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REVIEW ARTICLE

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Biopolymer-based composites for sustainable energy storage: recent developments and future outlook

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Abstract

Over the past decade, biopolymers made from renewable resources like plants, algae, seashell waste, and seaweed have become increasingly popular as industries strive to reduce their environmental pollution without compromising socioeconomic growth. Biopolymers are often regarded as a significant alternative to conventional materials due to their low weight, great strength, stiffness, biostability, and non-toxicity. Therefore, industries are beginning to adopt the use of biopolymers, including those dealing with packaging, agriculture, automobiles, healthcare, as well as energy harvesting. Supercapacitors and batteries are two examples of electrochemical devices for energy storage that can be made using bespoke biopolymers and their composites. Although biopolymers' potential uses are restricted, they are nevertheless useful when combined with other materials to create composites. This boosts the electrochemical efficiency of the biologically active molecules and also enhances their inherent physical features. This review focuses on recent developments, specifically the use of diverse biopolymers and composites for batteries and supercapacitor applications, followed by future perspectives.

Keywords Energy harvesting, Batteries, Supercapacitors, Biopolymers

Introduction

Micro- and nanoscale polymer composites have gained a lot of interest in the electronics industry particularly in energy storage and energy generation during the past few decades (S. Kumar, Yadav, Prakash, et al. 2022b). Polymer nanotechnology has seen rapid growth in the electronics industry as a result of its low production cost, light weight, high mechanical strength, flexibility, high breakdown strength, degradability, availability, low density, low cost, and ease of processing compared to ceramic and metallic materials (Rahul Singh and Rhee 2019) (Saraswat and Kumar 2023; Yadav et al. 2022). Polymer-based dielectric capacitors are used in numerous energy-related applications, including power transmission, advanced electronics, microelectronics, hybrid electric cars, microelectronic systems, and high-powered weapons (Anthony et al. 2021; Karki et al. 2021; Okonkwo et al. 2017). The polymer-based dielectric capacitors offer numerous advantages over their counterparts, such as increased power density, efficiency, durability, and affordability (Muthukrishnan et al. 2019; Siddiqui et al. 2019).

Modern electrochemical energy storage devices are both prohibitively expensive and limited in their scalability. Consequently, advances in electricity storage would allow for a more rapid adoption of renewable electricity in electrical power grids, cars, and mobile gadgets when off-grid. Even in remote areas, it has the potential to significantly alter the daily routines of the vast majority of people throughout the world who do not have access to power. As the use of intermittent electricity from wind turbines and solar photovoltaics continues to develop, it

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becomes increasingly important to find cost-effective and scalable materials for electrical energy storage that can be rapidly expanded to meet the demand (S. Kumar, Yadav, and Maiti 2022a).

Biopolymers have emerged in response to the demand for eco-friendly materials in recent years (N Patra et al. 2015; Niranjan Patra et al. 2016; Saini et al. 2017; Shchipunov 2012). Biopolymers are polymer-based compounds made up of individual units joined together by covalent bonding to form bigger molecules. This transition to renewable energy sources is vital because of the cumulative damage to the environment and the speed with which climate change has been exacerbated by the continued use of synthetic polymers derived from fossil fuels. As a result of being non-biodegradable and being synthesized using potentially dangerous substances or leading to the production of toxic by-products, synthetic polymers have a negative effect on the environment (George et al. 2022). In place of fossil fuel feedstocks, materials that can be replenished easily, like human and animal waste, are favored (Aziz et al. 2018; Gong et al. 2016; Mohiuddin et al. 2017; Muthumeenal et al. 2017; Rahul Singh and Rhee 2019). Composite polymers made from renewable sources are gaining attention as a superior replacement for petroleum-based polymers. Due to the availability of carbon resources on Earth and the low cost of generation, carbon produced from biomass is widely used as a precursor of carbon in supercapacitors (Jin et al. 2014; P. Kumar and Kim 2016; Lu et al. 2016). Carbon from natural resources can be utilized as a natural material for electrode material for supercapacitors to realize excellent properties due to carbon porosity, physicochemical activation, and heteroatom addition, all of which improve the material's specific capacitances (Illa et al. 2020; P. Kumar and Kim 2016; L. Ma et al. 2020; Y. Y. Wang et al. 2015; Zhou et al. 2018). Products based on biopolymer matrix are becoming essential because of the environmental benefits of matrix such as biodegradability, high electrical resistance, good acoustic and thermal properties, high specific strength and stiffness, and thermal recyclability are all beneficial pros of using biopolymer composites (Saini et al. 2017). Nanofillers like graphene, CNTs, fullerene, metal and metal oxide nanoparticles, and elongated rods provide biopolymer composites their semiconducting properties, while nanoreinforcements like clays and biofibers boost the composites' barrier resistance and mechanical strength (Zhou et al. 2018) (Ávila-Orta et al. 2018; Geng et al. 2018; Meng et al. 2013; Nilogal et al. 2021; Sanivada et al. 2020; Wei et al. 2016). Nanofillers with the ability to provide electric and dielectric properties are of immense use in electronics industries as they possess electronic transportation properties (Gnedenkov et al. 2015; Hu et al. 2017). This review presents comprehensive recent advances of the bionanocomposites used in energy storage devices, particularly batteries and supercapacitors (Fig. 1).

Biopolymers

Both biological processes and chemical synthesis employing bioresources can be used to create biopolymers of renewable and synthetic types. Cellulose, collagen, chitosan, sugar, starch, and lignin are all examples of naturally available biopolymers that may be extracted from live creatures and plants since these organisms already have the requisite metabolic system and access to a wide range of precursors (Ahmad 2018; Ghadam 2022; Gregory et al. 2021; N.BeMiller 2019; Okonkwo et al. 2017). Polylactic acid and polyvinyl acetate are two important examples of synthetic biopolymers that are made by first extracting bioresource precursors and then polymerizing them using standard chemical synthesis methods. Biopolymers can be categorized into three groups based on how they are prepared: first biopolymers made from genetically modified organisms like bacteria; second biopolymers made from fermentation or chemical synthesis of bioresources; and third biopolymers made from bioresources made in other ways (Senka Z. Popovic´, Vera L. Lazic´, Nevena M. Hromiš, Danijela Z. Šuput 2018). Biopolymers are classified according to their preparation techniques shown in Fig. 2.

Source of biopolymers

Cellulose, chitin/chitosan, and alginate are three common glycosidic-linked carbohydrates/polysaccharides. Proteins like silk, keratin, and collagen are all made up of different long chains of amino acids. As represented in Figs. 2 and 3 (Senka Z. Popovic´, Vera L. Lazic´, Nevena M. Hromiš, Danijela Z. Šuput 2018) (Xiong et al. 2018), these biopolymers can be harvested sustainably from a wide variety of natural sources, including plants, arthropod exoskeletons, skin, silkworm cocoons, spider webs, and hair.

Chitosan, derived from the polysaccharide chitin, is a naturally occurring carbohydrate modification. Numerous hydroxyl (-OH) and amino acid groups are present in the structure ($-NH_2$). Chitin that has been deacetylated forms a linear copolymer. A linear (1–4) copolymer is formed from 2-amino-2-deoxy-D-glucopyranos and 2-acetamido-2-deoxy-D-glucopyranose similar like cellulose structure. Absorbable sutures and wound dressings are just two of its many medical applications (Gregory et al. 2021). Incredibly widespread application of poly lactic acid polylactic acid is the main component of bioplastics (PLA). A different term for it is poly lactide. Linear thermoplastic polymer derived primarily

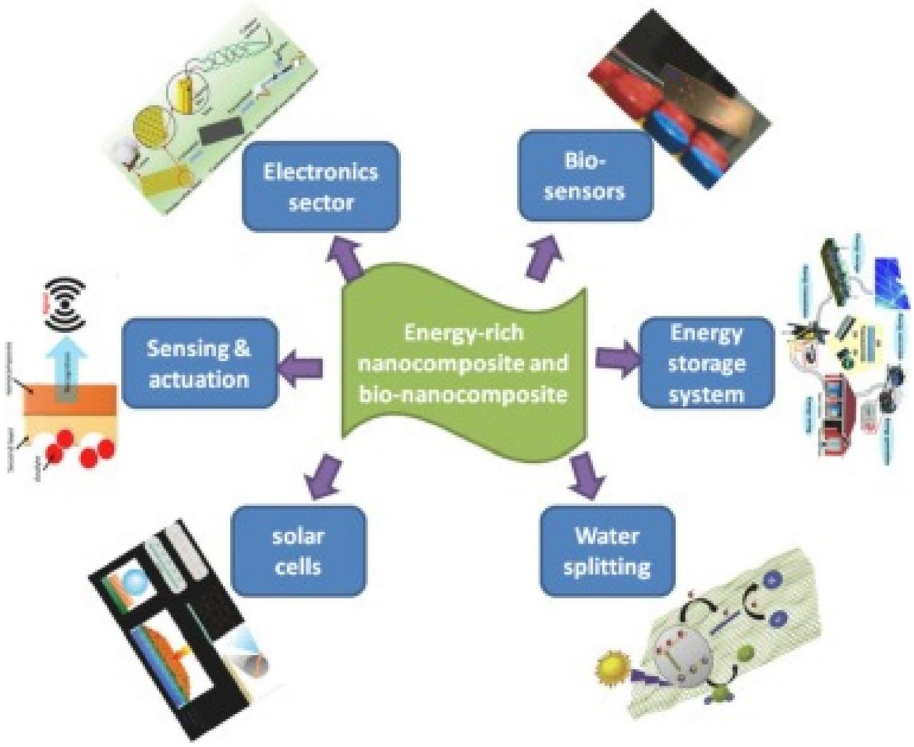

Fig. 1 Applications of bionanocomposites in energy sector (Adapted with permission from Elsevier (Karki et al. 2021))

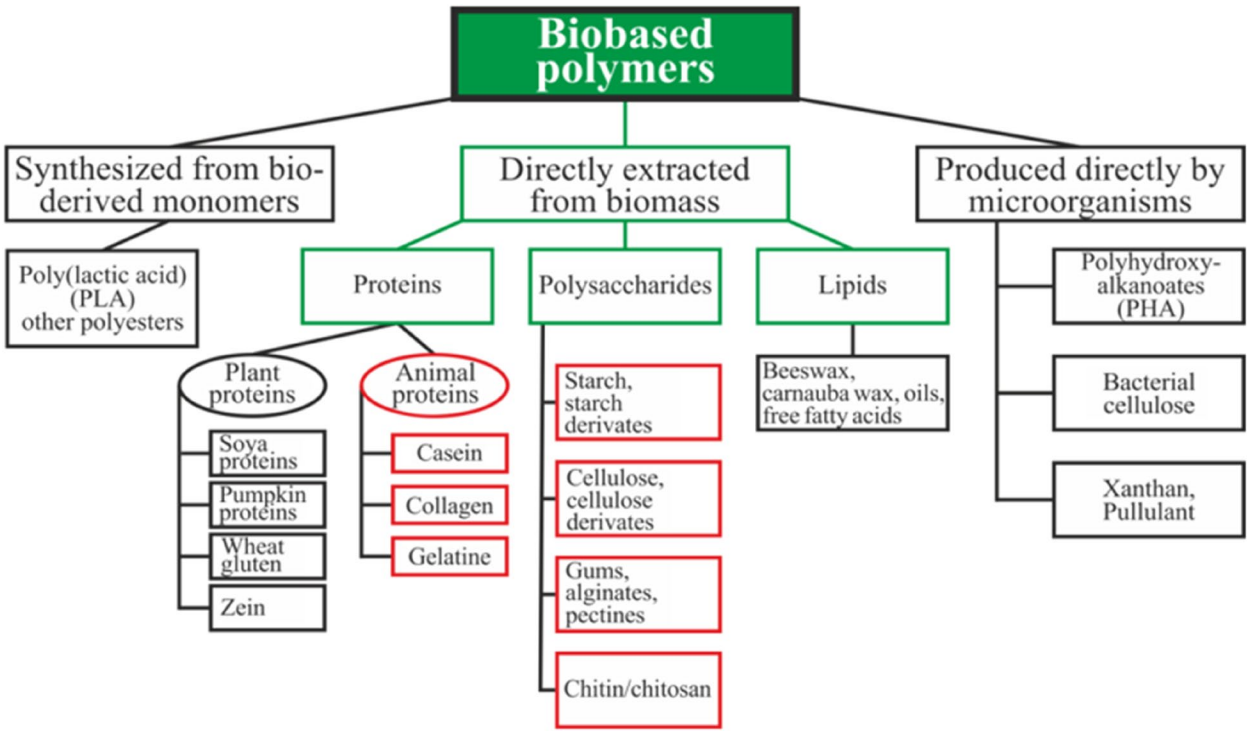

Fig. 2 Different sources of biopolymers synthesis (adapted with permission from Elsevier B.V. (Senka Z. Popovic´, Vera L. Lazic´, Nevena M. Hromiš, Danijela Z. Šuput 2018)

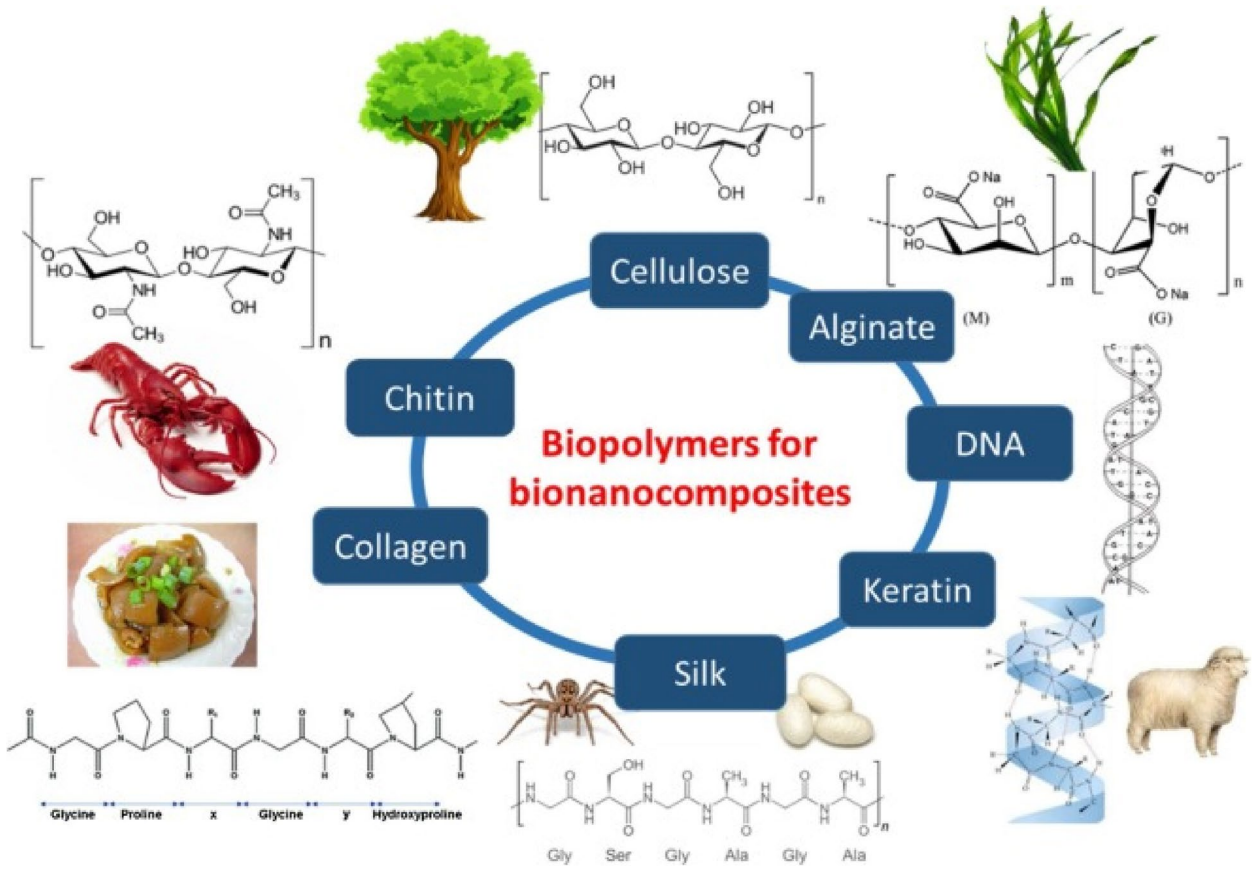

Fig. 3 Molecular structures and origins of naturally derived biopolymer constituents for bionanocomposites. (Adapted with permission from Elsevier B.V (Xiong et al. 2018))

from agricultural by-products such as corn and sugar beets. There are a wide variety of applications, including medical equipment, food, packaging, and textiles (Sanivada et al. 2020; Siqueira et al. 2010). Carbohydrates like starch or amylum have several glucose units linked together by glycosidic bonds. Starch consists of two different types of amylose and amylopectin. Starch is used as a source of energy storage mostly by plants and microorganisms. Potatoes, tapioca, maize, and wheat are the most common plant sources of starch (Ávila-Orta et al. 2018). Chitin is a kind of modified cellulose with a high molecular weight that is built from N-acetylglucosamine units. Its most important use is as a textile and dye binder (Butnaru et al. 2019; Huan et al. 2018). Polyesters derived from polyhydroxyalkanoates (PHA) are hydrophobic, biodegradable, and biocompatible, which are all desirable qualities. It can be found in a variety of various forms and is utilized by bacteria for the purpose of storing carbon or energy within the cell in the form of light-refracting granules.

Bionanocomposites

Bionanocomposites are a type of material that is created when biopolymers are combined with inorganic solids on the nanometer scale, and in recent years, they have received a great deal of interest from researchers. These organic–inorganic hybrids can be made from a wide range of biopolymers (polysaccharides, polypeptides and proteins, nucleic acids, etc.) and a wide range of inorganic solid particles (clay minerals, hydroxyapatite (HAP), silica, carbon nanomaterials and various metal oxides, etc.). The importance of bionanocomposites must be grasped to appreciate needs to be borne in mind that biopolymers are materials that are both biocompatible and biodegradable, and their composites are of interest for cutting-edge research in energy as well as other fields. Additional potential uses include, in terms of their ability to resist mechanical, thermal, and barrier breakdown, material class appealing because of its possible applications in membranes for food and drugs, controlled drug, and pesticide delivery preparation, sterilizing water for consumption, and creating oxygen-blocking films.

Materials show unique properties when their dimension is reduced to the nanometer range. Nanotechnology, the revolutionary scientific field of the twenty-first century, must be revisited from the ground up if we are to fully reproduce cutting-edge novel products in various scientific sectors. Composites are formed by adhering together two or more materials that have contrasting physical and/or chemical properties in their natural surroundings. Composites consist of different phases attached together by their physical or chemical linkages linkages. Hybrid composite materials have a number of benefits over pristine counterpart materials. To get beyond the limitations of biopolymer packaging, scientists have turned to nanocomposite technology in recent years. In comparison to regular composites and even pristine polymers, nanocomposites have superior mechanical strength, barrier qualities, self-healing behavior, and heat resistance. (Ahmadizadegan et al. 2018; Amin 2013; Dasan et al. 2017; Gregory et al. 2021; Kanmani and Rhim 2014; X. Ma et al. 2019; Park 2021; Patwekar et al. 2016; Sanivada et al. 2020). Nanocomposites are distinct from bulk polymers due to the presence of a sizable amount of nanomaterials incorporated in the matrix phase. The production of bionanocomposites can be accomplished in a wide variety of ways. Solutions mixing, in situ process, melt mixing, and template synthesis are all instances. Solution intercalation refers to the process by which a biopolymer, such as starch, is added to a solvent in which it dissolves completely.

Silicon oxide platelets are an example of an inorganic platelet. Water, chloroform, or toluenes are examples of polar solvents that might cause nanofillers to swell. As a result of mixing biopolymer with a nanoparticle swelling solution, polymer chains are able to intercalate within the silicate interlayer, which causes the displacement of solvent. After the solvent is evaporated, the biopolymer/layered silicate bionanocomposite retains its intercalated structure. The process of intercalative polymerization, in which polymerization occurs between intercalated sheets, is facilitated by dispersing the nanoparticle in a monomer. Polymerization can be triggered by introducing either heat or radiation, or by diffusing an appropriate organic initiator or catalyst. In a similar vein, melt intercalation is frequently used to create bionanocomposites made of polymer or layered silicate materials. In comparison to solution intercalation and in situ intercalative polymerization, this method has a number of advantages. When the polymer has reached the appropriate melting point, the nanoparticles can be mixed in. Inorganic replicas of bimolecular structures, subcellular components, and microbial cells can be fashioned using these source organisms as models. Nanoscale particles of bioorganics trapped in a mesoporous matrix serve as templates. This is a simple and easy method that has the potential for widespread application. Water-soluble polymers were required for this procedure, and it is possible that some unwanted by-products made it into the final product. Materials including cellulose, lignin, and hemicellulose from plants, animals, and microbes make up the bulk of bionanocomposites. Materials like chitosan, cellulose, starch, PLA, chitin, and polyhydroxyalkanoates are examples. Bionanocomposites are distinguished by their crucial biodegradability. Biodegradation, caused by microorganisms including bacteria, fungi, and algae, entails disintegration, deterioration, and sometimes degradation. The biodegradation of polymers is a complex process including enzyme-mediated hydrolysis and oxidation.

Bionanocomposites in energy storage

Energy storage is the process of storing energy in a device so that it can be used later upon requirement. Many different types of electrochemical devices have been brought to light as potential applications of biopolymers. Being biodegradable and biocompatible, biopolymers may be utilized to increase the effectiveness of other biologically active substances in a system. In this category, the most recent developments in devices for energy storage that make use of biopolymers; specifically, in batteries and supercapacitors are discussed.

Batteries

A chemical power cell, or battery, supplies electricity to a gadget. A battery's discharge capacity rate is proportional to the size of its individual cells, whereas the nominal voltage is governed by the number of cells linked in series. Traditional metals and inorganic materials have proven to be effective and reliable, indicating tremendous commercial potential. Nickel–cadmium and lithium-ion batteries are two of the most common types of batteries available today. However, there are concerns about the safety of these devices, including the possibility of poisonous leakage. Although several strategies have been implemented, such as recycling spent batteries, the most recent attempt is the use of biomaterials, particularly biopolymers, in the creation of eco-friendly batteries (Admassie et al. 2016; Mohiuddin et al. 2017; Rahul Singh and Rhee 2019).

Cellulose

When it comes to biopolymers, cellulose is one of the most popular options for usage in batteries. Cotton, maize, banana, corn cobs, and wheat are just a few examples of the many plant-based bioresources that can be mined for their natural cellulose fibers. Bacterial fermentation cellulose has gained attention as a potential

precursor for lithium-ion batteries capable of high rates of discharge, which may be accomplished by pyrolyzing the cellulose at elevated temperatures to create a material composed of carbonaceous fibers. Advantages of bacterial cellulose precursors over other types of precursors include their porous structure, increased surface area, high biodegradability, large-scale cellulose generation, and abundance of hydroxyl groups (Gregory et al. 2021; L. Ma et al. 2020; Muthumeenal et al. 2017). Due to its very strong interaction with various added chemicals and also the finely thinner morphology of bacterial cellulose compared to plant cellulose, bacterial cellulose is transformed into a tunable flexible scaffold appealing biomaterial that attracts widespread interest in forming a highly versatile three-dimensional carbon nanomaterials (Admassie et al. 2016; Anthony et al. 2021; Ghadam 2022; Illa et al. 2018; Y. M. Wang et al. 2021).

Using in situ free radical graft polymerization, Wei et al. (2016) developed a copolymer consisting of poly (acrylic acid sodium)-grafted-carboxymethyl cellulose (NaPAA-g-CMC) that serves as a good binder for Si anode in lithium-ion batteries as it is necessary to develop an innovative binder system as high-capacity silicon anodes typically experience a substantial volume change during charging and discharging cycle. The synthesis and the proposed mechanism are given in Fig. 4. The copolymer binder has a high binding strength for Si nanoparticles and copper current collectors, and it has helped to construct a stable solid electrolyte interface layer on the Si surface. The resulting NaPAA-g-CMC-based anode is very efficient in terms of both capacity and coulombics, and exhibits remarkable cycle stability. Enhanced electrochemical performance for NaPAA-g-CMC may be due to the existence of multi-point interaction with the Si surface. For high-capacity Si anodes, the NaPAA-g-CMC binder offers a competitive alternative due to its affordable cost, ease of synthesis, and compatibility with the environment. This binder can also be used with other types of high-capacity electrode materials.

Lignin

In recent years, lignin's potential as a biopolymer for use in batteries has garnered a lot of attention. Straw grains and vegetable stems are only two examples of the many plant-based bioresources from which lignin can be derived. Chen et al. (2016) used lignin in place of traditional polymer binder and carbon black additive to create a negative electrode that is both high-performance and long-lasting. The composite electrode was made by coating a mixture of silicon nanoparticles and lignin, a cheap, renewable, and readily available biopolymer, over a copper substrate using the standard slurry mixing and

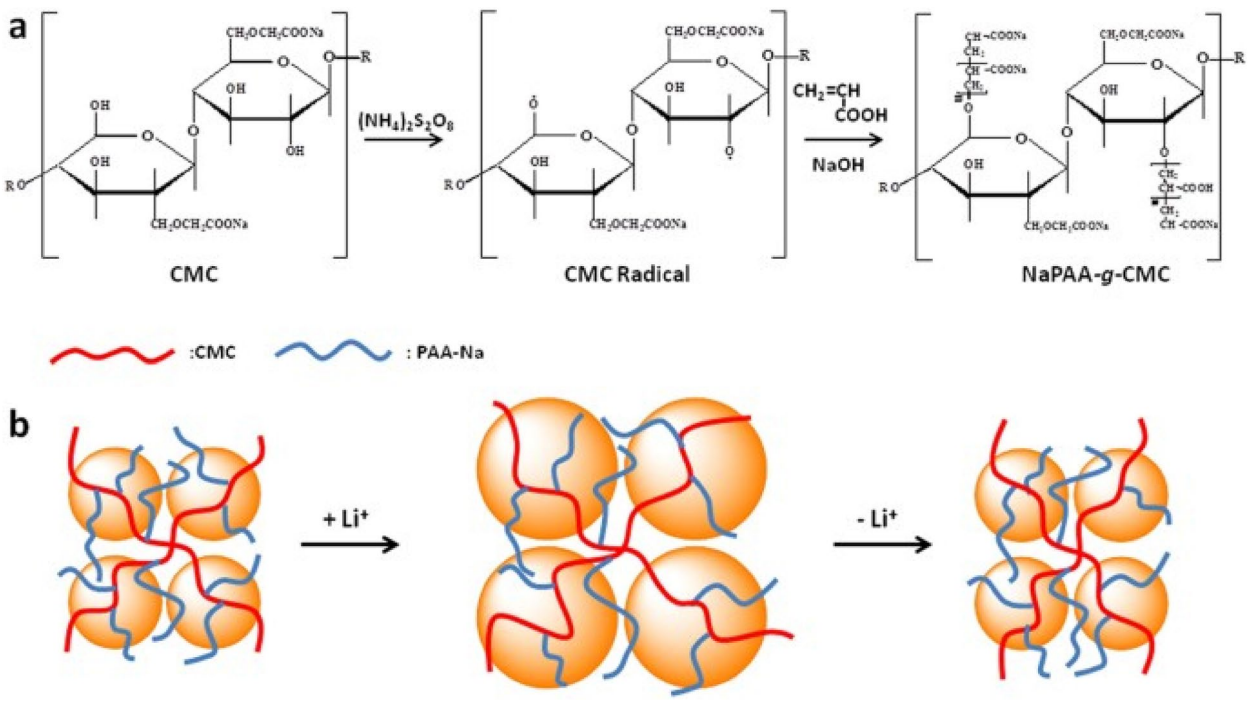

Fig. 4 **a** Scheme of synthesis for free radical covalent grafting of NaPAA onto CMC backbone; **b** proposed mechanism of NaPAA-g-CMC binder to accommodate the larger volume change of Si particles during cycling. (Adapted from (Wei et al. 2016) nature publishing, unrestricted use with proper citations)

coating procedure, and then heating the mixture (Fig. 5). With a maximum initial discharge capacity of 3086 mAh g^{-1} and a retention of 2378 mAh g^{-1} after 100 cycles at 1 A g^{-1}, the composite electrode demonstrated good electrochemical performance. In spite of the comparatively large areal loading of 1 mg cm^2, an areal capacity of 2 mAh cm^2 was nevertheless attained. In a full-cell configuration, the composite electrode likewise performed exceptionally well in terms of its rate capabilities. This study concludes that the optimal heat treatment temperature of 600°C for Si/lignin electrodes was determined at which the lignin goes through a complex compositional change in which the trade-off between increasing conductivity and maintaining polymer flexibility is realized.

Pectin

One further biopolymer frequently used in battery studies is pectin. Natural pectin can be found in abundance in the peel of many fruits, especially apples and oranges.

Biodegradable pectin electrolyte with magnesium nitrate salt $Mg(NO_3)_2$ made by solution casting was studied by Kiruthika et al. (2019) (Fig. 6). The XRD results show that the biopolymer electrolyte composed of 50% pectin and 50% $Mg(NO_3)_2$ was the most amorphous. DSC measurements show that an increase in

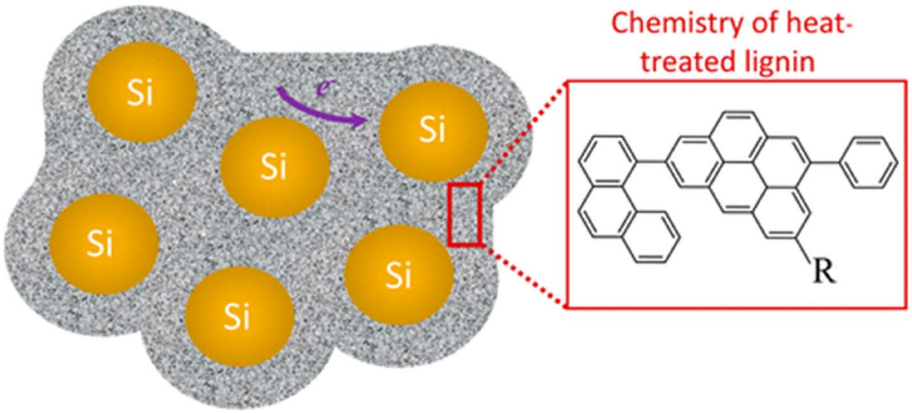

Fig. 5 Schematic depiction of the heat-treated silicon-lignin composite. (Adapted with permission from American Chemical Society (Chen et al. 2016))

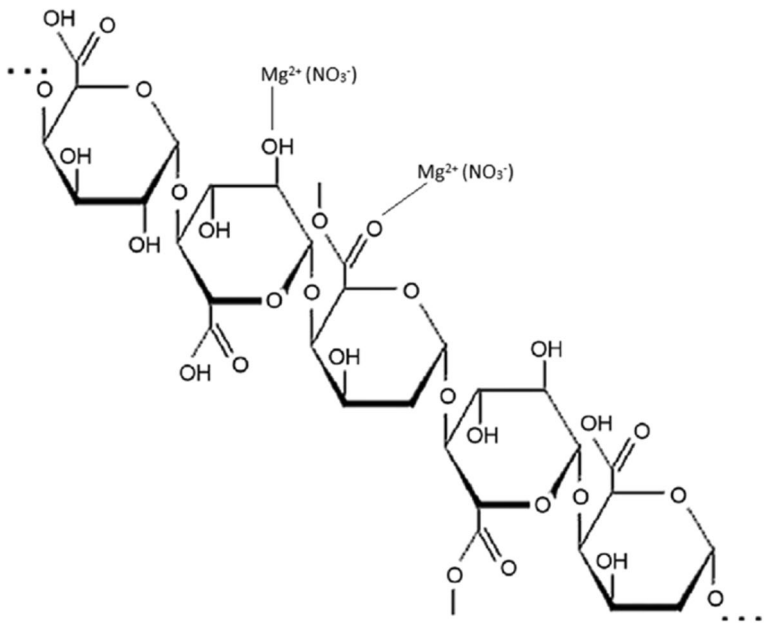

Fig. 6 Pectin probable interaction with magnesium salt $(MgNO_3)_2$ (adapted with permission from Springer Verlag (Kiruthika et al. 2019))

salt concentration causes an increase in the glass transition temperature as a result of less dipole interaction in the homopolymers. The maximum ionic conductivity of 7.70 104 Scm^1 at room temperature was found in a 50 M wt% pectin:50 M wt% $Mg(NO_3)_2$ biopolymer electrolyte, as measured by AC impedance spectroscopy. The dielectric analysis proves that electrolytes are non-Debye characteristics. Furthermore, using Evan's polarization method result reveals that in the 50 M wt% pectin:50 M wt% $Mg(NO_3)_2$ sample, the transference number of Mg^{2+} ions is 0.29. A value of 3.8 V is determined to be the electrochemically stable potential for the sample with the best conductivity. The pectin and magnesium in the primary Mg battery are both diluted to a weight of 50 mg/kg (NO_3).

Perumal et al. (2019a, 2019b) used pectin and lithium perchlorate ($LiClO_4$) as the solid biopolymer electrolyte. With AC impedance analysis, they found that at room temperature, the maximum ionic conductivity is on the order of 5.15105 Scm^{-1}, which is three orders of magnitude greater than that of pure pectin polymer membrane. Analysis of transport properties shows that cations have more mobility than anions. Natural pectin electrolyte is a superior solid electrolyte for lithium polymer batteries, as shown by impedance and transport study.

Chitosan

Chitosan, another prolific biopolymer utilized in battery applications, is another option. Shellfish bioresources like prawns, crabs, lobsters, and even some types of mushrooms are common sources of waste chitin. Carbon electrode materials for energy storage have been created from a wide range of biomass, including chicken eggshells, human hair, and ox horns; nevertheless, their restricted availability prevents their widespread use. Chitosan biomass, which is highly plentiful, is used as a cheap and easily accessible material necessary for the production of N-doped porous carbon material (PNCM) investigated by Wang et al (2015). According to structural characterizations, this PNCM is highly N-doped (4.19%) and has a hierarchical pore structure (abundant macro/micropores). Additionally, research is being done on the electrochemical properties of PNCM as electrode materials for supercapacitors and lithium-ion batteries. Synthesized at 700°C for use in a supercapacitor, the PNCM shows great power and energy density in both aqueous and organic electrolytes, and has remarkable cycle stability with only 1.3% capacitance loss over 11,000 cycles. In a 1 mol L1 H_2SO_4 electrolyte, the material's specific capacitance can reach up to 220 F g1. The research shows that PNCM has remarkable Li storage characteristics when used as an anode material for Li-ion batteries, including a high specific capacity (around 460 mA h g^1 at 50 mA g^1) and better cycle stability (no capacity decline even after 1100 cycles).

Polysaccharides

Biopolymer-based batteries also make use of polysaccharides which are obtained from tree gum or seeds. Tree exudates natural gum polysaccharides such as karaya gum or seeds of tamarind are also considered useful bioresources. Bie et al. (2017) studied the potential of karaya gum (KG) as a binding agent for Si-based anodes wherein said electrodes have demonstrated better cycle and rate characteristics compared to sodium carboxymethyl cellulose (CMC) and sodium alginate (SA). For KG electrodes, we get a reversible capacity of 1336 mA h g^1 at 4.0 A g^1 and a specific capacity of 2421 mA h g^1 at 1.5 A g^1 with 80% capacity retention after 150 cycles. In addition, with a fixed lithiation capacity of 4.0 A g^1, over 1200 cycles of current reversal at 1000 mA h g^1 are possible with KG electrodes. KG electrodes, when used in the Si/graphite composite electrode, demonstrate superior stability over 100-cycle runs, while other electrodes wear out quickly after only a few cycles. Branched structure and numerous polar groups serve as bonding sites, uniform SP dispersion creates an efficient conductive network, and the protein component helps maintain a stable electrode structure, all of which contribute to the enhanced electrochemical performance of KG electrodes. Additionally, KG's natural availability, low cost, eco-friendliness, and mature industrial process make it an attractive binder for Si-based anodes.

The solution casting method was used to create a Mg-ion conducting solid electrolyte based on the natural biomacromolecule tamarind seed polysaccharide (TSP) and the magnesium perchlorate ($Mg(ClO_4)_2$) salt (Bie et al. 2017). The TSP containing a 0.25 mol percent (m.m.) $Mg(ClO_4)_2$ salt combination showed excellent thermal stability and a wide electrochemical window (Fig. 7). At room temperature, the TSP doped with 0.25 (m.m.%) $Mg(ClO_4)_2$ has a maximum Mg-ion conductivity of 5.66 104 Scm^1 and a maximum ionic transference number of 0.43. The most conducting Mg-ion sample was used to build a primary Mg-ion battery, the functionality of which was investigated. At normal temperature, the open circuit value of 2.36 V was measured for the improvised battery.

Carrageenan

Battery applications also include carrageenan, another biopolymer. Most food packaging is comprised of carrageenan, a seaweed-based substance, despite the fact that waste carrageenan is typically scarce (N.BeMiller 2019). The advancement of electrochemical devices relies heavily on lithium-conducting materials. Mary et al.

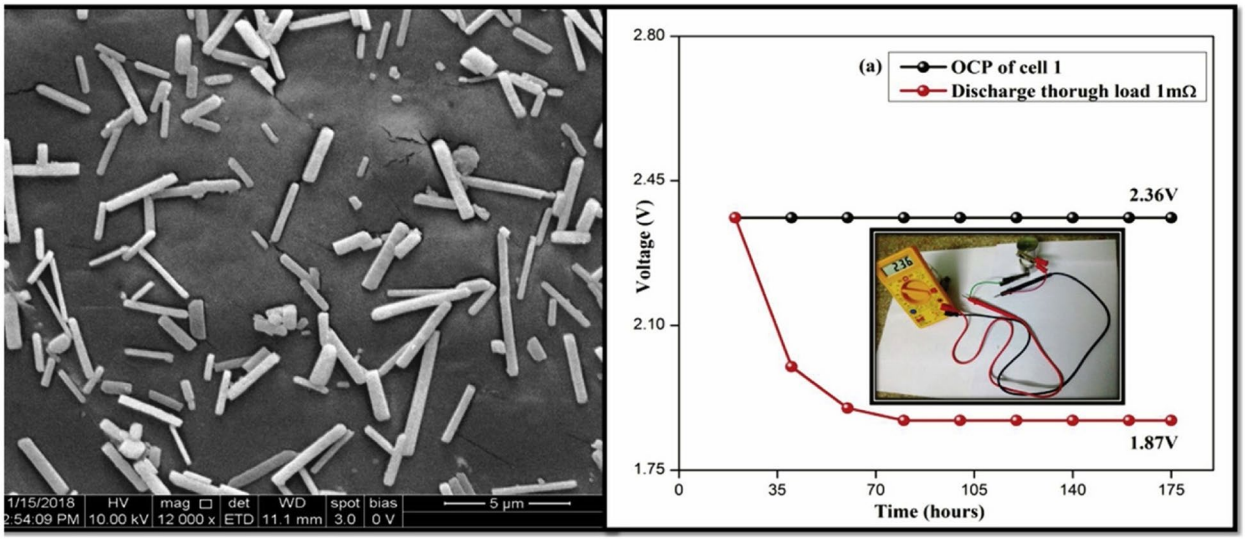

Fig. 7 Micrographs taken with a field emission scanning electron microscope showing **a** TSP containing 0.25 (m.m.%) $Mg(ClO_4)_2$ and **b** OCP of cell 1 (inset shows the photo image of cell 1). (Adapted with permission from Elsevier (Perumal et al. 2019a, 2019b))

(N.BeMiller 2019) prepared a lithium-ion conducting membrane using a solution casting method using lithium bromide (LiBr)-containing K-carrageenan membranes of varying concentrations. The powder XRD data show that 0.5 wt% LiBr in 1 g of K-carrageenan yields the maximum conductivity at room temperature (3.43 103 Scm^1) and a highly amorphous character. The glass transition temperature of the best conducting polymer electrolyte is 44.55°C. Ionic conductivity is demonstrated through the DC polarization method. The most efficient electrolytic biopolymer membrane has been used to create a lithium-ion battery, and its output voltage determined.

Supercapacitors

Supercapacitors are a type of chemical energy harvesting device that works by storing and discharging energy by means of ion adsorption and desorption over an electrolyte–electrode contact. Electrochemical pseudocapacitors and electrical double-layer capacitors are the two most common kinds of supercapacitors (EDLCs). Electrochemical double-layer capacitors (EDLCs) absorb charged ions at the interface between the electrolyte and the conducting electrodes to store electrical energy. Pseudocapacitors are able to store electrical energy because of the reversible oxidation–reduction that occurs between the electrode and electrolyte. Hybrid supercapacitors combine the charging and discharging strategies of pseudocapacitive and double-layer capacitive devices (Rahul Singh and Rhee 2019) (Okonkwo et al. 2017). The high energy conversion rate, fast charge speed, and better recyclability of supercapacitors indicate that they may one day prove to be invaluable as energy storage devices. However, conventional binders employed in supercapacitors are poisonous and feature subpar mechanical qualities. Considering their biodecomposition and high electroconductivity, biopolymers have emerged as a sustainable replacement.

Tammela et al. (2015) prepared polypyrrole-coated *Cladophora* sp. algal nanocellulose hybrid materials (PPy-cellulose) and extremely porous carbon nanofiber-based materials (C-nf) developed from the PPy-cellulose hybrid materials which have been shown to be effective building blocks for asymmetric energy storage devices incorporating aqueous electrolytes. The latter material, generated by heating the PPy-cellulose in inert gas flow, kept the porosity typical of the PPy-cellulose composites. PPy-cellulose and C-nf materials have very different electrochemical performances, and combining them in asymmetric supercapacitor cells is favorable. Thus, at 1.6 V, such an asymmetric cell yielded a capacitance of 25 F/g, which is equivalent to an energy gravimetric density of 33 J/g. An energy density of 14 J/g is the maximum achievable for a symmetric PPy-cellulose supercapacitor device, yet this number is much greater than what would be predicted for a cell that is symmetric with two C-nf electrodes. The greater capacitance related to the surface-limited redox processes on the PPy-cellulose electrode was clearly demonstrated by the specific gravimetric capacitances of 59 and 146 F/g for the C-nf and PPy-cellulose electrode materials, respectively. With a current density of 156 mA/cm^2, the asymmetric cell is shown to have a capacitance of 11 F/g. To top it all off, the supercapacitor was stable enough in an aqueous electrolyte to be charged and discharged galvanostatically for

1000 repetition at a current density of 20 mA/cm^2 within a potential window of 1.6 V (i.e., 2 M NaCl). This work is a huge way ahead in the development of organic, natural resources material-based energy harvesting devices with economically feasible mass loadings, capacitances, and energy densities.

Mo et al. (2018) used a simple wet-spinning method for producing a high-performance cellulose nanofiber-reinforced graphene/polypyrrole microfiber. The cellulose nanofibers not only supply significant hydrogen bonds to augment the interlaminar force, but they also inhibit the restacking of the graphene sheets, playing the dual role of "enhancer" and "spacer." The microfiber has a tensile strength of 364.3 MPa, making it stronger than the vast majority of other fibers used in supercapacitors. The specific capacitance of the built fiber-shaped supercapacitors is 334 mF cm^2 in liquid electrolyte and 218 mF cm^2 in solid electrolyte at the current density of 0.1 mA cm^2. Suitable for use as a guide in the design of future wearable and portable energy storage devices, the described method combines the industrially viable wet-spinning technology with a well-designed structure for the production of high-performance ternary fiber-shaped supercapacitors.

In situ crosslinking of cellulose/ionic liquid solutions with bisphenol A epoxy resin employing ring opening processes with cerium ammonium nitrate and halloysite nanotubes (HNTs) as the ionic conducting promoter yielded a high-performance halloysite nanotube (HNT)-doped liquid crystalline bionanocomposite ionogel (Guo et al. 2018). The scheme of synthesis is represented in Fig. 8. Due to the liquid crystal phases created by the assembly of anisotropic HNTs nanoparticles, these ionogels with HNTs display significantly increased ionic conductivity compared to that of pure ionogel without the inclusion of HNTs. Shearing can enhance the ionic conductivities of ionogels, which are on the order of 1 mS/cm at room temperature, and they rise with increasing HNT concentration. Nanocomposite ionogels showed considerable enhancements in mechanical characteristics and thermal stability in comparison to pristine ionogel without HNTs. After testing ionogel as a gel electrolyte for a supercapacitor, the device's measured specific capacitance was stable for up to 5000 charge–discharge cycles, demonstrating its durability (Fig. 9). The innovative bionanocomposite liquid crystalline ionogels possessing the aforementioned high ionic conductivity, mechanical strength, and flexibility are highly desired for application in bendable energy harvesting devices.

Another important naturally available biopolymer that has been extensively studied for its potential in supercapacitor device is chitosan. Carbon nanofibers have historically underperformed in charge storage applications due to its inert surface and high graphitic content. Punde et al (2019) developed a method of preparation of a ternary nanocomposite (CNF-CS/Co) consisting of carbon nanofibers, chitosan, and cobalt nanoparticles. The nanocomposite materials showed that it possessed a sizable

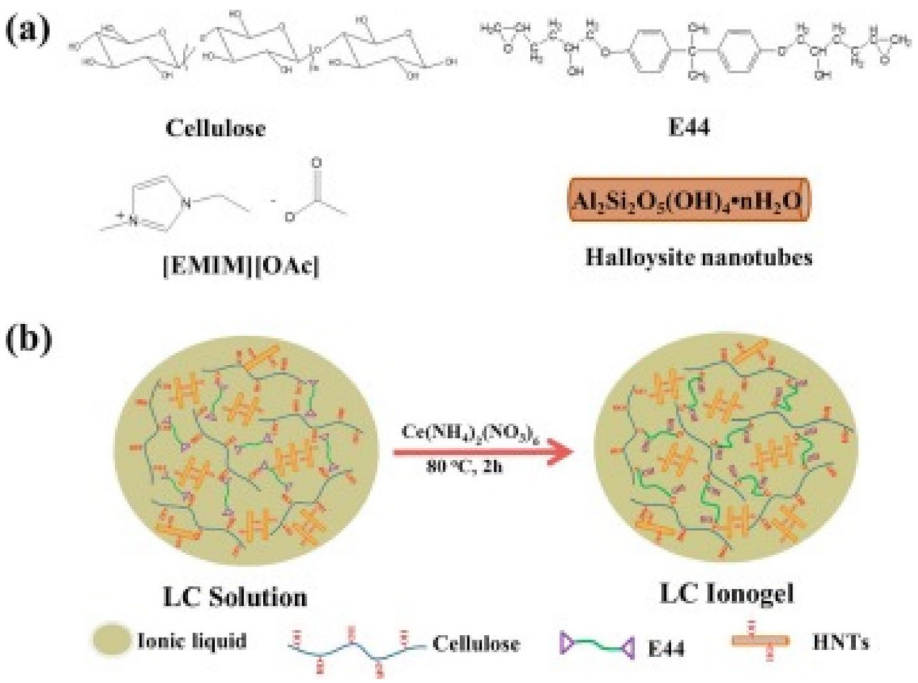

Fig. 8 A scheme depicting the synthesis of liquid crystalline ionogels. (Adapted with permission from Elsevier BV (Guo et al. 2018)

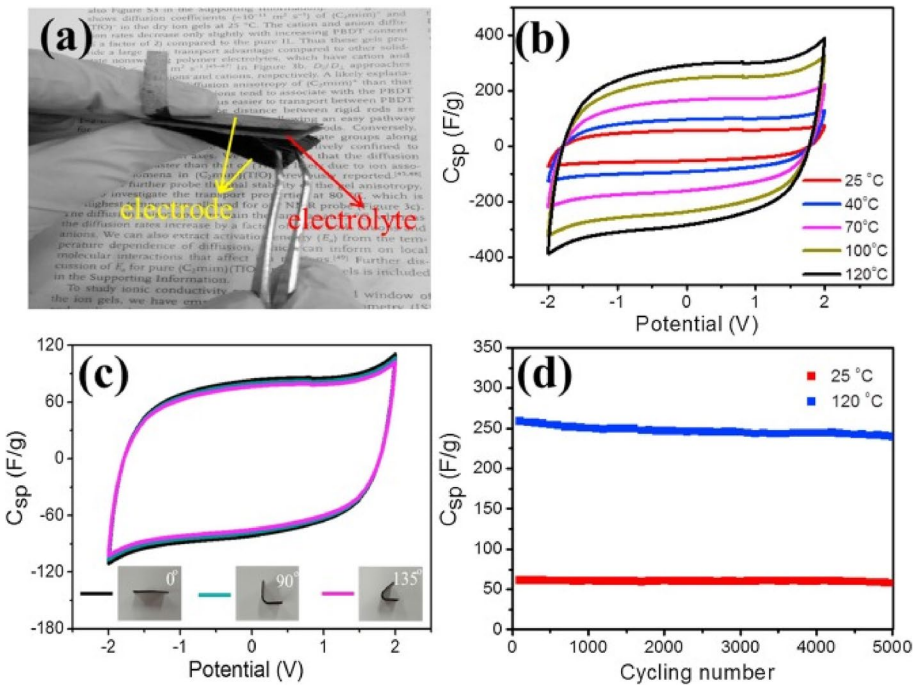

Fig. 9 **a** Electrochemical activity of ionogels. The supercapacitor's sandwich design. Ten millivolts per second cyclic voltammogram of the supercapacitor as a function of temperature (b). At 10 mV/s, (**c**) the cyclic voltammetry curves of a bendable supercapacitor with varying curvatures. Performance of the supercapacitor device during cycling at varying temperatures (**d**). (Adapted with permission from Guo et al. (2018))

surface area of 188.3 m^2 g^{-1}. The specific capacitance of CNF-CS/Co at 1 A g^{-1} is 438.6 F g^{-1}, which is significantly higher than the specific capacitances of pure CNF (81.3 F g^{-1}), Co (129.7 F g^{-1}), and CNF-CS (187.2 F g^{-1}). Over the course of 5000 charges and discharges, the electrochemical stability of the 1.2 V symmetric supercapacitor made using CNF-CS/Co was 95.6%, and the energy density was 38.2 Wh kg^{-1}. The CNF-CS/Co nanocomposite shows promise for high-performance supercapacitors due to its simple synthesis and promising results.

Highly mesoporous carbon fibers produced from lignin were used to create flexible electrodes with supercapacitance (Ago et al. 2016) (Fig. 10). As a sacrificial polymer, polyvinyl alcohol (PVA) made it possible to electrospin lignin from aqueous solutions. Importantly, with extremely high surface area (> 2000 m^2 g^1) and mesoporous volume (0.7 cm^3 g^1), PVA generated phase-separated domains. Following the migration of PVA to the gas phase and carbonization of the as-spun lignin-based fibers, a flexible, freestanding carbon network was formed, thanks to the optimized sequential thermal treatment, which initially included stabilization at 250°C. An extremely high specific capacitance (205 F g^1 in 0.5 M Na_2SO_4 electrolyte) was measured, making this electrode one of the best produced from biopolymer precursors. In addition, the electrical conductivity of the carbon fiber

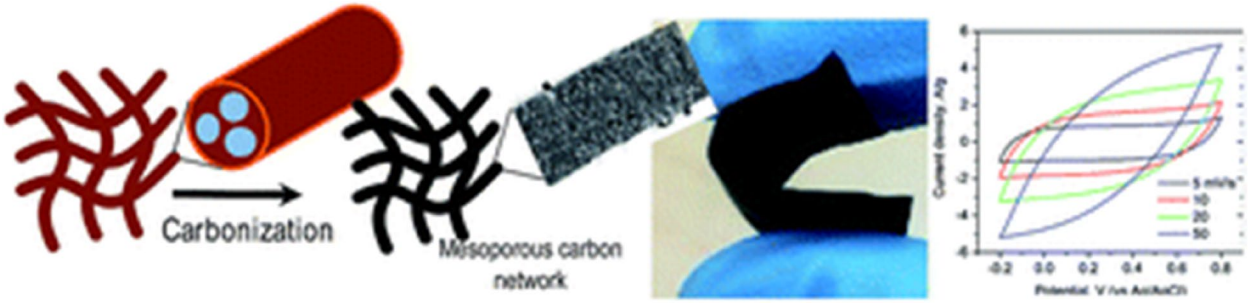

Fig. 10 Scheme of morphology of electrospun fiber, mesoporous carbon, and flexible carbon mat after carbonization and the cyclic voltammetry results.(Adapted from (Ago et al. 2016) Creative Commons license open access. (No permission needed)

network was found to be 386 S m^1, which is a whopping two orders of magnitude greater than the 2.47 S m^1 measured for the precursor (non-fibrous, powder) sample. Soft-templating from the phase-separated sacrificial polymer yielded the robust network shape and mesoporosity responsible for the outstanding performance of the produced electrodes. It is proof that lignin can be used in innovative ways in high-tech materials.

Chemical reactivity under elevated temperatures

Elevated temperatures can accelerate the chemical reactions within biopolymer-based energy storage devices, affecting both the biopolymer matrix and the incorporated conductive materials. Biopolymers, being organic in nature, often contain functional groups that are sensitive to heat. At higher temperatures, these functional groups can undergo degradation, leading to the breakdown of the polymer chains. For instance, cellulose, one of the most commonly used biopolymers, begins to degrade at temperatures above 200°C, leading to a reduction in mechanical strength and flexibility (Gregory et al. 2021). This thermal degradation can also produce volatile by-products, which may react with other components within the device, further compromising the overall stability.

Moreover, elevated temperatures can enhance the chemical reactivity of conductive fillers such as carbon nanotubes (CNTs) or metal nanoparticles within the biopolymer matrix. While this can sometimes improve conductivity by promoting better interaction between the polymer and filler, it can also lead to unwanted side reactions. For example, at high temperatures, the oxidation of metal nanoparticles can occur, leading to the formation of insulating oxides that reduce the overall conductivity of the composite (Ahmad 2018). Additionally, the thermal expansion mismatch between the biopolymer and the filler materials can induce stress at the interface, potentially leading to micro-cracking and delamination, further reducing the device's efficiency (Xiong et al. 2018).

Impact on energy efficiency

The energy efficiency of biopolymer-derived energy storage devices is closely tied to the stability of the materials used and their ability to maintain performance under varying environmental conditions. Elevated temperatures can have a dual effect on energy efficiency: they can initially increase ion mobility and improve charge transfer rates, but prolonged exposure can lead to detrimental effects that ultimately decrease efficiency. In the short term, higher temperatures can enhance the ionic conductivity of the electrolyte and reduce the internal resistance of the device. This can lead to faster charge and discharge rates, improving the overall power density of the energy storage device (Punde et al. 2019). For instance, biopolymer-based electrolytes, which often suffer from lower ionic conductivity at room temperature, may perform better at elevated temperatures, providing a temporary boost in energy efficiency (Perumal, Christopher Selvin, et al 2019b). However, prolonged exposure to elevated temperatures can lead to the degradation of the biopolymer matrix, as discussed earlier, which in turn can affect the structural integrity and functionality of the energy storage device. This degradation not only reduces the mechanical stability but also affects the electrochemical properties of the device. As the polymer chains break down, there is a loss of uniformity in the material, leading to increased resistance and reduced charge storage capacity. Over time, this can cause a significant drop in energy density and efficiency, as the device becomes less capable of storing and releasing energy effectively (Belaineh et al. 2022). Elevated temperatures can also impact the cycling stability of biopolymer-based energy storage devices. Repeated cycling at high temperatures can exacerbate the degradation processes, leading to faster capacity fade and reduced cycle life (Anthony et al. 2021). For instance, in lithium-ion batteries, elevated temperatures can accelerate the formation of a solid electrolyte interface (SEI) on the anode, which is typically beneficial at lower temperatures but can become unstable and thick at higher temperatures, leading to increased impedance and reduced efficiency (Liu et al. 2018). In the context of biopolymer-based systems, similar effects can occur, where the biopolymer may react with the electrolyte or other cell components under high-temperature cycling, leading to performance degradation.

Environmental and economic aspects and material stability

The development of biopolymer-based composites for sustainable energy storage holds immense promise, yet several challenges and limitations must be addressed to fully realize their potential. One of the most pressing challenges facing biopolymer-based composites is their long-term stability and durability, particularly in the context of energy storage applications (Ghadam 2022; Siwal et al. 2006; Xiong et al. 2018). Unlike conventional fossil fuel-derived polymers, biopolymers are often more susceptible to environmental degradation due to their natural origin. Factors such as humidity, temperature fluctuations, and exposure to UV radiation can significantly affect the mechanical and chemical properties of these materials (Ahmad 2018; Siwal et al. 2006). This degradation can lead to reduced conductivity, compromised structural integrity, and ultimately, a decline in the performance of energy storage devices over time.

Moreover, the electrochemical environment within batteries and supercapacitors can be harsh, often involving repeated cycles of charging and discharging. This cycling can induce mechanical stress, swelling, or even dissolution of biopolymer components, further exacerbating stability issues. Although efforts are being made to chemically modify biopolymers or reinforce them with inorganic fillers to enhance stability, these solutions may introduce new complexities, such as increased brittleness or reduced biodegradability (Karki et al. 2021). Therefore, ensuring that biopolymer-based composites maintain their desired properties throughout the lifecycle of an energy storage device remains a significant hurdle.

Another critical challenge is the economic feasibility of producing biopolymer-based composites at scale. The cost of raw materials, synthesis processes, and the need for specialized equipment can make these composites more expensive than their fossil fuel-derived counterparts. While the environmental benefits of biopolymers are clear, the higher costs associated with their production can be a barrier to widespread adoption, particularly in industries where cost-effectiveness is a primary concern. Additionally, the scalability of biopolymer production is not yet on par with conventional polymers (Liu et al. 2018). Many biopolymers are derived from renewable resources such as plants, which require agricultural land, water, and other resources. The availability of these raw materials can be affected by factors such as climate change, seasonal variations, and competition with food crops, leading to fluctuations in supply and price. Furthermore, the processes for extracting, purifying, and modifying biopolymers are often less mature and less efficient than those for fossil fuel-derived polymers, posing additional challenges for large-scale production. Efforts to overcome these challenges include developing more efficient synthesis methods, utilizing waste biomass as a feedstock, and improving the economies of scale in biopolymer production. However, until these challenges are addressed, the cost and scalability of biopolymer-based composites will remain significant limitations to their broader application in energy storage.

While biopolymer-based composites are often touted as environmentally friendly alternatives to fossil fuel-derived materials, a comprehensive assessment of their environmental impact reveals a more complex picture. One of the main advantages of biopolymers is their biodegradability, which reduces the accumulation of plastic waste in the environment. However, the rate and completeness of biodegradation can vary significantly depending on the specific biopolymer and the conditions under which degradation occurs. For instance, some biopolymers may require industrial composting facilities to fully degrade, limiting their environmental benefits in regions without access to such infrastructure. Moreover, the environmental impact of biopolymer production must be considered. The cultivation of crops for biopolymer production can lead to deforestation, loss of biodiversity, and increased greenhouse gas emissions if not managed sustainably. The use of fertilizers, pesticides, and irrigation in agriculture can also contribute to environmental degradation. A life cycle assessment (LCA) of biopolymer-based composites is essential to understand their true environmental footprint, from raw material extraction to end-of-life disposal. Finally, while biopolymer-based composites are intended to be more sustainable, the incorporation of inorganic fillers or other non-biodegradable components to enhance performance may reduce their overall environmental benefits. These additives can complicate the recycling or composting process, potentially leading to the release of harmful substances into the environment. Therefore, balancing the performance enhancements with the environmental integrity of biopolymer-based composites remains a significant challenge.

Conclusions and future perspectives

Industrial progress has prompted an increased focus on sustainability, which has contributed to the exponential growth of current technical solutions. In the rapidly evolving power and energy sector, materials originating from a supply that is neither renewable nor biodegradable are losing importance in the dynamic energy and power industry, and continuous efforts are underway to replace synthetic materials with a wide range of natural and renewable biopolymers. Bionanocomposites are being explored as potential materials for energy harvesting applications. Research in bionanocomposites has been given a boost by their use in a variety of contexts. Mass production is now possible at a cheap cost without harming the environment, whereas in the past, there were several limitations on the development of the fabrication techniques for bionanocomposite. Bionanocomposites are widely available and are finding potential use in a wide range of technologies. This means that we may create the necessary materials by carefully considering the manufacturing costs of the desired qualities and then picking the most suitable, feasible, readily available, and inexpensive material. Bionanocomposites have proven to be superior materials across the board due to their degradability and compatibility with bio-organisms. Bionanocomposite materials will be crucial in the coming era of nanotechnology. For the advancement of nanotechnology and the development of novel nanodevices, biopolymer-based composites hold great promise as future nanotechnology building blocks. Research into a flexible material made from natural polymer composites that can be used

for energy storage is now the most important area of study.

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Authors' contributions
NiranjanPatra: conception, data analysis, manuscript writing and composition. PrathipatiRamesh: literature survey. Donthu Vaishnavi: literature survey. AkilAhmad: manuscript review, discussion.

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Declarations

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