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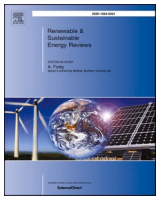

Recent advancements in utilizing biomass materials for aqueous electrolytes in rechargeable batteries

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ABSTRACT

In spite of the rising demand for rechargeable batteries and competing advancements in the field, aqueous electrolytes still hold advantage over other types of electrolytes because of their inherent safety, ease of fabrication, feasibility, and environmental friendliness. Addressing the leakage issue of liquid aqueous electrolytes, gel polymer aqueous electrolytes are attracting increasing attention. They have ionic conductivities between liquid and solid electrolytes, are mechanically and thermally stable, have high flexibility and corrosion resistance, and can accommodate volume expansion. However, the transition from synthetic polymers to natural polymers is still in the research phase. This review links biomass materials and aqueous electrolytes of rechargeable batteries by summarizing and analyzing the potential of a wide array of natural polymers (e.g., cellulose, alginate, carrageenan, natural gums, etc.) from various sources (plants, animal, algae). The properties, composites, and electrolyte fabrication techniques of each material are discussed, and future perspective is presented. Results show that the most used fabrication technique is solution casting while the highest ionic conductivity demonstrated is 96.89 mS/cm by a cellulose-based electrolyte. Additionally, natural gums show the highest capacity retentions (100 % even after 3300 cycles). By reviewing a wide range of materials and fabrication techniques, we aim to offer valuable insights into the development of innovative energy storage solutions that are sustainable, safe, and feasible.

Abbreviations:

AM	Acrylamide
CA	Cellulose acetate
CAB	Cellulose acetate butyrate
CMC	Carboxymethyl cellulose
FMEE	fatty methyl ester ethoxylate
LBG	Locust bean gum
PAA	poly(acrylic acid)
PAM	polyacrylamide
PAN	polyacrylonitrile
PEG	polyethylene glycol
PEGDA	Poly(ethylene glycol) diacrylate
PEO	polyethylene oxide
PMC	polyacrylamide-poly (ethylene glycol) diacrylate-carboxymethyl cellulose
PMMA	poly(methyl methacrylate)
PVAc	polyvinyl acetate
PVDF	polyvinylidene fluoride
PVP	Polyvinylpyrrolidone
SEI	solid electrolyte interface
SPI	soy protein isolate
TEOS	tetraethyl orthosilicate
WIS	water-in-salt electrolyte

(*continued on next column*)

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1. Introduction

The surging demand for power and the transition of the energy sector towards sustainability put pressure on advancing rechargeable batteries which form the heart of portable electronic devices, electric vehicles, and energy storage systems. Developing rechargeable batteries not only

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brings sustainable energy solutions, but also socio-economic improvements [1]. Although the market of rechargeable batteries is dominated by Li-ion batteries, this type of battery is not guaranteed to keep pace with the future demand and specifications. In addition to Li-ion batteries, other types of rechargeable batteries that are being investigated and developed include various metal-ion batteries such as Zn-ion, Mg-ion, Al-ion, Na-ion. Metal-air and metal-sulfur batteries are also promising [1]. Despite their relatively high energy density and long life cycle, the development of rechargeable batteries on large scales is hindered by challenges such as cost and safety issues. For example, the use of organic solvents in the electrolytes make the battery extremely flammable. Other safety issues include thermal runaways, dendrite growth, and stability of the electrode-electrolyte interface [2–4].

The electrolyte plays a critical role in determining the safety and stability of the battery. Many approaches are being studied to improve the aforementioned aspects such as developing electrolyte composites that contain additives to reduce their flammability, using less flammable and less volatile solvents such as ionic liquids or water, or using polymer gel materials that are less susceptible to leakage [5–11]. Aqueous electrolytes are advantageous in tackling the safety issue because they are water-based. Using water as the solvent not only levels up the safety of the battery, but also decreases its costs as compared to using organic solvents or ionic liquids. Other advantages of aqueous electrolytes are their higher ionic conductivities, convenience of fabrication under ambient conditions, and absence of the SEI (solid electrolyte interface) layer problem [12–14]. However, these merits come with limited energy density and low operational voltage.

Aqueous electrolytes include all liquid and gel electrolytes that are water-based. They can be acid-based, neutral, or alkaline [15,16]. Gel and polymer electrolytes were developed to overcome the leakage issue that liquid electrolytes suffer from. Although they are safer and can stay intact, their ionic conductivities tend to be lower than their liquid counterparts. Another advantage of gel polymer electrolytes is their mechanical stability that makes the use of a physical separator unnecessary [17,18]. Additional advantages of polymer electrolytes include their flexibility, corrosion resistance, and ability to accommodate volume expansion [17,19]. There are many commonly used polymers in the industry such as polyethylene oxide (PEO), poly(methyl methacrylate) (PMMA), polyacrylonitrile (PAN), polyvinyl acetate (PVAc), and polyvinylidene fluoride (PVDF) [19]. However, such synthetic polymers are not sustainable nor environmentally friendly because their fossil-fuel-based sources are not renewable and they are not biodegradable.

Researchers are investigating green biopolymer alternatives from various natural sources as potential materials for aqueous electrolytes of rechargeable batteries. Studies reported using many biopolymers (e.g., cellulose, alginate, agar, etc.) as host matrices with varying metal salts and additives. Natural biomass such as plants and algae are promising sustainable feedstocks that can replace the use of fossil-based materials in rechargeable batteries, and thus help decarbonize this industry.

The purpose of this paper is to review and analyze the trends and recent advancements in using biomass-derived materials for aqueous electrolytes in rechargeable batteries. The potential of different biomass materials to be used in aqueous electrolytes is discussed in separate sections including each material's characteristics, the relevant electrolyte composites and fabrication techniques, and final product properties. Results are compared and analyzed, and future perspective is presented. Since the focus of this paper is on aqueous electrolytes of rechargeable batteries, studies involving non-aqueous electrolytes even if biomass materials are used are not considered. Studies on energy storage materials other than rechargeable batteries (e.g., supercapacitors) were also excluded. Unlike other reviews available in the literature which either discuss one type of biomaterial or a particular type of battery, the novelty and unique contribution of this study lies in its specific focus on the use of biomass materials in aqueous electrolytes of rechargeable batteries. This approach not only allows more room for material exploration, analysis, and comparison, but also aims to help researchers advance aqueous electrolytes using sustainable methods without the need for organic solvents or other toxic materials.

2. Biomass for rechargeable batteries

Biomass is a sustainable and environmentally friendly feedstock for various materials that can be used in different parts of rechargeable batteries such as electrodes and electrolytes. Using biomass materials brings several advantages and facilitates adherence to the principles of green chemistry [20]. The most critical advantage is that biomass is renewable, meaning it can be replenished naturally without worrying about depleting the natural sources for future generations. But sustainability goes beyond just the source, the whole manufacturing to consumption process should be environmentally friendly. Similarly, the end-of-life procedures should be safe whether they are recycled or biodegraded [21].

Current commercial rechargeable batteries do not meet the sustainability and green chemistry standards. From the initial exploitation of raw materials in underprivileged countries to the synthesis process and design choices involving delicate separators and flammable electrolytes, the journey of traditional batteries is full of risks. Overheating hazards, combined with the formation of the solid electrolyte interface (SEI) layer, further complicate these dangers. Additionally, the recycling procedures for conventional batteries remain unclear, leading to potential emissions and environmental concerns at the end of their lifespan [22–24]. Although the production of conventional rechargeable batteries is monitored and regulated to meet the safety standards of the market, their designs are not inherently safe.

The ideal carbon cycle of a sustainable rechargeable batteries should be a full loop that does not produce CO_2 emission, as shown in Fig. 1. First, biomass is harvested and used to fabricate sustainable batteries. Then, these batteries are used to provide clean energy, and the CO_2 emissions produced in this stage are then captured by the biomass to regrow and provide new feedstock. Finally, the batteries either

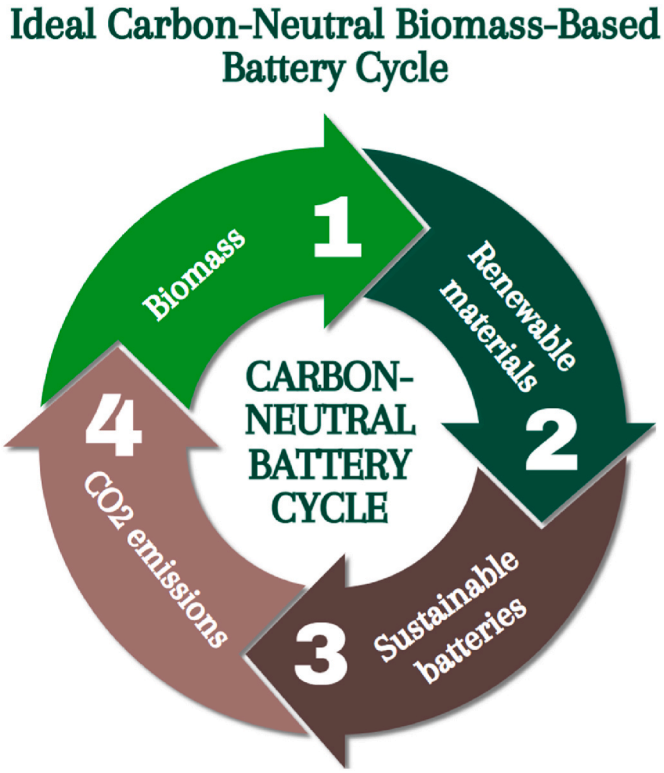

Fig. 1. Ideal carbon-neutral biomass-based battery cycle.

biodegrade upon disposal or they get recycled [20]. By implementing the use of biomass for sustainable batteries, a greener and more circular approach to energy storage can be achieved.

Biomass-derived materials are diverse and can be used in all parts of the battery. For example, forest and agricultural crops and residues provide a rich source of porous carbon, doped carbon, cellulose, silica, polymers, etc. The main difference between crops and residues is that crops such as cotton and other plants are consumable by humans while residues are waste biomass such as sawdust and pulps and peels of fruits. Residue biomass is more sustainable as it does not impact the food industry. Biomass materials can also be extracted from industrial, marine, and domestic wastes. Since industrial waste is mass produced, using it as a renewable feedstock would also reduce pollution. For example, chitosan is a valuable biopolymer that is recovered from the wastes of food industries that use seafood because it is found in chitin which is extracted from the shells of shrimps and other sea animals. Algae, which can be considered as an example of marine waste, also produce a wide range of gel-like polymers such as alginate and carrageenan which can be used in rechargeable batteries [20,25–28].

Mechanical, physicochemical, and electrochemical properties of each biomass-derived material make it suitable for use in a particular battery component. For example, carbon-rich materials are used in electrodes, specifically anodes. Interestingly, carbonaceous materials from biomass have unique porous and doped structures that give them desired electrochemical properties and high stability [28,30–33]. Additionally, their porous structures allow enhanced volume expansion accommodation which increases the safety of the battery [34]. Reviews about the use of biomass carbon-based materials as electrodes for specific types of batteries such as Li-ion batteries are available elsewhere in literature [28,35,36]. For example, Fig. 2 illustrates the diverse biomaterials that can be extracted from algae and used to develop anode composites for Li-ion batteries [29]. Biopolymers are another type of material that can be extracted from biomass and are typically used in electrolytes, separators, and binders. Polymer electrolytes are gaining increasing interest as they eliminate the leakage safety concern while maintaining high ionic conductivities [37–39]. Good mechanical properties and flexibility of biopolymers also make them suitable candidates for separator and binder fabrication [19,40]. Interestingly, biomass materials can be used in multiple battery components simultaneously. However, each component should be investigated and developed independently before it gets incorporated with other novel biomass-based components in a rechargeable battery. Studies have investigated the use of the same biomass materials in more than one battery component. For example, Kasprzak et al. studies the use of cellulose as a membrane, electrode, and binder altogether in a cellulose-based capacitor. Another study by Ref. [29] reports and compares the use of various biomass materials extracted from algae in electrodes, electrolytes, separators, and binders of Li-ion batteries. It seems possible that research and development will eventually reach the stage of combining more than one biomass-based battery component with more progress and

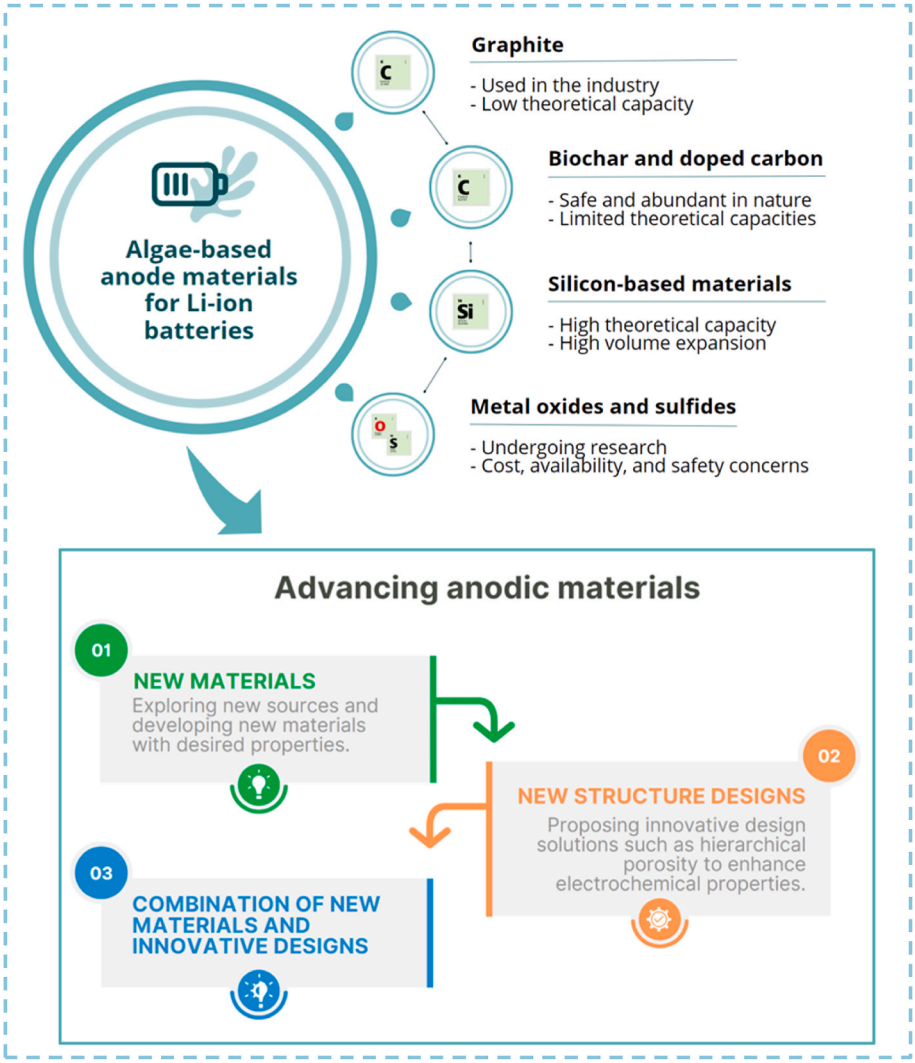

Fig. 2. *Illustration of the diverse materials that can be obtained from algae and used for anodes of Li-ion batteries* [29].

cumulative efforts.

3. Biomass for aqueous electrolytes of rechargeable batteries

In recent years, researchers have been exploring the potential of aqueous electrolytes, seeking to overcome the limitations of conventional non-aqueous electrolytes and harness their advantages. The motivation behind this pursuit is to overcome the limitations associated with non-aqueous counterparts while capitalizing on the inherent advantages of aqueous systems. In this context, biomass materials have emerged as promising candidates for aqueous electrolytes, primarily due to their sustainable nature and eco-friendliness. Their renewable and abundant supply makes them an attractive resource for the development of aqueous electrolytes, both in the form of liquid and gel composites.

Among the different types of aqueous electrolytes, gel polymer electrolytes have received considerable attention, driven by their distinctive ability to combine the desirable attributes of solid electrolytes, such as safety and stability, with the enhanced ionic conductivity and wettability characteristics of liquid electrolytes. Notably, polymers that have hydrophilic groups, such as -NH_2 and -OH, exhibit strong potential as candidates for aqueous electrolytes because they have high affinity towards water and thus high electrolyte wettability [19].

Studies have shown that biomass can be utilized not only as the main building block of aqueous electrolytes but also as a separator or a membrane layer that protects the electrolyte and electrode from mixing and having undesired side reactions [19,41]. For example, Fig. 3 shows a chitosan-based protective layer that is used in a Zn-based battery to protect the Zn anode and reduce the dendrite formation [42]. Additionally, aqueous electrolytes are typically used with acid-based batteries because unlike alkali metals, they are not reactive towards water [20].

Although alkali metal batteries such as Li-ion and Na-ion batteries do not commonly use aqueous electrolytes, a few studies proved the possibility of this combination through two main techniques. The first technique is using a layer that separates the reactive electrode from the aqueous electrolyte to avoid unwanted reactions. The second approach is the so-called water-in-salt (WIS) electrolyte. WIS is an emerging technique that uses high concentrations of salt as compared to water, resulting in different properties such as improved stability [20,43].

Interestingly, inorganic salts can also be extracted from natural sources, though studies on this topic are relatively limited. For example, Li et al. [44] investigated the compositions of various water bodies in and around the Kushui Lake in China and found that many samples are rich in lithium, sodium, and potassium salts. The rich lithium content in the Kushui Lake is mainly a result of geological factors and weathering

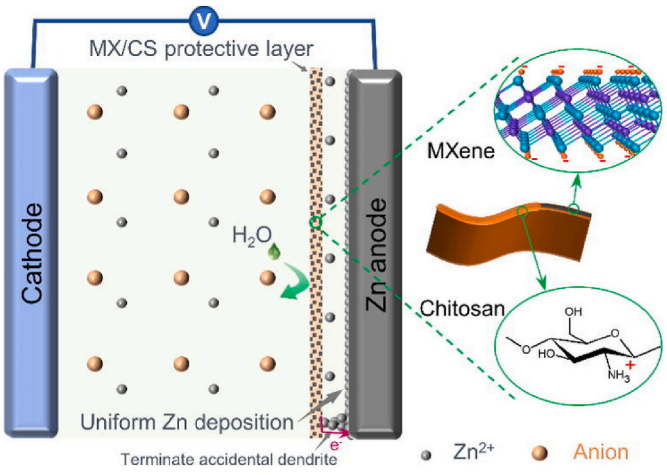

Fig. 3. *A schematic that shows the use of a chitosan-based protective layer to minimize dendrite formation on the Zn anode* [42].

influences such as the presence of volcanic rocks, water evaporation, and precipitation of salts. Similarly, Kudryavtsev [45] studied the reserves of lithium in nature. According to their estimates, around 70 % of the world's natural lithium reserves are found in Bolivia (21.4 %), Chili (17.8 %), USA (15.9 %), Argentina (15.4 %), China (12.1 %). Another study by Wang et al. [46] investigated the presence and extraction of potassium salts in straw ash. They were able to recover potassium salts making up to 78.1 % of the total recovered content, mainly in the forms of K_2SO_4, KCl, and K_2CO_3.

Examples of batteries that can benefit from biomass-based aqueous electrolytes include sodium-ion batteries, potassium-ion batteries, and zinc-ion batteries. These battery types are considered attractive alternatives to lithium-ion batteries due to their lower cost and abundance of raw materials. By incorporating biomass materials into the electrolytes of these batteries, researchers aim to enhance their performance, safety, and overall sustainability.

4. Plant-derived materials

4.1. Cellulose

Cellulose, the most abundant polymer on earth, is finding its applications in various industries. It is a natural polymer that is produced by plants and other organisms such as fungi, algae, and bacteria. The content of cellulose in some plants like cotton can reach up to 90 % [37]. Studies suggest that cellulose is a good candidate for aqueous electrolytes fabrication because of its numerous advantages. For example, it is renewable, biodegradable, and has favorable flexibility and mechanical strength. In addition, cellulose is hydrophilic because its chains have many free hydroxyl groups which can get attached to water. Fig. 4 shows the chemical structure of cellulose. These hydroxyl polar functional groups also allow simple modification through processes such as etherification and esterification to obtain new cellulose derivatives with tuned properties.

One of the challenges of using cellulose polymer is its amphiphilicity which makes its solubility in water limited. To overcome this limitation, cellulose is modified to obtain derivatives with better water solubility and ionic conductivity. Cellulose acetate (CA) is a commonly used cellulose derivative. The versions of CA available in the market are usually non-aqueous, however, studies proved that it can be prepared in an aqueous manner [47]. Cellulose acetate butyrate (CAB) is another derivative that is soluble in water because of its functional groups: hydroxyl, acetyl, and butyryl. Cellulose ethers such as methyl cellulose, ethyl cellulose, and hydroxyethyl cellulose are soluble in water [37].

To improve the performance of cellulose-based hydrogels, studies are developing novel crosslinking methods and agents. For example, Mittal et al. [48] proposed a transient and degradable electrolyte design based on crosslinked Na-CMC (sodium carboxymethyl cellulose) and agarose for Zn-ion batteries. The addition of agarose increased the hydrophilicity of the electrolyte because of its abundant -OH groups. Although the electrolyte is fabricated using the simple solution casting technique, agarose requires a temperature of 80 °C to dissolve completely. The final sample demonstrated an ionic conductivity of 87.57 $\times$ 10^{-3} S/cm. In another study [49], a multiple crosslinked hydrogel electrolyte containing polyacrylamide-poly (ethylene glycol) diacrylate-carboxymethyl cellulose (PMC) was prepared through a one-pot synthesis method and demonstrated exceptional properties. The 3D structure made possible by the unique crosslinking method showed in Fig. 5 resulted in high ionic conductivity (30.24 mS/cm), long lifecycle (5000 h), and excellent coulombic efficiency (99.5 %). The detailed electrochemical performance of the PMC electrolyte in a Zn-based battery is also demonstrated in Fig. 5.

Final properties of the cellulose-based electrolyte depend on the source of cellulose, its modification, and other additives used in the electrolyte composite. Poosapati et al. [50] used wood-derived cellulose nanofibers to fabricate an electrolyte made of cellulose, gelatin, poly

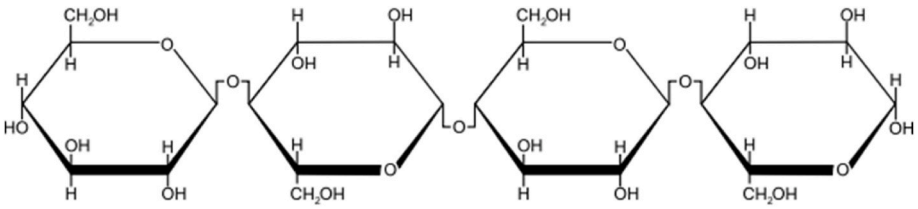

Fig. 4. Chemical structure of cellulose.

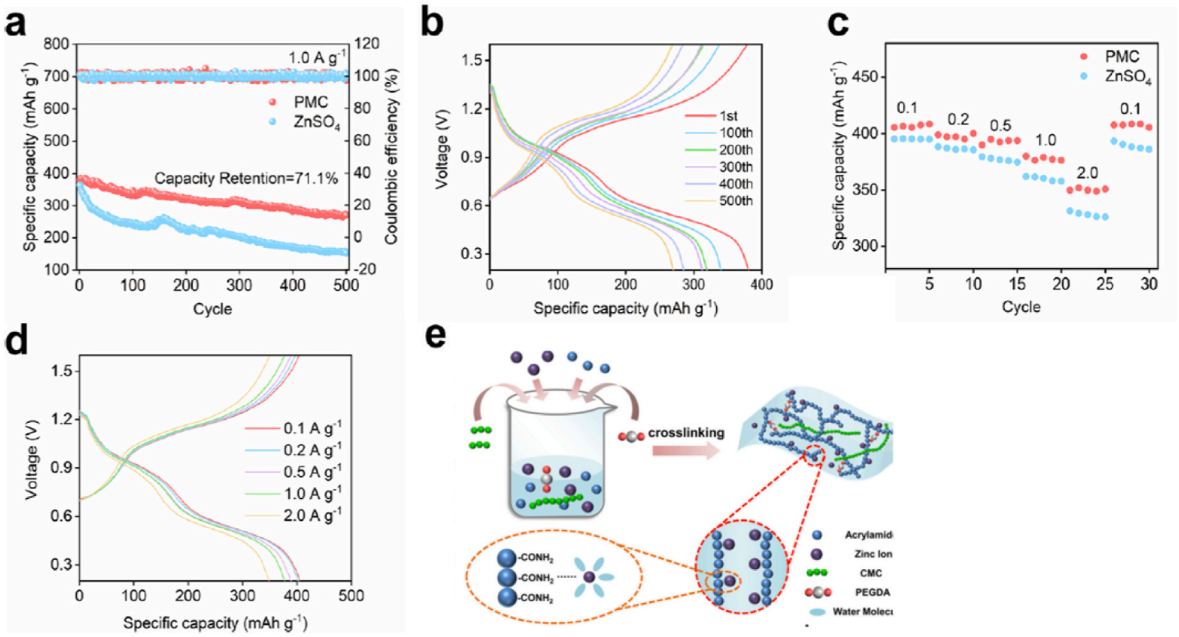

Fig. 5. *(a-d) Electrochemical performance of the PMC electrolyte in a Zn-based battery. (e) Crosslinking of PMC gel electrolyte* [49].

(acrylic acid) (PAA), and KOH. The advantages of using wood cellulose are its availability, safety, and ease of modification. Additives were added to improve the mechanical stability and crystallinity of the electrolyte while KOH was added to enhance the ionic conductivity. The final ionic conductivity of the electrolyte was 96.89 mS/cm, surpassing the values of pristine cellulose by a few orders of magnitude. On the other hand, Yue et al. [51] used bacterial cellulose, which has larger fiber diameters and better mechanical properties than plant cellulose, to prepare an aqueous gel polymer electrolyte. They modified it through a sulfonation process to enhance its ionic conductivity and combined it with polyaniline to achieve an electrolyte with an ionic conductivity of 5.2×10^{-3} S/cm.

Cellulose-based aqueous electrolytes and electrolyte-membranes were used in different types of batteries, mainly zinc-based and lithium-based batteries. For example, Glatz et al. [52] proposed the use of a cellulose-based membrane to inhibit the dissolution and diffusion of the electrode active material into the aqueous electrolyte ($ZnSO_4$) in a Zn-ion battery. Cellulose was treated through processes of sulfuric acid hydrolysis and sonication to achieve enhanced hydrophilicity, which in turn means high wettability. The final product proved effective separation between the aqueous electrolyte and the active cathode material. It also achieved a capacity retention of 82 % after 500 cycles, proving the stability of the cellulose-based membrane.

Ma et al. [53] used cellulose in a Zn-ion battery for a similar purpose yet in a different mechanism. They fabricated a cellulose-based dry membrane that was then soaked and allowed to swell in an aqueous Zn-containing electrolyte. The final electrolyte composite achieved a capacity retention of 84.7 % after 1000 cycles. Mo et al. [54] fabricated an electrolyte made of cellulose and polymerized zwitterionic sulfobetaine monomers for a Zn–MnO_2 battery. The composite had high flexibility, biocompatibility, water retention, stretchability (920 %), and capacity retention (90.24 % after 1200 cycles). Similarly, Chen et al. [55] developed a cellulose-based hydrogel electrolyte for aqueous Zn–MnO_2 batteries. Their sample consists of cellulose obtained from cotton through alkali pretreatment, $Si(OH_4)$ obtained from tetraethyl orthosilicate (TEOS) through hydrolysis, and glycerol. The fabricated electrolyte demonstrated good ionic conductivity of 32.3 mS/cm at 20 °C and 19.4 mS/cm at −40 °C. Moreover, it exhibited good tensile strength, elasticity, adhesion, heat resistance, and self-healing properties. It also had a low freezing point and dendrite formation suppression at −40°C–60 °C.

Cellulose-based electrolytes were also investigated in lithium-based batteries such as Li-ion and Li-S batteries. However, the number of studies available is limited since alkali metals are typically reactive and unstable with water-based electrolytes. Wan et al. [56] used cellulose from cotton pulp to fabricate an aerogel membrane layer that can maintain its thermal stability up to 120 °C. It was prepared through a process of dissolving the cotton pulp with an aqueous solution, then regeneration of the hydrogel membrane, and finally supercritical CO_2 drying to achieve an aerogel with high porosity and excellent flexibility. The ionic conductivity was measured as 5.76 mS/cm. In another study [57], cellulose from spruce pulp was treated, polymerized, and mixed with methacrylate to prepare a polymer matrix as an electrolyte for a Li-S battery, as shown in Fig. 6. However, this electrolyte was not aqueous. Table 1 summarizes the studies mentioned in this section.

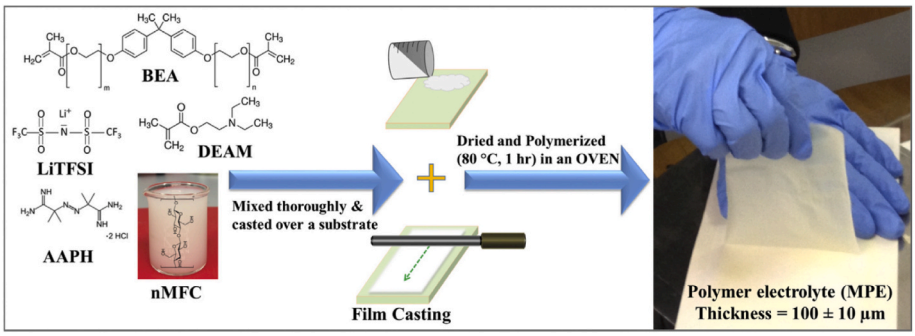

Fig. 6. *Process of making a cellulose-based electrolyte by* [57].

Table 1
Studies using cellulose in aqueous electrolytes of rechargeable batteries.

Source	Composite	Battery	Process	Properties	Ref.
Beetroot and celery	Zwitterionic sulfonated cellulose	Zn-MnO_2	Polymerization	Ion conductivity: 24.6 mS/cm Capacity retention: 90.24 % after 1200 cycles	[54]
Cotton	Cellulose, tetraethyl orthosilicate (TEOS), glycerol	Zn-MnO_2	Alkali pretreatment of cotton and hydrolysis of TEOS	Ion conductivity: 32.3 mS/cm at 20 °C and 19.4 mS/cm at −40 °C	[55]
Wood	Cellulose, gelatin, PAA, KOH	–	Simple steps at ambient conditions	Ionic conductivity: 96.89 mS/cm	[50]
–	Cellulose	Zn-ion	Sulfuric acid hydrolysis of cellulose	Capacity retention: 82 % after 500 cycles	[52]
Bacteria	Sulfonated bacterial cellulose, polyaniline	–	Chemical modification (sulfonation)	Ionic conductivity: 5.2 × 10^{-3} S/cm	[51]
Cotton pulp	Cellulose, aqueous AmimCl solution	Li-ion	Dissolution, regeneration, and supercritical drying	Ionic conductivity: 5.76 mS/cm, high porosity and thermal stability	[56]
–	Cellulose	Zn-ion	Cellulose membrane swelling in Zn-containing aqueous electrolyte	Capacity retention: 84.7 % after 1000 cycles at 4 A/g.	[53]
Spruce pulp	Cellulose, methacrylate-based polymer matrix	Li-S	Polymerization	*non-aqueous	[57]
–	Sodium carboxymethyl cellulose, agarose, glutaraldehyde	Zn-ion	Solution casting	Ionic conductivity: 87.57 × 10^{-3} S/cm	[48]
–	Acrylamide (AM), Poly(ethylene glycol) diacrylate (PEGDA), carboxymethyl cellulose (CMC)	Zn metal	One-pot synthesis method	Ionic conductivity: 30.24 mS/cm Coulombic efficiency: 99.5 % Lifecycle: 5000 h	[49]

4.2. Lignin and lignocellulose

Lignocellulose is another type of biomass that is found in plant sources such as wood. It consists of straight chains of cellulose, hemicellulose, and irregular lignin polymer structure, as shown in Fig. 7. Using lignocellulose without further separation requires lower costs and less process steps. Lignocellulose is also advantageous in terms of high surface area, wettability, and mechanical properties [58].

Song et al. [60] investigated the use of lignocellulose and polyethylene glycol (PEG) to make an electrolyte membrane for Li-ion batteries. The membrane, shown in Fig. 8, was created using a solution casting technique, PEG was added in different amounts, and the membrane was immersed in a liquid electrolyte that contains lithium hexafluorophosphate ($LiPF_6$). The addition of 30 % PEG was proven to enhance the mechanical properties by 3–4 times, achieve an electrolyte uptake up to 267 %, an ionic conductivity of 3.22 mS/cm, a stability window up to 5.3 V, and good thermal stability. Song et al. [59] also fabricated a lignocellulose-based electrolyte through a solution casting technique for Li-S batteries. They used lignocellulose with a lithium-containing solution and electrolyte uptake of 428 % and a

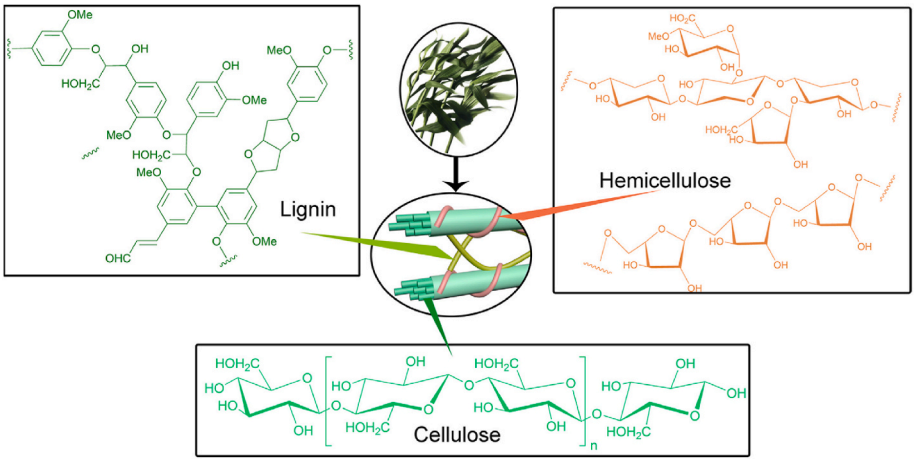

Fig. 7. *Chemical structures of the constituents of lignocellulose* [59].

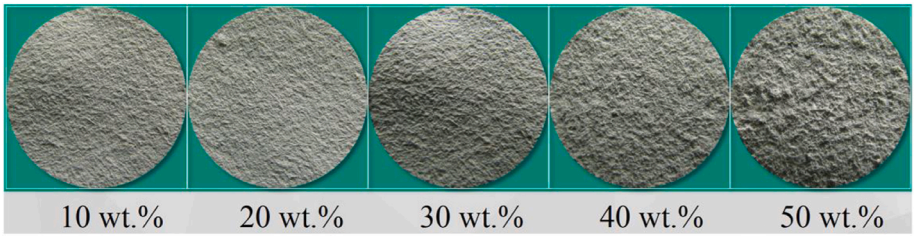

Fig. 8. *Lignocellulose-based electrolyte membranes containing different weights of PEG* [60].

stability window up to 5.3 V. Additionally, it demonstrated good thermal stability and mechanical strength. Lignocellulose was also investigated for the use in electrolytes of aqueous Zn-ion batteries. For example, an aqueous electrolyte fabricated by Zheng et al. [61] demonstrated an ionic conductivity of 6.3 mS/cm at 30 °C. The electrolyte was prepared through a simple solution casting technique and was tested in a Zn-based battery and a supercapacitor. The use of kraft lignin as an electrolyte base was also explored for Zn-ion batteries [62].

On the other hand, Gong et al. [63] investigated the use of lignin for a gel polymer electrolyte that is compatible with lithium electrodes. The electrolyte films, shown in Fig. 9, were made of lignin and immersed in EC/DMC/EMC (ethylene carbonate/dimethyl carbonate/ethyl methyl carbonate). They demonstrated an electrolyte uptake of 230 %, an ionic conductivity of 3.73 mS/cm, an electrochemical stability up to 7.5 V, and good thermal stability and flexibility.

4.3. Starch

Starch is a naturally occurring and abundant polysaccharide. It is found in high amounts in plants such as potato, corn, and rice. According to its chemical structure, starch consists of unbranched amylose and branched amylopectin, as shown in Fig. 10. However, starch types are conveniently classified and referred to based on their source (e.g., potato starch). The chain lengths and molecular weights vary depending on the starch source. In addition to being a renewable, biodegradable, non-toxic feedstock, starch is hydrophilic, fairly soluble and adhesive. In rechargeable batteries, starch shows potential as an aqueous electrolyte material due to its high molecular weight, branched structure, and ionic conduction. However, challenges including lower ionic conductivity and mechanical stability, temperature and humidity sensitivity, compatibility with electrode materials, and limited shelf-life necessitate further research for practical implementation [64–66]. Studies investigated the addition of plasticizers and other additives such as glycerol, cellulose, PVA, and many more to improve the properties of starch for different types of electrolytes [41,67].

In terms of starch in aqueous electrolytes for rechargeable batteries, there is limited literature. Teoh et al. [67] studied different ratios of corn starch, $LiClO_4$ as the salt, and water as the solvent for the fabrication of aqueous electrolytes. The thin films obtained by the solution casting technique demonstrated relatively good ionic conductivities, among which the highest was 1.28×10^{-4} S/cm at 353K when 40 wt% of salt and 60 wt% of starch were used. This study demonstrates the effective plasticizing properties of $LiClO_4$ when combined with corn starch. Corn starch was also used with additives such as PVP (Polyvinylpyrrolidone) [69].

4.4. Pectin

Pectin is a natural polysaccharide found in plant cell walls, primarily sourced from fruits and vegetables, such as apples, citrus fruits, and carrots. Different types of pectin are classified based on their degree of esterification, influencing their gelling and thickening properties. As a renewable resource derived from plant-based materials, pectin aligns with sustainable and eco-friendly practices.

Pectin exhibits gelling and stabilizing properties, making it widely used in the food industry for fruit preserves, jams, and jellies [70,71]. Pectin has also shown potential in applications beyond the food sector, including in batteries as an electrolyte component. When combined with other polymers and ionic salts, pectin-based electrolytes have demonstrated good ionic conductivity, mechanical stability, and adhesion properties. For example, Kiruthika et al. [70] used pectin with magnesium nitrate salt to fabricate a polymer electrolyte which demonstrated an ionic conductivity of 10^{-4} S/cm and stability up to 3.8 V. However, the sample was created for a magnesium primary battery. So far, limited literature is available about the use of pectin in rechargeable batteries.

4.5. Soy protein

Soy protein is a protein-based compound derived from soybeans, a renewable and sustainable plant source. Its chemical structure consists of amino acids linked together, providing favorable sites for ion transport and contributing to improved ionic conductivity in electrolytes. Generally, soy protein is known for its nutritional value and is widely used in the food industry as an alternative to animal-based proteins. Beyond culinary uses, soy protein is attracting interest as a potential material for polymer electrolytes in rechargeable batteries. Blended with other polymers and ionic salts, soy protein-based electrolytes have shown promise in terms of ionic conductivity, mechanical stability, and adhesion properties.

Zhu et al. [72] used an electrospinning method to fabricate a fibrous membrane that is activated by an aqueous electrolyte for Li-ion batteries. The electrospinning solution contained a soy protein isolate (SPI), PVA, glacial acetic acid and water and was spun using a 10 cm^3 syringe. The optimum SPI-to-PVA ratio of 3:1 showed an ionic conductivity of 3.8×10^{-3} S/cm, excellent electrochemical stability, and good compatibility with lithium electrodes.

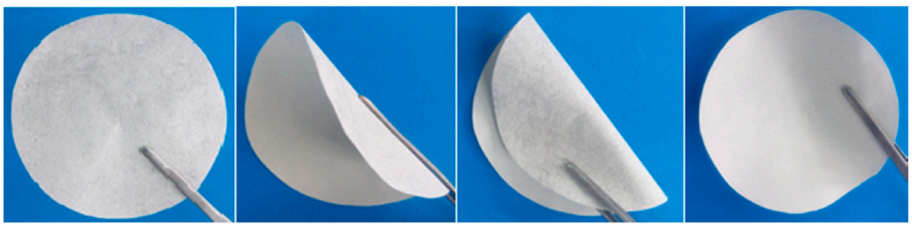

Fig. 9. *Flexibility of a lignin-based electrolyte film by* Ref. [63].

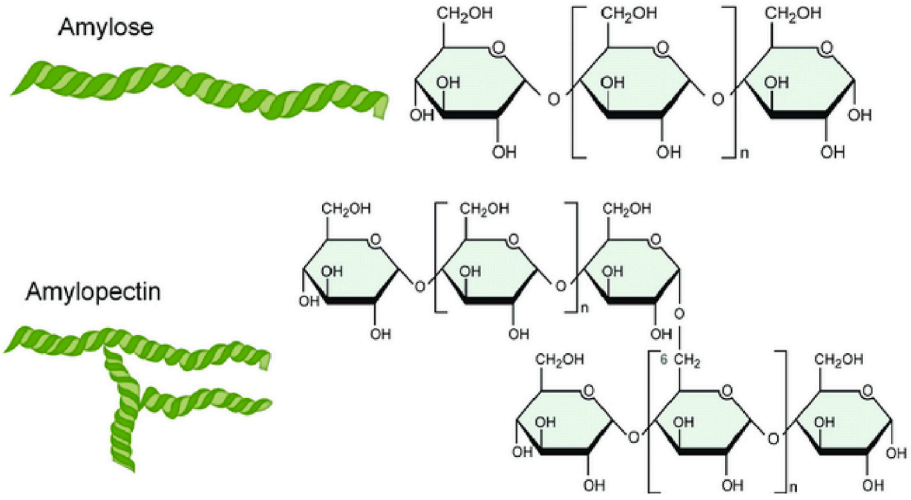

Fig. 10. *Chemical structures of the building blocks of starch: amylose and amylopectin* [68].

4.6. Silica

Silica, also known as silicon dioxide, is a naturally occurring compound commonly found in the form of quartz and sand. Silica is also produced by some types of algae. Its chemical structure consists of silicon and oxygen atoms arranged in a crystalline lattice. As a renewable resource, silica is abundant in nature and can be derived from renewable plant-based sources like rice husk ash, bamboo, and even algae [73–76]. Silica exhibits high thermal stability, excellent mechanical strength, and resistance to moisture and chemicals, making it suitable for use in diverse industries, including construction, electronics, and healthcare [77–82]. In the context of electrolytes for rechargeable batteries, silica-based materials have been explored for their potential in enhancing mechanical and thermal stability, improving the safety and performance of rechargeable batteries.

One of the limitations of using pure silica is its high thixotropic nature which leads to short gelling time and low ionic conductivity. To overcome this challenge, Hoang et al. [83] mixed fumed silica with the non-thixotropic β-cyclodextrin to make an aqueous electrolyte for a rechargeable Zn/$LiMn_2O_4$ battery. In comparison with a reference battery whose electrolyte does not contain silica, the proposed composite achieved 8 % higher capacity retention, 10 % enhanced reversible capacity, less dendrite formation, better thermal tolerance, and less corrosion. This presents promising possibilities for the use of silica in rechargeable batteries. In another study, Hoang et al. [84] demonstrated that using fumed silica with fatty methyl ester ethoxylate (FMEE) in an aqueous electrolyte containing $ZnSO_4$ and $MnSO_4$ achieves a capacity retention of 99.8 % after 100 cycles.

Addressing the same issue of high thixotropy, Mitha et al. [85] investigated the impact of incorporating poly(ethylene) glycol (PEG) as a non-thixotropic gelling agent in an aqueous electrolyte based on fumed silica for a Zn-ion battery. In their study, PEG also acted as a corrosion and dendrite growth inhibitor. The addition of 1 M.wt% PEG and 5 M.wt % silica to a reference aqueous electrolyte decreases its corrosion current density by 27 %. Additionally, it increased the battery's capacity retention by 39 % after 1000 cycles. Fig. 11 shows the SEM images of the electrodes of the fabricated batteries when different percentages of the gel electrolytes are used. In another study, results by Nguyen et al. [86] demonstrated the efficacy of combining fumed silica, an aqueous electrolyte (Li_2SO_4 and $ZnSO_4$), and a glass mat separator in improving the cyclability and capacity retention of the zinc-based secondary batteries.

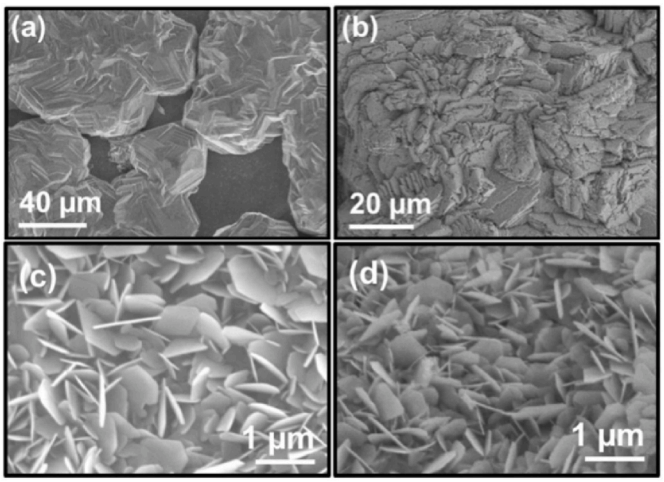

Fig. 11. *SEM images of the zinc electrodes when the (a) reference electrolyte (b) 5 % fumed silica (c) 4 % fumed silica and 1 % PEG (d) 3 % fumed silica and 2 % PEG are used* [85].

4.7. Natural gums

4.7.1. Guar gum

Guar gum is a hydrophilic polysaccharide derived from the seeds of the guar plant (*Cyamopsis tetragonoloba*). The ability of guar gum to dissolve readily in water and form a stable, viscous solution makes it a promising material for use in aqueous electrolytes of rechargeable batteries. It is also salt tolerant and acid resistant. In battery applications, guar gum can serve as a thickening and stabilizing agent, improving the overall performance and safety of the electrolyte. Its ability to create a stable gel-like matrix can enhance the adhesion between the electrode and electrolyte, ensuring better ionic conduction and preventing electrolyte leakage. Moreover, guar gum's biocompatibility and biodegradability align with the growing demand for sustainable and environmentally friendly energy storage technologies.

Wang et al. [87] and Huang et al. [88] explored the use of guar gum for electrolytes of Zn-ion batteries. In Ref. [88], guar-gum-based electrolyte was used in a Zn-ion battery as shown in Fig. 12 (left). The use of guar gum showed enhanced mechanical stability, excellent flexibility and bending durability, and reduced dendrite formation in the Zn-ion battery. It demonstrated an ionic conductivity of 1.07×10^{-2} S/cm and a capacity retention of 100 % after 1900 cycles and 85 % after 2000

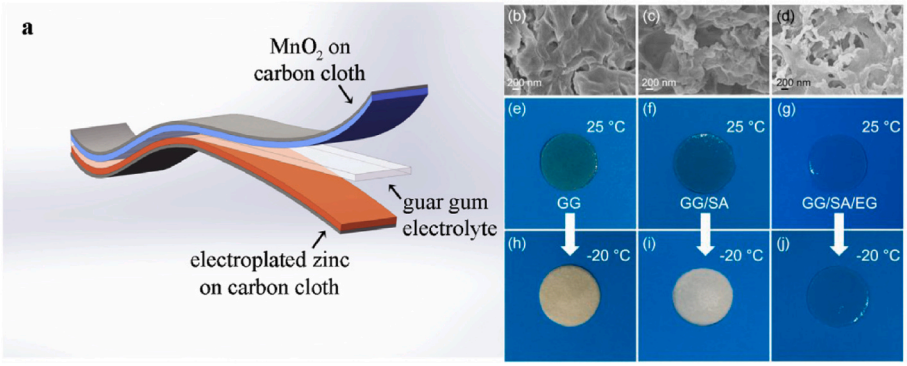

Fig. 12. *(a) Composition of the battery used by* Ref. [88] *to test the guar-gum electrolyte. (b-j) Electrolytes made of (e, h) guar gum, (f, i) guar gum and sodium alginate, (g, j) guar gum, sodium alginate, and ethylene glycol at ambient temperature and sub-zero temperature* [87].

cycles at 6.0 A/g. This electrolyte was stable in temperatures between 5 °C and 45 °C. In Ref. [87], the authors addressed the issue of freezing at sub-zero temperatures which limits the functionality of the battery. They developed an anti-freezing polymer electrolyte composite made of guar gum, sodium alginate, and ethylene glycol. Addition of the anti-freezing ethylene glycol improved the performance at low temperatures, however reducing the capacity retention. Without ethylene glycol, the electrolyte showed an ionic conductivity of 25.37 mS/cm at room temperatures and a capacity retention of 91.52 % over 1000 cycles. On the other hand, with ethylene glycol, the electrolyte achieved an ionic conductivity of 6.19 mS/cm at −20 °C and a capacity retention of 80.39 % over 100 cycles. Fig. 12 (right) shows the hydrogel electrolytes fabricated in this study.

4.7.2. Locust bean gum

Locust bean gum (LBG), derived from the seeds of the carob tree (Ceratonia siliqua), is a hydrocolloid with high water solubility and excellent thickening and stabilizing properties. When introduced into the aqueous electrolyte, locust bean gum creates a gel-like structure that enhances mechanical integrity and interfacial adhesion between the electrode and the electrolyte. This improvement in adhesion not only facilitates better ionic conductivity but also effectively prevents the dissolution or leakage of the electrolyte, thus contributing to the overall stability and safety of the battery system. Furthermore, the biocompatibility and biodegradability of locust bean gum align with the growing interest in green biomass alternatives for energy solutions. Guar gum is used in various applications including culinary, pharmaceutical, cosmeceutical, battery binders, etc.

In the context of aqueous rechargeable batteries, Liu et al. [89] explored the use of LBG with an aqueous $ZnSO_4$ solution for the fabrication of a gel polymer electrolyte. The sample, prepared as shown in Fig. 13, achieved high ionic conductivity of 33.57 mS/cm, capacity retention of 100 % after 3300 charge-discharge cycles, and 80.74 % after 1000 bending cycles which indicated high flexibility for wearable devices. The sample also inhibited dendrite formation and was stable within 0–40 °C.

4.7.3. Xanthan gum

Xanthan gum is a biopolymer produced by the bacterium *Xanthomonas campestris*, and it exhibits remarkable water solubility, salt tolerance, and strong thickening capabilities. When incorporated into the aqueous electrolyte, xanthan gum forms a stable and viscoelastic matrix that enhances the mechanical integrity and adhesion between the electrode and electrolyte. This feature promotes improved ionic conductivity and prevents the electrolyte from leaking or deteriorating over time, thus contributing to the overall stability and safety of the battery system. Xanthan gum was used by Zhang et al. [90] to make an electrolyte for a Zn–MnO_2 battery. The battery was made of xanthan gum and sulfate salts and resulted in a relatively high ionic conductivity of 1.46×10^{-2} S/cm, a 90 % capacity retention over 330 cycles, and a high bending durability. These results make xanthan gum an attractive option for further exploration in the field of rechargeable batteries electrolytes.

5. Algae-derived materials

5.1. Alginate

Alginate, a natural polysaccharide commonly extracted from brown algae, shows great potential as a sustainable material for aqueous electrolytes in energy storage and conversion systems. Its exceptional water solubility enables efficient ion transport in homogeneous solutions, facilitating electrochemical processes. Its biocompatibility and biodegradability make it a good fit for bioelectronics and medical devices that can be implanted in the human body. Since alginate can form films, it becomes possible to create flexible and versatile electrolyte composites that enhance the safety and reliability of wearable

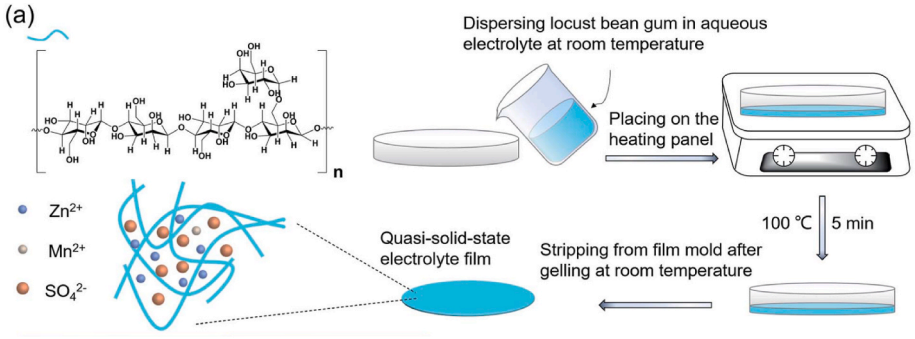

Fig. 13. *Process of fabricating a locust bean gum electrolyte by* Ref. [89]. Available under a CC-BY-NC license.

electronics. Another benefit is that brown algae, the source of alginate, is abundant and renewable, which makes it a sustainable choice. Although alginate has some challenges, like lower ion conductivity compared to certain other electrolytes, ongoing research aims to overcome these limitations, making way for wider applications in clean energy technologies.

In a study by Dong et al. [91], the use of a sodium alginate-based electrolyte to stabilize the cathode of Zn-ion batteries proved highly advantageous, as it successfully addressed the challenge of cathode dissolution, leading to a high capacity retention of 96 % after 1000 cycles at 2 A/g and a superior ionic conductivity of 2.92×10^{-2} S/cm. The entrapping of sodium ions within the alginate after gelation played a crucial role in enhancing its mechanical properties and limiting the dissolution of the electrode. The utilization of sodium alginate to tackle dendrite growth and metal anode consumption in Zn-ion batteries was also investigated by Tang et al. [92] and yielded promising results, in which the alginate-based electrolyte demonstrated good mechanical properties and ionic conductivity (1.83×10^{-2} S/cm). The graphs in Fig. 14 demonstrate the detailed electrochemical performance of the electrolyte. Fig. 14 also shown the crosslinking process and the flexibility of the fabricated sample. The confinement of ions effectively mitigated the issue of anode degradation. In another study [93], sodium alginate was used in combination with other additives (mentioned in Table 2) to fabricate an anti-freezing electrolyte that withstands temperatures as low as −50 °C and has an ionic conductivity of 10.38 mS/cm at −20 °C. Zhang et al. [94] investigated the use of modified alginate in aqueous electrolytes of Zn-ion batteries. They used tannic acid modified sodium alginate to prepare an electrolyte that demonstrated an ionic conductivity of 2.42×10^{-2} S/cm and a capacity retention of 94.5 % after 900 cycles at 2 A/g.

Alginate-based electrolytes were also explored in zinc-iodine batteries by Shang et al. [95]. Their successful application of an alginate-based polyanionic hydrogel electrolyte in zinc-iodine batteries resulted in a remarkable capacity retention of 97.6 % after 200 cycles at 0.2 A/g and addressed corrosion and dendrite growth issues. These findings highlight the effectiveness of alginates in addressing crucial challenges within different zinc-based battery systems.

In addition to zinc-based batteries, the application of alginate electrolytes was also investigated in Li-ion batteries. Fuzlin et al. [96] studied the relation between alginate as a base material for the electrolyte and $LiNO_3$ as the electrolyte salt. Among combinations of different percentages, the optimum ionic conductivity value was found to be 1.14×10^{-4} S/cm, indicating a positive potential of alginate to be used as an electrolyte material for Li-ion batteries and a good inorganic salt encapsulater. Table 2 summarizes the studies discussed in this section.

5.2. Agarose

Agarose, a versatile polysaccharide typically derived from red seaweed, holds promise in the field of rechargeable battery electrolytes. Its unique properties, including elasticity, flexibility, biocompatibility, nontoxicity, ion conduction, and reversible gel formation, make it a compelling candidate for battery applications. Fig. 15 shows the chemical structure of agarose. Additionally, agarose exhibits excellent film-forming ability, enabling the creation of stable and transparent free-standing films. Beyond battery research, agarose finds its applications in various industries, such as gelling agents in food, dental molds, biomedical materials, and electrophoresis [97–101]. Notably, several studies have explored the potential of agarose-based electrolytes, utilizing diverse materials such as acetic acid, lactic acid, $LiClO_4$, $KClO_4$, NiO, glycerol, and formaldehyde to enhance its performance in rechargeable batteries [102–104].

Raphael et al. [105] developed an agar-agar-based electrolyte, in combination with glycerol, formaldehyde, and acetic acid, resulting in transparent free-standing films. These films demonstrated ionic conductivity of 1.1×10^{-4} S/cm at ambient temperature and 9.6×10^{-4} S/cm at 80 °C. Notably, the films showed excellent stability up to 50 °C, with transparency levels ranging from 70 % to 92 %. The study revealed that ionic conductivity and transparency were influenced by the percentage of acetic acid. The ionic conductivity increased until the content of acetic acid reached 50 %, then started decreasing as shown in Fig. 16 (left).

Selvalakshmi et al. [104] and Park et al. [106] explored agarose-based electrolytes for zinc-based batteries. Selvalakshmi et al. [104] investigated agar-agar doped with ammonium thiocyanate (NH_4SCN), with the optimal ratio being 50 % agar-agar and 50 % NH_4SCN. This composition demonstrated ionic conductivities of 1.03×10^{-3} S/cm at ambient temperature and 3.16×10^{-3} S/cm at 343K, with cations playing a crucial role in enhancing the ion transport. Park et al. [106] developed an interphase layer using agarose crosslinked with citric acid, glycerol, and sodium hypophosphite monohydrate. This layer effectively prevented direct contact between the anode and aqueous electrolyte, suppressed dendrite growth, and facilitated zinc ion distribution. The interphase layer exhibited ionic conductivity of 9.52×10^{-3} S/cm and an impressive capacity retention of 94 % after 3000 cycles. Additionally, the transparent layer which is shown in Fig. 16 (right) and 17 (a) had a thickness of 300 μm. Additionally, Fig. 17 (b) shows the difference in electrochemical performance of a Zn-based battery with and without the interface.

Furthermore, Hazaana et al. [107] and Edward et al. [108] explored agarose-based electrolytes for lithium-based batteries. Hazaana et al. [107] utilized a solution-casting process with a ratio of 20 % of agar-agar and 80 % of LiCl, resulting in an aqueous electrolyte with amorphous structure and an ionic conductivity of 3.12×10^{-2} S/cm. The electrolyte showcased good cycle stability in Li-ion batteries. In Ref. [108], an electrolyte made of chitosan, agar-agar, polyethylene glycol (PEG), and $LiClO_4$ demonstrated an ionic conductivity of 4.56×10^{-4} S/cm and a stability up to 3.3 V. Remarkably, the electrolyte was utilized in Li-ion batteries, which are not commonly used with aqueous electrolytes. Table 3 summarizes the discussed studies which highlight agarose's potential as an electrolyte material in rechargeable batteries. Agar was also used with other types of salts such as NH_4Br [109] but with other types of energy storage systems.

5.3. Carrageenan

Carrageenan, an anionic polymer derived from algae, presents considerable promise as an electrolyte material for rechargeable batteries, specifically Li-ion and Zn-ion batteries. Iota, kappa, and lambda-carrageenan are three main types of carrageenan. They differ based on the number and positions of sulfate groups in the monomer unit. Fig. 18 shows the structures of these types of carrageenan.

Carrageenans are highly biocompatible, making them suitable for various applications in the food industry, pharmaceuticals, cosmetics, and now, energy storage systems. Their anionic nature facilitates ion transport, while their flexible and moldable properties enable thin and stable film formation. In the context of electrolytes for rechargeable batteries, carrageenan-based films have shown noteworthy ionic conductivities, demonstrating promise for safe and stable energy storage solutions.

The majority of the studies explored the use of carrageenan-based electrolytes for Li-ion batteries, as compared to other types of batteries [111–114]. In a study by Mobarak et al. [111], carboxymethyl iota-carrageenan and kappa-carrageenan were investigated for Li-ion aqueous electrolytes. Each type of carrageenan was combined with lithium nitrate ($LiNO_3$) and acetic acid in different percentages. The highest conductivity was achieved at 5.85×10^{-3} S/cm for a sample containing 20 wt% Li-salt with iota-carrageenan. However, the sample containing kappa-carrageenan exhibited greater transparency than iota-carrageenan films, as shown in Fig. 19. In another study by Mary et al. [115], kappa-carrageenan was used with the same Li-salt ($LiNO_3$)

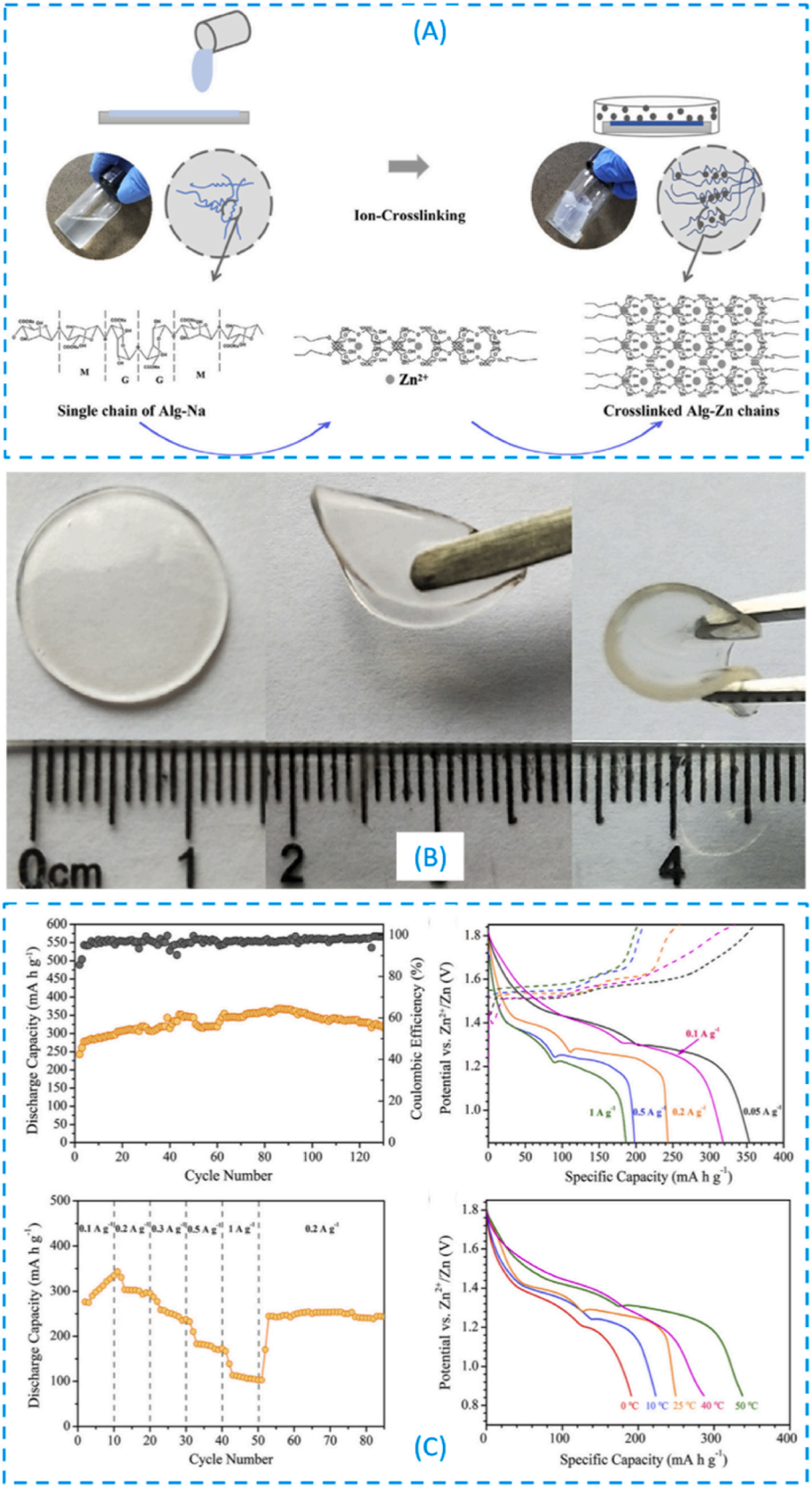

Fig. 14. *(A) creation of alginate-zinc chains via ion crosslinking. (B) Flexibility and transparency of the alginate-based electrolyte film. (C) Electrochemical performance of the fabricated electrolyte.* [92].

Table 2
Studies using alginate in aqueous electrolytes of rechargeable batteries.

Composite	Source	Battery	Process	Properties	Ref
Alginate and $LiNO_3$ salt	–	Li-ion	Solution casting	Ionic conductivity: 1.14×10^{-4} S/cm	[96]
Sodium alginate and polyacrylamide	Brown algae	Zn-ion	Crosslinking	Capacity retention: 96 % within 1000 cycles at 2 A/g. Ionic conductivity: 2.92×10^{-2} S/cm	[91]
Sodium alginate	–	Zn-ion	Ion crosslinking	Ionic conductivity: 1.83×10^{-2} S/cm	[92]
Sodium alginate, glycerol	–	Zinc-iodine	Ion crosslinking	Capacity retention: 97.6 % after 200 cycles at 0.2 A/g	[95]
Sodium alginate, [2-(Methacryloyloxy)ethyl] dimethyl-(3-sulfopropyl) (SBMA), acrylamide (AM)	–	Zn-ion	One pot polymerization method	Ionic conductivity: 10.38 mS/cm at −20 °C Anti-freezing up to −50 °C Capacity retention: 72.6 % after 1000 cycles at 1 C Coulombic efficiency: ~100 %	[93]
Tannic acid modified sodium alginate	–	Zn-ion	Solution casting	Ionic conductivity: 2.42×10^{-2} S/cm Capacity retention: 94.5 % after 900 cycles at 2 A/g Coulombic efficiency: ~100 %	[94]

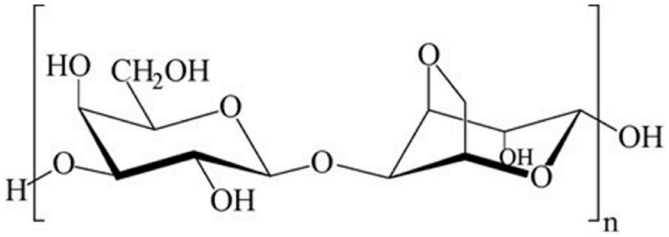

Fig. 15. *Chemical structure of agarose* [105].

without any additives and achieved an ionic conductivity of 1.89×10^{-3} S/cm (1 g kappa-carrageenan and 0.65 wt% of $LiNO_3$).

Chitra et al. [112] only used iota-carrageenan in combination with LiCl in a simple solution casting technique to obtain an electrolyte with an amorphous structure. The ionic conductivity in this electrolyte was found to be dominated by lithium ions, and the best value achieved was 5.33×10^{-3} S/cm with the composition of 1 g of carrageenan and 0.3 g of LiCl. On the other hand, Mary et al. [116] used kappa-carrageenan with the same salt (LiCl) and obtained an ionic conductivity that is one order of magnitude higher than the former study. The best ionic conductivity 1.15×10^{-2} S/cm was obtained using 1g kappa-carrageenan:0.5g LiCl.

Chitra et al. [113] focused on iota-carrageenan electrolytes with $LiClO_4$ and succinonitrile (SN) as an additive. The addition of SN improved the conductivity and flexibility of the electrolyte. The highest ionic conductivity observed was 3.33×10^{-3} S/cm for a sample containing 0.3 wt% SN, while without SN, the highest conductivity was 3.75×10^{-4} S/cm. Iota-carrageenan was also studied with lithium triflate ($LiCF_3SO_3$) using a solution casting method [114]. A sample comprising 1 g of carrageenan and 0.4 wt% $LiCF_3SO_3$ demonstrated an ionic conductivity of 1.27×10^{-3} S/cm and stability up to 3.52 V.

On the other hand, Huang et al. [117] explored kappa-carrageenan-based electrolytes for Zn-ion batteries. Their electrolyte sample consists of kappa-carrageenan, $ZnSO_4$, $MnSO_4$, and rice paper. The flexible films exhibited a high ionic conductivity of 3.32×10^{-2} S/cm and good cycling stability. The addition of rice paper enhanced the mechanical properties of this sample while the polymer allowed high ion transport. The study proposed a non-toxic, high-ionic conductivity, low-cost, and environmentally friendly flexible electrolyte for Zn-ion batteries. The Zn-ion battery using this electrolyte showed high capacity, fast charging, and good cycling stability. It showed good stability even with bending, making it suitable for wearable devices. Table 4 summarizes the studies mentioned in this section. It is noted that carrageenan has a good ability to encapsulate inorganic salts, especially Li salts within its polymer matrix [111–116].

6. Animal-derived materials

6.1. Gelatin

Gelatin is a protein-based substance derived from collagen, a structural protein found in animal bones and tissues. Its chemical structure is composed of amino acids linked together, providing favorable sites for ion transport. However, ion conductivity of pure gelatin is relatively low [118]. As a protein-based material, gelatin is sourced from renewable and sustainable animal by-products, aligning with the principles of

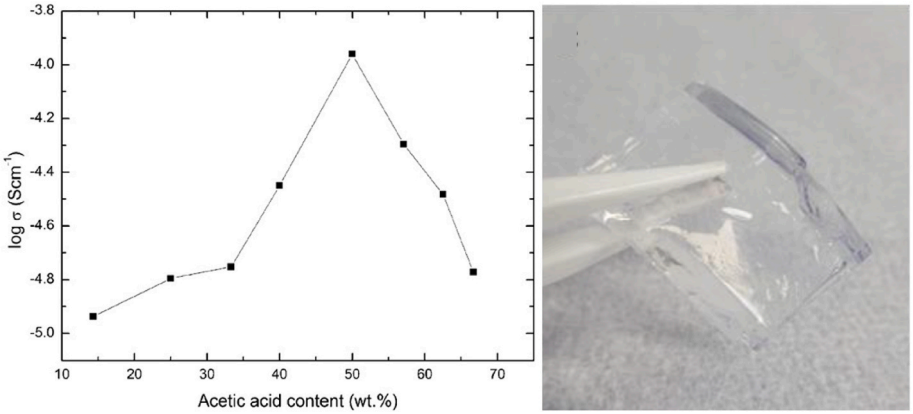

Fig. 16. *Left: Ionic conductivity of an agarose-based electrolyte composite as a function of acetic acid percentage by* Ref. [105]. *Right: An agarose-based film by* Ref. [106].

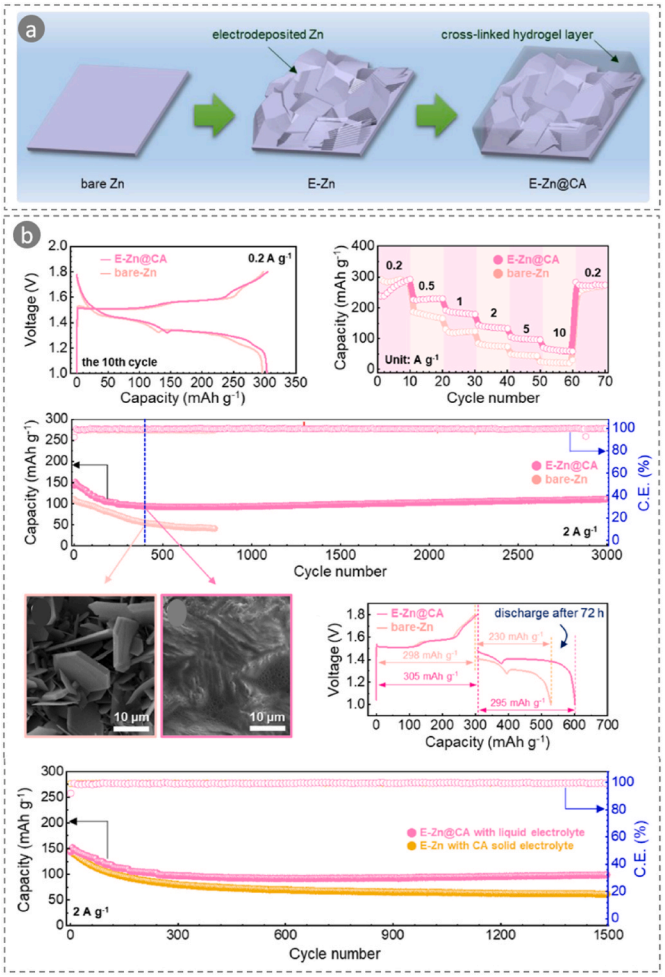

Fig. 17. *(a) an illustration of the agarose-based interface developed by* Ref. [106]. *(b) electrochemical performance of the Zn-based battery with and without the interface* [106].

environmental conservation. Additionally, it has high water and inorganic salt solubility [119].

In general, gelatin exhibits gel formation and film-forming abilities, making it suitable for various applications, including food, pharmaceuticals, and cosmetics. In the context of rechargeable batteries, gelatin has garnered interest as a potential material for polymer gel electrolytes. Studies explored the use of gelatin in combination with other components like polymers and nanomaterials to improve the electrochemical properties of gelatin-based electrolytes.

In the context of aqueous rechargeable batteries, Li et al. [119] developed a polymer electrolyte made of gelatin and polyacrylamide (PAM) for aqueous Zn-ion batteries. Their sample was made by grafting PAM on gelatin in a polyacrylonitrile (PAN) network and resulted in a 3D porous structure with high water retention, mechanical stability, and ionic conduction. The flexible electrolyte exhibited an exceptionally high ionic conductivity of 1.76×10^{-2} S/cm and high capacity retention of 97 % after 100 cycles at 2772 mA/g. Its high stability allowed it to perform well even when washed or put on fire, making it a promising option for wearable devices. Gelatin can also be used to fabricate biodegradable electrolytes, as proposed by Zhou et el [120]. In their study, they investigated the use of a gelatin-silk film as a humidity-sensitive biodegradable electrolyte for transient Zn-ion battery. Their transient battery demonstrated a specific capacity of 311.7 mAh/g, a capacity retention of 94.6 % after 100 cycles and 82.5 % after 80 bends, and full electrolyte degradation in 45 days under enzyme digestion. Fig. 20 shows the degradation mechanism and ion migration within the proposed battery.

6.2. Chitosan

Chitosan is a naturally occurring biopolymer derived from chitin which is found in the exoskeleton of crustaceans such as shrimp, crab, and lobster. As a renewable resource, chitosan is sourced from abundant crustacean waste, making it environmentally sustainable. The polycationic nature of chitosan gives it favorable ion transport properties.

Chitosan exhibits excellent film-forming ability and biocompatibility, making it applicable in a wide range of fields, including agriculture, medicine, and biotechnology [121–123]. In rechargeable batteries, chitosan has attracted attention as a potential material for polymer gel electrolytes. Blended with other polymers and lithium salts, chitosan-based electrolytes have demonstrated promising ionic conductivity, stability, and adhesion properties. Edward et al. [108] used a

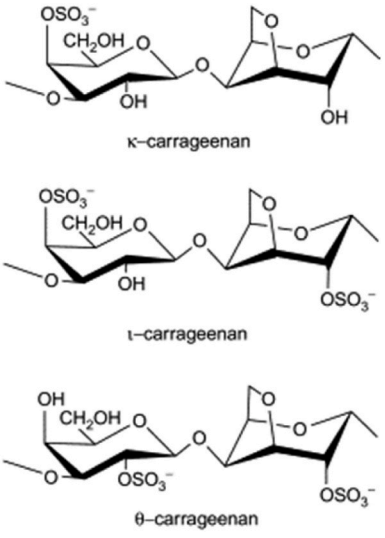

Fig. 18. *Chemical structures of carrageenans* [110].

Table 3
Studies using agarose/agar-agar in aqueous electrolytes of rechargeable batteries.

Composite	Source	Battery	Process	Properties	Ref
Agar-agar and LiCl		Li-ion	Solution casting	Ionic conductivity: $3.12 \pm 0.11 \times 10^{-2}$ S/cm	[107]
agar-agar, ammonium thiocyanate (NH_4SCN)	Algae	Zn-based	Solution casting technique	Ionic conductivity: 1.03×10^{-3} S/cm at ambient temperature and 3.16×10^{-3} S/cm at 343 K	[104]
Agar-agar, glycerol, formaldehyde, acetic acid	–			Ionic conductivity: 1.1×10^{-4} S/cm at room temperature and 9.6×10^{-4} S/cm at 80 °C	[105]
Chitosan, agar-agar	–	Li-ion	Solution casting technique	Ionic conductivity: 4.56×10^{-4} S/cm Stable up to 3.3 V	[108]
Agarose, glycerol, citric acid, $NaH_2PO_2 \cdot H_2O$	Seaweeds	Zn-ion		Ionic conductivity: 9.52×10^{-3} S/cm Capacity retention: 94 % after 3000 cycles	[106]

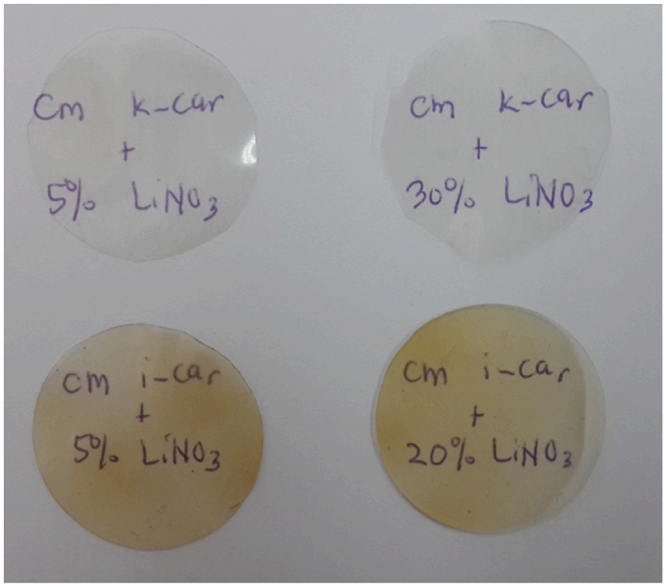

Fig. 19. *Electrolyte films consisting of carboxymethyl iota-carrageenan and kappa-carrageenan* [111].

Table 4
Studies using carrageenan in aqueous electrolytes of rechargeable batteries.

Composite	Source	Battery	Process	Properties	Ref
Carboxymethyl carrageenan, $LiNO_3$, acetic acid	Kappa-carrageenan from *Eucheuma cottonii* and iota-carrageenan from *Eucheuma spinosum*	Li-ion	Solution casting	Ionic conductivity of 5.85×10^{-3} Stability up to 3.0V	[111]
Iota-carrageenan, LiCl	–	Li-ion	Solution casting	Ionic conductivity of 5.33×10^{-3} S/cm	[112]
Iota-carrageenan, $LiClO_4$, succinonitrile (SN)	–	Li-ion	Solution casting	Ionic conductivity of 3.33×10^{-3} S/cm Stability up to 3.1 V	[113]
Iota-carrageenan, $LiCF_3SO_3$	Edible seaweed	Li-ion	Solution casting	Ionic conductivity of 1.27×10^{-3} S/cm Stability up to 3.52 V	[114]
Kappa-carrageenan, LiCl	–	Li-ion	Solution casting	Ionic conductivity: 1.15×10^{-2} S/cm	[116]
Kappa-carrageenan, $LiNO_3$	–	Li-ion	Solution casting	Ionic conductivity: 1.89×10^{-3} S/cm Stability up to 3.2 V	[115]
Kappa-carrageenan, $ZnSO_4$, $MnSO_4$, rice paper	–	Zn-ion	Solution casting	Ionic conductivity of 3.32×10^{-2} S/cm Flexibility and good cycling stability	[117]

combination of chitosan from black tiger shrimps, agar-agar, PEG, and $LiClO_4$ to fabricate a polymer-based electrolyte for Li-ion batteries. The amorphous nature and functional groups of chitosan contributed to the good mechanical properties, stability (up to 3.3 V), and ionic conductivity (4.56×10^{-4} S/cm). Chitosan was also used with polyaspartic acid in a process illustrated in Fig. 21 to fabricate an electrolyte that helps inhibit dendrite growth in Zn-based batteries [124]. This electrolyte demonstrated good biodegradability as shown in Fig. 22. Interestingly, it has the ability to degrade in the presence of bacteria and convert into plant nutrients.

7. Environmental and economic feasibility of using aqueous electrolytes from biomass sources

7.1. Environmental feasibility

The environmental feasibility of using aqueous electrolytes from biomass sources relies primarily on the availability and sustainability of biomass sources, whether direct raw biomass, or wastes and residues. Current sources of waste biomass include agricultural and industrial residues, such as sawdust, fruit peels, and pulp, along with other biomass materials derived from plants, algae, and marine creatures. If the huge amounts of biomass waste generated annually are not reused or recycled properly, they end up disposed of into the environment, leaving behind air, land, and water pollution [125]. Therefore, recycling biomass waste into battery components fits in as an advanced example of waste upcycling.

In addition, this approach not only reduces the environmental impact of biomass waste disposal but also minimizes the need to harvest nonrenewable fossil-based minerals that are conventionally used in batteries. Also, most renewable materials are naturally biodegradable, meaning the end-of-life disposal of such batteries does not harm the environment. However, some natural renewable materials still have poor biodegradability. Performing full cradle-to-grave life cycle analyses remains challenging at this point because the related technologies and processes are still in their early development phases. Even in relatively more mature fields of battery development and fabrication, proper assessment of environmental impacts is restrained by the inhomogeneity of life cycle assessment procedures adopted by the research community, which makes comparisons difficult [126,127].

Luckily, developments and innovations in the field of waste valorization are progressing rapidly, leading to better accessibility of biomass value added products [127]. Additionally, technological advancements in biomass extraction and processing could further reduce carbon footprints since research efforts aim to reduce chemical usage and reduce energy consumption. Moreover, public awareness and consumer demand for sustainable products could drive further environmental benefits, as more industries adopt greener practices and consumers opt for eco-friendly products. However, further comprehensive studies should be performed to analyze the supply of natural renewable materials versus the demand of the batteries industry. As a rule of sustainability, the demand should be high enough to deplete nature from its resources.

7.2. Economic feasibility

The economic feasibility of using aqueous electrolytes from biomass sources is shaped by several factors, including the cost of material extraction and processing, market potential, and scalability.

The cost of sourcing biomass depends on its availability, pretreatment, and transportation costs. If agricultural and industrial processes increasingly focus on waste management systems, sourcing could become more economical. The processing costs are influenced by the pretreatment technologies and energy required to convert raw biomass into useable battery components, with a focus on eco-friendly methods that could reduce these costs. However, since biomass materials are newly introduced to the batteries industry and are diverse in nature,

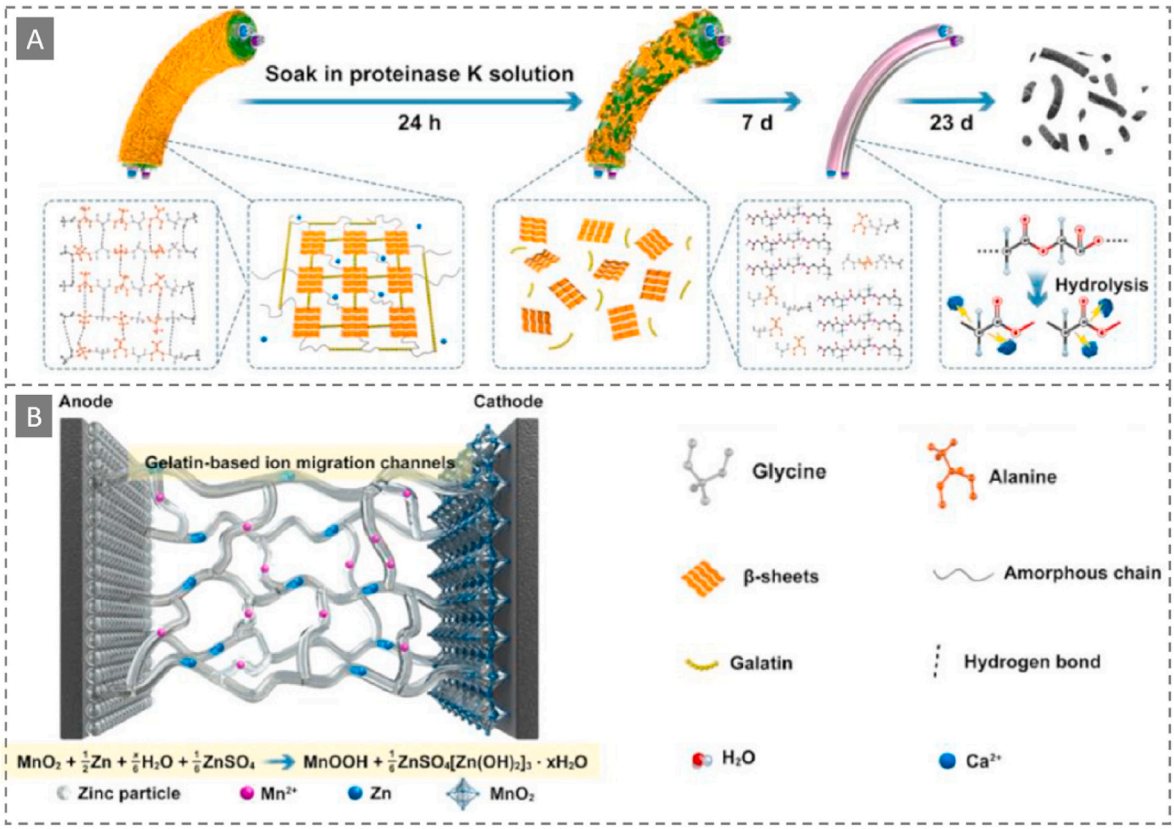

Fig. 20. *(A) Degradation mechanism and (B) ion migration of the transient Zn-ion battery containing a gelatin-silk electrolyte* [120].

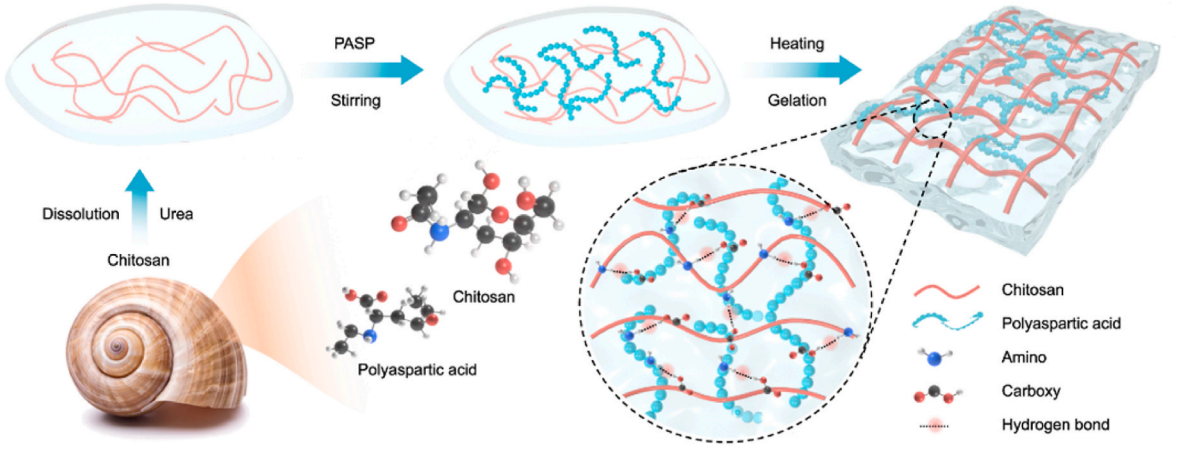

Fig. 21. *Preparation process of a chitosan-based electrolyte for an aqueous zinc battery* [124].

techno-economic and cost analysis studies are yet to be performed. Generally, researchers are starting to study the environmental and economic impact of biomass-based battery components, especially anodes. For example, Trotta et al. [128] performed a techno-economic and lifecycle analysis of biomass-based anodes to compare between Li-ion and Na-ion batteries and found that sodium-ion half-cells have lower environmental footprint and better techno-economics. Another study discusses techno-economic analyses of lignin-based products for different applications, not specifically for battery components. Overall, limited literature is available on the biomass material extraction and pretreatments for use in aqueous electrolytes of rechargeable batteries.

Market potential for aqueous electrolytes from biomass sources is expanding due to the demand for sustainable and green energy storage solutions. As electric vehicles and renewable energy storage systems become more popular and standardized, the scalability of these materials will be crucial. Business models supporting this market must consider the broader environmental impact and potential economic benefits, such as job creation, waste management, controlled battery biodegradation, and consumer safety. The long-term economic impact may also depend on government incentives and regulations promoting sustainable alternatives. However, additional information on the initial setup costs, the long-term sustainability of biomass sources, and the evolving regulatory landscape could provide a more comprehensive view of the economic feasibility.

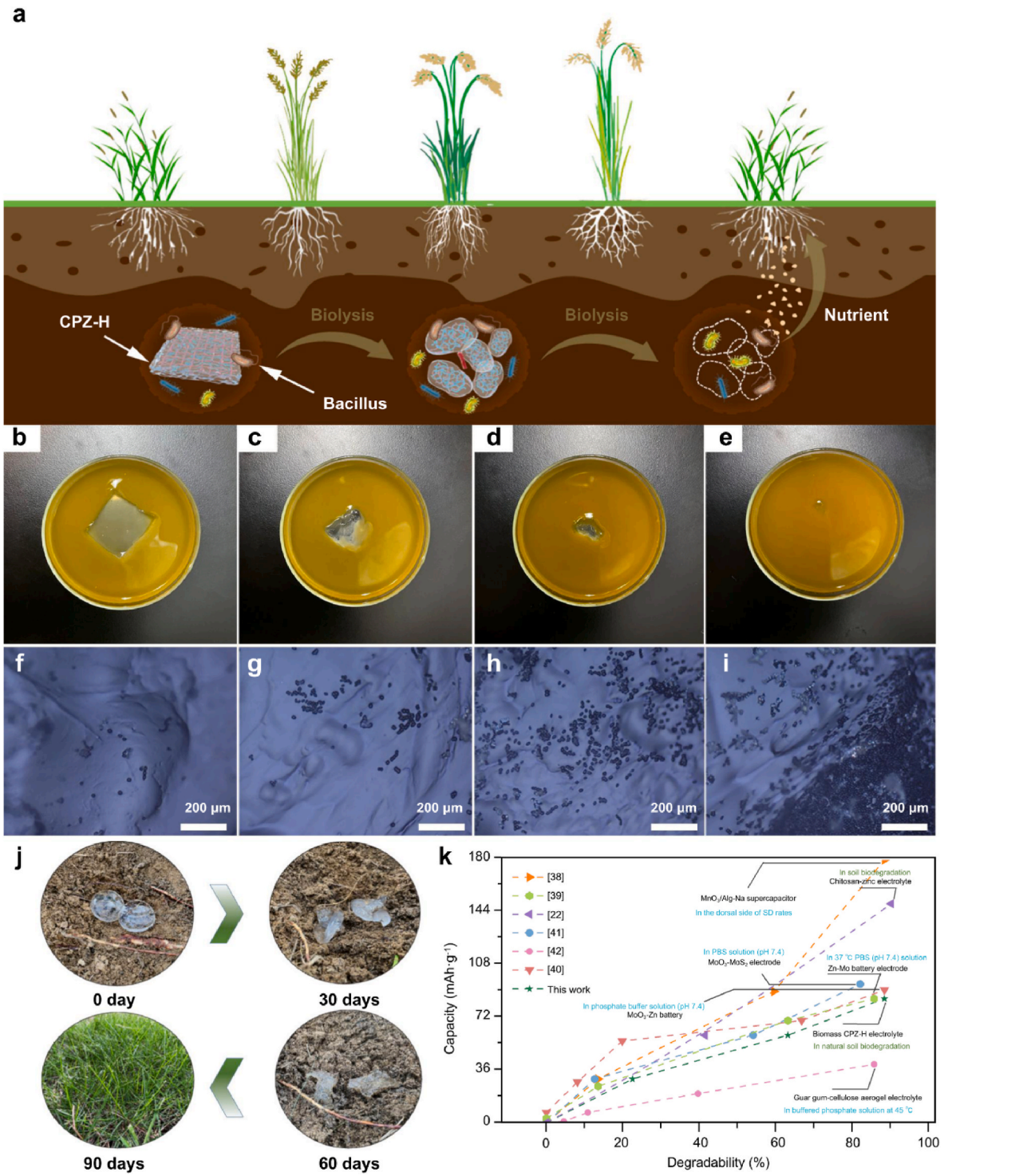

Fig. 22. *Degradation mechanism of the chitosan-based electrolyte* [124]. (a) Illustration of the degradation of the electrolyte into plant nutrients. (b–j) Degradation of the electrolyte over a 90-days period in a *Bacillus* culture fluid. (k) Degradation rate of the fabricated electrolyte compared to other biodegradable counterparts.

8. Discussion

Biomass from various sources and feedstocks such as lignocellulosic crops, algae, and the waste of seafood industry provide attractive sustainable materials that can be used in the aqueous electrolytes of rechargeable batteries. The exploration of biomass materials for rechargeable batteries holds significant promise in advancing sustainable and eco-friendly energy storage technologies. Utilizing renewable biomass resources as electrolyte components can reduce the environmental impact and reliance on finite natural sources such as fossil fuels, paving the way for greener and more sustainable battery solutions to address the growing demand for clean energy storage.

Biomass materials that were investigated for aqueous electrolytes of rechargeable batteries specifically are.

(1) Cellulose, lignin, lignocellulose (from lignocellulosic crops)
(2) Alginate, agarose, carrageenan (from algae)
(3) Starch

(4) Gelatin (from animal by-products)
(5) Chitosan (from crustaceans' shells)
(6) Pectin (from fruit skins)
(7) Soy protein
(8) Silica
(9) Guar gum, locust bean gum, xanthan gum

Fig. 23 illustrates the biomass materials discussed in this paper along with some highlights. Most of the studies used Zn-ion and Li-ion batteries, however some also used metal sulfur and other zinc- and lithium-based rechargeable batteries. It is worth noting that some studies also developed similar composites for primary batteries but since they are not rechargeable, they were not included in this review. Non-aqueous electrolytes were also excluded.

Polymer electrolytes combine the advantageous mechanical properties of solid electrolytes while providing higher ionic conductivities closer to liquid electrolytes. Additionally, they address numerous challenges such as cost, stability of the electrode-electrolyte interface, dendrite growth, thermal stability, flammability, and safety. Most biomass materials are soluble in water, thus eliminating the need to use flammable organic solvents and reducing the costs. Moreover, the flexibility and biocompatibility of polymer electrolytes make them an attractive option for wearable devices. However, to achieve the commercial stage, researchers are trying to find the best materials with the optimum design and electrochemical properties without sacrificing the sustainability, biocompatibility, and mechanical properties of the green material.

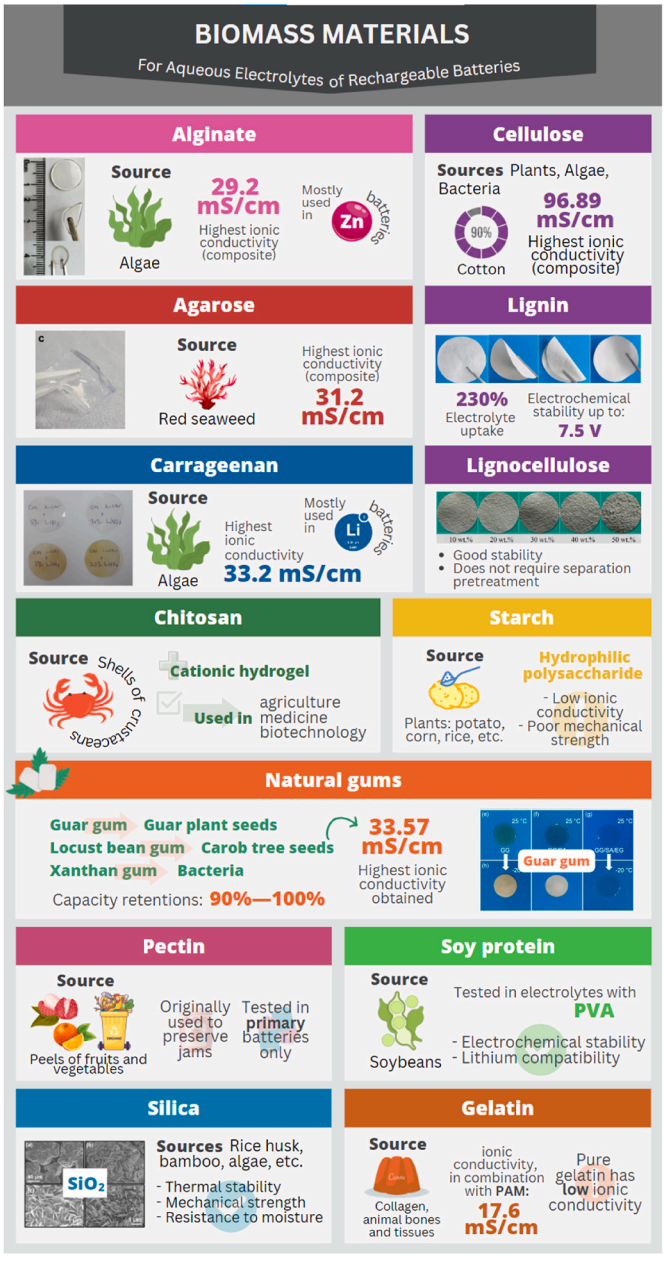

Fig. 23. Highlights of biomass materials for aqueous electrolytes of rechargeable batteries.

The most studied biomass materials in this field are lignocellulose, alginate, agarose, and carrageenan. They are sourced from plant crops and algae, which makes them readily available in nature. However, lignocellulosic materials should be obtained from crop residues rather than fresh plants to avoid affecting the food industry and negatively impacting the environment. Materials from algae are also preferably collected from algal blooms rather than deep ocean algae forests to avoid negative impact on the life under water. The mentioned polymers have favorable gelling properties that allow the fabrication of mechanically intact electrolytes which do not leak, can inhibit dendrite formation, and have good ionic conductivities. The typical fabrication technique used in most of the studies is creating films using a simple solution casting technique, ionic crosslinking, and/or immersion of polymer membranes in aqueous electrolytes. Some materials such as carrageenan can simply be used with salts to make the electrolyte while others require the addition of enhancers such as glycerol.

On the other hand, the least studied materials are starch, chitosan, pectin, soy proteins, and natural gums. These materials should be studied and developed further to exploit their full potential as they have shown a few promising results. For example, a guar-gum-based electrolyte achieved an ionic conductivity of 25.37 mS/cm and an electrolyte made of locust bean gum achieved an excellent ionic conductivity of 33.57 mS/cm.

Fig. 24 shows the highest ionic conductivities discussed in this review along with their corresponding biomass materials. Table 5 shows the composite that achieved the highest capacity retentions. Note that the electrolytes were not purely made of biomass materials, but the details of the composites are not included in the visual representations for simplicity. Each study was discussed in detail in previous sections. Comparisons show that cellulose has the highest potential in terms of its ionic conductivity while natural gums provide the highest capacity retentions. Better understanding of the ion transport mechanisms within the chemical structures of the natural gums will help to enhance their properties and to choose the compatible additives that can enhance their electrochemical properties. Similarly, for all types of biomass materials, more investigations should be done to achieve the optimum composites that deliver high electrochemical performance while maintaining the

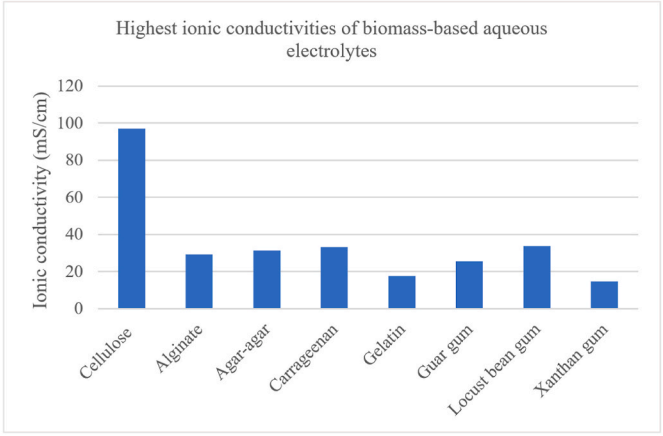

Fig. 24. Highest ionic conductivities reported in this study.

Table 5
Highest capacity retentions reported in this study.

Biomass material	Capacity retention
Cellulose	90.24 % after 1200 cycles
Alginate	97.6 % after 200 at 0.2 A/g
Agar-agar	94 % after 3000 cycles
Guar gum	100 % after 1900 cycles
Locust bean gum	100 % after 3300 cycles
Xanthan gum	90 % after 330 cycles

mechanical properties and sustainability of the biomass materials.

9. Future perspective and recommendations

According to the current literature, future trends for the use of biomass materials in aqueous electrolytes of rechargeable batteries involve enhancing electrochemical performance, developing advanced technologies and materials, scalability, and application in innovative areas. Below are key focus areas and recommendations for future research and development.

Better electrochemical performance: To meet the demands of modern rechargeable batteries, research trends are focused on improving the electrochemical performance of rechargeable batteries containing biomass-based electrolytes. This involves enhancing ionic conductivity, broadening the electrochemical stability window, and achieving a higher thermal stability [41]. Researchers are encouraged to explore novel additives, new electrolyte formulations and structures, and enhanced manufacturing techniques to boost overall battery efficiency.

Advanced composites and structures: One promising approach to improving electrochemical performance is through advanced composites and structures [41]. This includes the incorporation of nanofillers, blends of biopolymers with synthetic polymers, and the design of novel architectures. Future research should investigate how these composites can provide better mechanical properties and stability while maintaining eco-friendly characteristics.

Overcoming interfacial resistance: Interfacial resistance remains a significant obstacle in developing fully efficient batteries. Future efforts should focus on minimizing this resistance in order to improve the electrochemical performance. Strategies that are being investigated involve enhancing electrolyte-electrode contact and exploring new fabrication techniques to reduce interfacial resistance and ensure efficient ion transport [20,106].

Anti-freezing solid and semi-solid state batteries: The development of anti-freezing solid-state electrolytes offers new possibilities for batteries in cold environments. Researchers are exploring materials and structures that maintain functionality at low temperatures. This can be achieved through the use of specialized polymers and additives designed to prevent freezing and ensure consistent battery performance [87,93]. Since thermal stability in cold conditions is as important as in hot conditions, it is important to develop anti-freezing electrolytes that tolerate extreme conditions.

Pre-treatments, modifications, and functionalization: Pre-treatments, chemical modifications, and functionalization are crucial for enhancing the properties of biomass-based gel polymer electrolytes. When obtaining raw biomass from nature, it is typically necessary to perform pretreatment processes and even chemical modifications according to the need of the targeted battery. One of the most commonly functionalized biomass materials is cellulose, as it can change properties and functions easily with simple surface modifications [19,37]. Future trends may involve tailoring the surface chemistry of biopolymers, using cross-linking agents to strengthen mechanical properties, and exploring chemical functionalization to improve conductivity and stability. These approaches can lead to greater durability and performance in various battery applications.

Application in flexible and wearable devices: Polymer-based batteries are promising in the biotechnological field due to their flexibility and biocompatibility with human skin. As the demand for flexible and wearable devices grows, research is working on developing gel polymer electrolytes suitable for these applications. This includes improving the flexibility, biocompatibility, and safety of the electrolytes [41]. Researchers are encouraged to explore biopolymer-based materials and novel designs that facilitate integration into wearable technology.

Reversibility of Zn batteries using gel electrolytes: Aqueous electrolytes made of biomass materials are highly compatible with Zn-based batteries. According to the current research trends, they facilitate the reversibility of zinc anodes which is crucial for the longevity and efficiency of zinc-based batteries [92,129]. Future research is expected to focus on improving the reversibility of zinc anodes when using polymer electrolytes. This involves exploring new electrolyte compositions, optimizing gel properties, and studying the underlying electrochemical mechanisms to ensure consistent and reliable performance.

Diverse battery compatibility: Other than Zn-based batteries, biomass-based aqueous electrolytes are being studied in various battery types such as Li-ion batteries. Although the focus is slightly shifted towards Zn-based batteries, future research is expected to put more effort in investigating other types of batteries. For example, as discussed in Section 5.3, it is noted that carrageenan-based electrolytes are rather compatible with Li-ion batteries. Therefore, it is important to test each material and validate its compatibility with various aqueous battery types.

Commercialization and scaleup: Although the technologies regarding the use of biomass-based materials in aqueous electrolytes are still in their infant stage, it is important to keep an eye on the possibility and feasibility of scaleup when developing a new process or composite. Future research should aim to develop cost-effective manufacturing processes and scalable production techniques since commercialization and scalability are key challenges in the transition from lab-scale research to practical applications. This involves exploring new fabrication methods, reducing costs, and optimizing production processes for mass manufacturing.

10. Conclusion

This paper explored the potential of using biomass-derived materials for aqueous electrolytes of rechargeable batteries by reviewing and analyzing the relevant studies. Results show that renewable materials from various biomass sources such as lignocellulosic crops, gums, algae, and animal by-products have the potential to be used as an electrolyte base material, especially in gel polymer electrolytes. As discussed, gel polymer electrolytes combine the advantages of safety and relatively high ionic conductivities, which are challenging to obtain in liquid electrolytes (prone to leakage) and solid electrolytes (low conductivities). Additionally, aqueous gel electrolytes are less flammable since their base is water rather than organic solvents. Biomass-derived materials that have shown a positive potential to be used in aqueous electrolytes are cellulose, lignocellulose, alginate, agarose, carrageenan, starch, gelatin, chitosan, pectin, soy protein, silica, and natural gums. The highest ionic conductivity achieved was 96.89 mS/cm by a cellulose-based electrolyte and the highest capacity retention of 100 % after 3300 cycles was achieved by an electrolyte containing locust bean gum. However, these materials should be enhanced further, tested thoroughly, and optimized to achieve industry-level specifications. Further environmental and economic assessments should also be performed to analyze the viability of using biomass materials in aqueous electrolytes of rechargeable batteries.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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