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REVIEW ARTICLE

Latest advances in ionic liquids promoted synthesis and application of advanced biomass materials

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Abstract The utilization of sustainable resources provides a path to relieving the problem of dependence on fossil resources. In this context, biomass materials have become a feasible substitute for petroleum-based materials. The development of biomass materials is booming and advanced biomass materials with various functional properties are used in many fields including medicine, electrochemistry, and environmental science. In recent years, ionic liquids have been widely used in biomass pretreatments and processing owing to their "green" characteristics and adjustable physicochemical properties. Thus, the effects of ionic liquids in biomass materials generation require further study. This review summarizes the multiple roles of ionic liquids in promoting the synthesis and application of advanced biomass materials as solvents, structural components, and modifiers. Finally, a prospective approach is proposed for producing additional higher-quality possibilities between ionic liquids and advanced biomass materials.

Keywords biomass materials, functional materials, ionic liquids, synthesis, structure-property relationship

1 Introduction

The excessive production of petroleum-based chemicals has caused non-renewable resource shortages and serious environmental pollution and has correspondingly promoted the concept of sustainable and green production. Biomass materials derived from biomass (such as straws, fruit shells, shrimp shells, cellulose, chitin, alginate, and lignin) have attracted wide attention owing to their availability, renewability, biocompatibility, and abundance. The emergence of biomass materials has provided solutions to the crises of increasing shortages of oil resources, increasing environmental pollution, and large emissions of greenhouse gases [1]. Currently, researchers in many fields are committed to developing various biomass materials for medicine [2], electrochemistry [3], engineering technology [4], environmental science [5], and energy and fuel science [6]. In this study, the keyword "biomass materials" was searched on the core database of the Web of Science. The results involved 129 research fields and showed that the number of articles increased annually, indicating that research on biomass materials is getting increased attention (Fig. 1). Under the premise of meeting basic application requirements, advanced biomass materials with special properties are gradually emerging to optimize practical applications. These properties include thermal stability, frost resistance, antibacterial properties, and electrical conductivity. However, the diversity and complexity of the structures of biomass raw materials [7] and resistance of the biomass itself to chemical, physical, and biological decomposition have seriously hindered the synthesis of advanced biomass materials [8].

In recent years, the problem of biomass conversion has been greatly resolved with the widespread use of important functional fluids—ionic liquids (ILs). ILs are organic molten salts at temperatures below 100 °C and are usually composed of large asymmetric organic cations and organic or inorganic anions. They have excellent chemical stability, thermal stability, electrochemical stability, non-flammability, and negligible volatility [9]. Owing to their unique properties, ILs are widely used in chemical reaction processes as solvents [10,11], extractants [12,13], catalysts [14,15], reaction media [16,17], or their combinations [18]. In the synthesis of advanced biomass

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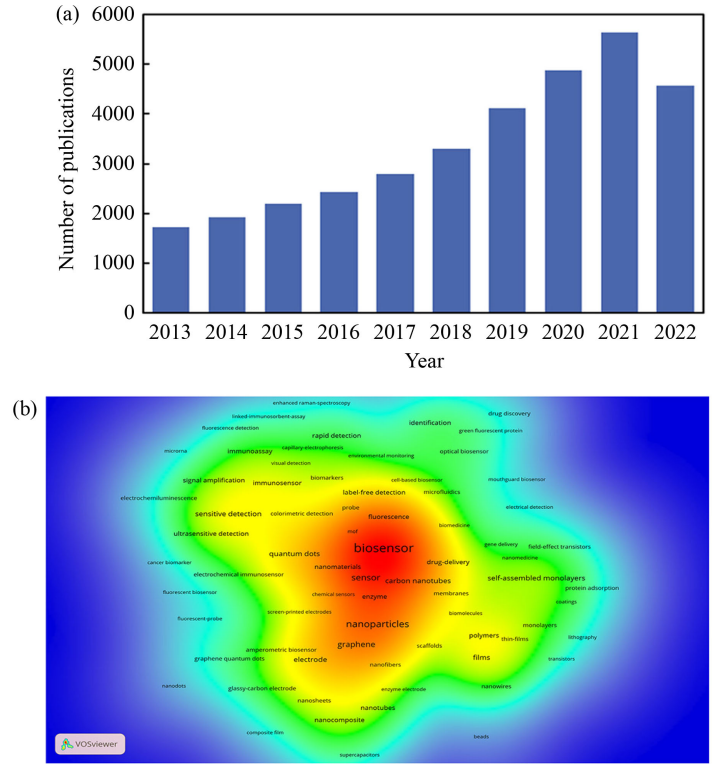

Fig. 1 (a) This statistical graph is based on published data retrieved on the Web of Science by the keyword "biomass materials", Nov. 19th, 2022; (b) this heat map is based on data from Web of Science, Nov. 19th, 2022, and analyzed by a VOS viewer.

materials, ILs can not only participate in the chemical reaction process but also be used as structural components to optimize the properties of the biomass materials by virtue of their biocompatibility, electrical conductivity, antibacterial properties, and other advantages [19–22]. In addition, ILs can be used as modifiers with specific charge groups or functional groups to improve or enhance the performances of biomass materials by introducing groups into the active reaction sites of the biomass [23] (Fig. 2).

In this mini-review, we reviewed the latest research progress of ILs acting on advanced biomass materials based on the three important roles of ILs. The content was divided into four parts: (1) ILs used as solvents to synthesize advanced biomass materials; (2) ILs used as structural components to strengthen advanced biomass materials; (3) ILs used as modifiers to modify advanced biomass materials; (4) a summary of the research.

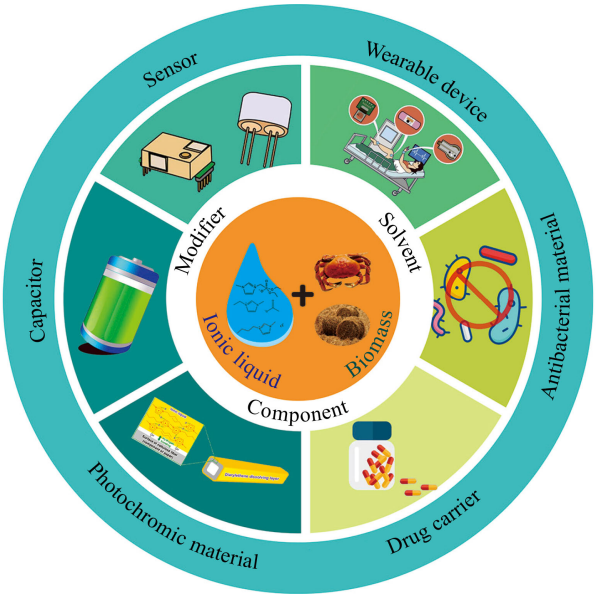

Fig. 2 Schematic diagram of ILs promoting the synthesis and application of advanced biomass materials.

2 ILs as solvents for advanced biomass materials synthesis

The complex structures and compositions of most biomass make them insoluble in ordinary solvents, hindering their deconstruction and utilization [24]. Thus, solvents able to dissolve biomass will make an outstanding solvation contribution to synthesizing value-added advanced biomass materials. To resolve the shortcomings of traditional organic solvents and mineral acids/bases such as their harsh conditions, high toxicity, flammability, low recovery, and poor stability [25], researchers are committed to finding environmentally-friendly and efficient alternatives. The explored solvents include biomass-derived resources, water, supercritical CO_2, and ILs [26].

As one type of task-specific media, the unique physicochemical properties of ILs compensate for the shortcomings of most organic solvents and provide comparable solvent properties. The negligible vapor pressure of ILs alleviates the environmental pollution caused by volatile organic solvents and the high melting point and thermal stability can withstand high-temperature and high-pressure reactions (ILs are not flammable in most cases) [27]. More importantly, based on the structural characteristics of ILs, appropriate combinations of anions and cations can be designed according to application requirements [28]. This feature overcomes the limitation of traditional organic solvent selection and has won ILs the title of "design solvent" [29].

2.1 Biomass extraction by ILs

Biomass is mainly distributed on land, in oceans, and in deep underground environments [30]. Lignocellulose represents most of the terrestrial biomass. Its most important components include cellulose, hemicellulose and lignin and a small number of other organic compounds such as pectin and protein [31,32]. Owing to the highly complicated 3D amorphous network surrounding the outer layer of polysaccharide fibers [33,34], lignin is regarded as one of the main obstacles to the deconstruction of lignocellulose biomass. Delignification can help produce cellulose-rich materials (CRMs) and high-purity hemicelluloses; these are the raw materials for conversion into various advanced biomass materials, biofuels, or valuable chemical products [6,35]. However, this process requires complex mechanical grinding and chemical purification. The large amount of "black liquor" produced by chemical treatment causes wastes of resources and environmental pollution. In recent years, the application of ILs in lignocellulose pretreatment has aroused widespread concern in the scientific community. Figure 3 summarizes the typical pretreatment routes of lignocellulose by ILs. Owing to the different solubility of the various components in lignocellulose, the composition and structure of the biomass change after the IL treatment and promote a subsequent upgrading process [36].

In studying the adsorption of heavy metal ions by cellulose, Shen et al. [37] found that the adsorbent prepared by CRMs extracted from wood by IL 1-ethyl-3-methylimidazolium acetate ([Emim][OAc]) had a stronger adsorption capacity for Cu^{2+} and Pb^{2+} than that directly prepared from poplar powder. This was owing to the low specific surface area of lignocellulose in raw wood and insufficient exposure of the –OH adsorption sites. The IL treatment broke down the structure of the cell wall layer to expose the cellulose nanofibers (CNFs), resulting in a larger specific surface area and richer accessible –OH. In addition, a small amount of lignin remaining in the CRM was extracted by the IL. The porous structure supported by the lignin caused the CRM to have additional hydroxyl sites and voids [38], allowing the CRM to be further magnetized and carboxymethylated. An experiment showed that the adsorption capacity of the functionalized CRM adsorbent for heavy metal ions was higher than that of a similar cellulose adsorbent; the maximum adsorption capacities for Cu^{2+} and Pb^{2+} were 0.144 and 0.161 $mmol \cdot g^{-1}$, respectively.

In addition to the abundant renewable resources on land, chitin is the main component of crustacean shells and second-largest biopolymer on earth. It has the

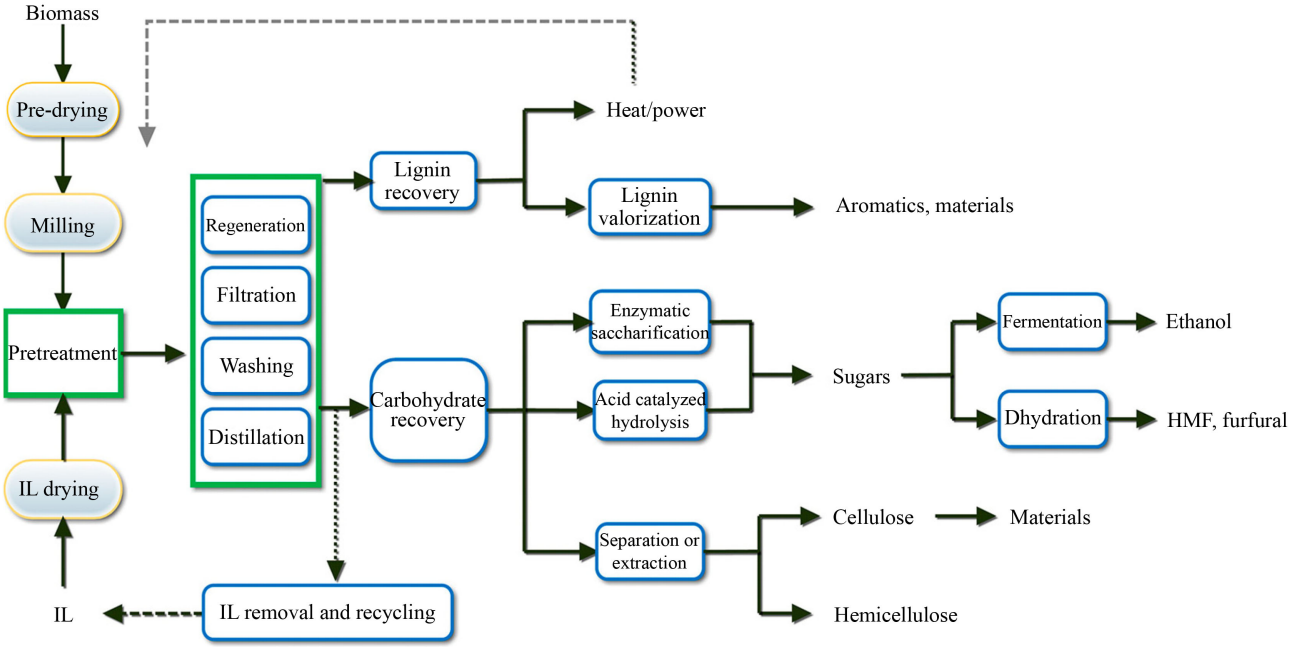

Fig. 3 Typical routes for the pretreatment of lignocellulosic biomass with ILs. Reprinted from Ref. [36].

potential to supplement lignocellulose biomass resources for producing renewable nitrogen-derived chemicals [39]. Owing to its unique properties such as its being a cationic polyelectrolyte, multi-functional reactions, antibacterial, biocompatibility, and biodegradability, it is widely used in medicine [40], food [41], and the environment [42]. At present, chitin extractions commonly use hydrochloric acid to remove inorganic salts, and then treat it with sodium hydroxide at temperatures up to 160 °C for several days to remove protein [43]. This causes many problems, such as high energy consumption, high emission, and equipment corrosion. Moreover, the strong acids and bases may lead to the depolymerization of the chitin and reduce its degree of acetylation, hindering the functional role of the chitin in materials [44]. Current green extraction technologies were developed to overcome the environmental problems associated with acid-base treatments. Studies using ILs to directly extract chitin from raw materials have shown great reliability [45]. Hydrogen bonds are formed between the anions of the ILs and acetamide groups and hydroxyl groups of the chitin, weakening the intramolecular and intermolecular hydrogen bonds of the chitin and leading to the deformation and peeling of the chitin molecular chain. Simultaneously, the cations of the ILs penetrate the gaps between the chitin molecular chains to prevent the recovery of the crystalline phase after shedding. Finally, the chitin chain is dispersed in ILs in the form of completely separated molecules. In this way, the ILs extract chitin from marine biomass in a relatively mild way [46]. Qin et al. [47] used [C_2mim][OAc] to extract 94% of the available chitin from shrimp shells in one step. The excellent extraction rate contributed to the conversion of chitin to advanced functional materials such as fibers, beads, and membranes. Microbeads are a promising material structure for biomedical and cosmetic applications [48,49]. In recent years, the environmental concerns owing to petroleum-based microbeads have led some countries to enact legislation to restrict or eradicate them [50]. Renewable biomass-based beads are emerging as a sustainable alternative to petroleum-based beads. King et al. [51] compared the abilities of chitin extracted by [C_2mim][OAc] from waste shrimp shells with that of commercially available chitin used in the production of cosmetic microbeads. The results showed that the latter beads had no bead formation at all and lacked uniformity in the shape or size of the product, whereas the IL-extracted chitin formed uniform beads with a smooth surface and internal porosity (Fig. 4); thus, they could be used to encapsulate and release active compounds with different chemical structures. Approximately 90% of the active substance was released after 7 h, providing a stable, prolonged release. This was owing to the low molecular weight of the commercially available chitin. It did not allow sufficient polymer chain entanglement when solidifying, thereby preventing the formation of beads. The chitin extracted using the IL had a higher molecular weight and could better meet the conditions for the formation of high-quality microbeads. However, the relatively high cost of [C_2mim][OAc] and its complex recycling process limit its large-scale application. Therefore, some researchers have proposed extracting chitin from shrimp shells based on one-pot pulping method using cheap IL [NH_3OH][OAc] made from highly acidic and alkaline ions [43]. This cheap pulping method has the same or even higher chitin yield and purity than the [C_2mim][OAc]-based extraction method with a 10-fold increase in the biomass loading, leading to better economic potential.

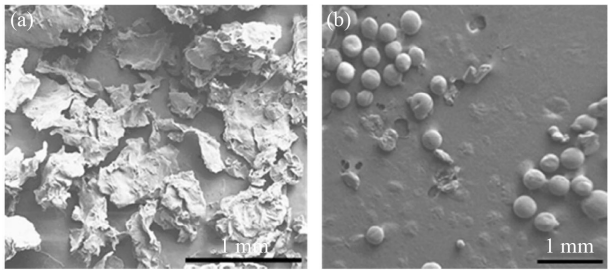

Fig. 4 Scanning electron microscopy (SEM) images of beads prepared from (a) commercially available chitin and (b) IL-extracted chitin (The beads size fraction used for imaging was > 250 μm). Reprinted with permission from Ref. [51], copyright 2017, American Chemical Society.

2.2 Biomass dissolution by ILs

Cellulose is a D-glucose polymer connected by *β*-(1-4) glycosidic bonds. It is the main component of the cell wall of an important protective structure of plant cells [52]. Cellulose has beneficial properties such as its hydrophilicity, biocompatibility, active hydroxyl group, and stereoscopic regularity, making it an important resource for derivative products such as chemicals, fibers, composites, films, and biofuels [53]. However, the existence of a large number of intramolecular and intermolecular hydrogen bonds in cellulose leads to high crystallinity and low dissolvability, hindering its value-added applications [54]. In 1934, a US patent by Graenacher first proposed the use of 1-ethylpyridinium chloride to dissolve cellulose [25]. However, because the melting point of 1-ethylpyridinium chloride was too high (approximately 118 °C) and the concept of ILs was not proposed at that time, this discovery was not taken seriously. In 2002, Swatloski et al. [55] conducted the first study on dissolving cellulose in ILs and found that the IL 1-butyl-3-methyl-imidazoliumchloride [Bmim][Cl] could dissolve cellulose, opening up a new field for the development and research of cellulose solvent systems. As mentioned in most of the current research, the hydrogen bond interactions between the anions of the ILs and cellulose are the keys to the dissolution of the cellulose in ILs. These hydrogen bonds change the

orientation of the cellulose hydroxyl groups, making the surface chains swell in the solution. Meanwhile, the roles of the cations cannot be ignored. The heterocyclic structures and alkyl chain lengths of the cations have important effects on the interactions between the anions and cellulose. Through the synergistic actions of the cations and anions, the cellulose chains are gradually peeled from the crystal and cellulose forms less uniform aggregates with the ions until fully dissolved in and surrounded by the ILs [56,57].

The principle of a circular economy has enabled people to turn their attention to waste utilization. Waste paper usually contains 90%–99% cellulose derivatives and is an ideal choice for recycling and value-added products [58]. Jeong et al. [59] used the IL 1-allyl-3-methylimidazolium chloride ([Amim][Cl]) as a solvent to dissolve waste paper by destroying the cellulose structure. Then, an anti-solvent (anhydrous ethanol) was added for cellulose regeneration. The IL was separated according to the principle that IL interacts more strongly with anti-solvents than with cellulose. The recovery rate of IL was more than 96% and the recovered IL maintained a high dissolution efficiency. Finally, using the regenerated cellulose as a raw material, regenerated cellulose carbon dots (RCCDs) were synthesized by microwave-assisted method. Although the mechanism of carbon dot (CD) production from regenerated cellulose is not clear, existing research suggests that pretreatments such as ILs, enzymes, or acid treatments are often required during cellulose syntheses of CDs [15,60,61]. The structure of the pretreated cellulose fiber decomposes and its surface area increases; these conditions are beneficial to the conversion process. The dissolution of IL provides these conditions for the formation of CDs. In the study by Jeong et al. [59], the prepared RCCDs showed blue fluorescence under 365-nm UV light (Fig. 5). The red-shift phenomenon was observed by gradually changing the excitation wavelength from 340 to 440 nm. In addition, the RCCDs had good biocompatibility and could be further applied to fluorescence biotechnology.

Chitosan, a product of the complete or partial deacetylation of chitin, is the second-most abundant natural nitrogenous biopolymer after protein [62]. Chitosan has great potential in biomedical applications because of its low toxicity, antibacterial activity, hygroscopicity, biocompatibility, and biodegradability. In the traditional process, acetic acid is used to dissolve chitosan [63], but the effects of the acid residue on skin, mucosa and tissue have hindered the application of chitosan in the field of biomedicine. Recently, Li et al. [64] used the [Emim][OAc] to dissolve chitosan. After a solvent exchange with water, the chitosan solution was prepared into a brimonidine tartrate (BT)-loaded chitosan film for loading the antiglaucoma drug. The chitosan-BT film was transparent and stable with good adhesion and biocompatibility. During the dissolving process, the hydrogen bonding network of the chitosan was destroyed by the IL to release the amines involved in the hydrogen bonding. The solvent exchange step led to the protonation of chitosan, which in turn promoted the solvation of the chitosan in clear water. This method eliminated the solvent effect and skipped the lengthy post-dissolution process. Chitosan has one amino group and two hydroxyl groups in the repeated hexosamine residue. Under suitable conditions, chitosan can be used as a natural amino functional polymer to fix CO_2 [65]. However, traditional acid hydrolysis makes the H^+ combine with the amino group, limiting the application of chitosan in this field. Xie et al. [66] used [Bmim][Cl] to dissolve chitosan. By destroying the intermolecular hydrogen bonds, the IL completely exposed the amino group in the chitosan without the constraint of the hydrogen bonds. This was conducive to the participation of the amino group in the reaction. The reaction process is shown in Fig. 6. The mild dissolution of IL overcame the shortcomings of traditional solvents. It was proven that

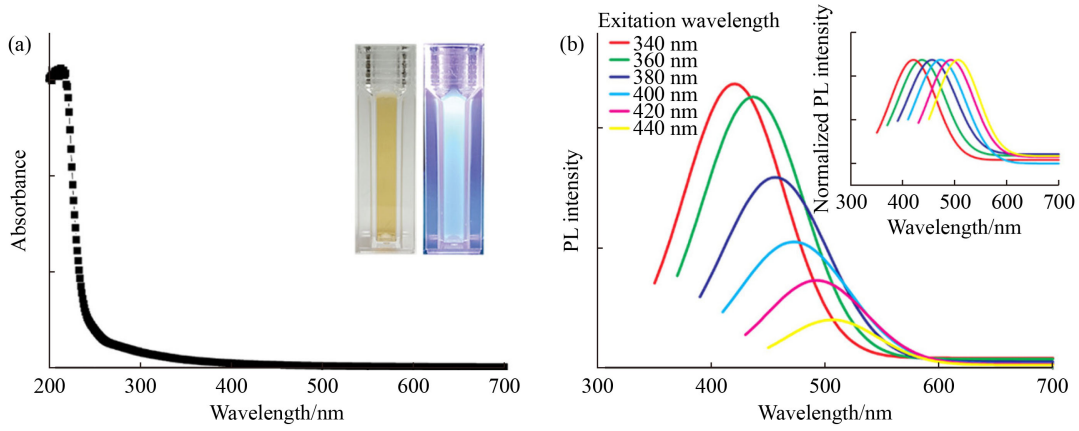

Fig. 5 (a) Absorption spectra of RCCDs (Inset: optical images under daylight (left) and 365-nm UV light (right)); (b) excitation-dependent emission photoluminescence (PL) spectra of RCCDs at different wavelengths, increasing from 340 to 440 nm with 20 nm increments (Inset: normalized PL emission spectra). Reprinted with permission from Ref. [59], copyright 2018, American Chemical Society.

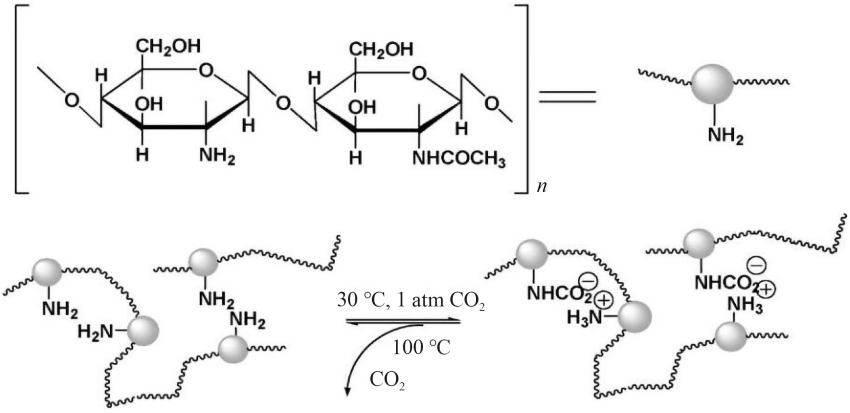

Fig. 6 Reversible covalent chemistry between CO_2 and amines linked to the chitosan polymer chain. Reprinted with permission from Ref. [66], copyright 2006, Royal Society of Chemistry.

almost all amino groups of chitosan were involved in CO_2 capture. Thus, chitosan had high adsorption and desorption rates and could be used for CO_2 adsorption/desorption recycling.

At present, ILs have been used as solvents to prepare a series of advanced biomass materials. However, for practical large-scale applications, ILs still have certain defects. For example, some ILs are expensive (e.g., [Bmim][OAc]), some are corrosive, and some have high viscosity (e.g., [Bmim][Cl]). To alleviate such defects, researchers have proposed using ILs together with organic solvents for the pretreatment of biomass [67]. Some inexpensive organic solvents such as dimethyl sulfoxide (DMSO) and *N*,*N*-dimethylformamide have been used as co-solvents for ILs [68]. The addition of a co-solvent can reduce the amount of ILs, thereby reducing the cost of pretreatment. In an experiment for making organic solvent nanofiltration membranes with date seeds, Alammar et al. [69] used a mixture of [Bmim][OAc] and DMSO as a solvent to successfully dissolve the date seeds. As a non-edible by-product, date seeds are produced at more than 1 million tons per year and are a very rich source of lignocellulos [70]. The robust and insoluble nature of lignocellulosic biomass make them a promising candidate for fabricating advanced materials for application in harsh environments. Hardian et al. [71] used the same binary solvent system to dissolve a cellulose−chitosan mixture and cast the solution to prepare organic solvent nanofiltration membranes. With the addition of the co-solvent, the viscosity of the IL decreased and the interaction between IL and biomass was enhanced; meanwhile, the co-solvent did not participate in the interaction, so the mixed system promoted the dissolution of the biomass to a certain extent [72]. As environmental substitutes for petroleum-based membranes, these biofiltration membranes have strong separation properties, chemical and thermal stability, and biodegradability. The combination of cheap organic solvents and ILs makes the production process cost-efficient.

2.3 Dispersion of ILs in advanced biomaterials

Biomass nanomaterials are commonly used as fillers to enhance the properties of composites, such as their mechanical, flame-retardant, antibacterial, and thermal stability properties [73–75]. Biomass nano-fillers are considered as the most promising alternatives to many carbon-based or metal-based nano-fillers [76]. However, biomass nanomaterials often cause composites to perform poorly owing to excessive agglomeration [77]. To solve this problem, conducting ultrasonic dispersion or adding dispersants are usually employed in practice [78,79]. Considering the excellent abilities of ILs to dissolve biomass, they have been used to facilitate the dispersion of biomass nano-fillers and thereby improve the uniformity of composites. Generally, there are two ways commonly used to perform this task, based on whether the ILs directly or indirectly disperse the biomaterials.

For direct dispersion, Wang et al. [80] interpreted the dispersion effect of IL [Emim][OAc] on chitin nanowhiskers (CNW) as the anion of the IL interacting with the polar partial domain of the CNW and intercalating between the chitin molecules (Fig. 7). This allowed a high concentration of nanofiller to be dispersed in the polymer, thereby improving the mechanical properties of the composite. The dispersion of cellulose nanocrystals (CNCs) in ILs was also proven; it is caused by the blocking effect of the ILs between CNCs and inhibits the formation of intermolecular hydrogen bonds in the CNCs [81].

In indirect dispersion, ILs indirectly promote the dispersion of other functional molecules in a composite by interacting with the matrix of the biomass. For instance, enabled by IL 1-butyl-3-methylimidazolium bis (trifluoromethanesulfonyl)imide ([Bmim][NTf_2]), Koga et al. [82] successfully dispersed the photochromic molecule diarylethene uniformly on the surface of a cellulose substrate and developed a stimulus-responsive colored paper device. The cations of [Bmim][NTf_2] showed a hydrogen bond donor capacity to form hydrogen bonds with the abundant hydroxyl groups of

cellulose, thereby promoting the diarylethene dissolved in the IL onto the cellulose substrate surface (Fig. 8). Compared with the paper device without [Bmim][NTf_2], the IL-based device exhibited a more uniform color and more vivid photochromic properties.

In practical applications, ILs are most commonly used as a dispersant for carbon nanotubes (CNTs) and graphene (Gr) [83,84]. Coupled with the effective dissolution of biomass by ILs, the application of bio-composites containing CNTs and Gr in many conductive materials has been further broadened. CNTs tend to agglomerate owing to their high aspect ratio and surface energy. The sporadic scattering affects the conductivity of the CNTs [85]. Gr requires dispersants for decomposition owing to its inherent insolubility, smooth surfaces of its atoms, and a strong tendency to aggregate [86]. Therefore, providing good dispersion of the CNTs and Gr is a key challenge in the application of polymer Gr composites [87]. One study concluded that ILs with a large dielectric constant could effectively shield the π–π interactions between CNTs, enabling the CNTs to be well-dispersed in the ILs [88]. Singh et al. [89] used an IL as a dissolving agent for chitin and dispersant for CNTs, respectively. The biocompatibility of the chitin and conductivity of the CNTs were used to prepare biocompatible scaffolds for nerve growth. Wu et al. [90] developed a temperature-pressure sensor for composite fabrics based on functionalized silk coiling and weaving techniques. The stacking of CNTs with a conductive material led to an increase in temperature, enhanced transmission of electrons between the CNTs through contact points, and a decrease in resistance, ultimately resulting in a decrease in temperature sensitivity. The addition of the IL [Emim][NTf_2] dispersed the CNTs to a certain extent, changing the charge transfer mode of the CNTs from direct contact to an IL-mediated charge transfer (Fig. 9), thereby increasing the temperature sensitivity of the sensor. Zhou et al. [87] used 1-ethyl-3-methylimidazolium diethyl phosphate ([Emim]

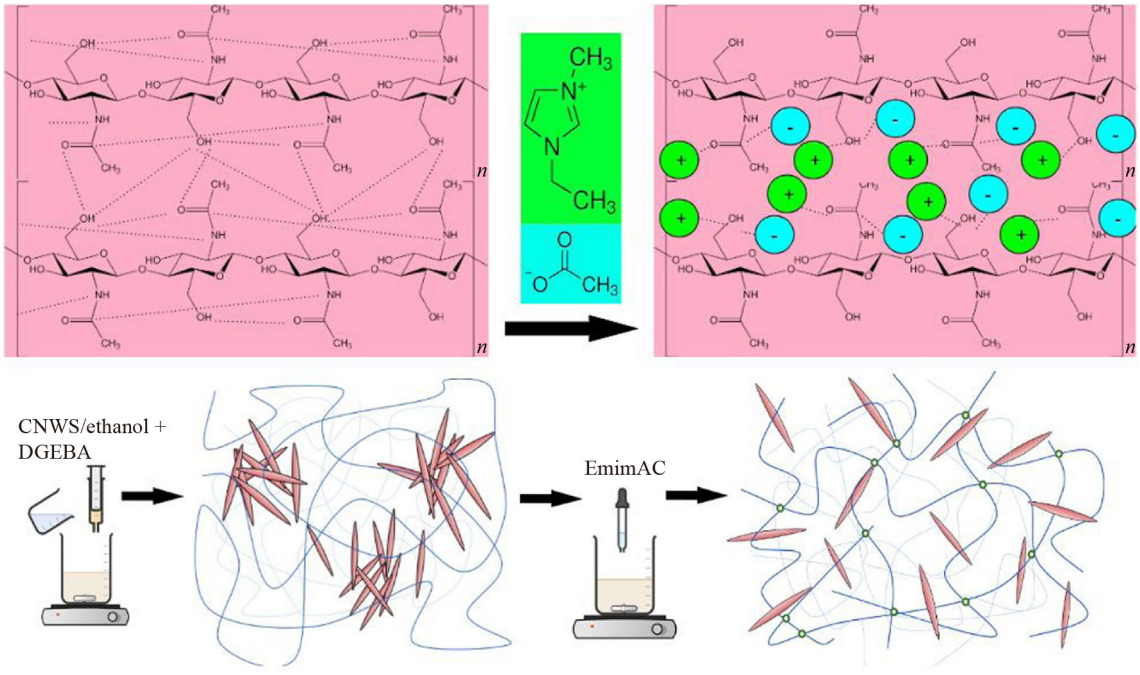

Fig. 7 Chemical scheme explaining the dispersing role of the IL. Reprinted with permission from Ref. [80], copyright 2020, Elsevier.

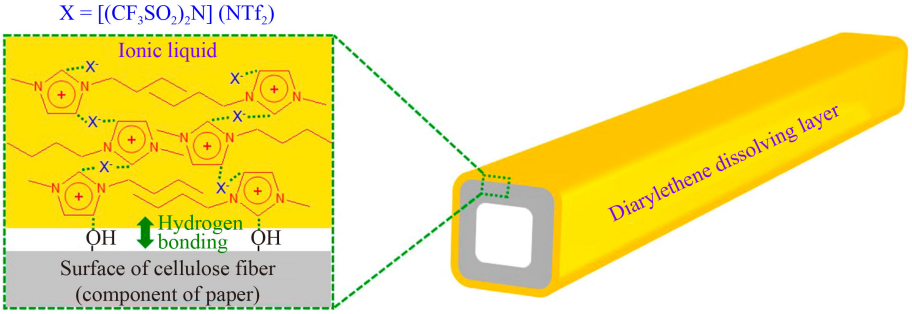

Fig. 8 Schematic diagram of photochromic diarylethene dispersed on cellulose pulp fibers by IL [Bmim][NTf_2]. Reprinted with permission from Ref. [82], copyright 2017, American Chemical Society.

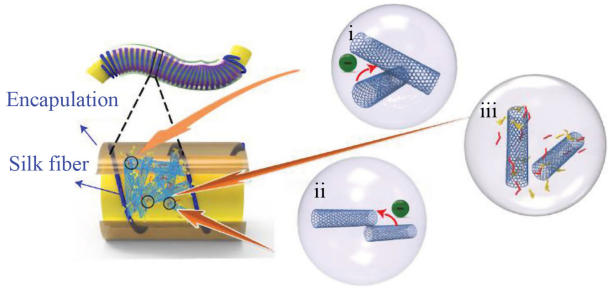

Fig. 9 Three different conducting mechanisms for composite CNT temperature sensors. (i–iii) Three different electron/ions transport modes in CNT and [Emim][NTf_2] composite sensing materials: (i) the transport of charges via the body-body contacts among CNTs; (ii) the transport via the end-to-end contacts of CNTs; (iii) the transport via ILs. Reprinted with permission from Ref. [90], copyright 2019, Wiley.

[DEP]) to disperse graphene nanoplates, multi-walled carbon nanotubes (MWCNTs), and dissolved cellulose to prepare a composite conductive film. In general, when the limited dispersion capacity of ILs limits the amount of filler added, researchers have found that binary IL systems can be used for larger-scale dispersion. Wang et al. [91] found that the conductivity of a membrane increases with the increase of MWCNTs during preparation of cellulose/MWCNTs composite membranes. However, excessive MWCNTs will cause the [Emim][DEP] originally used to dissolve cellulose in the system to become insufficient for completely dispersing the MWCNTs, making it difficult to form a film. Nevertheless, further improved dispersion of MWCNTs can be achieved by adding another IL, i.e., 1-hexadecyl-3-methylimidazolium bromide ([C_{16}mim][Br]). [C_{16}mim][Br] is a type of amphiphilic IL whose hydrophobic structure interacts with MWCNTs and hydrophilic structure interacts with [Emim][DEP], forming a double-layer cylindrical shell structure outside the MWCNTs and effectively preventing their aggregation. Thus, it plays positive roles in regulating the conductivity and tensile strengths of conductive materials.

3 ILs as structural components to enhance advanced performances of biomass materials

In addition to using ILs as solvents for biomass processing, they can also be integrated into the system to interact with the biomass and produce biomass-based advanced functional materials. In this context, ILs can be fixed in the materials by a variety of methods, such as *in situ* introduction by the sol–gel process or immersion of a previously prepared system in ILs [92]. The addition of the ILs not only adjusts the physicochemical properties of the biomaterials for mission-specific applications but also provides other attractive features such as high electrical conductivity, wide electrochemical windows, thermal stability, bactericidal capabilities, and catalytic properties [93–95]. However, most of the current focus is on ionogels and capacitor electrolyte applications. The combination of IL and biomass materials represents a promising research direction with undeveloped opportunities.

3.1 ILs as structural components to enhance performances of biomass-based ionogels

As promising functional materials, ionogels have attracted remarkable interest for various applications including flexible electronics, energy storage, and biomedical applications. They can be prepared by immobilizing ILs in a gel network. Owing to the presence of the ILs, ionogels offer additional advantages over conventional hydrogels [28]. In addition, the solvent capacity of the ILs allows for the addition of biomass to prepare sustainable biomass ionogels. Biomass-based ionogels have additional advantages of degradability, low toxicity, and biocompatibility, and are currently used in antibacterial dressings [94], biosensing [96], drug delivery [97], harmful gas adsorption [98], and many other fields.

3.1.1 Antibacterial dressing

Skin wounds are vulnerable to bacterial attacks causing infection. A bacterial infection is one of the most common clinical diseases; it can endanger human health and even threaten human life [99]. The ideal wound dressing should prevent further wound damage and infection and accelerate the healing process [100]. A hydrogel is a potential candidate for a wound dressing owing to its unique characteristics, such as its sufficient flexibility, elasticity, biocompatibility, high water content, and high sensitivity to the physiologic environment [101]. Different types of antibacterial agents including antibiotics, metal and metal oxide nanoparticles, and ILs have been integrated into hydrogels to enhance the antibacterial effects [102–104]. However, the development of drug resistance owing to the inappropriate use of antibiotics remains a serious challenge to public health, and the impacts of nanoparticles on the immune system and toxicity caused by their proliferation and degradation have not been clarified [105,106].

Current studies have found that certain ILs have good antibacterial effects on common bacteria, cocci, and fungi, but have good biocompatibility with the normal cell structure [107]. First, based on the electrostatic interaction, ILs are adsorbed on the negatively charged cell membrane. Then, the interaction between the ILs and membrane phospholipids leads to the disintegration of the phospholipid bilayers, resulting in irreversible damage [106]. This damage changes the potential, fluidity, viscoelasticity, and other properties of the membrane. It

has a serious impacts on the membrane functions, such as the molecular transport, recognition, migration, adhesion, and mechanical transduction [108]. According to the bactericidal characteristics of ILs, Li et al. [94] prepared an antibacterial wound dressing based on 1-vinyl-3-butylimidazolium ([VBIm][Br]) and COOH-modified gum arabic. The gum arabic provided the adhesion of the dressing and [VBIm][Br] acted as an antimicrobial agent bound to the hydrogel by covalent junctions. Antibacterial tests were performed on *E. coli*, *S. aureus*, and *C. albicans*. The untreated bacterial cell wall was smooth and intact, whereas after the ionogel treatment, the bacterial surface appeared folded and the bacterial morphology was significantly damaged (Fig. 10). This antibacterial activity can be interpreted as an electrostatic interaction between the imidazolium cationic groups in the IL and negative phosphoric acid groups in the microbial cell wall. The hydrophobic segment of the compound inserts into the hydrophobic region of the bacterial lipid membrane, resulting in membrane leakage and eventual microbial death [109]. Zhang et al. [22] designed and synthesized a novel lignin/polyionic liquid composite ionogel dressing, demonstrating good antibacterial activity and self-healing properties. In general, the polymerization of ILs results in the formation of ionic polymers called polyionic liquids or polymeric ILs (PILs). In most cases, PILs can provide the features of ILs with excellent adaptability, even better than ILs [110]. The positive ions of PILs destroy the phospholipid bilayers of bacteria and inactivate them [111]. The extensive physical and chemical effects of the formation of phenolic hydroxyl groups in the lignin are helpful to improving the mechanical properties of the ionogel [112]. In addition, owing to the existence of phenolic hydroxyl groups, methoxy groups, aromatic rings, and epoxy functional groups in the lignin network, the antioxidant performance of ionogel also improves by the addition of lignin [113]. However, with the increase of lignin, the positive ions of the PILs in the dressing are gradually neutralized by the negative ions of lignin, leading to a decrease in antibacterial activity. Therefore, it is necessary to design the appropriate ratio of lignin to PIL according to the desired mechanical properties, antioxidant properties, and bactericidal effect of the dressing.

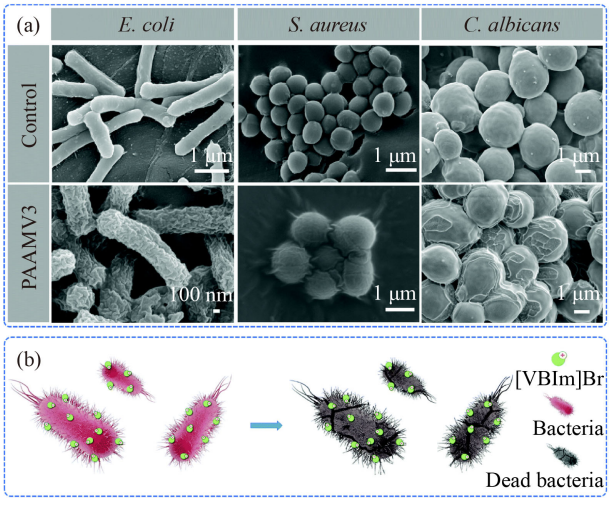

Fig. 10 (a) SEM images of *E. coli*, *S. aureus*, and *C. albicans* before treatment (control) and after treatment using the ionogel; (b) schematic diagram of the antibacterial mechanism of ionogel. Reprinted with permission from Ref. [94], copyright 2021, Royal Society of Chemistry.

In contrast to the traditional drug bactericidal mode, the physical bactericidal effect of ILs does not release any antibacterial agents into the body. This antimicrobial property opens up new vistas for overcoming the current challenges associated with combating antibiotic-resistant pathogens.

3.1.2 Biosensors

Hydrogel-based strain sensors have a massive advantage because of their attractive properties such as stretchability, skin compliance, and ion mobility [114]. However, compared to natural tissues, they have low mechanical strength, unstable sensing properties when swelling, and relatively low response rates owing to their loose cross-linked networks [115]. The addition of certain biological substances such as lignin [116], cellulose [117], and silk fibroin [118] has been used to improve these defects and give hydrogels better biocompatibility and biodegradability. Sensors based on these bio-conductive materials can be used to alleviate the electronic pollution caused by traditional pressure sensors. However, owing to the inevitable evaporation of water and low conductivity of the biomass, the durability and conductivity of hydrogel sensors can become severely hampered. At present, researchers are using the non-volatile properties and high electrical conductivity of ILs to improve the performance of hydrogels. Guan et al. [21] designed humidity sensors based on an IL-loaded biomass. The synergism of carrageenan and gelatin was used to construct a stable ionogel. The IL 1-ethyl-3-methylimidazolium chloride ([Emim][Cl]) was loaded into K-carrageenan and gelatin using an *in situ* cross-linking reaction. Carrageenan is a type of hydrophilic biopolymer extracted from red algae. There are many types of carrageenan depending on the degree of acidification. Among them, a K-carrageenan solution can form a hydrogel after cooling, i.e., the ideal material for a gel [119]. However, its main limitation is its low electrical conductivity [120], which can be improved by adding ILs. The cations and anions of ILs in an ionogel can move freely, providing the ionogel with good electrical conductivity [28]. In a comparison experiment, a sensor without an IL did not respond in a low-humidity environment (e.g., RH: 5%–35%), whereas the sensitivity of an IL-loaded sensor significantly improved with the increase of IL content (i.e., 0%, 4%, 8%,

12%, and 16%). This phenomenon might be attributed to the improved hydrophilicity and ionic conductivity of the ionogel.

3.1.3 Flexible wearable device

Wearables for daily healthcare and self-diagnosis are on the rise. Compared to organic or inorganic filler/elastomer composites, biomass hydrogels have been widely studied in the field of wearable devices in recent years owing to their good mechanical properties, stretchability, and biocompatibility. However, it remains a challenge to provide self-healing without the help of external forces [121]. Current studies showed that the self-healing properties of hydrogels can be improved by integrating dynamic covalent bonds (e.g., Schiff base bonds [122], borate ester bonds [123], or imine and acyl hydrazones bonds [124]) into the hydrogel network. Functional groups of certain biomass, such as the hydroxyl and amino groups on chitosan [125] and hydroxyl groups on cellulose [126], can be used as reactive functional groups to participate in the formation of dynamic covalent bonds. Yao et al. [96] alkylated IL 1-vinylimidazole with 4-(bromomethyl)phenylboronic acid (PBA) and introduced the PBA group to design a PBA-IL. An ionogel was prepared in a system where the poly-acrylamide (PAM), PBA-IL, and CNFs coexisted (Fig. 11). The hydroxyl group of the CNFs combined with the PBA group to form a dynamic borate ester bond which could be dynamically rebuilt and autonomously healed after destruction. In addition, the noncovalent bonding (hydrogen bonds and ionic interactions) between the CNFs and PBA-IL endowed the ionogel with better mechanical properties. The prepared ionogel had remarkable tensile, self-healing, and electrical conductivity properties, allowing it to be used as a wearable sensor to detect and distinguish various human movements in real-time with high sensitivity, stability, and repeatability.

3.2 ILs as structural components to enhance performances of biomass-based supercapacitors (SCs)

The porous carbon materials formed by biomass carbonization have been paid wide attention by industry owing to their abundant resources, low cost, and large specific surface area. A considerable number of studies have used physical and chemical methods to prepare porous carbon from biomass such as bamboo [127], pine [128], and algae [129], e.g., as adsorption materials and carbon electrode materials. ILs have attracted great attention in the field of electronic device manufacturing owing to their high ionic conductivity, wide electrochemical window, and excellent electrochemical stability [130]. SCs with excellent electrochemical performance are considered as potential energy storage devices for increasing power demands [131]. The specific surface area and porosity of the electrode materials affect the capacitance of SCs [132]. Biomass conduit and screen channels contribute to the carbonization process to form additional voids and are good carbon electrode materials. The electrochemical window is mainly determined by the electrolyte as an important part and ion source of the SC. The wide electrochemical windows and high electrochemical stability of ILs meet the requirements for SCs for high-performance electrolytes [133]. SCs based on IL electrolytes and biomass-based electrode materials have broad prospects for green energy storage applications. Dhakal et al. [134] prepared an SC using natural lotus calyx as a porous carbon electrode material and the IL 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide ([Emim][TFSI]) as an electrolyte. The surface area of the lotus calyx carbon electrode was 798 $m^2 \cdot g^{-1}$ and the specific capacitance at 1 $A \cdot g^{-1}$ was 223 $F \cdot g^{-1}$. It also had very high cyclic stability. After 50000 charge-discharge cycles in the KOH electrolyte, its capacity level remained at 97% of the initial amount. The IL electrolyte promoted the energy density of the SC to approximately

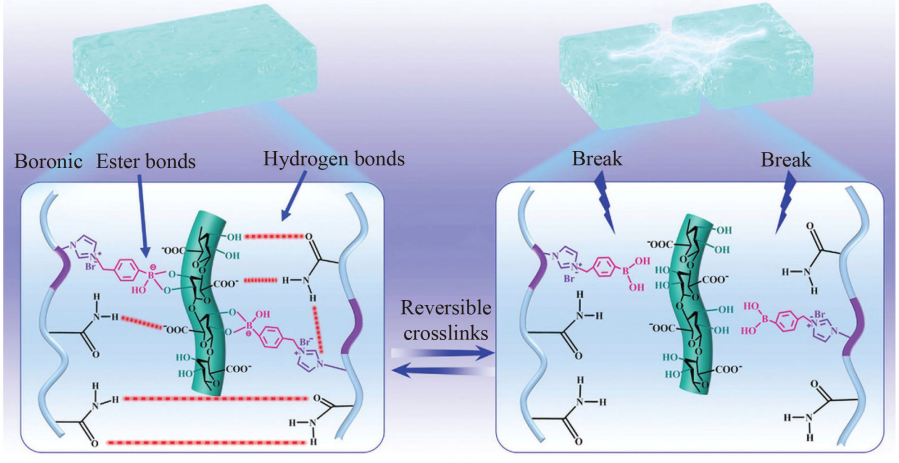

Fig. 11 Schematic diagram of the self-healing mechanism of the PAM/PBA-IL/CNF ionogel. Reprinted with permission from Ref. [96], copyright 2022, Wiley.

10-fold of that of the water-based electrolyte (Fig. 12). Considering the potential toxicity of the cations in certain ILs, Migliorini et al. [93] reduced the amount of ILs and used a naturally derived ionogel made from 2-hydroxyethyl cellulose mixed with the IL lactate choline as an electrolyte. This biomass-based electrolyte increased the biocompatibility and degradability of an SC, showing promise as a biodegradable energy storage component in wearable and environmental sensors.

3.3 ILs as structural components to enhance performances of biomass-based catalysts

In the past few years, research on the capture and utilization of CO_2 into valuable chemicals including methanol, urea, lactones, various heterocycles, biodegradable polymers, and carboxylated structures has increased exponentially [135]. However, the carbon in CO_2 is in the most stable valence state, and thus CO_2 is a very stable molecule. Therefore, catalysts are commonly used to improve the conversion efficiency of CO_2, e.g., using metal−organic framework-based materials [136], enzymes [137], and ILs [138].

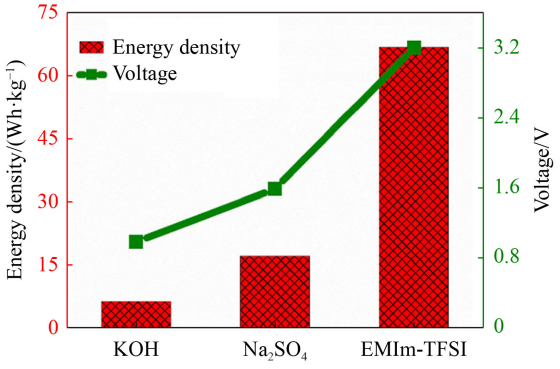

Fig. 12 Electrochemical voltage and energy density of different electrolytes. Reprinted with permission from Ref. [134], copyright 2022, Elsevier.

ILs are considered as promising media for CO_2 processes not only for efficiently absorbing CO_2, but also for smoothly catalyzing CO_2. As versatile catalytic systems for CO_2 conversion, ILs have advantages such as realizing a single-component homogeneous system without an additional co-catalyst, meeting the requirements of high conversion and high selectivity under mild solvent-free conditions, and achieving the catalytic reaction without the presence of expensive transition metals [135]. In addition, ILs can be immobilized onto inorganic carriers or organic functional polymers with large surface areas for easy separation, including porous silica [139], graphene [140], chitosan [141], and carboxymethyl cellulose [142].

Although immobilized ILs provide excellent recyclability, their catalytic performance is generally unsatisfactory relative to those of bulk ILs [143]. To solve this problem, Sun et al. [141] used chitosan-supported IL 1-ethyl-3-methyl imidazoliumhalides ([Emim][Br]) as catalysts for a CO_2 cycloaddition reaction. In the heterogeneous catalytic system, in addition to the catalytic activity provided by the anions of the IL, the hydroxyl groups of the chitosan provided effective cooperative sites in accelerating the ring-opening of the epoxide, and the tertiary nitrogen atom of the chitosan coordinated with the CO_2 reversibly to activate the CO_2 (Fig. 13). The synergistic catalysis of the hydroxyl and tertiary amine groups in the chitosan made up for the weakening of the catalytic performance of the heterogeneous catalytic system. Under the same reaction conditions, the catalytic performance of the synthesized CS-[Emim][Br] heterogeneous catalyst was similar to that of the homogeneous [Emim][Br] catalyst. The catalyst was easy to recover and had high activity and selectivity after being reused five times.

Wu et al. [142] chose protonated carboxymethyl cellulose (HCMC) with high thermal stability and rich hydroxyl and carboxyl groups as biomass carriers. IL

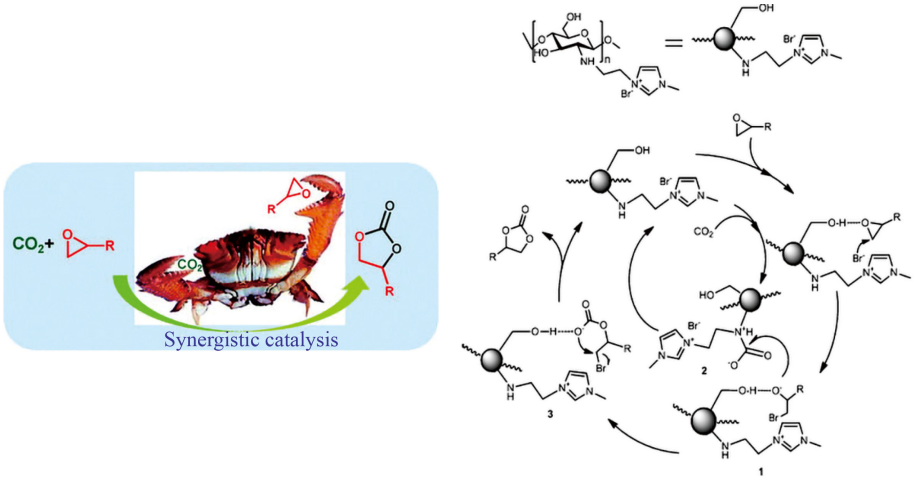

Fig. 13 The proposed mechanism for CS-[Emim][Br] catalyzed reaction. Reprinted with permission from Ref. [141], copyright 2012, Royal Society of Chemistry.

1-hydroxypropyl-3-*n*-butylimidazolium chloride ([HBim][Cl]) and Lewis acid ($NbCl_5$) were immobilized on HCMC by co-impregnation. The activation of epoxide molecules by a strong Lewis acid site (Nb site) and the hydroxyl group of the HCMC cooperated with the ring-opening effect of Cl^- to make the reaction proceed smoothly. Notably, the catalyst could be continuously operated in a continuous fixed-bed reactor for over 100 h, showing high activity and catalytic stability.

4 ILs as modifiers for modifying advanced biomass materials

Chemical modification is an attractive way to expand the upgrading use of biomass for materials by altering or enhancing certain important functions of the biomass [144]. The modified biomass can be further used to produce advanced functional materials with designability [145]. Generally, the chemical composition of a biomass is complex and usually has multiple reaction sites, such as the 2, 3, and 6 hydroxyls of each glucosinyl ring of cellulose [146], acetylamino of chitin, and amino group of chitosan [147] (Fig. 14). Chemical modifications with new functions such as esterification, amination, sulfation, acetylation, and phosphorylation can be imparted to biomass [148]. These modifications are used to improve the functional properties, such as the solubility, hydrophobicity, flame retardancy, adsorption, and antibacterial properties. However, the strong intermolecular hydrogen bond network has seriously hindered the evolution of certain biomass. The low solubility of biomass in common solvents has led to the chemical modification of biomass by means of heterogeneous systems in industry. These types of methods have many problems, such as uneven substitution, serious side reactions, and biomass degradation [146]. New solvent systems (such as those based on ILs) can now be used to circumvent these obstacles. Many studies have shown that ILs can be used as reaction medium to reach a homogeneous system in the process of biomass modification [144,149]. However, the study of ILs as both solvents and modifiers for biomass has not received widespread attention.

Based on the adjustable structural characteristics of ILs, it is easy to design different anion and cation combinations [150], or to introduce functional groups to modify the ionic structure [151]. ILs with specific charge groups or functional groups cannot only dissolve biomass but can also introduce groups to improve or enhance the properties of certain biomass. The flammability of cellulose involves safety hazards in transportation and operation, reducing its commercial feasibility [152]. The main method for forming a cellulose flame-retardant material is to functionalize the cellulose with nitrogen or phosphorus [153]. Al Hokayem et al. [154] reacted with cotton cellulose hydroxyl groups by using two phosphate-based ILs, 1,3-dimethylimidazolium methylphosphonate ([Dimim][(MeO)(H)PO_2]) and 1-ethyl-3-methylimidazolium methylphosphonate ([Emim][(MeO)(H)PO_2]), resulting in the phosphorylation of hydroxyl groups of cotton cellulose. A flame-retardant functional cotton fiber was obtained after the phosphorylation modification. With the increase in phosphorus content, the heat release of the cotton cellulose decreased, the carbonization degree increased, and the flammability significantly decreased. Considering the extraction rate and purity of biomass polymers, Nishita et al. [155] directly prepared flame-retardant thermoplastics using a hydroxyl phosphorylation of plant biomass (bagasse, cedar, and eucalyptus) via ILs. Notably, the hydroxyl substitution rate of biomass by this method was less than 14%, and most of the remaining active hydroxyl groups retained the potential for the further functionalization of biomass.

Deep eutectic solvents (DESs) are generally considered as a class of ILs because they share many of the same general characteristics, including high thermal stability, low volatility, low vapor pressure, and adjustable polarity [156,157]. DESs comprise mixtures of a hydrogen-bonded donor (HBD) and hydrogen-bonded receptor (HBA) bound strongly to each other through hydrogen bonds [158]. Similar to ILs, DESs with different properties can be obtained through the combination of

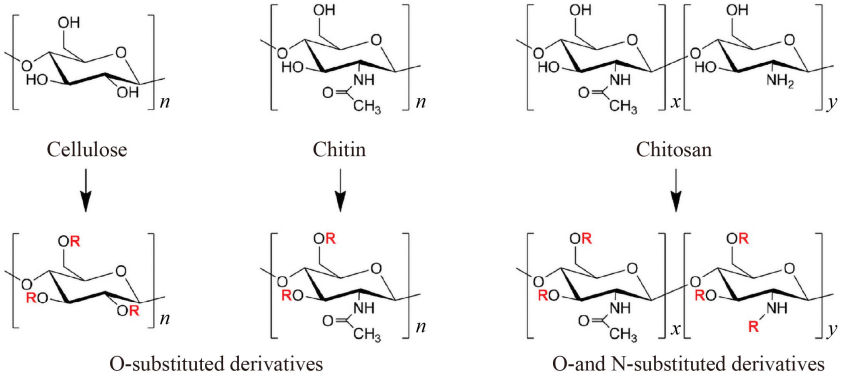

Fig. 14 Molecular structures of cellulose, chitin, and chitosan and their potential reaction sites for chemical modification. Reprinted with permission from Ref. [147], copyright 2019, Wiley.

different HBDs and HBAs. DESs with various functional groups and modification abilities can be used as modifiers to impart better properties to a material [159]. For example, researchers developed a way to use sulfonated CNFs to remove heavy metals from wastewater [160]. Traditionally, sulfuric acid is often used to obtain CNFs with a sulfate group. However, this highly acidic environment will lead to the hydrolysis of the cellulose [161]; the corresponding low sulfate content will affect the decontamination effect of the CNF [162]. At present, reactive DESs based on amino sulfonic acid and urea are used as possible sulfating agents for cellulose [163]. A significantly high sulfate group content can be obtained by using DESs with a moderate amount of sulfated chemicals under external solvent-free conditions. In the process of preparing CNF from carboxylic acid-based DESs, the hydroxyl group of cellulose is esterified with the carboxyl group in the DESs. As hydrophobic functional groups, the ester bonds can improve the dispersibility and compatibility of the CNFs in other polymer matrices to enhance the properties of the composites [164]. Yang et al. [165] cationized cellulose monomers by using a choline chloride/urea DES to adsorb anionic reactive dyes in water. Under alkaline conditions, the chlorinated part of chlorocholine chloride in the DES was nucleophilic with the alkali-activated hydroxyl groups of the cellulose to bind the trimethylammonium groups to the cellulose (Fig. 15). This DES-modified cellulose monomer exhibited a strong adsorption capacity and could bind quickly and strongly to anionic dyes. In addition, it maintained excellent performance with high adsorption capacity and fast adsorption/desorption rate during five consecutive cycles.

5 Conclusions and perspective

In this review, the multiple roles of ILs in the synthesis and application of advanced biomass materials were analyzed by illustrating a variety of cases. The first part introduced the solvation roles of ILs in the synthesis of advanced biomass materials. ILs can extract target components from the original biomass in a pretreatment process by effectively or partially dissolving the whole biomass. In addition, ILs can also uniformly disperse biomass materials to solve the problem of material performance degradations caused by filler agglomeration. The second part discussed the promising functions of ILs as structural components for adjusting the properties of advanced biomass materials. Specifically, the inherent properties of ILs including their high conductivity, wide electrochemical window, thermal stability, sterilization, and catalytic properties, enable biomass materials with the specialized functionalities required for applications of SCs, antibacterial media, biosensors, and biobased catalysts. The third part summarized the chemical modifications of biomass by ILs to endow or strengthen various functions of advanced biomass materials. A variety of functional ILs, including DESs, allow biomass to be chemically modified in homogeneous systems. The chemical modifications taking place on the active reaction sites of the biomass matrix, such as esterification, amination, sulfation, acetylation, and phosphorylation, generally afford the normal biomass new functions or facilitate the transformation of the normal biomass into advanced materials.

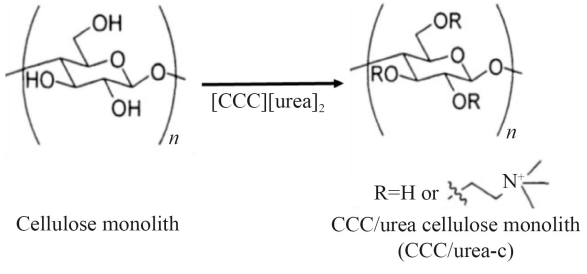

Fig. 15 Schematic diagram of the functionalization of cellulose monolith by choline chloride/urea DES. Reprinted with permission from Ref. [165], copyright 2019, Elsevier.

Despite the existing research progress, there remains significant room for further exploration. First, although the number of scientific publications describing IL concepts related to biomass materials is increasing annually, most of the studies are conducted on a laboratory scale. Further investigations on the large-scale synthesis of advanced biomass materials to gain insights into the scale-up effects, from the perspective of chemical engineering are still needed. Second, the toxicity and high prices of some imidazolium-based ILs would lead to strict requirements on the recovery rates of these ILs. However, owing to the relatively low vapor pressure of ILs, it is difficult to efficiently recover them by traditional vacuum distillation. To address this issue, membrane-based technologies such as pervaporation and electrodialysis can be considered, whose performances could be regulated by surface and/or structural modification. Third, as mentioned above, although the technical economy of the process can be improved through the use of co-solvents, the volatilization and pollution of the environment by these organic solvents cannot be ignored. Therefore, a high recovery rate as well as reuse of the solvents would be mandatory when conducting scale-up processing. Moreover, if a small amount of ILs exists in the materials as a residue, especially when the materials are used for medical applications, the cytotoxicity and histocompatibility of the ILs must be further evaluated and confirmed. Recently, some studies have used biomass-based ILs or natural ILs (e.g., choline-based ILs, amino acid-based ILs) as a green substitute for traditional ILs. However, the synthesis of these biomass-based ILs also consumes a certain amount of environmentally unfriendly and non-renewable reagents. As the research

on biomass-based ILs remains at its preliminary stage, there are many aspects that remain to be explored in the near future.

ILs are typically recognized as designable solvents, and their solvation effects have been extensively discussed in most current reports. As structural components, the applications of ILs have mainly been focused on ionogels, capacitor electrolytes, biosensors, and biobased catalysts. As modifiers, ILs can promote most of the chemical modification of biomass by affording a homogeneous system rather than directly reacting with the biomass. With significant breakthroughs in the fundamental and application research of ILs, there are many opportunities for further exploration. As ILs have great potential for design and functionalization by changing the cations and anions, it is suggested that future research should focus on both the micro-interaction mechanism of the biomass/IL system and structure-property relationships of ILs, so that the biomass processing can be theoretically guided toward both economy and efficiency. Also, expanding the application range of advanced biomass materials and exploring more possibilities of ILs for advanced biomass materials are complementary approaches.

Conflicts of interest There are no conflicts to declare.

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