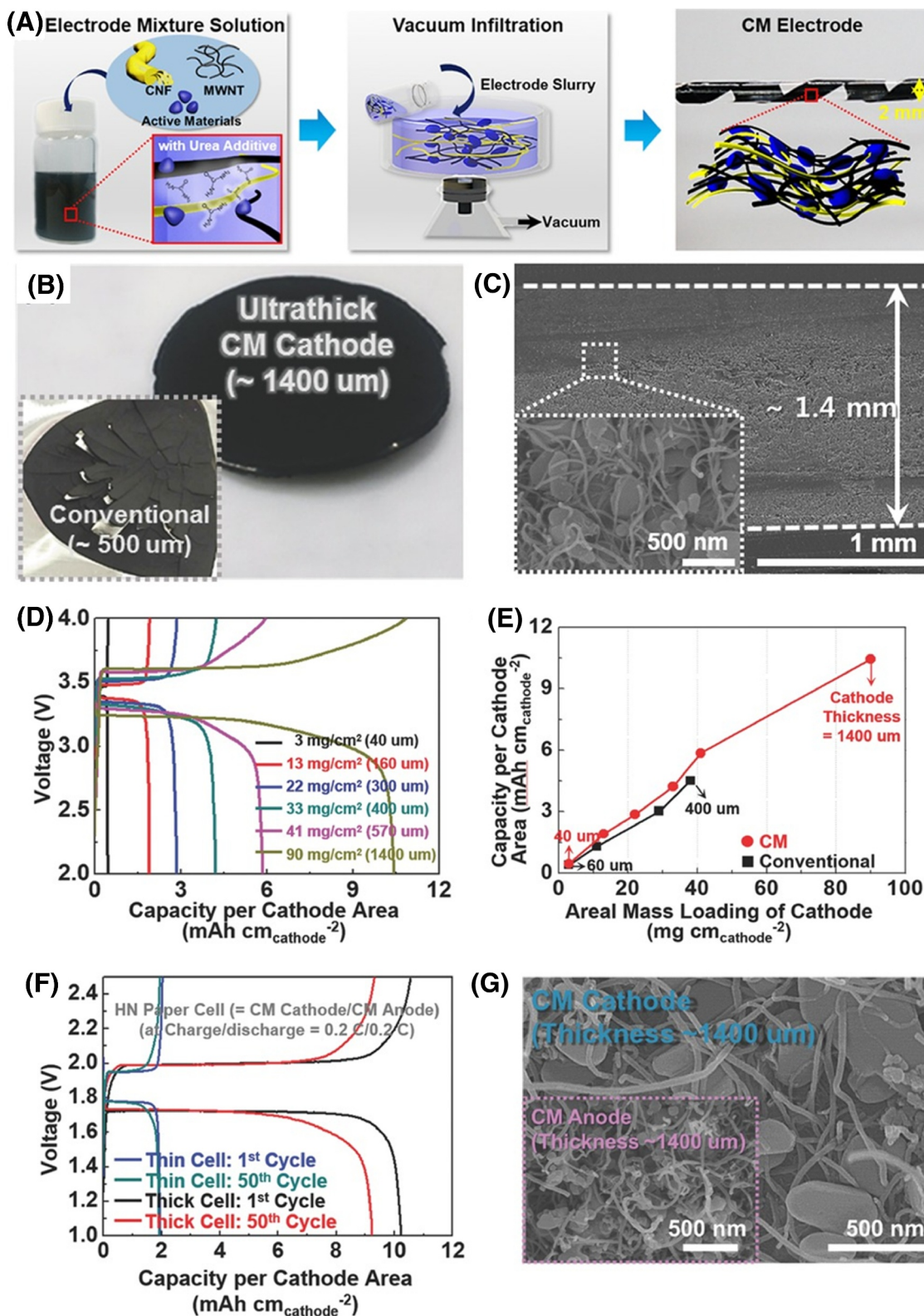


FIGURE 18 (A) Pictures and schematics showing the entire CM electrode fabrication process. Electrochemical performance and morphology of CM electrodes with varying thickness. (B) A picture showing the CM cathode, which is self-supporting and dimensionally acceptable, with a thickness of approximately 1400 μm. The conventional cathode (thickness ≈500 μm) exhibits poor structural stability, as seen in the inset image. (C) An SEM cross-section of the ultrathick CM cathode with the high-magnification view shown in the inset image. (D) The charge/discharge profiles of CM LiFePO_4 cathodes; (E) a comparison of CM and conventional cathodes' capacity per cathode area ($\text{mAh cm}_{\text{cathode}}^{-2}$). (F) HN paper full cells' cycling performance in relation to electrode thickness. (G) Following the cycling test, SEM pictures of the CM cathode and CM anode in the HN paper full cell. Reproduced with permission.¹⁸⁸ Copyright 2015, Wiley-VCH Verlag GmbH & Co. CM, cellulose-mediated; HN, hetro-nanonet; SEM, scanning electron microscopy.

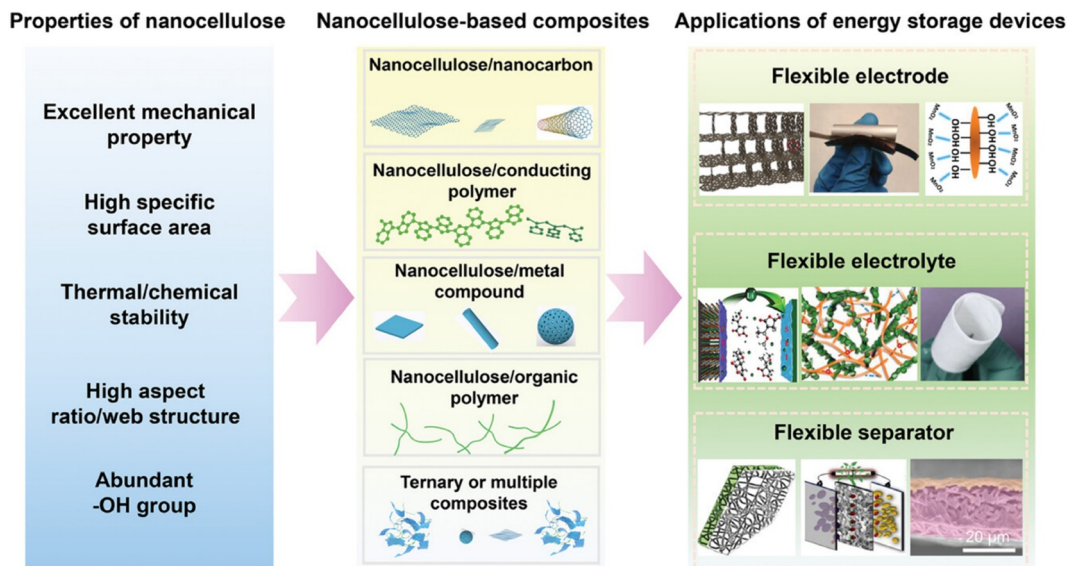


FIGURE 19 Demonstrates the development of NC-based composites as flexible electrodes, separators, and electrolytes in the field of EES. Reproduced with permission.¹⁸⁹ Copyright 2021, Wiley-VCH GmbH. EES, electrochemical energy storage; NC, nanocellulose.

have strong thermal stability, high ionic conductivity, and insulating qualities. High ionic conductivity, temperature tolerance, and a wide voltage window are usually necessary for polymer-based electrolytes. Furthermore, in flexible ESDs, gel-state or solid-state polymer-based electrolytes perform the dual roles of an electrolyte and a separator.¹⁸⁹

In order to address water-induced corrosion and uncontrolled dendritic growth in Zn metal anodes within aqueous ZIB, a composite membrane combining CNC and graphene was developed. The research demonstrated that by dispersing cations and blocking anions, the CNC-graphene interface layer—which is characterized by a negative surface charge—could cause a deionization shock. Zn deposition was redirected as a result, paralleling the (0002) Zn plane.¹⁹⁰

Many natural biopolymers have recently been extracted, such as chitosan, collagen, silkworm silk, spider silk, gelatin, and cellulose. These materials show promise for use in many flexible devices (such as portable ESSs and flexible sensors).¹⁹¹ Cellulose, is produced by certain microbes like *Acetobacter xylinum* and is present in a wide variety of plants. Printing paper, packaging, textiles, medical supplies, and medications have all been made with it on a large scale.^{137,192}

8 | CONCLUSION AND FUTURE CHALLENGING

In this review article, we concluded cellulose is a highly adaptable polymer that is easy to produce and extract. Every community on earth creates a significant

number of CB wastes, which represent a resource that is still substantially under-utilized. Solvents have played and will continue to play an essential role in cellulose research and development. Nonetheless, cellulose is soluble in a range of solvents with middling properties, including NMMO, DESs, and ILs. In many organic solvents and water, cellulose is insoluble. It can also dissolve in water at both high and low pH levels, particularly when combined with a cosolute with an intermediate polarity.

Numerous characteristics of hydrogels include strength, transparency, and swelling ratio. Cellulose hydrogels are regarded as useful biocompatible materials that can be used in medical devices that support, replace, or treat any tissue organ. These hydrogels also show significant promise for usage in innovative materials, agriculture, and other beneficial industrial applications. The main emphasis of this study is the energy storage technologies, such as batteries, supercapacitors, and solar cells. Despite extensive research on cellulose- and cellulose-derived hydrogels, several obstacles must be addressed before commercialization. The investigation of the hydrogel synthesis method is maturing due to the quick growth of flexible materials and wearable electronic materials. Creating materials such as cellulose, which are resourceful and environmentally friendly and greatly enhance hydrogel performance, is critical. Ionic conductivity changes brought on by solvent water evaporation can impact the electrolyte's lifespan. Excessive alteration of polymer chains may make production more expensive and complicated, reducing industrial output.

CB materials offer great potential for sustainable applications due to their renewability and

biodegradability. Yet, challenges persist in enhancing properties through functionalization and integrating cellulose into advanced materials while ensuring scalability and commercial viability. Environmental impact, safety, and regulatory compliance are vital considerations. While cellulose improves mechanical properties such as tensile resistance in hydrogel electrolytes, it fails to lower their freezing point. To address this, researchers incorporate antifreeze agents such as glycerol. However, this limitation severely hampers hydrogel electrolyte performance in low-temperature conditions. Overcoming these hurdles demands interdisciplinary collaboration and innovative approaches to exploit CB materials for sustainable global solutions fully.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

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