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DOI

[10.1016/j.molliq.2024.126834](https://doi.org/10.1016/j.molliq.2024.126834)

Publication date

2025

Document Version

Final published version

Published in

Journal of Molecular Liquids

Citation (APA)

Mahmood, H., Khan, A., Shakeel, A., Uroos, M., Dhakal, H. N., Al-Khazaal, A. Z., & Moniruzzaman, M. (2025). Ionic liquids assisted processing of animal resources for fabrication of sustainable composite materials. *Journal of Molecular Liquids*, 421, Article 126834. <https://doi.org/10.1016/j.molliq.2024.126834>

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Review

Ionic liquids assisted processing of animal resources for fabrication of sustainable composite materials

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ARTICLE INFO

Keywords:

Animal derived polymers
Ionic liquids
Pretreatment
Biopolymer
Sustainable development

ABSTRACT

It is a global ambition to realize sustainable development goals (SDGs) by 2030 following the aspiring climate change and pollution alleviation, and thus, the notion of sustainable biorefinery has emerged as an indispensable asset for worldwide sustenance and economy. Animal derived materials are biopolymers that are produced in nature during the life cycles of animals and usually considered as versatile, biocompatible, non-toxic, stable, and cheap feedstock with a promising closed-loop life cycles in the industrial ecology of future materials. Such materials could produce a severe hazard to the environment and its occupants if not ditched appropriately. Conversely, animal derived wastes are complex molecules in which manifold structure and heterogeneous networks of inter- and intra-molecular clutches endure unresolved challenges to sustainable processing and purification of these valuable waste matrices into biomaterials with numerous applications. Utilizing ionic liquids (ILs), which are environmentally safe and recyclable alternatives to organic solvents, greatly facilitates the handling and processing of biopolymers obtained from animals. ILs have been used progressively for the use of biopolymers. Compared to customary techniques, ILs assisted processing of animal-based biopolymers is superior as ILs are noncorrosive, possess extremely low vapor pressure, exhibit high thermal solidity and superb dissolution capabilities under relatively moderate conditions. Currently, a broad spectrum of various techniques has been studied to further tune up the proficiency of ILs processing of animal derived biopolymers for sustainable product development. The primary aim of this review is to illustrate the latest advancements in technology wherein the ILs leverages as processing media for animal-based biopolymers for the manufacturing of a plethora of sustainable materials have been progressively recognized. In addition to examining how various ILs serve as solvents, reagents and/or (co)solvents, we also look at the larger process and system context in which the ILs are

Abbreviations: [C₂C₁IM][Ac], 1-ethyl-3-methylimidazole acetate; [C₄C₁IM][Br], 1-butyl-3-methylimidazole bromide salt; [DIPEA][Ac], diisopropylethylammonium acetate; [DIPEA][P], diisopropylethylammonium propionate; [C₂C₄N][Ac], dimethylbutylammonium acetate; [CH][Ac], Choline acetate; [CH][Prop], Choline propionate; [CH][Hex], Choline hexanoate; [C₄C₁IM][HSO₄], 1-butyl-3-methylimidazolium hydrogen sulphate; [C₂C₁IM][OAc], 1-ethyl-3-methylimidazolium acetate; [C₄C₁IM][Cl], 1-butyl-3-methylimidazolium chloride; [AllylC₁IM][OAc], 1-allyl-3-methylimidazolium acetate; [C₂IM][Cl], 1,3-dimethylimidazolium chloride; [C₁IM][Cl], 1-H-3-methylimidazolium chloride; [C₄C₁IM][OAc], 1-butyl-3-methylimidazolium acetate; [C₁IM-COOH][HCl], 1-carboxymethyl-3-methyl-imidazolium hydrochloride; TBPH, Tetra-butylphosphonium hydroxide; CH, Choline hydroxide; [DMEA][HCOO], Dimethylethanolammonium formate; [C₆C₁IM][Cl], 1-hexyl-3-methylimidazolium chloride; [C₁₀C₁IM][Cl], 1-decyl-3-methylimidazolium chloride; [C₄C₁IM][NCN²], 1-butyl-3-methylimidazolium dicyanamide; [C₃C₁₆P][Sal]p, Tributylhexadecylphosphonium salicylate; [AllylC₁IM][Cl], 1-Allyl-3-methylimidazole chloride; [VinylC₁IM][Cl], 1-vinyl-3-butylimidazolium chloride; [VinylC₄IM][Cl], 1-vinyl-3-butyl imidazolium chloride; [AllylC₁IM][Cl], 1-allyl-3-methylimidazolium chloride; [VinylC₁₂IM][Br], 1-dodecyl-3-vinylimidazole bromide; [C₄C₁IM][BF₄], 1-butyl-3 methyl imidazolium tetrafluoroborate.

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Received 8 August 2024; Received in revised form 23 November 2024; Accepted 30 December 2024

Available online 3 January 2025

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used. It is anticipated that the current review will infuse new thoughts and reconciliation in ILs-mediated processing of animal waste resources for a promising paradigm of sustainable biorefinery.

1. Introduction

Acquiring comprehensive and sustainable methods for producing, pretreating, extracting, purifying, and functionalizing diverse biodegradable materials and polymers obtained from conversion and processing of animal and other renewable resources is receiving much attraction today [1]. Over the years, some extraction technologies have been scaled up after process optimization. Separation methodologies for the extraction of particular components from biodegradable resources are generally categorized as chemical, mechanical, biochemical, high pressure, microwave, and ultrasound assisted. Extracting the target polymer using such technologies is complex, and achieving high yields and purity levels entails considerable labor. Generally, extraction and separation steps should be combined and completed within a single pot. Nevertheless, current extraction or separation techniques suffer numerous deficiencies including poor selectivity, low extraction efficiency, prolonged processing time and intensive energy demand provoking higher final product cost. In addition, severe operating conditions and noxious volatile solvents are usually used. These limitations have provoked the researchers to explore alternative, environmentally friendly separation and purification techniques, with the utilization of ILs emerging as a consideration due to their greener and more sustainable attributes [2] (see Table 1).

ILs are molten salts with structures that only have ionic compositions and with melting points less than 100 °C, while this temperature restriction is becoming more often regarded as illusive. In the presence of strong ionic electrostatic interaction, Ionic liquids typically contain structural components that shield the charged centers, thus inhibiting crystal formation and enabling them to achieve their characteristic low melting temperatures [3–5]. To create ILs with unique characteristics, it is advantageous to adjust the anion and cation structures to control the attributes of ILs [6]. In contrast to other conventional solvents that evaporate into the atmosphere and pose significant environmental risks, ILs exhibit low or even negligible vapor pressures, exceptional chemical, thermal, and electrochemical stability, and, in most cases, minimal flammability [7–9]. ILs could be a promising substitute for conventional solvents to avoid such environmental and process problems and historically are referred to as green solvents [10,11]. In the past two decades, ILs are frequently praised for their remarkable solvation capacity for a wide range of polymeric materials sourced from renewable and natural resources [12]. Additionally, they are recognized as excellent processing media for proteins, nucleic acids, enzymes, and other substances [13–15]. Furthermore, their exceptional solvation ability offer more efficient extraction of target components from biopolymer matrices allowing enhanced extractions [16,17]. ILs due to their unique solvating power assist in processing of animal resource by disrupting the hydrogen bond network of biopolymers. Ionic liquids increase the thermal stability of enzymes and provide greater reaction rate with shorter reaction time at high substrate concentration thus the extraction efficiency is improved compared to traditional solvents. Additionally, ionic liquids stabilize both enzymes and substrates hence increase product yield [14,15,18,19].

1.1. Ionic liquids and sustainable development goals

The concept of sustainable and environmentally friendly production has gained attraction in the twenty-first century, driven by concerns about climate change and effective energy management. In 2015, the United Nations introduced a new development strategy titled “Transforming Our World: The 2030 Agenda for Sustainable Development,” comprising 17 Sustainable Development Goals (SDGs) that address

various issues, many of which recognize the importance of sustainable and green chemistry and engineering. Organic solvents are indispensable for many extraction, refining, and cleaning processes, often necessitating large quantities to achieve sufficient product purity. Industrial-scale production of organic solvents is approximately 20 million metric tons annually [5]. The excessive use of hazardous solvents perpetuates negative environmental impacts and serves as a clear example of unsustainable practices [20,21].

The Sustainable Development Goals are a trade-off and a crucial instrument that many industrialized countries have used to eradicate and restrict the use of toxic organic solvents; one approach is to replace them with an ecologically benign solvent. Over a decade, ILs have had enormous impact on numerous applications due to their extraordinary attributes and ideally as a “green” processing media [22]. ILs have been applied in various fields, such as adsorbents [23], batteries [24], biomass conversion [25], lubricants [26], stationary and mobile phases of chromatography [27], nanocomposites [28], fluid additive [29], catalysts [30], CO₂ capture [31], metal separation [32] and pharmaceutical/biomedical applications [33,34]. Previous studies illustrated the possibility of these ILs for the extraction of biopolymeric components from renewable resources [35,36]. ILs must meet numerous requirements in order to dissolve biodegradable compounds effectively. Low melting point of ILs are indispensable to restrict biopolymer degradation; other beneficial qualities include high stability and storability, simple recovery and recyclability of ILs, non-toxicity, odorlessness, and biodegradability, and inexpensive synthesis. Several patents have been filed for the applications regarding extraction and processing of lignocellulose and other biopolymer matrices for value-added production [37]. ILs can offer a sustainable platform to improve chemical processes for efficient implementation of biorefinery and circular economy concepts by exploiting renewable materials along with abating energy demands, anticipating waste and reducing emissions [20].

1.2. Important parameters influencing processing of animal-based polymers using ILs

The large-scale utilization of ILs to process animal resources in the sustainable biorefinery context demands consideration of numerous parameters which unfortunately have rarely been explored. It's of paramount importance to optimize both the processing conditions and ILs properties for commercial employment of ILs to produce material products from animal derived polymers (see Table 2).

Cationic & Anionic structure of ILs: The dissolution capability of an IL strongly depends on the nature of its cationic and anionic groups. The anions that are strong hydrogen acceptors such as acetate, chloride, phosphate, bromide, sulfonate and imide and are believed to be the effective solvents for biopolymer processing [38]. Conversely, the peculiar conjunction of cation and anion as an electron donor–acceptor (EDA) complex strongly influences the interactive mechanism between the ILs and biopolymer matrix. Although cations also take part in the biopolymer dissolution process, the influence of cation structure on its dissolution power is more intricate. Indeed, both anions and cations disintegrate the complex hydrogen bonding network of biomolecules via intermolecular synergy [39].

Toxicity and corrosivity: Toxicity of ILs is of extreme interest particularly in commercial applications where human interact with ILs during the process thus triggering the safety concerns. Issues arise in situations where ILs are part of the end product or flee into the environment. The presence of some constituents of ILs in the soil in the vicinity of a landfill in Newcastle, England was found which could be

Table 1

ILs assisted extraction of chitin, chitosan, keratin, collagen and silk from different animal based resources.

Entry	IL	Raw Material	Extraction conditions	Loading (%)	Fabrication Technique	Yield (%)	Ref.
Chitosan							
1	[EMIM][Ac] [BMIM]Br	White shrimp shells (Penaeus vannamei)	110 °C 24 h	Mixing ratio 3:2	Binary IL mixture method	35.72	[72]
2	[Bmin]Ac	Crab, squid pen	100 °C	4 %	Solution casting	–	[168]
3	[DIPEA]Ac [DIPEA]P [DMBA]Ac	Shrimp shells	110 °C, 36 h 110 °C, 30 h 110 °C, 30 h	30 g	Acid/base neutralization	14.8 12.1 13.7	[73]
4	Chlorine hydroxide Glacial Acetic acid Propionic acid Hexanoic acid	Komagataella pastoris and Aspergillus niger	30 °C–100 °C	27.56 % acetic acid 34.03 % propanoic acid 49.56 % hexanoic acid	Acid/base neutralization	58.5–60.8	[74]
5	[C ₄ MIM]HSO ₄	Crustacean biomass	350 °C–375 °C 24 h	10	–	79	[75]
6	[AMIM]Br	crab shells	60–100 °C 24 h	2	–	71.2	[169]
7	[AMIM]Cl [BMIM]Br [EMIM]Br	crab shells	100 °C 24 h	17–22	Mixing 1-chloro-1 propene, 1-bromobutane and 1-bromoethane	73–87	[71]
Chitosan							
8	[BMIMAc]	Silk hydrogels	95 °C 24 h	Mixing ratio (50:50, 50:70)	Gel formation	–	[83]
9	[BMIMAc]	Commercial	100 °C 2 h	–	Hydrothermal treatment	–	[83]
10	[C ₄ MIM]HCOO	Commercial	150 °C	100 g/L	Anion exchange method	–	[112]
11	[C ₄ MIM]CH ₃ CH ₂ COO	commercial	150 °C	100 g/L	Anion exchange method	–	[112]
12	[C ₄ MIM]HOC H ₂ COO	commercial	150 °C	100 g/L	Anion exchange method	–	[112]
Keratin							
13	[EMIM]Ac	Wool	80 °C 24 h	25:75 and 75:25	Dissolving-regeneration	65–70 %	[170]
14	[BMIM]Cl	Wool, hair and chicken feathers	48 h	–	One step process	80 %	[171]
15	[AMIM]Cl	Turkey feathers	130 °C 10 h	5 g/1g	Acid base neutralization	51 %	[172]
16	[HMIM]HSO ₄	Wool	Microwave treatment	solid liquid ratio 1:10	One step synthesis	67–94.2 %	[173]
17	[EMIM]Cl	Yak hair	120 °C 11 h	8 wt%	–	90.8 %	[174]
Collagen							
18	[EMIM]BF ₄ [EMIM]Ac	Calf skin	10 °C 48 h	10 %, 30 % and 70 %	Dilution method	–	[175]
19	Tributyl methyl phosphonium methyl sulphate (PMS)/Tributyl ethyl phosphonium diethyl phosphate (PEP)	Commercial	4 °C 3 h	ILs:collagen 1:0.05, 1:0.5, 1:5, 1:10	Precipitation method	–	[176]
20	Imidazolium based ILs	Commercial	100 °C 12 h	–	Precipitation method	47–52 %	[177]
21	[BMIM]Ac	Carp fish	100 °C 12 h	Equal volumes of both IL: carp fish	Precipitation method	32 %	[39]
22	[EMIM]Br [EMIM]Ac [EMIM]Cl	Grass carp skins	30 °C 2 h	–	Gel method	–	[178]
Silk							
23	[BMIM]Br [BMIM]I [BMIM]BF ₄ [DMBIM]Cl	Cocoon silk	100 °C 2 h	10 % and 12.24 % (w/w)	Extraction method	–	[179]
24	Switchable ILs	B. mori silkworms	90 °C	100 g silk in 5 g molten IL	–	–	[180]

hazardous to human health [40]. A major challenge in this field is that only a limited number of researchers have the necessary expertise in both biological and chemical methods for toxicological studies. More efforts are imperative to understand the toxicity of ILs as processing solvents in the biocomposite industry [41]. Moreover, the corrosivity of ILs is a crucial economic factor when selecting construction materials for vessels intended for large-scale biopolymer processing. Halogen-free ILs may serve as suitable candidates for non-corrosive IL solvents [42].

Viscosity, melting point and thermal stability: Viscosity is another crucial parameter from practical perspective, as a higher viscosity of an IL necessitates a higher dissolution temperature and increased energy for mixing and fluid transport. Additionally, this may lead to other potential side effects such as the occurrence of undesirable side reactions. The issue can be tackled by mixing the IL with cosolvents including polyethylene glycol, DMSO etc. to diminish viscosity [43]. ILs possessing low melting point exhibit utmost performance in modification and processing of biopolymer matrices as low melting point promotes dissolution and handling of biopolymers [44]. Conversely, thermal stability of IL significantly impacts its recyclability which is a substantial parameter for a feasible process economic [45].

Presence of moisture and salt contents: ILs assisted processing of animal-based biopolymers is perceptive to the presence of moisture in

both IL and biopolymer. Indeed, water inhibits biopolymer dissolution in IL due to competitive hydrogen bonding to biopolymer microfibrils restraining dissolution [46]. The melting point and viscosity of IL are also remarkably affected by presence of residual moisture contents in it. Therefore, both ILs and biopolymer must be thoroughly dried prior to the dissolution process [47]. On the other hand, moisture-tolerant ILs have been studied and a substantial auspicious impact on lignin dissolution and depolymerization in IL has been reported [48]. The effect of presence of salts such as Na_2SiO_3 and LiCl during deconstruction of biopolymers and boosting selective dissolution in IL has also been revealed [49,50].

1.3. ILs for biopolymer processing: A new target with proactive approach

The number of commercial industrial applications for ILs is not yet keeping up with the rate of scientific discovery; this is typical for the emergence of most new technologies. The hype cycle of ionic liquids implies that the field maturity (i.e., plateau of productivity) would be attained in the few coming years [4]. Therefore, this is the ideal time to inquire about the structural requirements for choosing more greener ILs for lignocellulosic and animal derived waste biorefinery [18]. The life cycle assessment approach underscores the importance of considering

Table 2

ILs assisted processing of gelatin, alginate, and xanthan gum.

Sr #	IL	Biopolymer concentration	Functional additive(s)	Fabrication technique	Application	Ref.
Gelatin						
1	[BMIM][DCA]	9.2 wt%	Water	Simple mixing	—	[187]
2	[P44416][Sal]	1.45 wt%	Avermectin	Complex coacervation	Controlled release of pesticides	[188]
3	[AMIM][Cl]	18–22 wt%	Triglycidyl naringenin	<i>In-situ</i> cross-linking	Ionic skin and wearable sensors	[189]
4	[EMIM][Ac]	10 wt%	Methylglyoxal	Chemical cross-linking	Stretchable electronic devices	[190]
5	[VBIM][Cl]	74–75 wt%	Sodium Alginate	3D printing	Tissue-engineered bio-scaffolds	[191]
6	[VBIM][Cl]	5 wt%	Quaternized chitosan, decellularized extracellular matrix	3D printing	Skin tissue engineering	[192]
7	[BMIM][DCA]	10 wt%	4-cyano-4'-pentylbiphenyl (LC)	Film coating	Gas-sensing applications	[233]
8	[AMIM][Cl]	1.3 wt%	CsSnCl ₃ quantum dots (QDs)	<i>In-situ</i> method	—	[234]
9	[Ch][Sal]	10 wt%	Water	Simple mixing	Food packaging	[183]
10	[EMIM][C ₁₂ H ₂₅ SO ₄], [AMIM][C ₁₂ H ₂₅ SO ₄], [BMIM][C ₁₂ H ₂₅ SO ₄], [EtMIM][C ₁₂ H ₂₅ SO ₄]	1 wt%	Water	Simple mixing	—	[235]
Alginate						
11	[C ₁₀ MIM][Br], [C ₁₂ MIM][Br], [C ₁₄ MIM][Br], [C ₁₆ MIM][Br]	0.001–0.01 wt%	Water	Simple mixing	—	[207]
12	[1-(4-ethoxy-1-butan-1-yl)sulfonate]-3-methylimidazole bromide], (triethylammonium hydrogen sulfonate), [1-(1-propylsulfonate)-3-methylimidazole chloride], (1-heptyl-3-methylimidazole methanesulfonate)	5 wt%	Water, 5-fluorouracil, indomethacin	Dripping method	Drug delivery	[208]
13	[EMIM][BF ₄]	2.43 wt%	Multiwalled carbon nanotube (MWCNT)	Simple mixing and drying	Smart artificial muscle	[210]
14	[PC ₆ C ₆ C ₆ C ₁₄][Br]	2 wt%	Powder activated carbon (PAC)	Dripping method	Phenol adsorption	[213]
15	[Hmim][HSO ₄]	4 wt%	Chitosan, <i>N,N</i> -dimethyl chitosan	Precipitation	Pb(II) adsorption	[236]
16	[PC ₆ C ₆ C ₆ C ₁₄][Br]	2 wt%	Agar	Dripping method	Phenol adsorption	[237]
Xanthan gum						
17	[BMIM][Cl]	1 wt%	Water	Simple mixing	Valorization of recalcitrant polysaccharide	[221]
18	[VBIM][Cl]	0.6–3.1 wt%	Poly (ethylene glycol) diacrylate (PEGDA), water	Photo initiated radical crosslinking copolymerization	Pressure sensor	[222]
19	[ImSi][PF ₆]	4.75 wt%	—	Mixing and heating	Heterogeneous catalyst	[223]

the environmental impact of IL synthesis as a whole. Consequently, when establishing guidelines for environmentally friendly ILs, conducting thorough evaluation of IL synthesis and evaluating the sustainability implications of specific IL structures becomes imperative [18].

The modular design of ILs could allow proactively select structural components for pretreatment of biodegradable polymers that boosts environmental sustainability without surrendering process efficiency. Fortunately, atomistic molecular dynamics simulations have transformed into a crucial tool to afford a molecular scale intuition into the structure–property relationships and virtual design of new ILs primarily due to the rise in computational power, availability of massively parallel architectures coupled with force fields and advanced modeling methods [51]. Nevertheless, high proficiency and affordable cost to extract and process valuable materials from biological resources will always be imperative prerequisites [18].

This comprehensive review examines the utilization of ILs as dissolution media for a variety of animal-derived waste materials, facilitating pretreatment, extraction, processing, and purification of biodegradable polymers for a broad range of sustainable applications. Various biopolymer types are explored, encompassing polysaccharides like chitin, chitosan, and xanthan gum etc, as well as protein-based polymers such as silk, collagen, keratin, and gelatin. By surveying the diverse IL-mediated methodologies employed thus far, the review provides a succinct overview of the advancements made in employing ILs within biorefineries utilizing animal-derived waste. Additionally, it addresses recent developments and key challenges that remain unresolved in the processing and separation of biopolymer matrices sourced from renewable resources using ILs. Ultimately, this review aims to establish a robust framework that bridges fundamental research in the physical sciences, engineering, and environmental technology with applied research endeavors aimed at mitigating environmental hazards and fostering the development of truly sustainable industrial practices, with ILs serving as a prime example.

1.4. Ionic liquids assisted processing of animal resources for sustainable development

In recent years there has been a noticeable increase in consumer preference for natural ingredients in pharmaceutical, cosmetic, and nutraceutical goods (such as biopharmaceuticals and naturally derived antioxidants). Animal derived biopolymers are processed using a variety of solvents other than ILs. Acetic acid and hydrochloric acid are common acidic solvents used in the processing of chitin and chitosan because they boost solubilization by deacetylation [52,53]. Strong basic solutions and supercritical CO₂ are also among the ordinary solvents for biopolymer extraction and processing [54,55]. In some systems, urea-based solutions aid in breaking down the protein's hydrogen bonds promoting its dissolution [56]. Conversely, organic solvent-assisted processing has been employed to process animal derived polymers for textile and medicinal applications [57]. Biological methods utilizing enzymes are also practiced under minimum temperature requirements [58]. Traditionally, aldehyde-based tanning agents such as glutaraldehyde, vegetable tannins, and chromium salts have been utilized in the leather-making process [59]. Hydrolysis, denaturation, and the breakdown of glycosidic or cross-linked bonds are the usual degradation processes of animal derived biopolymers [60,61]. The complex 3D natural structures of animal based polymers is usually disintegrated by processes of hydrolysis, denaturation, or the breakdown of glycosidic or cross-linked bonds under conventional treatment mechanisms which lowers their molecular weight and transforms them into more soluble forms [62].

Conventional biopolymer extraction techniques from renewable raw material, on the other hand, exhibit numerous shortcomings, including poor efficiency and selectivity, prolonged residence time, high energy input, and generally employ hazardous and volatile compounds. ILs have emerged as one of the most extensively researched alternative

solvents for sustainable processing of biopolymeric ingredients sourced from animals and other biological origins [21]. ILs, along with their fluid mixtures containing water or other co-solvents, serve as effective dissolution media for solid–liquid extraction processes aimed at recovering valuable components [18]. This section provides insights into the applications of ILs in the extraction, separation, purification, and functionalization of biobased compounds obtained from animal resources. It offers a comprehensive overview of existing studies on the development of processes mediated by ILs (see Fig. 1).

2. ILs assisted processing of chitin

Chitin, the second most prevalent compound after cellulose, offers biocompatibility and biodegradability. Utilizing biological sources for biofuel and high-value chemicals is seen as an alternative to traditional energy sources. With the rise in aquaculture and population growth, chitin from food processing waste has significant potential. Despite annual production in the range of 1012–1014 tons, the global disposal of marine trash overlooks the value of chitin polymers. Unlike cellulose, chitin with nitrogen shows potential for applications like synthesizing nitrogen-containing compounds, offering economic benefits and energy savings. Investing in the sustainable extraction of this renewable resource is crucial for its varied industrial applications. Incorporating chitin into food could positively impact human health, highlighting its potential in functional food and human health applications (Fig. 2).

Chitin is obtained by the utilization of chemical, biological, and physical techniques either individually or in combination [64–66]. Physical methods are frequently employed to facilitate the extraction process [67,68]. These methods have been devised to overcome the constraints linked with rigorous acidic or alkaline techniques. On the other hand, the employment of an environmentally sustainable biological approach offers several benefits including the preservation of chitin structure and enhanced resource utilization. However, the consideration of extraction time and costs are crucial factors in the context of industrial manufacturing [64,69]. In order to enhance the output, it is imperative to prioritize elements such as appropriate bioreactors, proficient operations, and energy utilization [64,66,70]. Recently, there has been an increase in innovative extraction methods which include the use of environmentally friendly solvents like ILs [64,71].

Yinzhe Jin et al. investigated the application of a combination of two ILs for chitin extraction from shrimp shells, specifically [C₂C₁IM][Ac] and [C₄C₁IM][Br] [72]. Several ratios of these ILs were tested including 0:1, 1:1, 3:2, 4:1, and 1:0. Using a conductivity meter, a viscometer, and a network analyzer (PNA-L), the basic properties of the IL were



Fig. 1. Schematic representation that ILs can boost the sustainable biorefinery by enabling the benign processing of animal derived polymers.

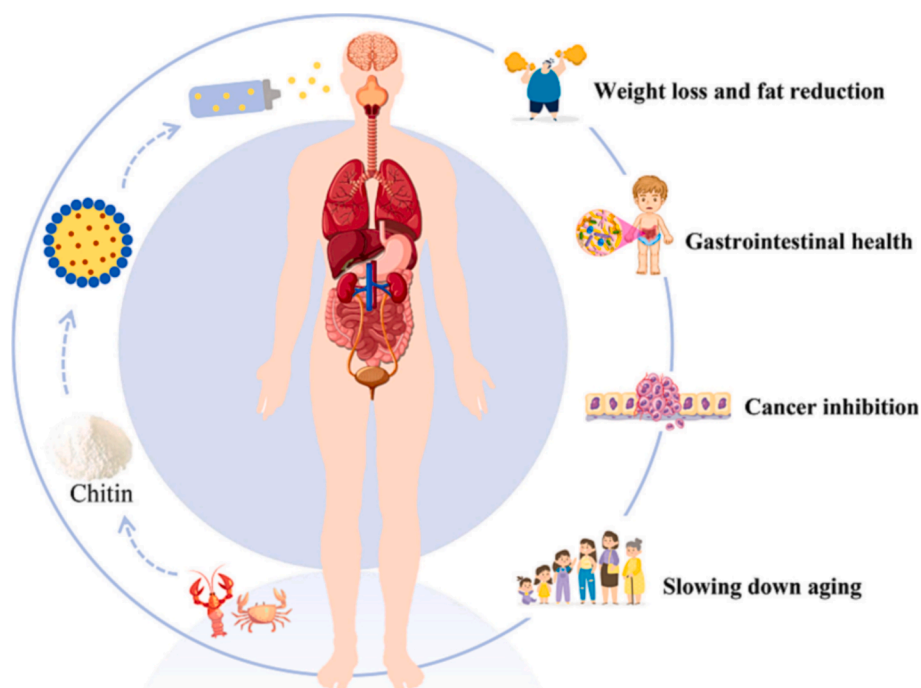


Fig. 2. Health beneficial effects of chitin on human body adapted from [63].

evaluated. Afterward, a comprehensive array of analytical approaches, such as Raman Spectroscopic analysis, Fluorescence Spectroscopic analysis, FTIR, DSC, AFM, TEM, SEM were applied to assess the properties of the derived chitin. This study marks the inaugural exploration of employing a binary ionic liquid mixture for chitin extraction from shrimp shells. Additionally, it demonstrates an inverse relationship between the viscosity and conductivity of ILs. The extracted chitin (3:2) exhibited absorption peaks in both IR and Raman spectra that were comparable to those observed in commercial chitin. When examined through AFM (Fig. 3), the sample exhibited a notable degree of crystallinity. Moreover, the chitin extracted using an ILs ratio of 3:2 displayed a finer and more uniform structure, resemblance to commercially

available chitin [72].

Ming Jer Lee et al. examined chitin production using ammonium-based ILs including [DIPEA][Ac], [DIPEA][P], and [C₁₂C₄N][Ac]. The obtained chitins were analyzed using SEM, FTIR, TGA, H NMR and XRD. Afterwards, the chitins underwent deacetylation to produce chitosan. The extracted chitins exhibited high extraction efficiency from shrimp shells as indicated by their low ash content, degree of deacetylation, and nitrogen content. These findings affirm the resemblance of the isolated product to commercially available chitin from shrimp shells [73].

Luísa A. Neves et al. investigated a novel environmentally friendly approach for dissolving the chitin-glucan complex (CGC) obtained from two fungal strains, *Komagataella pastoris* (CGCP) (KiOnutrime-CGTM)

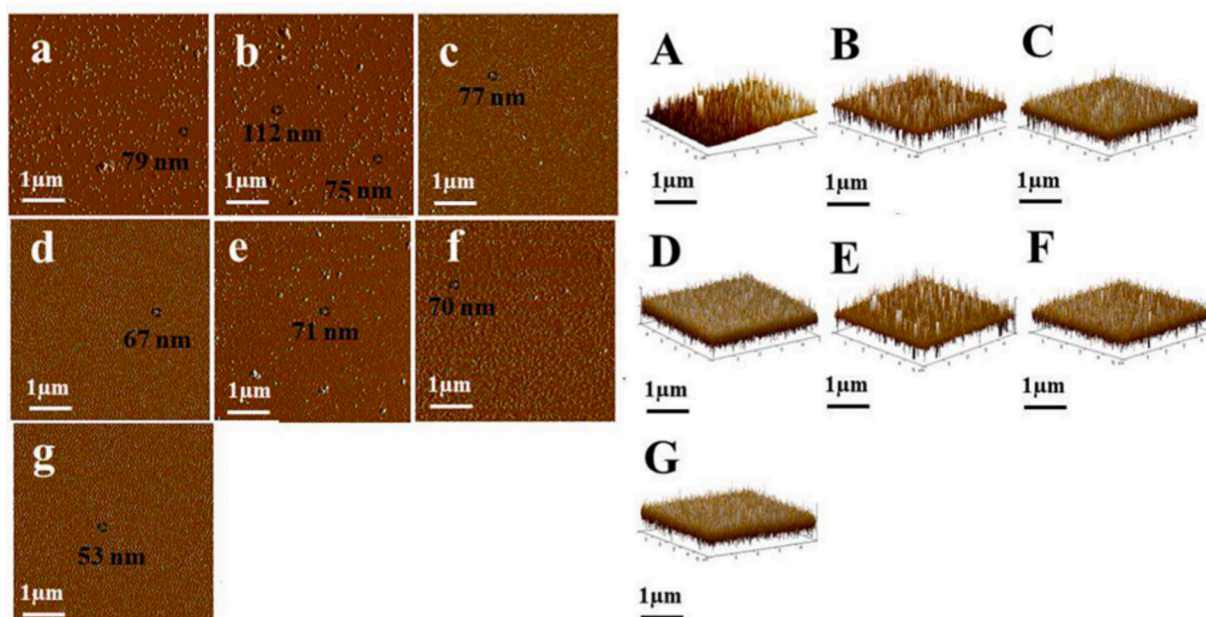


Fig. 3. Images obtained via AFM depicting Chitin (CT) and Polystyrene (PS). (a & A) Polystyrene, (b & B) Chitin (0:1), (c & C) Chitin (1:1), (d & D) Chitin (3:2), (e & E) Chitin (4:1), (f & F) Chitin (1:0), and (g & G) commercial chitin adapted from [72].

and *Aspergillus niger* (CGCKZ), using biocompatible ILs. Three cholinium-based ILs, [CH][Ac], [CH][Prop], and [CH][Hex] were chosen for extensive CGC extraction trials. The phase separation technique was applied to fabricate CGC polymeric structures resulting in the formation of gels or films (Fig. 4). Various analytical approaches such as SEM, FTIR, elemental analysis, water contact angle measurement, mechanical testing, swelling tests and rheological assessments were employed to characterize the chemically and physically synthesized biopolymeric structures. These biopolymeric structures, including films and gels exhibited significant potential for biomedical applications owing to their biocompatibility and the ability to adjust their physical and chemical properties by IL adjustments used for dissolving cellulose-based gel and film composites [74].

Shamshina et al. introduced a pioneering technique for the in-situ synthesis of discrete chitin-based nanowhiskers (ChNWs) extracted from crustacean waste. The use of [C₄C₁IM][HSO₄] facilitates the liberation of ChNWs while simultaneously removing the mineral and matrix components. This process eliminates the need for isolating and purifying the chitin polymer upfront. Moreover, this technique effectively reduces the amount of required chemicals and minimizes process waste, thereby eliminating the handling of boiling acidic solutions typically used in conventional ChNWs production. The ChNWs derived from biomass exhibited remarkable characteristics including a substantial aspect ratio of 75 and a high crystallinity index of 92 %. These NWs also demonstrated exceptional heat stability and were fully acetylated as evident from their breakdown temperature of 347.9 °C. A schematic representation of the structural hierarchy is illustrated in Fig. 5. The observations regarding the aggregation of ChNWs particles in non-diluted suspension were supported by AFM images consistent with the data obtained through TEM. When a mica substrate coated with an aqueous ChNWs suspension underwent water evaporation, imaging in air-repulsive mode revealed the presence of aggregated ChNWs as depicted in Fig. 6 (left). Conversely, by diluting the suspension, individual nanocrystals could be observed as shown in Fig. 6 (right) [75].

3. ILs assisted processing of chitosan

Chitosan, extracted from chitin present in the shells of crustaceans, is a natural polymer widely utilized for various purposes. Through the process of deacetylating chitin, a polymer with distinctive characteristics is obtained. Within the medical field, researchers are exploring the

potential of chitosan in wound dressings, drug delivery mechanisms, and tissue engineering due to its compatibility with biological systems and natural biodegradability. Additionally, it exhibits promise as a coagulant in the purification of water contributing to the elimination of contaminants. Despite assertions regarding its efficacy as a weight management aid, there remains a dearth of scientific substantiation. Within agriculture, chitosan serves as an eco-friendly insecticide and promotes the growth of plants potentially enhancing agricultural output. It serves as a natural preservative and clarifying agent in the food industry, prolonging shelf life and improving product quality. In cosmetics, its moisturizing and film-forming abilities benefit skin and hair [76–99]. Extraction methods for chitin and chitosan include chemical extraction, microbial fermentation, enzyme-assisted extraction (EAE), and microwave technology [52,53,70,76,100,101]. Chemical extraction, though established, raises environmental concerns due to harsh chemicals and waste production. Microbial fermentation offers cleaner production but requires precise control and longer fermentation times (Fig. 7). EAE reduces energy and chemical usage but may be costlier and require careful optimization. Microwave technology offers efficient heating, reducing chemical usage, but requires optimization for cost-effective production. Overall, chitosan's applications are vast, but its effectiveness and safety depend on factors like origin and extraction method, emphasizing the need for careful consideration in its use [64].

According to literature, the commonly utilized ILs comprise [C₂C₁IM][OAc], [C₄C₁IM][Cl], [C₂C₁IM][Cl], [C₂C₁IM][OAc], and [AllylC₁IM][OAc] [8,102,103]. The mentioned ILs have demonstrated efficiency in processing chitosan derived from squid pens, exoskeletons of shrimps and crab's waste. Fig. 8 illustrates the methodology employed in the chitosan extraction utilizing ILs. Following this, the thermal dissolving process was carried out under controlled conditions using a 100 °C oil bath. Chitin exhibited insolubility in the [C₄C₁IM][Cl] as well as ILs containing anions such as [CF₃SO₃], [Tf₂N], [BF₄], and [PF₆]. However, it shown solubility in [OHC₂C₁IM][Cl], and a partial solubility in [AllylC₁IM][Cl] and [C₄C₁IM][Cl] [104–106].

W. Xiao et al. synthesized a novel IL known as 1,3-dimethylimidazolium chloride [C₂IM][Cl] [107]. This IL exhibits lower viscosity and higher efficiency compared to existing ILs. The current IL in combination with another IL [C₁IM][Cl] was developed specifically to dissolve and regenerate chitosan. This unique combination of ILs was employed in the production of blended membranes and fibers comprising chitosan and cellulose. Fig. 9a and b represent a composite fiber and membranes

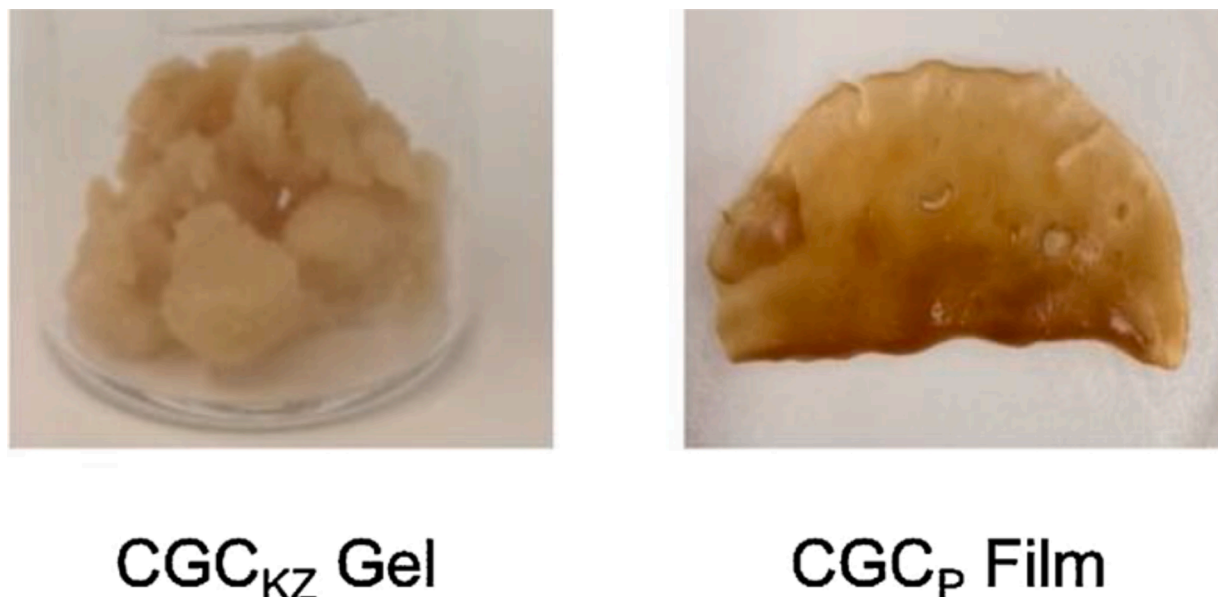


Fig. 4. Visual representations illustrating the resultant CGC polymeric configuration post-phase inversion with water adapted from [74].

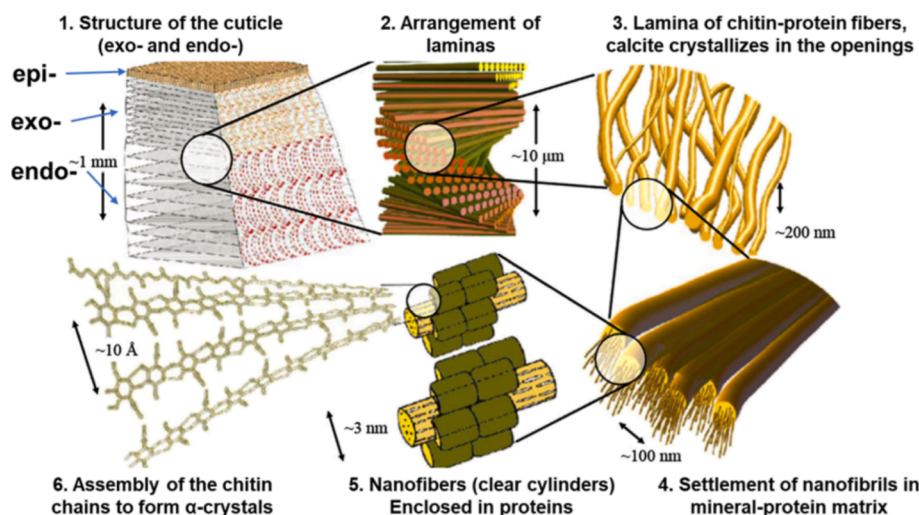


Fig. 5. Visual exploration of the intricate architecture of *Homarus americanus* exoskeleton material. (1) Bouligand pattern. Twisted plywood arrangement of exo- and endocuticles; (2) Interconnected chitin-protein planes; (3) chitin-protein fiber network; (4) nanofibrils embedded within the mineral-protein matrix; (5) protein-embedded chitin nanofibrils; (6) anti-parallel arrangement of 18–25 Polysaccharide Chains in Clusters, adapted from [75].

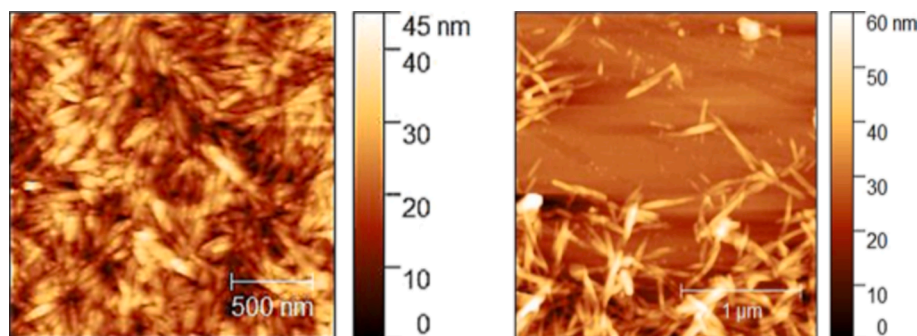


Fig. 6. AFM visuals illustrating (on left side) a concentrated and (on right side) a diluted ChNW suspension extracted from ChNWchitin. Visuals are cross-sectional examined using Gwyddion software adapted from [75].

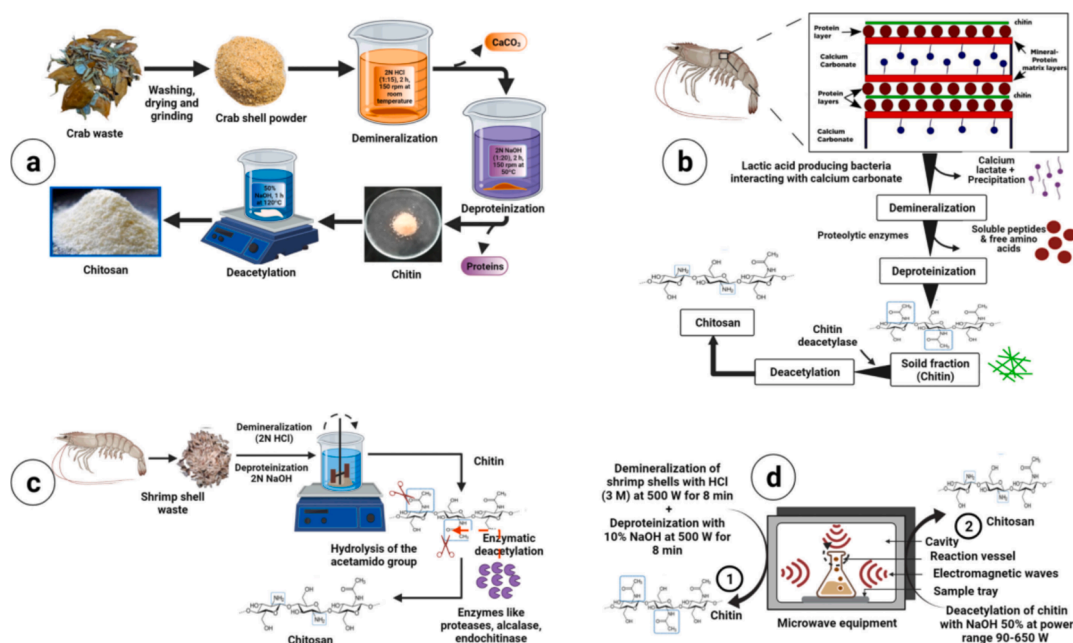


Fig. 7. Schematic diagram of (a) extraction of chitosan by chemical processing (b) extraction by microbial processing (c) extraction by enzymatic processing (d) extraction by microwave treatment adapted from [64].

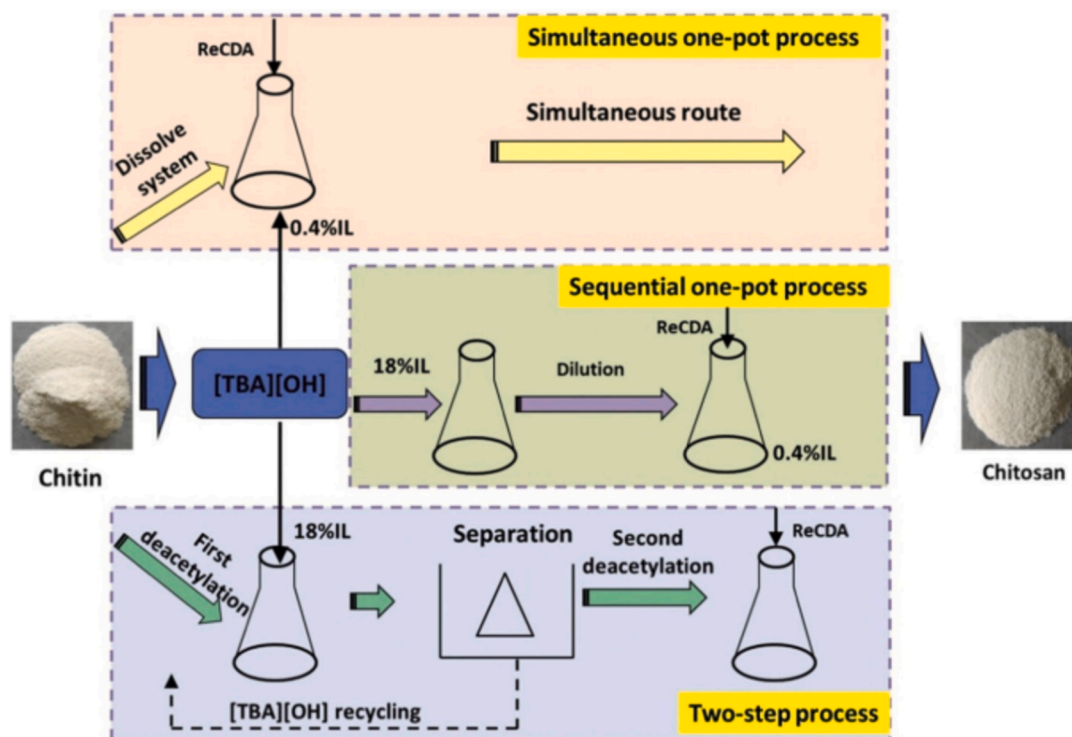


Fig. 8. The extraction of chitin and chitosan through ionic liquids involving the use of tetrabutylammonium hydroxide ([TBA][OH]) adapted from [64].

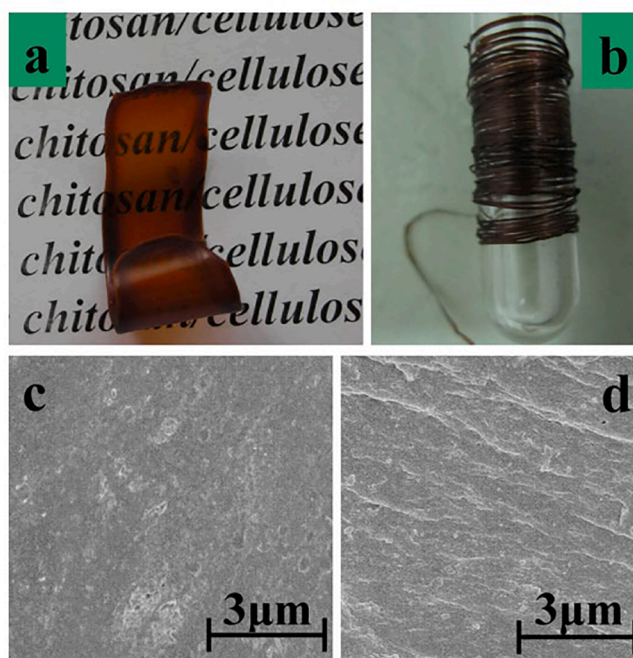


Fig. 9. Digital images displaying blended membranes (a) and fibers (b) composed of regenerated chitosan/cellulose (1:3, w/w) ratio, alongside SEM micrographs illustrating the surface characteristics of blended membranes (c) and fibers (d) with the same chitosan/cellulose ratio adapted from [107].

composed of chitosan and cellulose in a weight ratio of 1:3. The membrane and fibers have a brown coloration suggesting that chitosan has been evenly incorporated into the cellulose matrix. Fig. 9c and d depict SEM micrographs portraying the surface attributes of the 1:3 (w/w) blend of chitosan and cellulose membranes and fibers. The digital images of the composite membranes and fibers collectively exhibit a

uniform structure suggesting a level of compatibility among the constituent materials. In contrast to the regenerated cellulose from NaOH/H₂O [108] and rejuvenated chitin from [C₄C₁IM][OAc] [109], the blended membranes and fibers did not exhibit any porous structure.

Several ILs, including [C₂C₁IM][Cl], [C₄C₁IM][Cl], [C₂C₁IM][OAc], and [C₄C₁IM][OAc] have been investigated for their ability to dissolve chitosan [110]. The determination of chitosan solubility in ILs has been a common focus in numerous investigations primarily investigating the influence of temperature. Xie et al. showed in their investigation that [C₄C₁IM][Cl] has the ability to dissolve chitosan by rupturing the hydrogen bonds within chitosan molecules [111]. This disruption facilitates the complete exposure of chitosan's amino groups making it easier for them to react with other chemical reagents. Chen et al. investigated the impact of temperature and the ability of ILs to accept hydrogen bonds on chitosan dissolution aiming to explore the anionic structures of various synthetic imidazolium-based ILs affecting the process [112]. The research demonstrates that elevating the dissolution temperature (ranging from 50 °C to 150 °C) and enhancing the IL anions' capacity to disrupt hydrogen bonds within chitosan can augment its solubility in the investigated ILs. Additionally, owing to their impressive thermal stability, imidazolium-based ILs exhibit superior solvation properties for chitosan. As per the studies conducted by Sun et al. [113] and Cristina Stefanescu et al. [114], [BMIM][Ac] demonstrated the highest efficacy among ILs for dissolving chitosan. Feng et al. [115] initially reported the utilization of [C₁IM-COOH][HCl] as a highly promising and environmentally friendly solvent for chitosan. According to their findings, chitosan could maintain its crystalline structure and associated characteristics even after being dissolved in [C₁IM-COOH][HCl] for one hour at 50 °C.

4. ILs assisted processing of keratin

Keratin is widely acknowledged as an abundant protein biopolymer commercially available on a global scale [116–120]. This polymer is primarily found within the cells present in the outer skin of vertebrates having high protein percentage [121–124]. Keratin is present in

different forms such as α & β keratin. The α -keratin is typically found in soft tissues like fleece, dermal cells, and fur whereas β -keratin is mainly found in rigid tissues like bird's quill, hoof, nails and toenails. Typically, keratin biomass undergoes hydrolysis using acid, alkali, or enzymes in order to extract and isolate the keratin [118,125–129]. Due to its minimal toxicity, the keratin biopolymer finds utility in various domains such as biomedical, cosmeceutical, and industrial sectors. The utilization of keratin biopolymer, characterized by its substantial molecular weight, is prevalent in the field of skincare and various other cosmetic applications. The keratin has emerged as a prominent biological tool especially in the mechanism of drug deliveries. Keratinous hydrogels have found application in the field of tissue engineering such as bone tissues, eye reconstruction, healing of wounds, nerve reformation, and dermal treatments [118,120,126,130,131]. The process of dissolving and extracting keratin exhibits inherent challenges. The quest for an efficient, environmentally friendly and cost-effective solution holds paramount importance. In recent years, significant progress has been made in the extraction and characterization of keratin leading to an increase in the production of keratin-derived products. Various methods can be employed to extract keratin including chemical hydrolysis, enzymatic and microbiological treatment, microwave treatment, steam explosion and thermal hydrolysis [132–138].

S. Feroz et al. investigated the keratin generation from sheep wool fibers which presented itself as a promising biopolymer with the potential to serve as an affordable, environmentally friendly, and renewable material source for the development of many specialized products within the field of biomedicine [139]. In contrast to traditional approaches employed in keratin extraction, the utilization of environmentally sustainable and reusable ILs present a novel opportunity for the treatment of natural resources derived from livestock sectors. They developed a method for efficient dissolution of keratin using IL combined with probe sonication eliminating the need for external heating. Keratin was extracted by using aqueous ILs TBSh and CH, determining the optimal sonication time, adjusting the initial sample loading and optimizing the sonication process. Impact of varying sonication duration was examined while maintaining a constant amplitude of 30 %. To assess the effect of sonication process on wool breakdown in a specific IL, the study investigated variable sound levels ranging from 30 % to 60 % at 10 % intervals under consistent conditions of sonication time and initial loading. The breakdown rate of choline hydroxide reached 90 % at acoustic power levels of 40 % and 50 % over a 30-minute sonication period. Therefore, it was recommended to use a reduced acoustic power range, specifically 40 %, to avoid potential damage to the probe sonicator tip that may occur with higher power levels [139].

Yang and their coworkers conducted a comprehensive study on the processing and characterization of keratin derived from camel hair and goat cashmere [140]. The cashmere and camel hair fibers were subjected to dissolution in $[C_4C_1IM][Cl]$. Following this, an evaluation was conducted to discern the properties of the keratin that remained soluble and insoluble after the dissolution process. The investigation focused on examining the composition, organization, and characteristics of the unprocessed substance, as well as the soluble and insoluble forms of keratin. In comparison to the initial material, the soluble keratin exhibited chemical alterations, specifically a reduction in cysteine content, as well as small modifications in its structural composition. The initial in-vitro assessment of the biological characteristics included lactate dehydrogenase (LDH) analysis and a scratch test. These tests revealed that the keratin obtained from both camel hair and goat cashmere exhibited good bioactivity. The experiment results indicated that the presence of soluble keratin derived from cashmere and camel hair led to an increased rate of cell migration when cells were cultured [140].

Feathers are primarily composed of keratin and are abundantly present as a waste in numerous processes [141,142]. Following the appropriate techniques for the disintegration and subsequent regeneration of keratin, it is plausible that they may serve as a viable source for

keratin-polyamide membranes [143]. In the study conducted by Azila et al., a range of protic ionic liquids (PILs) containing the N,N-dimethylethanolammonium cation were examined as potential processing solvents for the turkey feathers [144]. The solubility of feathers in $[DMEA][HCOO]$ (Fig. 10a) was found to be approximately 150 mg/g of solvent. Reproduced keratinous material can be retrieved by introducing methanol. This investigation also involves IL recovery from the diffused keratin. The keratin was regenerated (Fig. 10b) through precipitation using methanol. The resulting fraction exhibits water insolubility. Initially, about 37 % of the material remains soluble in water and methanol, uniformly mixed within the IL. A distillation process was conducted at 122 °C, achieving a 99 % recovery rate for the IL, underscoring the significance of using PILs in this context. The regenerated material was further processed into a film, exhibiting a glass transition temperature (Tg) of approximately 93 °C [144].

5. ILs assisted processing of collagen

Collagen is predominant a protein scaffold in mammals constituting around 30 % of the total protein composition. It is widely distributed throughout various tissues and organs including but not limited to the skin, tendons, bones, cartilage, and ligaments [145–147]. There are currently 28 different forms of collagen known and described with type I collagen being the most prevalent. All forms of collagen are composed of three α -chains which can be either identical or different and possess at least one domain characterized by a triple helix structure [148,149]. In various in-vivo contexts, collagen exhibits a wide range of morphologies by self-assembly. Under specific conditions, it is possible to isolate a significant quantity of collagen molecules that retain their triple-helical structures. On the contrary, it's important to note that the extraction process of collagen can result in the disruption of both intramolecular and intermolecular covalent bonds, which are crucial for maintaining the stability of collagen fibrils. Additionally, the extraction process may also break the hydrogen bonds that contribute to stabilizing the collagen triple helix structure. Consequently, the resultant mixture of polypeptides obtained from this process is commonly referred to as gelatin which can be considered as a partially degraded form of collagen [150]. Collagen and gelatin exhibit numerous functional features associated with their protein side chains encompassing gelling, emulsifying, foaming, and film-forming characteristics. They are useful ingredients in many food systems because of all these characteristics (Fig. 11).

The extraction process for collagen is determined by its solubility characteristics. Common methods include precipitation-based extraction using sodium chloride or guanidine hydrochloride, though they often yield low extraction rates. Acid-based extraction employs acids like HCL, H_2SO_4 , etc. Enzymatic extraction involves the use of enzymes such as pepsin, papain and collagenase typically in conjunction with an organic acid like acetic acid. These methods aim to disrupt the collagen structure to make it soluble for further processing [151–156]. Previous research has suggested that inorganic acids including HCL and H_2SO_4 exhibit lower extraction ability as compared to that of organic ones. Enzymatic breakdown is widely utilized to remove non-helical regions of collagen thereby improving its solubility. Collagen extraction via enzymatic hydrolysis is common from various sources including skin, scales, and remnants of swimming bladders in teleostean fish species. One potential downside of this approach is the possibility of causing permanent alteration to the collagen structure through enzymatic breakdown. This occurrence was analyzed through FTIR [157]. The acid and pepsin soluble treatments are often employed methods because to their ability to yield high amounts of extracted substances. These treatments can be utilized alone or in combination with the aim of increasing the overall yield.

A study was conducted to examine the impact of IL concentrations and cationic chain length on type-I collagen utilizing multi-spectroscopic techniques and molecular docking analysis [158]. To accomplish this goal, various imidazolium ILs with a stable chloride

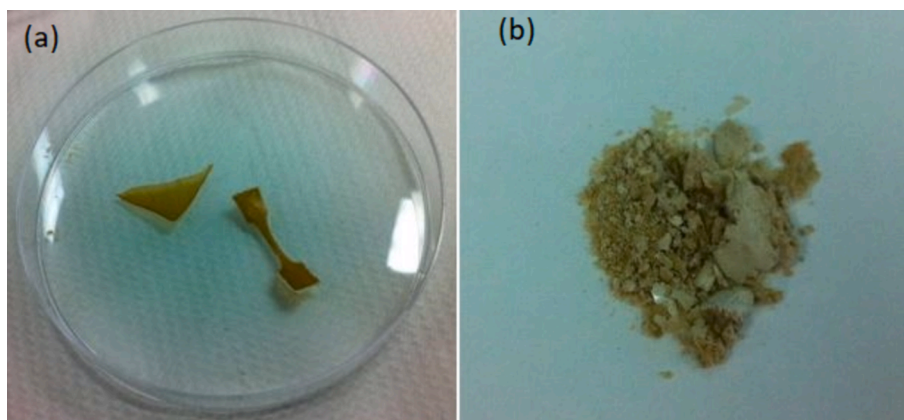


Fig. 10. (a) Keratin film prepared from feathers dissolved in ILs (b) regenerated feather Keratin adapted from [144].

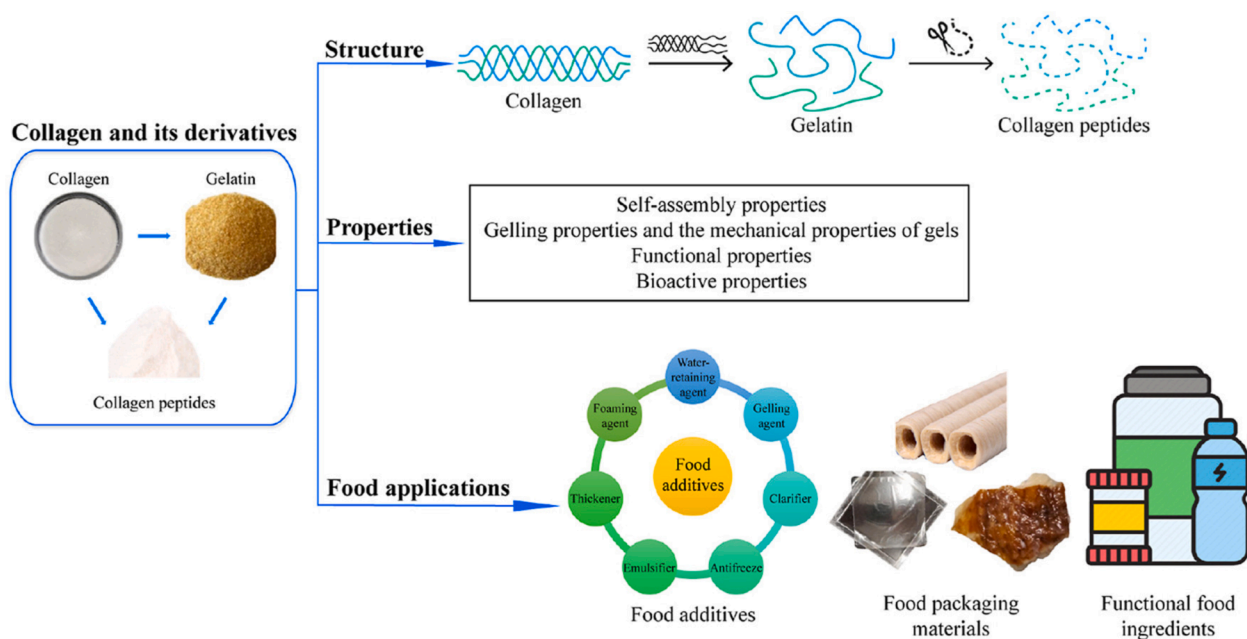


Fig. 11. Schematic diagram of collagen structure, properties and applications adapted from [150].

anion (Cl^-) and cations with different chain lengths were employed. Specifically ILs $[\text{C}_4\text{C}_1\text{IM}][\text{Cl}]$, $[\text{C}_6\text{C}_1\text{IM}][\text{Cl}]$ and $[\text{C}_{10}\text{C}_1\text{IM}][\text{Cl}]$ were examined. A range of analytical techniques including UV-visible spectroscopy, CD, TRF, FTIR, molecular docking approach etc. were utilized to assess the ILs impact on type-I collagen. The outcomes showed that collagen stability was affected by alkyl structures and IL concentration. Hydrophobic and hydrogen bonding interactions resulting from cationic species in ILs, alongside residual amino acid, significantly contributed to collagen unfolding and destabilization. As a result, the inclination of ILs to destabilize collagen is heightened as the chain length of ILs grows. Furthermore, it was observed that IL cationic species followed the Hofmeister classification when arranged according to their ability to induce destabilization in the collagen triple helix structure. Molecular docking analyses additionally validate the prevalence of hydrophobic interactions and the diminished occurrence of hydrogen bonding due to lengthy alkyl structures in IL cations. Docking outcomes provide data about location of the ligand binding site on the protein and are in agreement with the spectroscopic findings [158].

6. ILs assisted processing of silk

Silk is a versatile material obtained from silk-moth larvae [159]. The primary constituents of silk are sericin and fibroin. The major portion of silk is fibroin which makes up about 75 % of the material with molecular structure comprises heavy & light chain portentous material [160–162]. The structural organization of fibroin is categorized into two distinct regions: crystal and powder. The presence of a well-defined crystalline phase along the axis of the silk thread contributes significantly to its high tensile strength. In the presence of stress, the powdered silk has a notable capacity for energy absorption resulting in a heightened level of toughness [163,164]. The outstanding biological compatibility, controlled breakdown and ease of processing make fibroin a potential candidate for its widespread utilization in various industries including fabric sector, nutrition, biosensors, tissue formation etc. (Fig. 12). It has been utilized in diverse forms such as nanoscale fibers, membranes, scaffoldings and hydrogels etc. [165].

The pupa protein of the silkworm (*Bombyx mori*), known as SPP (Self-Assembled Polypeptide) is regarded as a valuable animal protein that offers significant nutritional advantages and health benefits [166]. In pursuit of an effective and environmentally sustainable extraction

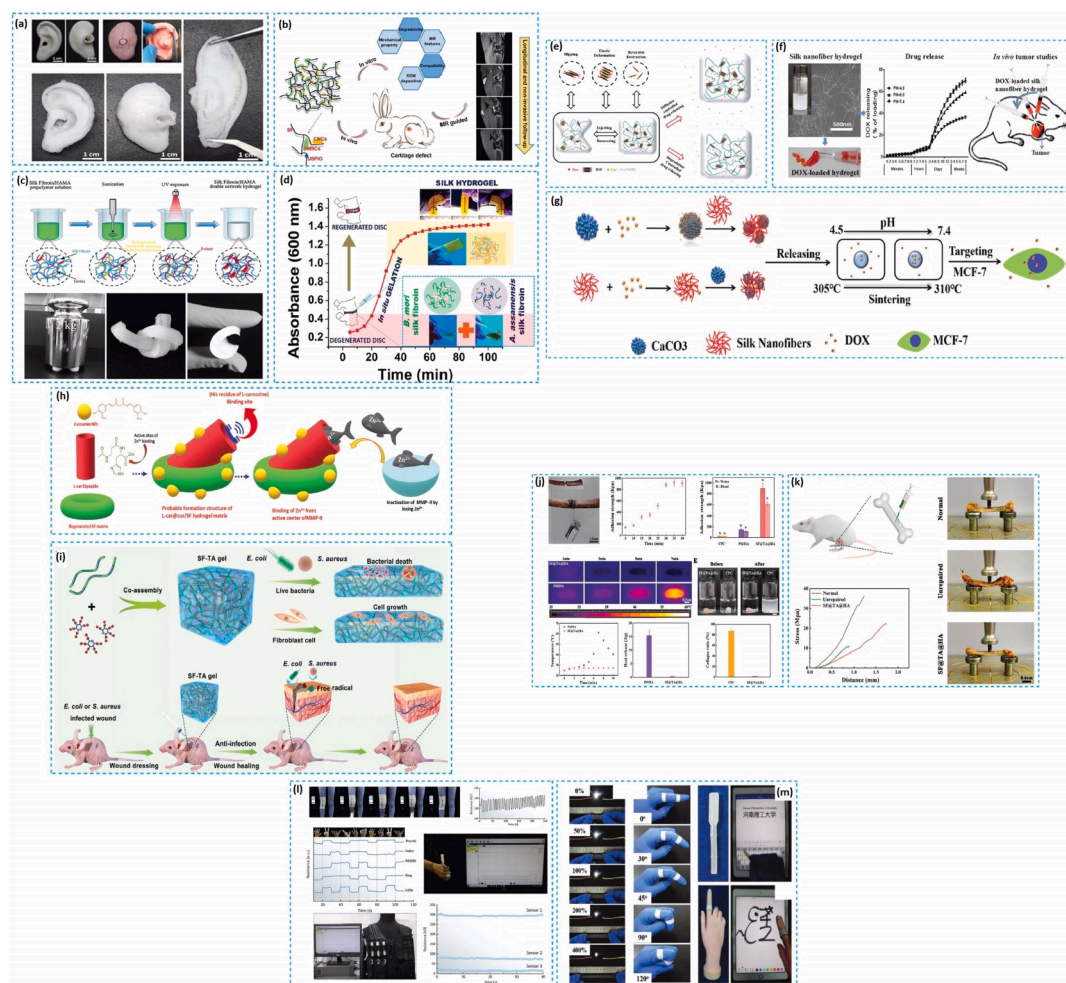


Fig. 12. (a) Silk fiber used for ear cartilage repair due to its high strength; (b) silk hydrogel undergoes in vivo degradation for non-invasive monitoring; (c) high strength silk aids in developing load-bearing tissue; (d) injectable silk gel facilitates invasive in situ disc regeneration; (e) injectable silk fiber combined with doxorubicin (Dox) and curcumin for cancer treatment; (f) pH-responsive silk fiber for cancer treatment; (g) Silk hydrogel having pH responsiveness and injectability for drug release; (h) Silk fiber hydrogel promotes wound healing in diabetic mice; (i) injectable silk protein with antioxidant and antibacterial characteristics; (j) silk fibroin hydrogel used for repairing fresh pig bones; (k) adhesive silk fiber repairing enhances the compressive strength of rat femur; (l) conductive silk fiber monitors human finger and knee movement and aids in grasping objects; (m) conductive silk fiber responds to finger movement and serves as electronic skin [165].

technique for SPP and CH-IL were employed as an extracting agent. Subsequently, a series of orthogonal experiments were conducted to optimize the extraction parameters. The results revealed that 3 % CH-IL, 1:30 g/mL solid-liquid ratio, at 40 °C and 1 h duration gave best outcomes. In comparison with standard acid settling- alkali dissolution procedure, the protein content increased by 12.14 % increment was observed in protein contents with the CH-IL extraction method. Analysis of the protein structure indicated a significant increase of 10.98 % in β -sheet content, along with a notable decrease of 16.4 % in the presence of disulfide linkages. The experimental findings suggested that the

protein extracted with CH-IL exhibited improved processing characteristics, including an 82.87 % increase in solubility, a 15.44 % increase in emulsification capacity, and an 18.97 % increase in foaming capacity. The physical properties of SPP demonstrated a notable enhancement in response to the increased elongation of the peptide chains. These results provide valuable insights that can contribute to the enhancement of pupal protein processing efficiency [166].

An innovative, cost-effective and biologically diversified IL (choline based) was explored to dissolve and reform fibroin acquired from bombyx mori. This procedure was further refined by optimizing various

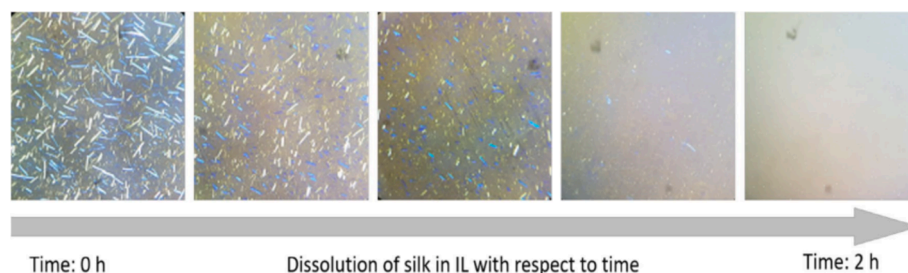


Fig. 13. Inverted light (optical) microscope images of dissolution of silk fibroin in chlorine hydroxide IL adapted from [167].

factors such as temperature, duration, stirring rate, IL selectivity, and the maximum solubility. Chlorine hydroxide was selected which was biocompatible as compared to other ILs. The dissolution process was evaluated both visually and with the aid of an optical microscope (Fig. 13). The experimental parameters that resulted in the highest dissolution rate, specifically 25 %, were attained by subjecting the sample to a temperature of 50 °C for a duration of 2 h [167].

7. ILs assisted processing of gelatin

Gelatin is a natural polymer derived from the partial hydrolysis of collagen. Its distinctive amino acid structure renders it suitable for various medical applications, thanks to its biocompatibility and degradability in the physiological environment [181]. Moreover, the molecular chains of gelatin possess many functional groups such as amide, hydroxyl, amino and carboxy groups, making them an ideal candidate for post-processing towards targeted applications [182]. Gelatin can be shaped into coatings and films with good mechanical strength and optical opacity. It also has inherent UV-blocking properties because of the abundant aromatic amino acids [183]. The thermo-responsive behavior of gelatin in an aqueous solution, i.e., melting above 40 °C and gelation below 30 °C, makes it a suitable material for 3D printing applications [184]. Furthermore, gelatin has many other applications such as cosmetics, food (frozen food or candy), and forensics as a thickener and emulsifier (Fig. 14) [185]. However, due to less soluble properties of

gelatin in conventional solvents, the highly toxic or corrosive nature of these solvents, and the need for the post processing treatment requires to develop sustainable and green solvents for gelatin processing [186].

Pabst et al. [187] investigated the properties of an ionic gel (Fig. 15) consisting of gelatin, water, and an ionic liquid (1-butyl-3-methylimidazolium dicyanamide $[C_4C_1IM][NCN^2]$). They utilized dynamic light scattering to monitor the re-orientational motion of ions and fluctuations of the gel matrix, dielectric spectroscopy to assess ion transport, and rheology to analyze the shear response of the material from temperatures exceeding room temperature to the glass transition temperature. The comparative results between IL-water mixture and IL-water-gelatin mixture showed that the presence of gelatin had no influence on the microscopic dynamics of ions, giving equal re-orientational time constants and DC-conductivities. However, additional dynamic processes were witnessed in light scattering and rheology measurements which were linked to the motions of gel matrix. In particular, a prominent broadening of low frequency behavior in shear modulus – similar to Rouse behavior – and depolarized light scattering was observed (Fig. 15), which was attributed to the slow motions of parts of gelatin network or ions. Moreover, the outcome of this study showed the absence of thermal reversibility of ionic jelly, which was associated to the ability of ions to form cross-links. In conclusion, this research provides an interesting correlation between microscopic ion dynamics and meso- or macro-scale rigidity of gel matrix.

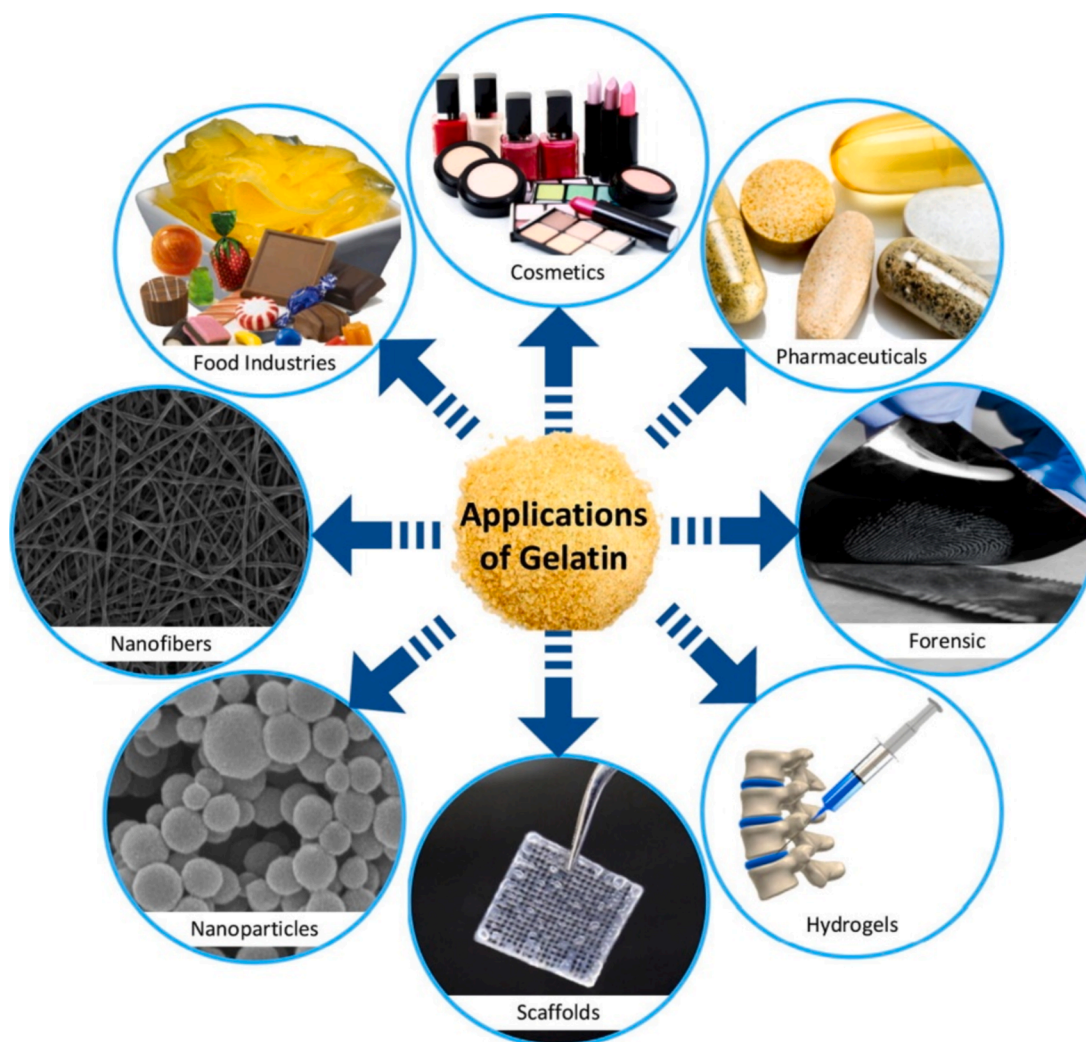


Fig. 14. Schematic illustration of different applications of gelatin adapted from [185].

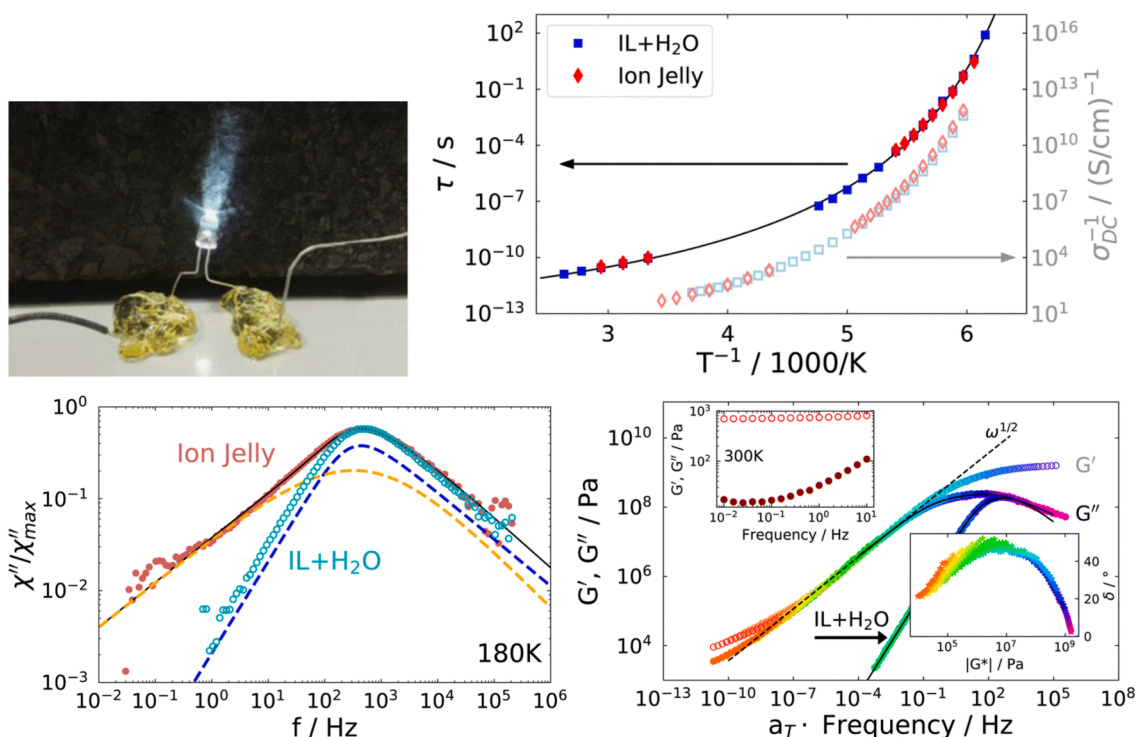


Fig. 15. The image depicts the prepared ionic gel, while the study includes a comparative analysis of DC-conductivities and rotational time constants between the ion gel and the IL–water blend. Additionally, it examines the light scattering spectra of the ion gel and the IL–water mixture at 180 K. A comprehensive plot illustrates the storage (G') and loss (G'') moduli of the ion gel and the IL–water mixture at a base temperature of 178 K. The solid black line represents the fitting using a combination of CD and HN functions, while the dashed line indicates the power law relationship in the low frequency domain. The upper inset shows the response of G' and G'' at 300 K, while the lower inset displays the vGP plot demonstrating excellent time–temperature superposition, particularly at lower temperatures adapted from [187].

In another study, phosphonium IL (tributylhexadecylphosphonium salicylate, $[C_{16}P][Sal]p$) was used as a functional additive to develop gelatin based microcapsules using complex coacervation methodology [188]. Initially, water was used to dissolve gelatin, which was then followed by adjusting the pH of the solution to acidic levels (pH = 4). Subsequently, Avermectin, a pesticide, was dissolved in an ionic liquid

(IL) and rapidly dispersed into a gelatin solution at high speed (8000 rpm). The resulting mixture was then elevated to a basic pH level of 9 by adding 10 % ammonia solution dropwise. Finally, the pH of the emulsion was readjusted to 6–7, followed by the addition of a 1 % transglutaminase solution to solidify the gelatin shell. The microcapsules were prepared by centrifuging the mixture, washing it with distilled

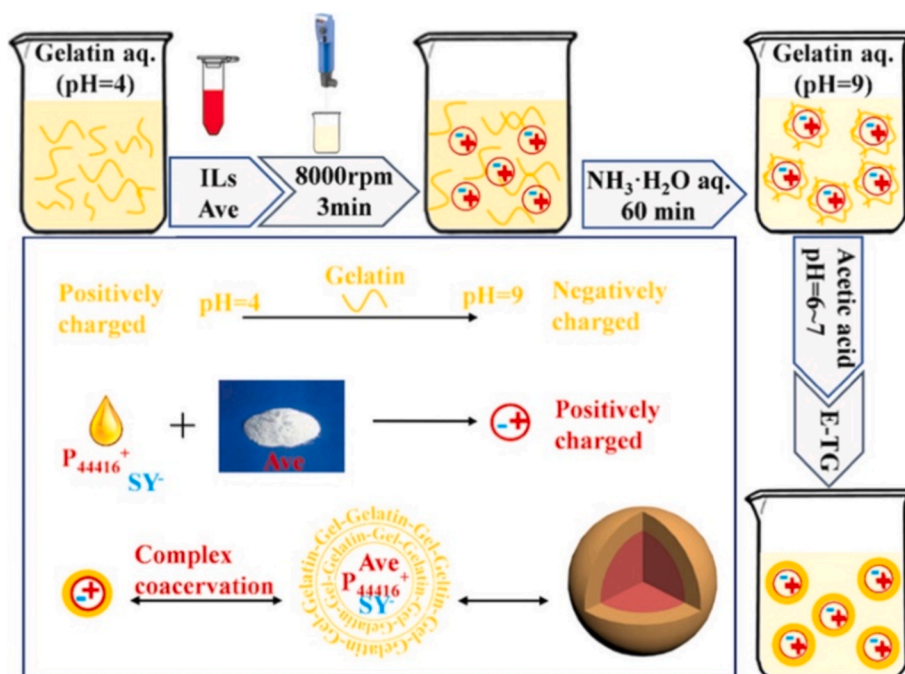


Fig. 16. Illustration outlining the synthesis process for IL-Gelatin-Avermectin microcapsules adapted from [188].

water, and drying it at a low temperature of -20°C . The findings indicated that the ionic liquid (IL) used in this process proved to be an effective cationic surfactant and eco-friendly processing solvent, which led to a remarkable entrapment efficiency of 98 % w/w. The synthesized microcapsules exhibited a higher degradation temperature in soil, good photostability, and remarkable nematocidal activity against *M. incognita*. Hence, the proposed methodology is an efficient, simple and sustainable approach to produce pesticide-based formulations with controlled release period and release rates (Fig. 16).

Most of the developed gels based on ionic liquids and biopolymers (gelatin) displayed low stretchability (<200 %), weak mechanical strength, gradual self-repairing mechanism (hours or days), and poor temperature-mechanical stability and elasticity due to their non-covalent weak interactions among ions within the ILs and biopolymeric chains, which restricts their window of applications. However, this hurdle was crossed by introducing chemical crosslinks into biopolymers based ionogels.

In this regard, Zhu et al. [189] reported the synthesis of a sustainable ionogel based on gelatin which was chemically crosslinked via novel bio-based epoxide. First of all, a bio-based crosslinker with three epoxide groups was prepared from grapefruit peel. Then, the ring-opening reaction between gelatin and epoxide in IL (1-Allyl-3-methylimidazole chloride $[\text{AllylC}_1\text{IM}][\text{Cl}]$ resulted in an ionogel. These chemical crosslinks' creation produced materials with strong mechanical strength, good durability, and thermo-tolerance in addition to excellent reversible elasticity. Conversely, the ionogel's ability to self-heal, stretch, and recycle was eventually improved by the triple-helix unit, hydrogen bonding, and ionic contact that the gelatin matrix supplied. Notably, because gelatin naturally blocks UV light, the produced ionogel also demonstrated exceptional UV-shielding properties (Fig. 17). The gelatin-based ionogel produced intriguing results when utilized as a wearable sensor. It exhibited a gauge factor of 1.03, with a working range spanning from 1 % to 500 %. Notably, it displayed exceptional temperature tolerance, a rapid response time of 102 ms, and exhibited good cycle stability and durability, enduring over 5000 cycles. Ultimately, this wearable sensor was employed to track human movements

in real-time, including walking, finger bending, and sequential waving and throwing motions (Fig. 17).

Likewise, a chemical crosslinking in ionogel was introduced by a Schiff base reaction between methylglyoxal and gelatin in IL (1-ethyl-3-methylimidazolium acetate $[\text{C}_2\text{C}_1\text{IM}][\text{OAc}]$ [190]. The results of rheological tests indicated a rise in the plateau storage modulus corresponding to increasing gelatin content. The developed ionogels exhibited excellent thermo-mechanical stability and the chemical crosslinks displayed stability up to 140°C . Furthermore, the prepared ionogels displayed an excellent set of properties including high stretchability, excellent reprocessability and self-healability, high elongation at break (368.6 %), and remarkable ionic conductivity at room temperature ($1.0 \times 10^{-3} \text{ S/cm}$). Hence, the developed ionogel is a potential contender for producing next-generation stretchable electronics.

In addition to the conventional processing methods, additive manufacturing techniques (i.e., 3D printing) have also been used to process and produce intricate shapes based on IL-biopolymers mixture. For instance, Zhang et al. [191] reported the 3D printing of bio-inks prepared from the mixture of gelatin, sodium alginate and poly ionic liquids (PILs) (Fig. 18). Three different PILs (PIL1, PIL2 and PIL3) were prepared by the crosslinking of the monomer (1-vinyl-3-butylimidazolium chloride, $[\text{VinylC}_1\text{IM}][\text{Cl}]$ with three other compounds. The findings revealed varying biological activity of the prepared 3D scaffolds for different PILs, with $\text{PIL1} < \text{PIL3} < \text{PIL2}$. This characteristic renders them an ideal candidate for tissue engineering applications.

Similarly, in another study, Xu et al. [192] developed a 3D scaffold using decellularized extracellular matrix (dECM), quaternized chitosan (QCS), and gelatin (gel), which was further modified by poly(ILs) (Fig. 19). The scaffold demonstrated remarkable properties such as efficient hemostatic performance, high biocompatibility and excellent antibacterial activity. The study showed that the developed scaffold had the potential for use in skin tissue engineering.

8. ILs assisted processing of alginate

Alginate forms through a linear copolymerization process involving β -D-mannuronic acid (M) and α -L-guluronic acid (G) residues, connected via (1 $\rightarrow$ 4) linkages [193]. The technological and physicochemical attributes of alginate primarily hinge on its structural characteristics notably the M/G ratio, length of G-block and molecular weight [194]. Research findings indicate that solely the carboxylate groups within the G block participate in crosslinking with divalent ions such as Mg^{2+} and Ca^{2+} to create hydrogels. Consequently, a higher G content yields stiffer hydrogels, while a higher M content leads to softer hydrogels [195]. Similarly, an increased molecular weight of alginate leads to a more viscous system during the gel formation process. Alginate is described by its biocompatibility, high viscosity, biodegradability, non-toxicity, and a broad range of technological and biological applications (Fig. 20). Alginate has been reported to be used as a fat substitute [196], texturizing agent [197], hybrid edible coatings [198], biopolymeric nanofibers [199], membrane development [200], emulsifiers [201], tenderizers for meat [202], encapsulating agents for drug delivery [203], and other biomedical applications, including biosensing, tissue engineering, wound healing, etc. [204]. Commercial extraction of alginate from brown seaweed typically follows a conventional method (Fig. 20), encompassing six stages: pretreatment of algal biomass, acidic treatment, alkaline extraction, precipitation, bleaching, and drying. Nonetheless, this process requires significant quantities of solvents and reagents, coupled with labor-intensive techniques, thereby prolonging the duration of extraction [205]. Therefore, novel extraction procedures are currently being established in the hydrocolloid industry that target to optimize the process sustainably.

De Freitas et al. [207] reported the influence of alkyl chain length ($[\text{C}_n\text{C}_1\text{IM}][\text{Br}]$ ($n = 10, 12, 14$, and 16)) and concentration of ILs and charge density and concentration of alginates (hydroxamic alginate (AHX) or sodium alginate (ALG)) on the aggregate formation in aqueous

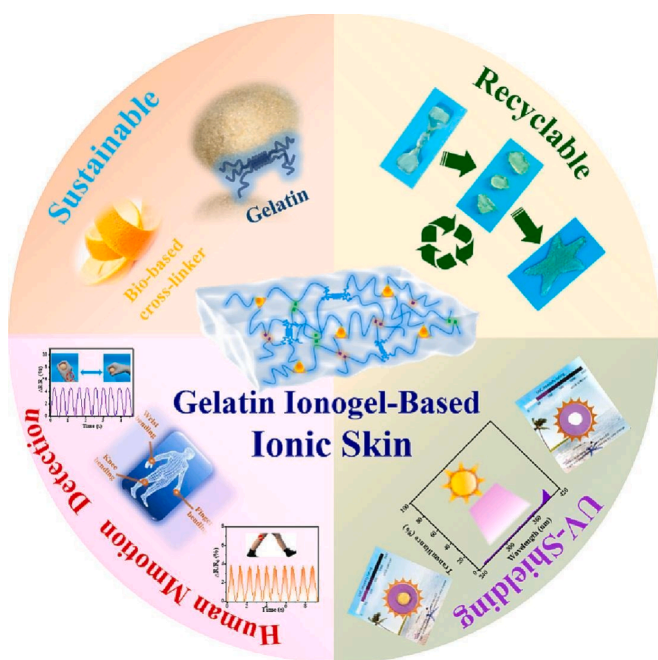


Fig. 17. Components of the prepared ionogel; images showing regeneration and shape molding of damaged sample using water-assisted approach; UV absorption of original UV Test Card and ionogel sample; pictorial illustration of real-time human motion sensing. Throwing and walking adapted from [189].

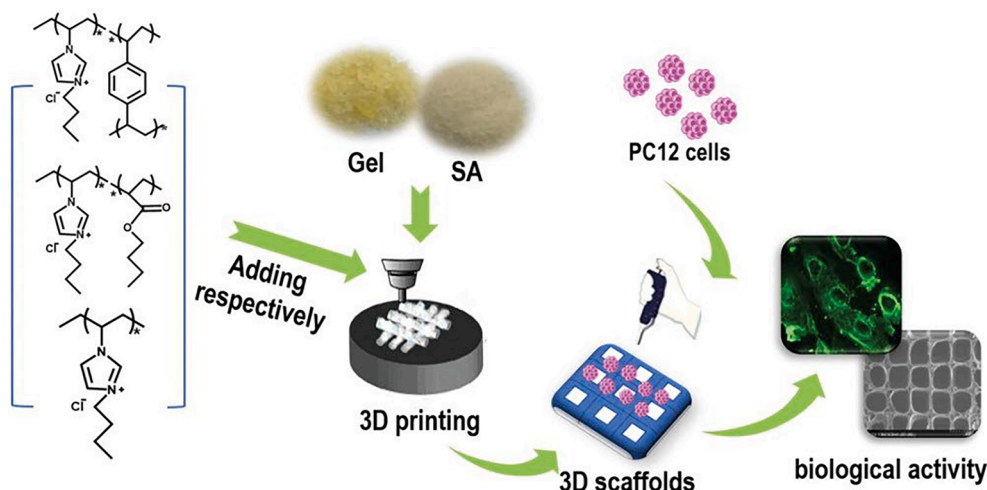


Fig. 18. Pictorial representation of 3D printing of bio-inks based on poly(ionic liquid), gelatin, and sodium alginate to form 3D scaffolds and their biological activity adapted from [191].

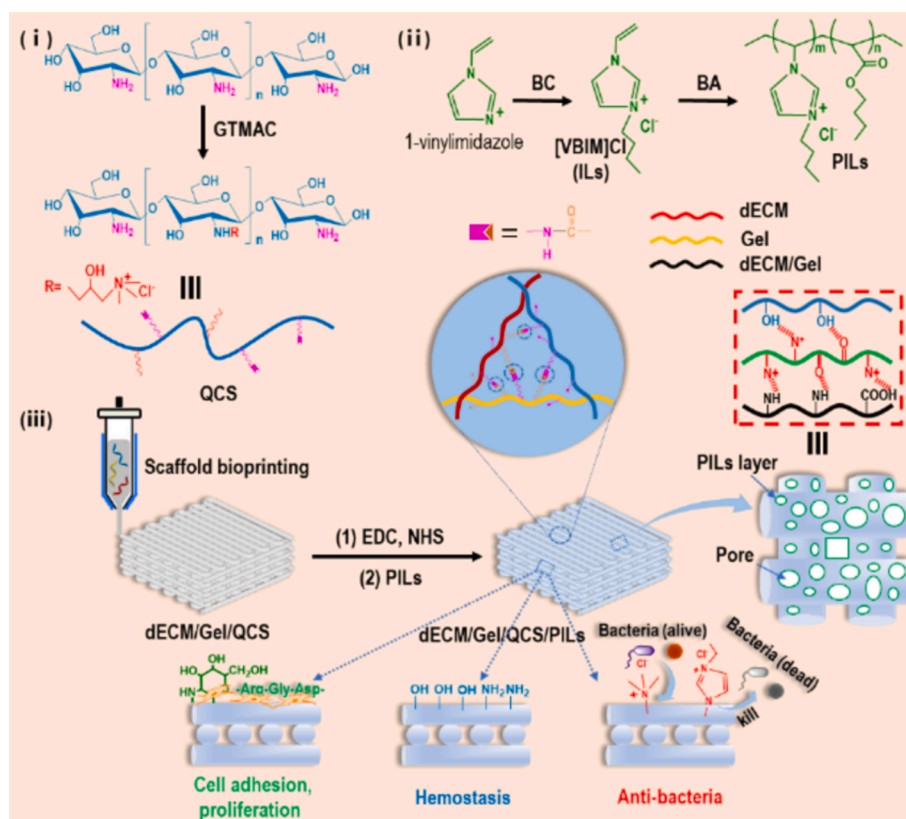


Fig. 19. Pictorial illustration of preparation of 3D scaffold; Preparation schemes of QCS polymer and PILs; printing of scaffold using bioink based on dECM, Gel, and QCS followed by the assembly with PILs; biocompatibility, hemostasis and anti-bacterial performance of prepared scaffold adapted from [192].

solutions. The results displayed an increase in IL-polymer interactions (i. e., aggregation) by increasing alkyl chain length of ILs, which was attributed to the cooperation between electrostatic and hydrophobic interactions. Moreover, the surface tension results showed the formation of thicker layer of adsorbed liquid–air interface for AHX/ILs systems than for the ALG/ILs systems. Likewise, the outcome of standard Gibbs energy indicated favorable interactions of $[C_{12}C_1IM][Br]$ with ALG as compared to interactions with AHX, which was linked to the higher polymer charge density. In contrary, $[C_{14}C_1IM][Br]$ and $[C_{16}C_1IM][Br]$ showed favourable interactions with AHX, a more hydrophobic

polymer. This study highlighted the function of ILs in developing novel and sustainable polymeric formulations.

Likewise, a novel application of ILs to process alginate was targeted by producing pH sensitive hydrogel beads using sodium alginate as a biopolymer and sulfonate ionic liquids (A1 [1-(4-ethoxy-1-butanedisulfonate)-3-methylimidazole bromide], A2 (triethylammonium hydrogen sulfonate), A3 [1-(1-propylsulfonate)-3-methylimidazole chloride] and A4 (1-heptyl-3-methylimidazole methanesulfonate)) as a crosslinking agent [208]. The hydrogel beads were formed by dripping method using A1 and A2 ILs (Fig. 21). Swelling studies revealed that at pH 6.0 and 7.4,

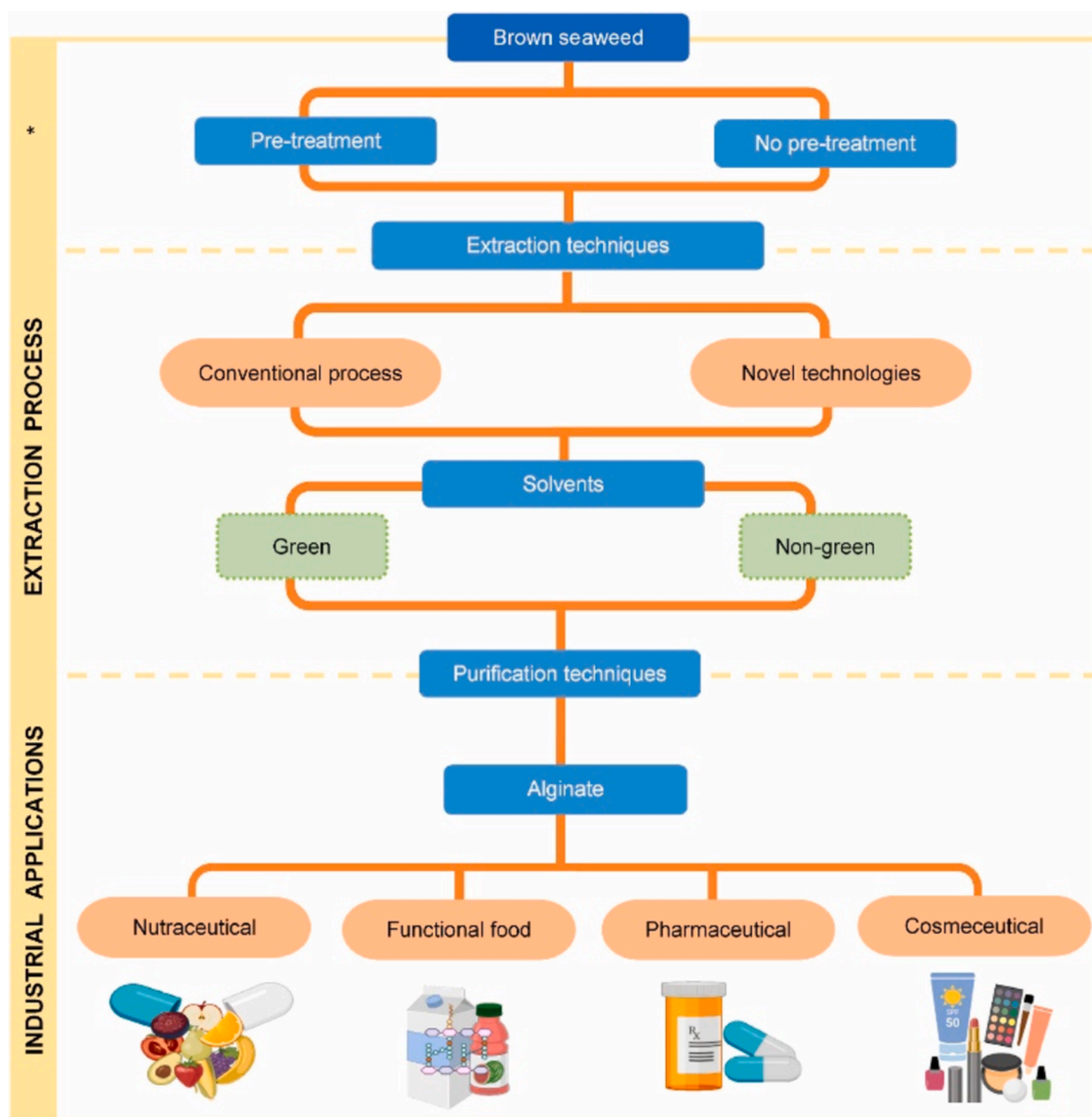


Fig. 20. Insightful illustration of alginate extraction processes and promising application prospects adapted from [206].

the beads disintegrated within minutes, whereas they remained stable for at least 7 h at pH 1.2. A comparison between IL/alginate-based beads and conventional bivalent ion-formed beads indicated a lower degree of swelling for IL/alginate-based beads over time. Additionally, drug release studies (indomethacin and 5-fluorouracil) demonstrated sustained release for a minimum of 8 h at pH 1.2. Furthermore, energy conformation calculations indicated stronger interactions between 5-fluorouracil and the hydrogel bead's structural network compared to indomethacin, causing in a lower release rate (9.84 % w/w) against 25 % (w/w). This research presents a compelling avenue for leveraging IL-based beads as precise carriers for drug delivery.

In another study, Abdullah & Al-Lohedan [209] reported the development and characterization of alginate-based materials for the demulsification of water-in-oil emulsions in heavy crude oil, utilizing PILs. First, sodium alginate was transformed into alginic acid followed by a conversion into corresponding polyester through esterification reaction. Second, the prepared polyester was exposed to a quaternization reaction in the presence of 1-bromononane or 1-bromohexadecane, which resulted in two PILs, (BN-ALG) and (BH-ALG), respectively. Characterization outcomes demonstrated the effectiveness of both PILs in demulsifying water-in-oil emulsions. Moreover, an increase in demulsification efficiency was found as a function of ratio of water in

emulsions and PIL concentration. BH-ALG PIL showed better demulsification efficiency as compared to the BN-ALG, which was associated with its higher hydrophobicity and, hence, its better efficiency to be dispersed as a continuous phase in crude oil emulsion.

Yang et al. [210] examined the impact of IL ($[\text{C}_2\text{C}_1\text{IM}][\text{BF}_4]$) doping technique on the electrochemical efficacy of Biomass Gel Artificial Muscle (BGAM) based on sodium alginate (Fig. 22). The investigated outcomes revealed that doping of IL improved the number of free-moving ions and the porous ionic channel inside BAGM, which eventually enhanced its electrochemical performance. Moreover, with the optimal IL doping content (4 ml), a porosity of 70.47 % was obtained for the porous ionic channels of BGAM, which was 1.4 times higher than the undoped BGAM (Fig. 21). Similarly, IL-doped BGAM exhibited minimal values for elastic modulus (1.871 MPa) and inner resistance (2.018 Ω), which were 56.02 % and 46.87 % lower, respectively, compared to those of undoped BGAM. Additionally, the specific capacitance of IL-doped BGAM reached a peak value of 126.98 mF/g, marking a 52.44 % increase over undoped BGAM. Moreover, IL-doped BGAM demonstrated optimal values for duty cycle (24.2 %), working life (1720 s, 19.5 times higher than undoped BGAM), tremble frequency (5.02 min), tremble amplitude (8.51 %), and steady force density (13.072 mN/g, 1.1 times higher than that of undoped BGAM). Furthermore, BGAM's force

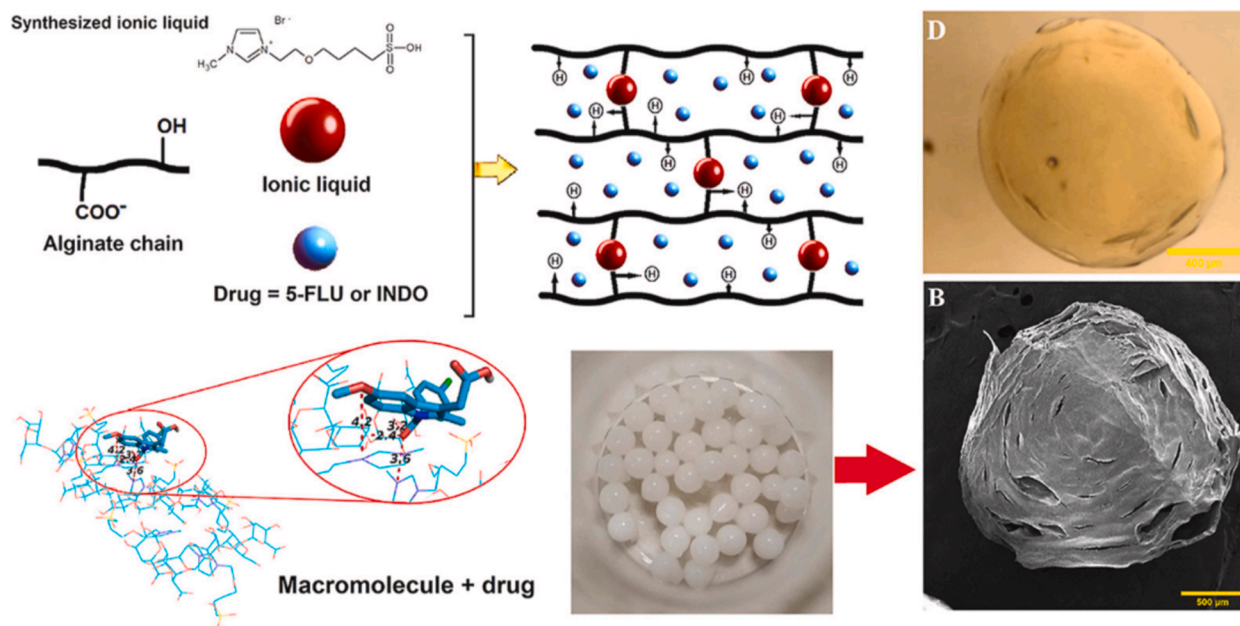


Fig. 21. Pictorial representation of formation of alginate beads and chain stability under pH of 1.2; lower energy interaction between A1-beads and indomethacin; SEM image for A1-beads showing cross section from the center; swollen A2-beads adapted from [208].

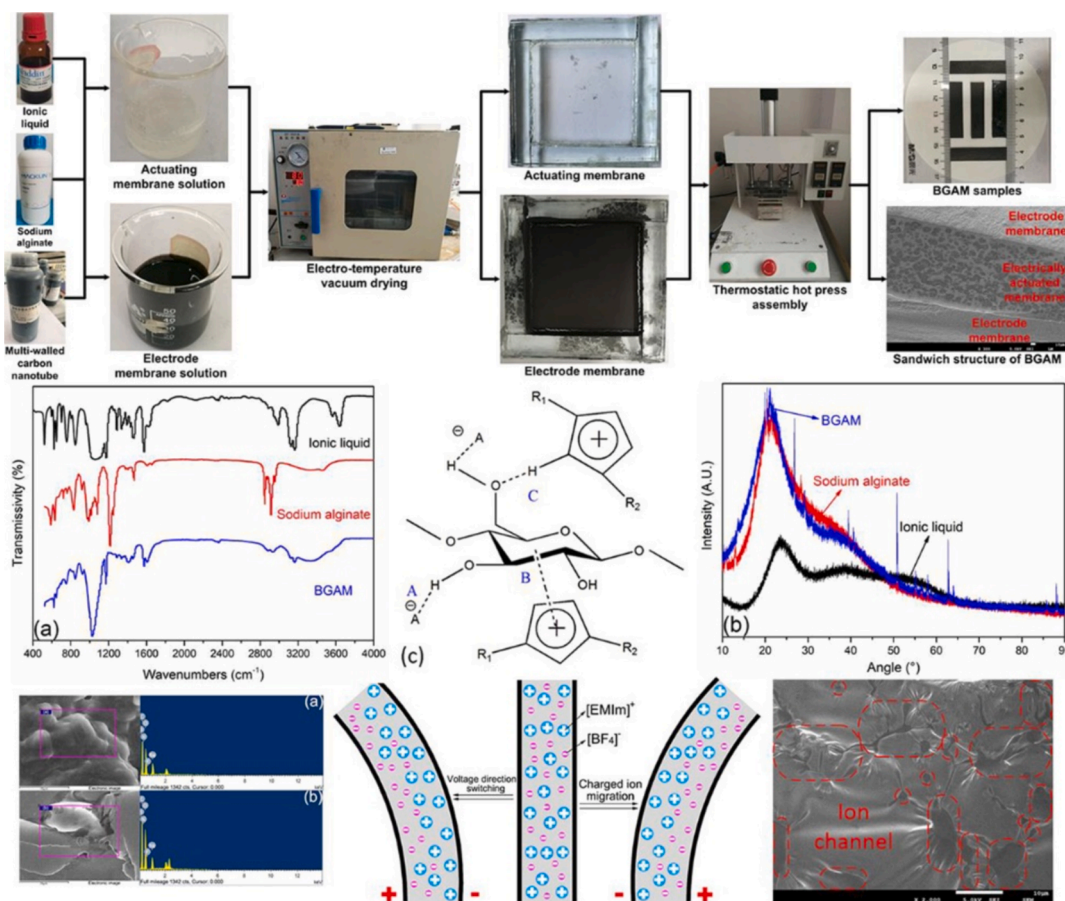


Fig. 22. A visual representation of the process used to create BGAM specimens; a diagram explaining how IL and sodium alginate react, using infrared spectrum and XRD pattern; polymer architecture; a comparison of the EDS spectra of BGAM specimens before and after performance tests that involved electrical actuation; electrical actuation mechanism of IL-doped BGAM in ambient conditions; microscopic analysis of BGAM specimen doped with 4 ml IL (2000× magnification) adapted from [210].

response exhibited a direct correlation with its electron current under a 4 V DC voltage, showcasing excellent reproducibility in electromechanical behavior and reversibility in air. Hence, this study provided useful knowledge regarding BGAM actuation and its commercial applications.

In addition to only single component systems based on alginate, different materials have been combined with alginate to alleviate its properties, referred to as multi-component systems or composites or hybrid materials. Alginate-based composites are commonly manufactured utilizing a range of materials including natural polymers like chitosan, collagen, and gelatin; synthetic polymers such as polypyrrole and polylactide; and inorganic compounds like hydroxyapatite (HA), tetraethylorthosilicate (TEOS), bioglass, ceramics, carbon nanotubes, and carbon powder [211,212]. For instance, Khan et al. [213] reported the development of IL-modified activated carbon along with sodium alginate (AC-Alg-IL) for phenol decontamination. The AC-Alg-IL beads were found to encapsulate the IL ([PC₆C₆C₆C₁₄][Br]) successfully, as indicated by the lower degradation temperature observed during Thermogravimetric analysis. Additionally, FTIR analysis showed the presence of new peaks after phenol adsorption, indicating the successful adsorption of phenol at the adsorbent surface. Compared to granular activated carbon (GAC) and activated carbon modified with sodium alginate (AC-Alg), the prepared beads (powdered activated carbon (PAC) and AC-Alg-IL) exhibited superior porous surfaces with numerous pores, resulting in higher maximum adsorption capacities (143.67 mg/g and 78.48 mg/g, respectively). The synthesized beads (AC-Alg-IL) demonstrated excellent recyclability for phenol adsorption. Additional findings revealed a strong interaction between IL and phenol, indicating efficient removal from aqueous media. This study developed a sustainable adsorbent based on IL, activated carbon and alginate polymer for efficiently and conveniently removing phenolic compounds from contaminated water.

9. ILs assisted processing of xanthan gum

Xanthan gum was discovered in the 1960 s from the bacterium *Xanthomonas campestris* [214]. The exciting set of properties of xanthan gum including rheological fingerprint, water solubility, and stability at a wide range of pH and temperature, make it a potential candidate as a stabilizing and thickening agent in the food industry [215]. Due to its fantastic synergy with other polymers, xanthan gum is extensively used as a thickening, suspending, film-forming, and emulsifying agent in drug delivery and pharmaceutical applications [216]. Several other application areas of xanthan gum include cosmetic products, such as facial creams, lotions, moisturizers, etc., tissue engineering, the petroleum industry, and water-based drilling fluids because of its gelling properties and shear thinning behavior [217]. Moreover, xanthan gum was also derivatized to enhance its properties for various biomedical and pharmaceutical applications [218]. However, the commercial application of xanthan gum involves a pretreatment and/or processing step to disturb both intra- and inter-molecular hydrogen bonding. The growing demand for environmental protection and sustainability motivates using greener pretreatment/processing methods of xanthan gum to reduce energy consumption and harmful byproducts [219]. ILs have been extensively utilized as “greener solvents” for pretreatment of polysaccharides and to assist succeeding biological or chemical modifications [220].

For instance, Liu et al. [221] reported the influence of aqueous IL (1-butyl-3-methylimidazolium chloride, [C₄C₁IM][Cl]) with different dilution ratios on the pretreatment and hydrolysis of xanthan gum. The results from hydrolysate analysis and structural characterization indicated that the presence of 1 % (v/v) IL successfully disrupted intra-molecular hydrogen bonds within xanthan gum. This disruption occurred via the binding of xanthan gum side chains with [C₄C₁IM] + . Incorporating IL could further substantially enhance the amount of superoxide and hydroxyl radicals in the system. Moreover, a small fraction of IL was needed to restrain the Maillard browning reaction, which

typically resulted in the degradation of xanthan gum. According to this research, xanthan gum can be hydrolyzed into a mixture with a high dextrose equivalent value of 29.16 % by using 0.75 % (w/v) H₂O₂, 0.4 M HCl, and 1 % (v/v) IL at 80 °C for 4 days. The hydrolysates of xanthan gum showed significant elicitor activity against soybean cotyledon, indicating its potential use in plant growth promotion. Additionally, the hydrolysates also exhibited remarkable antioxidant properties which could be beneficial in various applications. These results suggest that the hydrolysis of xanthan gum could be an effective and eco-friendly approach to valorize water-soluble polysaccharides.

Similarly, poly ILs were used to prepare an ionogel based on xanthan gum using a one-pot method, which resulted in a semi-interpenetrating network of both components in the final system [222]. 1-vinyl-3-butyl imidazolium chloride [VinylC₄IM][Cl] and poly (ethylene glycol) diacrylate (PEGDA) were photo-polymerized in the presence of xanthan gum, which produced a crosslinked network between [VinylC₄IM][Cl] and PEGDA and a crosslinked network between [VinylC₄IM][Cl] and carboxy groups of xanthan gum due to electrostatic interactions (Fig. 23). The results demonstrated that incorporating small amounts of xanthan gum (0.3–3.1 wt%) significantly improved the mechanical properties of the ionogel without compromising its ionic conductivity. Furthermore, the ionogel containing the optimal concentration of xanthan gum (2.2 wt%) exhibited exceptional ionic conductivity (0.63 S/m at 25 °C), outstanding compression strength (761.0 kPa), linear response (0–100 kPa), and a high sensitivity value of 6.86 kPa⁻¹. Hence, this study reports a novel technique for creating sustainable ionogels by using a semi-interpenetrating network. The resulting ionogels demonstrated exceptional antibacterial properties against both gram-negative and gram-positive bacteria. Moreover, the ionogels also exhibited pressure-sensitive characteristics. Therefore, this approach could pave the way for developing advanced materials with unique properties for various applications.

In another study, researchers developed a novel heterogeneous catalyst, [ImSi][PF₆]/xanthan, supported by xanthan gum and silica base. This recyclable and efficient catalyst was prepared through a simple methodology and utilized for the synthesis of acridine-1,8-diones [223]. The results indicated that the prepared catalyst displayed remarkable catalytic activity and excellent thermochemical stability. Furthermore, the developed catalyst demonstrated recyclability for up to 5 reaction cycles. The proposed methodology displayed some prominent characteristics such as cost-effectiveness, simple method, high product yield (88–96 %), and short reaction time (29–47 min) without using any toxic metals or hazardous solvent.

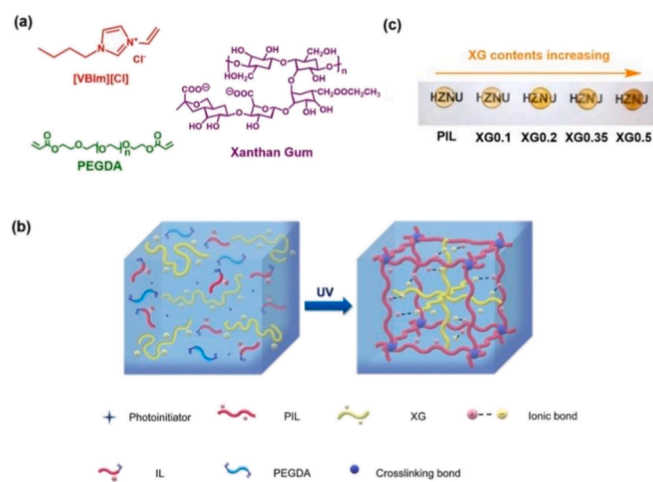


Fig. 23. Pictorial representation of preparation of ionic liquid/xanthan gum/PEGDA based ionogel using UV light. Photographs of the ionogels showing their transparency adapted from [222].

10. ILs assisted processing of leather

Leather is composed of collagen protein and is widely used to manufacture multiple products such as upholstery for automobiles, footwear, sports and garments equipment. Leather is typically obtained from animal hides/skin after several chemical processes. The pivotal chemical process in leather production is tanning which transforms animal skin into leather by stabilizing collagen fibers to prevent putrefaction [224]. However, this process is highly hazardous and results in toxic effluents due to the use of harsh chemicals during unhairing, fiber opening, tanning, re-tanning, and fatliquoring treatments. Various treatments involving chromium sulfate as well as combinations of synthetic or vegetable-based tannins and heavy metals have been investigated [225,226]. However, the chromium-based tanning method is still widely used due to its efficiency and cost-effectiveness. Various approaches have been developed to improve the chromium-based tanning process, which resulted in reduced water and chemical consumption, but the process is still an environmental concern due to the release of process effluent containing harmful levels of chromium. Moreover, unhairing and fiber-opening processes using sodium sulfide and calcium hydroxide also generate a significant portion of harmful effluent. In the literature, different sustainable approaches have been proposed based on enzymes, enzymatic and chemical crosslinking, bio-derived and ionic liquids-based polymers, and nanostructured materials as alternatives to harmful chemicals involved in the tanning process [227–229].

Liu et al. 2022 [230] reported using 1-allyl-3-methylimidazolium chloride [AllylC₁IM][Cl], and dispase enzyme (prepared from *Bacillus subtilis*) for cyclic unhairing and one-pot beamhouse for recycling of bovine waste (Fig. 24). The results revealed that the enzyme activity after using 5 times for the unhairing process in the presence of IL was higher than the enzyme activity used for unhairing in the presence of KCl, which was linked to the rapid penetration of IL-enzyme solution in cattle hides. The elastic modulus and tensile strength of IL-dispase cured leather were higher than that obtained from KCl-dispase treatment while the shrinkage temperature was comparable for both leathers. This process resulted in organic matter recovery from wastewater and non-toxic effluent due to 66.6 % lower total solids, 58.13 % lower wastewater discharge, and 97.23 % lower ammonia nitrogen. Hence, the reported process is a potential alternative to the conventional leather process to develop sustainable technology by decreasing environmental pollution.

Likewise, a PIL was synthesized to replace conventional fatliquoring

agents for the tanning process [231]. Free radical polymerization method was used to prepare a poly(ionic liquid) (p(DM-co-[DDVIM]Br) PS) from 1-dodecyl-3-vinylimidazole bromide [VinylC₁₂IM][Br], dimethylaminoethyl methacrylate (DM) and 1,3-propanesultone as monomers. The fatliquoring agent was produced by mixing industrial oil and poly(ionic liquid) in equal amounts, followed by the application of the fatliquoring agent at 50 °C on the re-tanning leather (Fig. 25). The research study suggested that a fatliquor agent based on PIL (polymerized ionic liquid) could enhance the process of fatliquoring by releasing leather fibers and improving the binding of anionic dyes. The study showed that the PIL-based fatliquor agent was highly effective, with absorption rates of 99.26 % and 99.01 % for fatliquoring and dye absorption, respectively. Potential candidates for the Cr-free tanned leather fatliquoring method were the dye and fatliquoring agent solutions, which were both transparent. Moreover, PIL-treated leather exhibited a higher K/S ratio, suggesting a more intense color on the leather surface, and demonstrated greater softness compared to leather treated with a commercial fatliquoring agent. The developed agent presents an innovative approach for producing cleaner leather, which can enhance the absorption rate of anionic compounds used in wet finishing and mitigate the environmental risks associated with chromium tanning treatments.

Alla et al. [232] explored the substitution of sodium sulfide and calcium hydroxide with a protease enzyme and an imidazolium-based IL (1-butyl-3-methylimidazolium tetrafluoroborate, [C₄C₁IM][BF₄]), for the unhairing (depilation) and fiber-opening processes of animal skin (Fig. 26). The results showed that the presence of ionic liquid ensured higher activity of enzyme which eventually reduced the time required for unhairing. Moreover, the fiber-opening and unhairing processes were performed in the same bath which resulted in less consumption of ionic liquid and water and a faster process. The proposed approach also avoided the use of deliming process which further eliminated the use of ammonium salts. The physical properties and strength of IL treated leather were observed to be similar to the leather obtained from standard procedure. However, a slight reduction in shrinkage temperature of IL treated leather was found which was attributed to the chaotropicity of the anion of IL. Hence, this study clearly shows that ILs have a great potential to be employed for unhairing process of animal skin by enhancing the activity of enzymes.

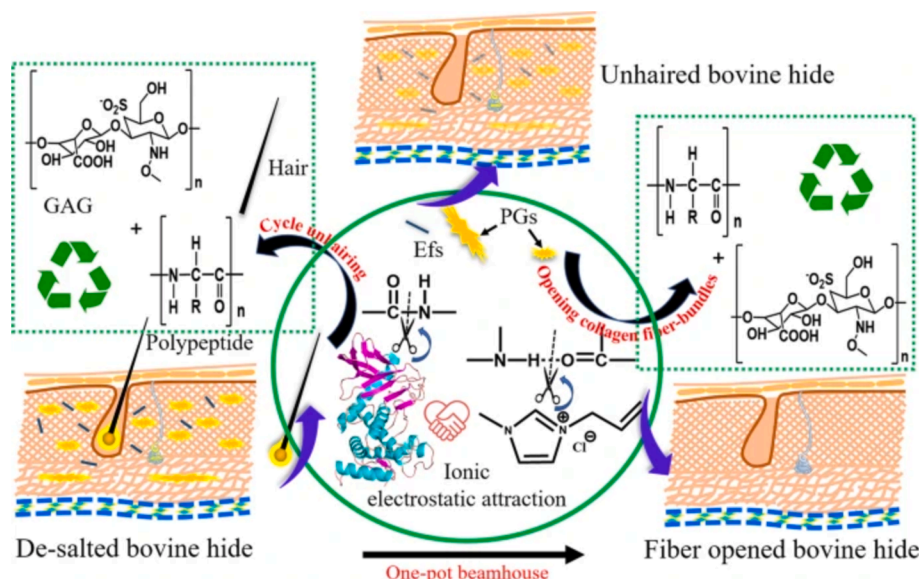


Fig. 24. Schematic illustration of IL-dispase synergistic one-pot beamhouse for cyclic unhairing and fiber-opening processes adapted from [230].

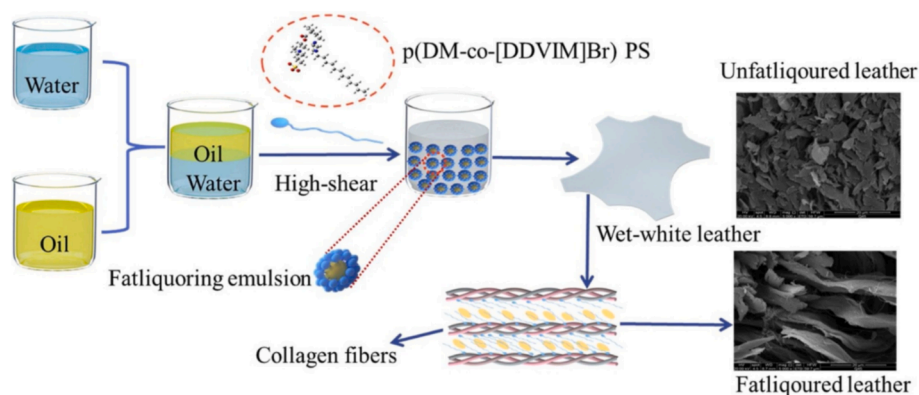


Fig. 25. Pictorial representation of the preparation and application of PIL-based fatliquoring agent adapted from [231].

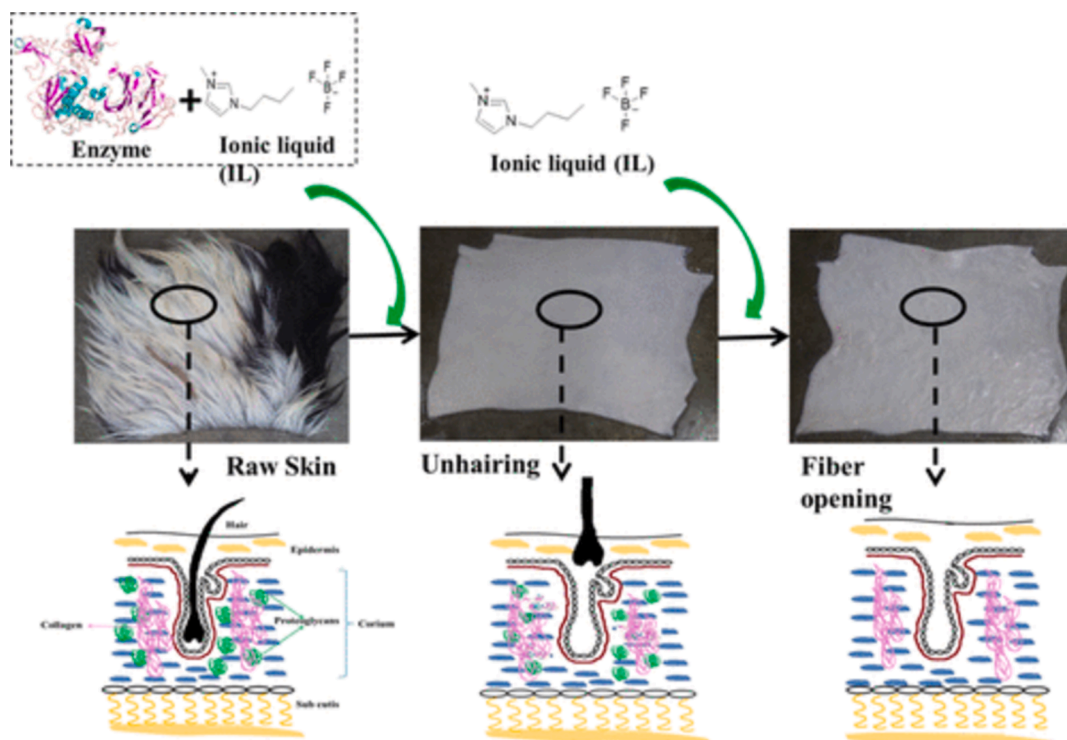


Fig. 26. Pictorial representation of the interaction mechanism between IL and animal skin adapted from [232].

11. Novel trends in ionic liquids for future development

11.1. Nanoconfined ILs

In many instances, the liquid state of ILs poses limitations on their application as high-tech materials and solvents, particularly in devices requiring a solid form. The fluidic nature of ILs presents various challenges, including high cost, unique characteristics such as high viscosity and low gas diffusivity, storage, portability, potential leakage, and difficulties in product refining and recycling. Encapsulating a solid and thin layer of IL within a highly porous matrix, allowing the entire IL mass to be utilized, can significantly decrease the amount of IL needed for a given application scale. Confining ILs within nanospaces ensures the activation of novel chemistries across diverse chemical and material domains, facilitating the easy transformation of homogeneous liquids into heterogeneous ones and reducing the distance between organic and inorganic chemistry [238].

11.2. Renewable and biocompatible ionic liquids

It has been reported that several ILs or their degraded constituents are toxic [239–241]. Conversely, the environmental impact of some ILs is observed to surpass the expected level when their entire life cycle was considered, particularly due to use of petroleum-derived building blocks in production [242]. One strategy to enhance the green feature of ILs is the employment of renewable raw materials in their synthesis. Various building blocks derived from primary renewable resources can serve as IL precursors, including amino acids, proteins-based amino alcohols, sugars from polysaccharides like cellulose, starch, chitin, lignin-derived aromatic aldehydes, and diverse matrices such as algae-derived oils or fatty acids from vegetables. Consequently, to establish a closed-loop biorefinery capable of sustaining its own demand for process solvents while producing bioderived platform chemicals, biobased ILs would ideally process their source materials. However, the stability of such ILs under relevant process conditions must be ensured for their suitability in a specific task.

11.3. Immobilization of ILs

While having enormous potential at the laboratory scale, applications of ILs employing homogeneous systems have not shown a significant promise in large-scale industrial processes because of various difficulties raised due to large amount of ILs used. Recently, the concept of supported IL phase materials has emerged, integrating IL with heterogeneous support materials to immobilize a film of IL on a solid matrix. This immobilization process aims to transfer the unique characteristics of ILs to a solid catalyst, thereby combining the advantages of ILs with those of heterogeneous support matrices and active species or catalytically functional groups. Several reviews have discussed the interactions of ILs with solid matrices and the utilization of supported ILs in various applications, providing a comprehensive introduction to their attributes and potential applications [240].

11.4. Multiscale studies on ionic liquids

A fundamental understanding of hierarchical structures and behaviors is essential for the continued advancement of IL-mediated biorefineries. This requires adopting multiscale approaches to facilitate integration among various length scales. Chemical engineers dream of having a consolidated design that combines the macroscopic process design using novel ILs with the microscopic molecular level. To better understand IL complex structures and unique characteristics and increase their industrial employment, the multiscale strategy is a potential method. Fortunately, advancements in computer modeling and simulation during the last 20 years have made it possible to conduct multiscale research, either in addition to or instead of experimental. Several modeling techniques have been developed in this respect, including micromechanics, Ginzburg-Landau theory, PDD BD, QM/MM, MD, CG, LB, and FEM. Actually, these studies show approaches at various length scales, including molecular (such as ions, atoms), mesoscale (such as

clumps, particles, conglomerates), and macroscale (such as unit operations, environmental impact, thermodynamic prediction, process optimization, LCA), and they have shown varying degrees of success in addressing various aspects of ILs [241,243] (Fig. 27).

11.5. Poly ionic liquids

Polymerized ionic liquids, also known as poly ionic liquids (PILs), are polyelectrolytes consisting of a polymeric backbone with ILs incorporated as recurring monomer units. In this manner, from monomers to oligomers then to high molecular weight matrix, distinguished characteristics of ILs are passed on to polymer chains. PILs afford structures and attributes that could not be achieved employing simple ILs. The concept of PILs has garnered increased attention, particularly in the last decade, owing to the design flexibility of ILs and the selective properties of polymer units, which enhance and modernize the characteristics and applications of PILs. By incorporation of numerous functional anionic and cationic IL structures including imidazolium, pyrrolidonium, pyridinium, and hexafluoro-phosphate into dynamic macromolecular polymeric matrices, a wide array of PILs have been produced broadening the structures, features, functionalities, and employment of PILs. The present emphasis of PIL research is towards better understanding of structure-property correlations, functional devices, energy and environment. It is anticipated that these activities will have a remarkable impact on PILs related research in the following years [242,244].

11.6. Temperature-responsive ionic liquids

The substitution of toxic organic solvents with ILs and their utilization as efficient catalysts in homogeneous systems have been extensively investigated [245–247]. The inherent drawbacks of homogeneous systems have led to the adoption of supported ILs. Nevertheless, challenges such as gas/liquid or gas/solid interface mass transfer limitations, as

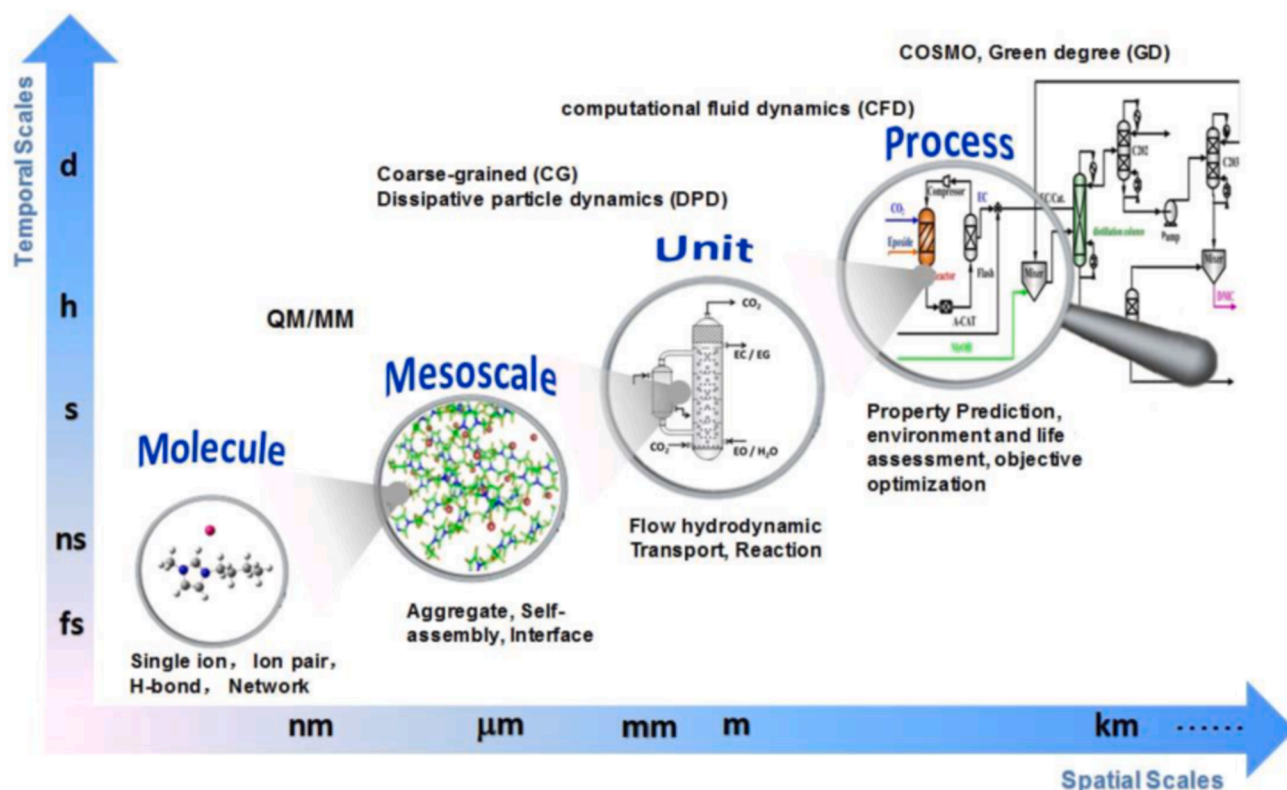


Fig. 27. Implementing a multiscale strategy with ILs addresses the need for a novel industrial design paradigm, encompassing the entire design spectrum from molecule to process adapted from [241].

well as leaching of active components in supported ILs systems, persist as significant issues. Conversely, ILs with thermoregulated properties, which maintain a homogeneous system at reaction temperature and transition to a heterogeneous phase upon temperature changes, offer a highly promising alternative to mitigate the challenges associated with conventional ILs systems. Owing to the facile tuning of reaction temperature to couple homogeneous reaction with heterogeneous separation, thermoregulated ILs can be recovered conveniently and reutilized many times. Therefore, a broad range of applications utilizing thermoresponsive ILs have been described in detail and the researchers look forward to extending the applications in other areas as well [248].

12. Key market inclinations and economic aspects of ILs technology

The commercial technological industry is driven by need and possible revenue, but it has also been influenced and permeated by the growing public awareness of sustainable products and the bio-based economy. This opens up a significant array of ILs, particularly as they may align with the requirements of the European Union's REACH (Registration, Evaluation, Authorization, and Restriction of Chemicals) law. Market landscape analyses and expansion projections indicate that the IL market is anticipated to grow at a compound annual growth rate (CAGR) of 10 %, reaching a value of 62.3 million US dollars by 2025 [18].

In a techno-economic analysis of IL pretreatment utilizing 1-ethyl-3-methylimidazolium acetate [C_2C_1IM][OAc] for a 113 million L/year cellulosic biorefinery process, it was found that the recovery of ILs constituted the most energy-intensive phase, accounting for approximately 88 % of the total energy requirement. Secondly, the main cause of operating expenses was the high cost of IL (\$2.5/kg). However, it was proposed that IL recovery progress would have a greater economic impact than IL cost reduction. Many ILs recycling methods have been documented to date, but significant energy costs and waste production continue to be the major obstacles towards their industrial adoption [249].

Prolonged synthesis times and expensive raw ingredients are needed for the aprotic ILs. Furthermore, low atom economies and laborious production of salted wastewater are brought on by salt metathesis. On the other hand, protic ILs can be made economically using straightforward acid-base neutralizations. The cost of a solvent is anticipated to nearly double with each successive step beyond its precursor. Because protic ILs are made of simple acids and bases and have small life cycle trees, they can be produced at a very minimal cost. The choice of anion has a major impact on an IL's dissolving capability for biopolymer matrices. Anions with strong fluorination, like $[OTf]^-$ or $[NTf_2]^-$, need a lot of work to synthesize and are expensive. Moreover, the high cost is also influenced by the anions' relative sizes.

13. Conclusions and outlook

- The ILs have augmented into a broad subject domain in the last decades and emerged as extremely attractive media for benign extraction and processing of animal based polymeric materials to comply with the SDGs. The up-to-date knowledge and latest advances regarding ILs-solvent mixture for separation, purification or modification of biopolymers derived from animal resources has been enumerated. The areas of application at the crossroads of polymer chemistry, materials and chemical engineering and ILs afford promise for future developments. Various ILs have been investigated with a range of animal-derived materials, with the focus primarily on imidazolium-based ILs. This preference likely stems from their well-established efficacy and widespread availability. In many applications, including the dissolution of protein constituents, imidazolium ILs with small chain substituents have consistently yielded excellent results. The characteristics of regenerated biopolymer fiber depend

on the IL employed for dissolution and the nature of the antisolvent. However, the lack of established procedures for pre-treatment, purification, extraction, and chemical alterations hinders the method of dissolving and regeneration of biopolymer matrices from IL solution which limits its widespread commercial output.

- Regenerating fiber matrices into more durable, well-organized, multi-level 1D materials with a wide range of applications will be made easier with an understanding of the mechanics underlying the dissolution and interaction of proteins in ILs. In between, the cost and sustainability of ILs need to be taken into account by chemical scientists and engineers for their next research projects. This means that new synthetic strategies for generating soft, polarizable, organic cations coupled with anions made from inexpensive, biorenewable raw materials like amines and amino acids are required, as well as completely biocompatible, if not entirely biodegradable, synthesized ILs for animal-based biopolymer dissolution.
- The role of ILs in process economics and circular bioeconomy based on animal-derived materials needs to be further developed to make industrial biomaterial production more economical and sustainable. However, solely relying on scientific progress and industrial efforts may not suffice for the successful realization of ILs-assisted bioeconomy involving biodegradable materials derived from animal biopolymers. Therefore, it is imperative to establish indigenous government policy frameworks that encompass the collection and supply of animal waste from its source to the respective industries and research institutions, potentially through subsidized payment schemes.
- The processing of animal-derived polymers by ionic liquids spans a wide range of remarkably diverse fields of study, and because sustainability is inherently interdisciplinary, collaboration amongst researchers is essential. Because of this, the long-term viability of ILs in biorefineries that use animal produced waste requires a comprehensive strategy to ensure that future expansion is maximized along the whole supply chain while maintaining or enhancing their environmentally friendly footprints. Compared to lignocellulosic waste materials, the varied fabrication of biopolymers from animal-based resources has not been thoroughly examined using ILs. Nonetheless, we firmly believe that ILs hold tremendous potential to facilitate the development of safer, more effective, and environmentally friendly methodologies and processes for sustainable development.

CRedit authorship contribution statement

Hamayoun Mahmood: Writing – original draft, Supervision, Project administration, Methodology, Data curation, Conceptualization. **Atif Khan:** Writing – review & editing, Writing – original draft, Methodology, Data curation. **Ahmad Shakeel:** Writing – review & editing, Writing – original draft, Methodology, Data curation. **Maliha Uroos:** Writing – review & editing, Writing – original draft, Methodology. **Hom Nath Dhakal:** Writing – review & editing, Writing – original draft, Methodology, Data curation. **Abdulaal Zuhayr Al-Khazaal:** Writing – review & editing, Writing – original draft, Data curation. **Muhammad Moniruzzaman:** Writing – review & editing, Writing – original draft, Supervision, Project administration, Conceptualization.

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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