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Ionic liquid-based antimicrobial materials for water treatment, air filtration, food packaging and anticorrosion coatings

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A B S T R A C T

Efforts to widen the scope of ionic liquids applications across diverse research areas have flourished in the last two decades with developments in understanding and tailoring their physical, chemical, and biological properties. The promising applications of ionic liquids-based materials as antimicrobial systems is due to their ability and flexibility to be tailored in varying sizes, morphologies, and surface charges. Ionic liquids are also considered as greener materials. Common methods for the preparation of ionic liquid-based materials include crosslinking, loading, grafting, and combination of ionic liquids with other polymeric materials. Recent research focuses on the tuning of the biological properties to design novel ionic liquids-based antimicrobial materials. Here, the properties, synthesis and applications of ionic liquids and ionic liquids-based materials are reviewed with focus on antimicrobial activities applied to water treatment, air filtration, food packaging, and anticorrosion.

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Abbreviations: AMIB IL, 1-aminoethyl-3-methylimidazolium bromide ionic liquid; [A336][NAA], aliquat 336-based methyltrioctylammonium naphthylacetate; BCILs, benzalkonium chloride ionic liquids; [EMIm]Cl, 1-ethyl-3-methyleimidazolinium chloride; GA-IL, gum Arabic-based ionic liquid; HATPS, aminothiazolyl-functionalized phosphonium; IL(Br)/PDA@PES, 3-(12-mercaptododecyl)-1-methyl-1H-imidazol-3-ium bromide ionic liquid/polydopamine@ poly(ethersulfone); IL-g-PA6, ionic liquid grafted polyamide 6; IL-g-SS, ionic liquid grafted stainless steel; IZD, inhibition zone diameter; PAILMs, polyamide ionic liquid membranes; PDA, polydopamine; PDDA, poly(diallyldimethylammonium chloride); PES-g-hbSPG-QPEI, poly(ether sulfone) grafted hyperbranched polyglycerol-quaternized polyethylenimine ionic liquid; PES-g-IL, polyethersulfone grafted ionic liquid; PIL(Br), poly(1-(2-methacryloyloxyhexyl)-3-methylimidazolium bromide) ionic liquid; PILMs, poly(ionic liquid)s membranes; PIL(Tf_2N), bis(trifluoromethanesulfonyl)amide poly ionic liquid; PIP-TMC, piperzine ~ trimesoyl chloride; PLA/PHB, polylactide/polyhydroxybutyrate; PMMA/IL-TiO_2, poly(methyl methacrylate)/ionic liquid-TiO_2; POM-ILs, polyoxometalate-ionic liquids; POM-SILP, polyoxometalate supported-ionic liquid phase; [Py1,4]TfO, 1-butyl-1-methyl pyrrolidinium trifluoromethyl sulfonate; RTILs, room temperature ionic liquids; SILMs, supported ionic liquid membranes; [VBIM][Cl], 1-vinyl-3-butylimidazolium chloride ionic liquid; [VBIm][BF_4] or IL (BF_4), 1-vinyl-3-butylimidazolium tetrafluoroborate ionic liquid; ZnO@IL, ionic liquid coated on ZnO nanoparticles.

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1. Introduction

Over the past decades, microbial infections have become a major public health issue that has diverted global attention towards the development of new drugs. Additionally, the increase in antimicrobial resistance has emerged as another challenge to tackle [1]. A global action plan has therefore been formulated by the World Health Organization (WHO) about antimicrobial resistance with a list of antibiotic-resistant priority pathogens [2]. More responsible use of antibiotics is recommended along with the development of effective and new drug treatments, which is not an economically suitable alternative to pharmaceutical companies due to the time and expenses involved [3]. Although it is practically unavoidable to completely stop the appearance of resistance towards the new drugs considering the fact that antibiotics might not be the only cause for the surge in resistance. Hence, attaining the contemporary knowledge of the microbiological challenge is essential for the effective future design of antimicrobial agents which can circumvent or reduce the occurrence of multidrug resistance.

Renewed efforts have been deployed to overcome the associated danger of antimicrobial resistance such as the use of material with inherent antimicrobial features namely carbon-based nanomaterials, silver nanoparticles, and polymers [4]. Among others, the application of antimicrobial polymers has sprung up as a promising and attractive system; a large number of polymers have been broadly studied owing to their wide spectrum of activity bearing different groups such as pyridinium, triazolium, quaternary ammonium, guanidinium, imidazolium, or phosphonium groups [5]. The main underlying mechanism basically involves the electrostatic interactions of the negatively charged bacterial membrane with cationic polymer moieties along with the contribution of hydrophobic segments which play an important role in membrane rupture and cell lysis [1].

Ionic liquids have emerged as a novel class of low temperature (<100 °C) molten salts which are composed of weakly coordinated cations and anions [6]. Although ionic liquids are discovered a long time ago, however, the researchers have been mainly interested in this appealing class as a low melting point 'fluid' salts in various industrial chemical processes as feasible substitutes to traditional volatile organic solvents as reaction media. The surprisingly diverse array of ionic liquids applications has been witnessed over the past decade [7]. The most distinct feature of ionic liquids includes low melting point, exceptional solvation potential, negligible vapor pressure, along with antibiofilm and antimicrobial activity. The chemical, physical, and biological properties can be tailored by the selection of different anions and cations. This distinctive, flexible, and unique characteristic in combination with the capability for numerous functionalities, could have the potential to offer innovative antimicrobial strategies in design which is needed to deal with the increasing challenge of bacterial resistance [8].

In recent years, versatile ionic liquids-based antimicrobial polymeric materials are mainly synthesized by combining ionic liquids by crosslinking, loading, and grafting them with other polymeric materials, nanomaterials, or solvents, which possess antimicrobial properties [1]. The ionic liquids-based antimicrobial materials have been used in various applications such as water treatment [9], air filtration [10], food packaging [11], anticorrosion [12], and coating processes [13] including biomedical applications (Table 1).

The antimicrobial properties of ionic liquids have been known for more than two decades, which mainly exhibits broad spectra of activity for Gram-negative and Gram-positive bacteria as well as fungi and mycobacteria. In general, the antimicrobial activity of ionic liquids increases with increasing lipophilicity, usually influenced by stretching the alkyl chain (chain-length effect) while the anions play a secondary role [8]. The combination of ionic liquids with other materials is known to enhance the intrinsic properties of pristine materials to destroy a wide range of microorganisms [14]. The previously published reviews mainly focus on the structural properties of ionic liquids [8] and poly(ionic liquid)s [1] and enlisted the alternative methods to obtain antibacterial materials based on ionic liquids. The lack of any detailed review related to this emerging topic prompted us to deliberate the environmental applications of ionic liquids-based antimicrobial materials.

This review presents several methods for the preparation of ionic liquids-based materials followed by a detailed discussion about their physicochemical and antimicrobial properties. Finally, the applications of various ionic liquids-based antimicrobial materials in the aforementioned environmental applications are highlighted with future perspectives.

2. Preparation of ionic liquids

The preparation of ionic liquids was initiated by Walden in 1914 with the synthesis of ethyl ammonium nitrate, $[EtNH_3][NO_3]$ by a reaction between concentrated nitric acid and ethylamine [25]. After a long gap, in between the 70s and 80s, the alkylimidazolium halogenoaluminate ionic liquids, termed "first-generation ionic liquids" were developed via reaction between aluminum halides and imidazolium halides [26]. In 1992, air and water-stable imidazole-based, ionic liquids with a wide range of applications were developed [27] and were termed "second generation ionic liquids". Assorted synthesis strategies such as photo-crosslinking, radical polymerization, radiation-induced graft polymerization, light-induced polymerization, reversible addition fragmentation chain transfer (RAFT), atom transfer radical polymerization (ATRP), electrospinning, condensation reaction, metal-free organocatalytic ring-opening polymerization, and solution blending, have been deployed for their preparation [1]. Based on the chemical composition, ionic liquids are categorized as simple ionic

Table 1
Various biomedical applications of ionic liquid-based antimicrobial materials.

Ionic liquid	Added component	Antibacterial activity against	Application	Ref.
Pyrrolidinium	Acrylonitrile, styrene	*Escherichia coli, Staphylococcus aureus*	Wound healing	[15]
1,3-Bis(2-hydroxyethyl) imidazolium bromide	Polypropylene glycol	*Escherichia coli, Staphylococcus aureus, Pseudomonas aeruginosa*	Wound healing	[16]
1-Vinyl-3-butylimidazolium bromide	PVA, acrylamide	*Escherichia coli, Staphylococcus aureus, Bacillus subtilis*	Wound healing	[17]
1-Vinyl-3-butylimidazolium bromide	Polyethylene glycol dimethacrylate, *N,N*-methylene-bis-acrylamide, methyl methacrylate, and acrylamide	*Escherichia coli, Staphylococcus aureus*	Wound healing	[18]
Pyrrolidinium	PVA (with/without tetrahydroxyborate anion)	*Escherichia coli, Staphylococcus aureus*	Wound healing	[19]
1-N-butyl-3-methyl imidazoilium bis (trifluoromethanesulfonyl)imide	BisGMA, TEGDMA, colloidal silicon dioxide	*Streptococcus mutans*	Orthodontic	[20]
1-Butyl-3-methylimidazolium hexafluorophosphate	PLGA or Soluplus®	*Staphylococcus epidermidis*	Periodontal	[21]
Poly(ionic liquid) graft copolymer	–	*Escherichia coli*	Drug delivery	[22]
Trimethylammonium	poly(meth)acrylates	*Escherichia coli*	Drug delivery	[23]
1-Butyl-3-methylimidazolium hexafluorophosphate	PLGA	*Staphylococcus epidermidis*	Drug delivery	[24]

Abbreviations: bisGMA; bisphenol A glycidyl methacrylate, PLGA; poly(lactic-*co*-glycolic acid), PVA; polyvinyl alcohol, TEGDMA; triethylene glycol dimethacrylate.

liquids such as $[EtNH_3][NO_3]$, having unary anion and cation as constituents, and binary ionic liquids such as imidazolium-based ionic liquids (Fig. 1a) [28].

Metathesis, the exchange of halide salts or anion and acid-base neutralization reaction are the two conventional routes for ionic liquids preparation [29]. Quaternization of imidazole, amine, phosphane, and pyridine salts to respective cationic forms such as imidazolium, ammonium, phosphonium, and pyridinium occurs during a single-step ionic liquids synthesis [30], while hindrance of the desired anion directly through quaternization reaction gives rise to the multi-steps ionic liquids synthesis [23]. Fig. 1b illustrates both, the single and multi-step ionic liquids synthesis procedures, exemplified with an ammonium salt.

The metathesis reaction is commonly employed for imidazole-based ionic liquids synthesis. Depending on the solubility of target ionic liquids in water, metathesis can be divided into two reaction categories as (i) silver (Ag) salts-based reaction, or (ii) free acids or alkali metals/ammonium salts reaction. During metathesis, 1,3-di-alkyl imidazolium cation reacts with anions derived from sodium (Na), potassium (K), or with Ag salts of CH_3COO^-, NO_2^-, NO_3^-, SO_4^{2-}, and PF_6^-. Ionic liquids synthesized by a reaction between imidazolium halide and Ag-salts in methanol are air and water stable [31]. This is because of the lower solubility of Ag halides, notably AgI, which facilitates the separation of ionic liquids, consequently producing water-miscible high-quality ionic liquids in a good yield. However, the higher costs of Ag salts and the large quantities of solid waste generation limits their wide range of applications. The sulfate, phosphate, or sulfonate-based ionic liquids with traces of imidazole salts or acids can be prepared by neutralization of a base with a Bronsted acid or by direct alkylation of alkylimidazole. Thus, high purity ionic liquids cannot be developed through this procedure [32]. The acidic ionic liquids are synthesized through an addition reaction between Lewis acid ionic liquids ($AlCl_3$ based salts) or metal halides such as $FeCl_3$, CuCl, and $InCl_3$, and halide salts [33]. The role of dimethyl carbonate for methylation as an alternate to alkyl halides during ionic liquids synthesis has been reported [34]. Avoidance of generation of by-products and absence of halide (as an impurity in ionic liquids) are the major merits of using dimethyl carbonate in ionic liquids synthesis. However, the presence of acid and ammonium salts restricts its use for ionic liquids synthesis [34]. A new class of highly viscous and conductive functionalized ionic liquids with low thermal stability has been generated by the reaction between zwitterions and acids [35]; zwitterions are formed by a single-step ring-opening reaction of sultones [36]. Along these conventional routes, some non-conventional solvent-free

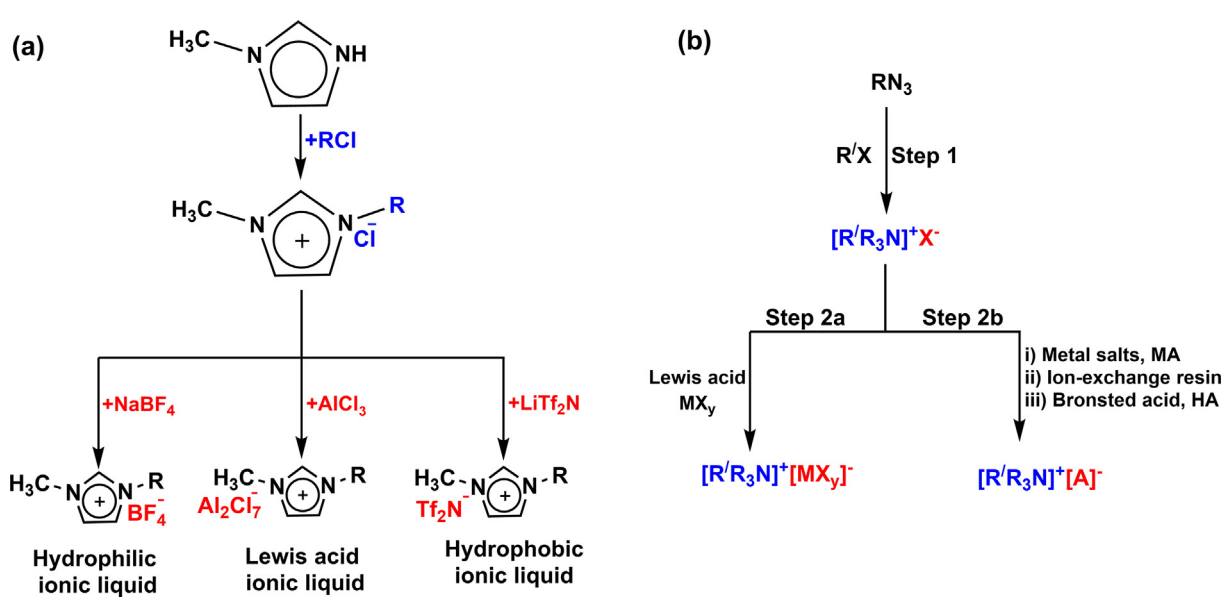


Fig. 1. The synthetic pathway for ionic liquids based on (a) imidazolium and (b) ammonium salt.

synthesis procedures assisted by microwave irradiations [37] and ultrasound waves [38] have also been reported for the synthesis of ionic liquids. Compared to conventional ionic liquids synthesis routes, these non-conventional routes are rapid, selective, and environmentally benign due to the absence of any solvents during their synthesis.

A series of picolinium-based ionic liquids were synthesized through microwave irradiations (in a controllable single-mode microwave reactor) and compared with the conventional alkylation procedures using toluene as a solvent [39]. Comparatively better ionic liquids yield (76–91%) was observed through solvent-free microwave-assisted synthesis route. Likewise, camphor-derived chiral imidazolium [40] and amino acid-based [41] ionic liquids were also synthesized through microwave irradiations and solvent-free ultrasound-assisted synthesis exemplified by *N*-methyl-2-pyrrolidinium hydrogen sulfate-based ionic liquid [42]. Highly pure 1-butyl-3-methylimidazolium salts-based ionic liquids with better yield were synthesized through the ultrasound-assisted synthesis procedure [43]. Also, the synergic use of microwave and ultrasound in the synthesis of second-generation ionic liquids has been reported by a reaction between chloroalkanes and heterocycle compounds (1-methylimidazole pyridine and 1-methylpyrrolidine) [44].

3. Physicochemical properties

In generally, ionic liquids are thermally stable, non-flammable, conductive, and barely exhibit vapor pressure [45]. Based on their wide range of applications, the physicochemical properties of ionic liquids can be tuned by modulating their molecular structure [46]. The specific physical and chemical properties of ionic liquids are exemplified and appended below with their significant properties being summarized in Table 2.

3.1. Melting point

The key criteria for ionic liquids are melting point (T_m), glass transition temperature (T_g), and decomposition temperature (T_d) which depend on a balance between ion structural symmetry, chain flexibility, and charge accessibility [56]. At standard atmospheric pressure, the ionic liquids in the temperature range 200–300 °C exist in the liquid state [57]; instead of melting point, most of the ionic liquids exhibit glass transition temperature. Also, some room-temperature ionic liquids such as 1-ethyl-3-methylimidazolium tetrafluoroborate, [C_2mim][BF_4], below the melting point, exist as a supercooled liquid [58]. The melting point of ionic liquids is different from typical salts. For example, sodium chloride (NaCl) and potassium chloride (KCl) have respective melting point magnitudes of 801 and 772 °C compared to chloride, Cl^- ions based ionic liquids with suitable organic cations like 1,3-dimethylimidazolium chloride, [MIMM][Cl] (with the melting point: 125 °C) and 1-butyl-3-methylimidazolium chloride, [BMIM][Cl] (with the melting point: 65 °C) [30]. In the case of NaCl and KCl salts, both ions (cation and anion) are small, spherical, and have closely packed geometry, which results in extremely strong Coulombic forces, consequently have a high melting point magnitude. Contrarily, the ionic liquids with Cl^- ion attached to large and asymmetric organic counter ions (cation) have low lattice energy, resulting in low melting point magnitude [30]. Likewise, protic ionic liquids with alkylammonium salts have melting point >100 °C. However, the incorporation of carboxylic acids to alkylammonium cation and most of the salts with an imidazolium cation have very low melting point magnitude [53].

3.2. Viscosity

The viscosity (η) of ionic liquids is mainly determined through their tendency to form a hydrogen bond, by the strength of van der Waals interactions among ions, and Coulombic interactions [59]. It is inversely proportional to diffusion rate. The increase in temperature and the presence of impurities (even in trace amounts) can drastically decrease the viscosity of ionic liquids. Highly viscous ionic liquids limit ions' movement compared to less viscous. Thus, for applications as a solvent, less viscous ionic liquids are desirable as they can increase mass transfer rates, while ionic liquids with higher viscosity favor lubrication and supported membrane separation applications. Generally, the viscosity of ionic liquids at room temperature ranged between 10 and 2000 cP [60]. However, ionic liquids of tri-alkylimidazolium and ammonium families displayed viscosity between 2000 and 3000 cP, while ionic liquids of the phosphonium family are highly viscous with viscosity reaching ~16,000 cP. In addition, the viscosity of the ionic liquid is directly proportional to the alkyl chain length of the cation. For example, the viscosity of 1-alkyl-3-methylimidazolium bistriflamide, [Cnmim][Ntf_2] ionic liquids where alkyl chain length (ranges between 2 and 8) increases from 29 to 108.4 cP with an increase in alkyl chain length [61]. Also, the viscosity depends on anions type, as observed among ionic liquids with the same cation chains and series of different anions. The observed viscosity is in the order: $[CH_3COO]^- > [PF_6]^- > [C_1SO_4]^- > [C_2SO_4]^- > [BF_4]^- > [OTf]^- > [NTf_2]^-$. This change in viscosity might be due to the ability to form weak hydrogen bonds with the cation chain. The $[NTf_2]^-$ based ionic liquids combine better charge delocalization with greater chain flexibility, which increases the chain mobility so that the viscosity is lower than that of other anion-based ionic liquids [61]. Thus, ionic liquids with anions of high structural symmetry or spherical shape are more viscous.

3.3. Conductivity

The inherent motion of ions present in ionic liquids under the electric field generates conductivity [62]. At room temperature, the conductivities (σ) of ionic liquids are typically ranged between 0.1 and 30 $mScm^{-1}$. The conductivity is directly proportional to temperature and mobility, and inversely proportional to the viscosity of ionic liquids [63]. The point was confirmed by a sharp increase in 1-ethyl-3-methylimidazolium tetrafluoroborate, [C_2C_1 im][BF_4] conductivity from ~16 to ~120 $mScm^{-1}$ with a rise in temperature from 25 to 150 °C

Table 2
Physical and chemical properties of some ionic liquids.

Ionic liquid	Melting point (T_m) (K)	Viscosity (η) (cP)	Conductivity (σ) ($mScm^{-1}$)	Density (ρ) (gcm^{-3})	Surface tension (γ) (mNm^{-1})	Reference
[C_2C_1 im][Cl]	361.75	48.3	2.49	11.124	–	[45]
[C_4C_1 im][Cl]	314.1	30.0	10.0	10.800	54.70	[47]
[C_4C_1 im][BF_4]	190.15	100	35	12.035	43.84	[48]
[C_4C_1 im][PF_6]	283.15	258	14.6	13.709	37.28	[49]
[C_4C_1 im][NTf_2]	270.15	49.0	39.0	14.397	37.0	[50]
[C_4pyr][NTf_2]	299.0	61.0	30.7	14.360	33.4	[51]
[$EtNH_3$][NO_3]	282.15	32	269.0	12.160	47.30	[52]
[$EtNH_3$][HSO_4]	313.15	128	44.0	14.380	56.30	[53]
[N_{2226}][NTf_2]	287.1	187	8.87	12.886	34.10	[54]
[P_{66614}][NTf_2]	206.1	336.7	–	10.661	30.90	[55]

[6]. The relationship between conductivity and viscosity has been well demonstrated by Walden plot [64]. Usually, an increase in alkyl chain length decreases the conductivity of ionic liquids; for example, the conductivity of protic ionic liquids with alkylammonium and alkylimidazolium cations decreases with an increase in alkyl chain length [53]. The conductivity behavior of acidic ionic liquids was analyzed by Vogel-Tammann-Fulcher Eq. [65]. The ionic liquids with tetraalkylammonium, pyridinium, pyrrolidinium, and piperidinium as cations are classified as lower (≤ 5 $mScm^{-1}$) conductivity ionic liquids, while ionic liquids with imidazolium cations are classified as the ionic liquids with higher (≤ 14 $mScm^{-1}$) conductivities [63].

3.4. Density

Density is a crucial parameter for the simulation of heat and mass transfer, and hydrodynamics calculations [60]. It can be significantly affected by the presence of impurities and is used as a factor to ascertain the purity of ionic liquids [66]. The density of ionic liquids is often measured by a U-shape vibrating tube densimeter operating in static mode [67]. Under ambient conditions, the densities of ionic liquids generally varied between 0.8 and 3.3 g cm^{-3} [68]. The ionic liquids' density varies by the choice of anion and cation. The cations containing hydroxyl or ether groups form high-density ionic liquids, while high molecular masses anions generally led to higher density ionic liquids with the same imidazolium cation [69]. Also, the densities of ionic liquids with similar anionic parts progressively decrease with an increase in the alkyl chain length of the cationic part. The alkyl chain length elongation increases the free volume within the ionic liquid, accordingly lowering its density [60].

3.5. Surface tension

Surface tension is a powerful means to explore a variety of arrangements and orientations of free ions at the surface through a tensiometer [6]. It also measures the cohesive energy present at an interface and is usually quantified as a force/length measurement. Generally, the surface tension magnitude of most of the ionic liquids ranged between 1.55 and 65 mNm^{-1}, which is lower than the surface tension of water (i.e. 71.97 mNm^{-1} at 25 °C) and higher than the surface tensions of many organic solvents [57]. The surface tension of many ionic liquids exhibits a linear decrease with the increase in temperature can be expressed by Eötvos Eq. [70]. Based on their properties, Table 3 summarizes the advantages and shortcomings of ionic liquids.

Table 3
Merits and demerits of ionic liquids [71].

Merits
• Number of imaginable solvents – Up to 10^{18} number of possible ionic liquids.
• Tunability – Based on applications can be tuned by varying functionalities and alkyl chain length.
• Vapor pressure – Under normal conditions low or negligible vapor pressure. Thus, non-volatile.
• Flammability – Non-flammable.
• Detachability – Volatile compounds can be easily separated.
• Stability – Stable for a wide temperature and electrochemical decomposition potential ranges.
Demerits
• Monetary value – About 5–20 times expensive than molecular solvents.
• Viscosity – Pure ionic liquids are highly viscous.
• Vapor pressure – Low vapor pressure hinder solvent separation during distillation.
• Synthesis – Generally, tiresome, costly and multi-step synthesis.
• Sustainability – Though considered as green solvents, but often toxic, non--biodegradable and non-sustainable.
• Hygroscopicity – Most of the ionic liquids are highly hydroscopic.
• Corrosiveness – Highly corrosive, thus, require special containers during commercial storage.

4. Toxicity of ionic liquids

Ionic liquids have been claimed as a greener and environmentally benign replacement for volatile and flammable organic solvents. However, their miscibility/solubility in water could be a potential threat to the aquatic ecosystem. Studies have found a strong relationship between ionic liquids' chemical structure and toxicity. Cationic moiety and the length of the alkyl chain significantly influences their toxicity level while an anion is a secondary contributor to toxicity [72]. Phosphonium and imidazolium-containing ionic liquids are highly toxic, while the least toxic ionic liquids comprise thiophenium and pyrrolidinium ions. Thus, based on cations, the toxicity of ionic liquids is as follow: phosphonium > ammonium > imidazolium > pyridinium > morpholinium > pyrrolidinium > thiophenium [73]. A reduction in imidazolium-based ionic liquids toxicity has been observed by the incorporation of oxygen or chlorine atoms into its long chain or by replacement of the alkyl chain with hydrogen atom [74].

The toxicity of imidazolium (1-octyl-3-methylimidazolium nitrate and 1-octyl-3-methylimidazolium chloride) and pyridinium (N-octyl-3-metylpyridinium chloride and -octyl-3-metylpyridinium bromide) containing ionic liquids on a marine diatom, *Phaeodactylum tricornutum* has been investigated; significant inhibition in growth and photosynthesis was observed [75]. The toxic effect of chiral enantiomers of 1-alkyl-3-methylimidazolium tartrates (D-(+)-RMIM T and L-(+)-RMIM T) on algae, *Scenedesmus obliquus* has been examined; chiral ionic liquids caused oxidative stress in *S. obliquus*. In addition, the L-(+)-RMIM T enantiomer was comparatively more toxic than D-(+)-RMIM T enantiomer [76]. The toxicity of butyl- and octyl-substituted ionic liquids was assessed on *Chlorella vulgaris*, *Scenedesmus quadricauda*, and *Botryococcus braunii* where octyl-substituted ionic liquids were comparatively more toxic to algae than butyl- counterparts [77]. The toxic effects of ionic liquids with different cations viz. *N*-ethyl-pyridine alanine, tetraethyl phosphine l-alpha-amino propionic acid salt, 1-ethyl-3-methyl-imidazolium alanine, and tetraethyl ammonium 1-alpha-amino propionic acid salt have been assessed on *Triticum aestivum* seedlings. Cation type and ionic liquids solution concentrations significantly influence the growth, chlorophyll content, and nutrient uptake of *Triticum aestivum* seedlings, while *N*-ethyl-pyridine alanine contributed to relatively stronger toxicity [78].

In another study, silkworm larvae were used as an in vivo model to assess the biotoxicity of 1-octyl-3-methylimidazole chloride, *N*-octyl-3-methylpyridien chloride, and 1-octyl-3-methylimidazole tetrafluoroborate. It was observed that ionic liquids could induce lipid peroxidation and cellular damage, which may be the primary reason for their toxicity [79]. The potential hazardous effects of ionic liquids on the human subject was studied by the evaluation of cytotoxicity through in vitro studies of imidazolium-based ionic liquids on the human tumor cell line HeLa. Results showed 4–5 fold lower EC_{50} values compared to common organic solvents [80].

5. Antimicrobial activity of ionic liquids

The World Health Organization (WHO) is working to establish a globalization program to combat microbial resistance and has compiled a list of priority pathogens for antibiotic resistance [81]. To counter the threat of antimicrobial resistance, various systems have been developed as alternatives to antibiotics, including the introduction of antimicrobial and promising ionic liquids [82]; their performance is comparable to that of ordinary antibiotics. Conventional antibiotics generally target bacteria with their antibacterial activity [83] and their mechanisms of action can be categorized as follow: (i) interfering with the cell wall synthesis, (ii) inhibiting the protein synthesis, (iii) inhibiting nucleic acid synthesis, (iv) interruption with the structure of the bacterial membrane, and (v) inhibition of metabolic pathways/bacterial enzymes [84].

In contrast, ionic liquids mainly disrupt the bacterial membrane by interacting and creating pores, cell leakage and death occur with their rapid action [85]. These membrane alterations, which lead to bacterial death, can be traced by the effect of ionic liquids on membrane properties. Ionic liquids can alter membrane properties such as membrane potential, viscoelasticity, phospholipids alignments, and ultimately the membrane fluidity. The latter affects the transport of molecules, their distinction, migration, adhesion and mechanical transport, and even the diffusion rate and stability of proteins within the membrane. The combination of these changes lead to bacterial cell death [86].

Operational viewpoint of the antibacterial effect of ionic liquids entails their easy attachment to the bacterial wall (mainly constructed by peptidoglycan), and complex cross-linked cell walls have occurred. In bacterial studies, it should not be overlooked that prokaryotic cells, unlike eukaryotic cells, do not have the uptake ability of external substances through endocytosis or pinocytosis, but uptake occurs through active/passive penetration into the cell wall [87]. Therefore, ionic liquid-based materials can easily act as a binding agent to the cell wall to produce antibacterial properties. In a study, the 1-vinyl-3-dodecyl imidazole as a cationic ionic liquid and chlorin e6 (containing carboxyl functional groups) as an anionic segment of an assembled compound was reported with dual antibacterial function. The cationic part of the ionic liquid attaches to the peptidoglycan wall and the anionic part is deprotonated in acidic media, illustrating an enhanced effect for eliminating the bacterial infection [88].

Besides the mentioned conditions, various structural features of ionic liquids such as chain length can affect their antibacterial performance. Increasing the alkyl chain length is an important and influential factor for ionic liquids, often up to a certain threshold or critical point, thus enhancing the antibacterial efficacy [89]; enhanced hydrophobic nature of the ionic liquid ensue wherein longer carbon chains (excess of 10 carbon atoms) destabilizes the cell membrane by penetrating the phospholipid bilayer and inducing membrane disruption [90]. In one study, the antibacterial properties of alkylimidazolium and alkoxyimidazolium lactates with various alkyl chains, viz.,11–12, and 12–14 carbon atoms respectively, were investigated, which also confirm the mentioned effective factor [91]. In a nutshell, the threshold value (i.e. the best chain length) for a specific ionic liquid depends on its structure and the final platform (i.e. hydrophobicity, crosslinking density of the final fabricated polymer, and composite) [89]. Deployment of such ionic liquids with lengthy alkyl chains, have a tendency to aggregate in solution resulting in amphiphilic micelles development [92].

The antimicrobial efficacy and antibiofilm activity of imidazolium-based ionic liquids against Gram-positive and Gram-negative bacteria (e.g., *Staphylococcus aureus*, *Escherichia coli*, *Pseudomonas aeruginosa,* and *Enterococcus faecalis*) depends on the length of the alkyl chain and adjusting the head group, leading to enhanced lipophilicity, culminates in interaction and disruption of the bacterial membranes [93]. However, it reduces the surface tension with enhanced expectation of the interaction with the cell membrane, followed by infiltration of the compound into the bacterial membrane for efficient growth inhibition [94].

Other ionic liquids, such as those contain fumarate anion, are soluble in both polar organic solvents and aqueous media as well as in the common ionic liquids, e.g. the case of [BMIM][MRF] ionic liquid. The antibacterial activity here is related to the properties of fumarate anions, and the alkyl chain length and depends on the assessed microorganisms, (e.g., alkyl group containing 4 carbon atoms exhibit maximal activity against Gram-negative *Escherichia coli*, and the activity decreased with additional increase in the chain length. On the other hand, in the case of Gram-positive *Bacillus subtilis* higher alkyl chain length display better antimicrobial activities. These ionic liquids are more active relative to the commercially available potassium sorbate as a reference for antimicrobial activity in the case of zone diameters of the growth inhibition [91]. Combining the hydrophobic and hydrophilic properties of fumarate anion ionic liquids, allows it to have high antibioactivity along with appropriate water solubility. The hydrophobicity of such ionic liquids can be adjusted by altering the alkyl group chain length of fumarate while hydrophilicity of the ionic liquids is determined by the carboxylate end. Thus, the hydrophilic and hydrophobic parts of the fumarate anion deal with interaction among the ionic liquids and the bacteria cell walls [91].

The ionic liquids and organic salts containing ammonia hydrolysates of penicillin G and amoxicillin reveal antibacterial properties against sensitive and resistant *Escherichia coli* and methicillin-resistant *Staphylococcus aureus* strains [95]. Additionally, their ease of synthesis is credible for their future industrial production and can be an effective mean to challenge the emerging bacterial antibiotic resistance [95].

The redox-active ionic liquids may serve as photosensitizers, which is a promising option with antibacterial properties and no detectable resistance. Ionic liquids with optical sensitizer effect can disrupt pathogenic bacteria through singlet oxygen. The thought-provoking challenges of most photosensitizers are (i) their hydrophobicity and their aggregation in the physiological environment, (ii) structural instability in the acidic environment due to acidosis of inflammatory tissue, and (iii) their ineffectiveness against Gram-negative bacteria. The negatively charged cell wall of these bacteria prevents them from being used [88]. The mechanism of destruction of cell walls from Gram-negative bacteria and Gram-positive bacteria by ionic liquids is depicted in Fig. 2.

6. Biodegradability of ionic liquids

Biodegradation, an important principle of green chemistry, is a method to determine and predict the interacting behavior of ionic liquid with its surrounding environment [96]. The biodegradation assessment of ionic liquids under biotic conditions is mainly based on standardized assays implemented by the Organization of Economic Co-operation and Development (OECD) and the International Organization for Standardization (ISO) [96]. Dissolved organic carbon die-away test (OECD 301A), CO_2 evolution (OECD 301B), MITI (I) (OECD 301C), closed bottle test (OECD 301D), and CO_2 headspace test (ISO 14593) are some of the commonly standardized assays applied to determine the biodegradability of ionic liquids [97]; remaining ionic liquids that fared poorly in these procedures tend to have a greater potential to bioaccumulate in the environmental system. Additionally, the chemical structure of ionic liquids also influences their biodegradability. Reduction in mineralization of ionic liquids by microbes from activated sludge highlights the potential risk of these salts persisting in wastewater and, consequently, contaminating the aquatic and soil systems and living organisms [97].

7. Environmental applications of ionic liquid-based antimicrobial materials

Ionic liquids are often liquid at room temperature and possess unique properties which give them the potential ability to be used in various arena especially as green solvents [98]. Ionic liquids are selected as green solvents, a fascinating alternative for volatile organic solvents, because of their excellent physicochemical properties including non-flammability, negligible vapor pressure, non-volatility, solubility, and miscibility [99].

Worldwide interest in sustainable chemical developments compatible with the green chemistry principles has led to the innovative design of antimicrobial ionic liquid-based materials. As mentioned before (Section 5), many factors affect the antibacterial activities of ionic liquids. Optimization of these factors can lead to the production of targeted ionic liquids for use in water treatment, air filtration, anti-corrosion coatings, among other appliances. To improve the physical, chemical and mechanical properties of ionic liquids, they are mixed with other materials, including metallic/non-metallic nanoparticles and polymers. In the following sections, several applications of antimicrobial materials based on ionic liquids are summarized for use in diverse fields such as water purification, air filtration, food packaging, and coating.

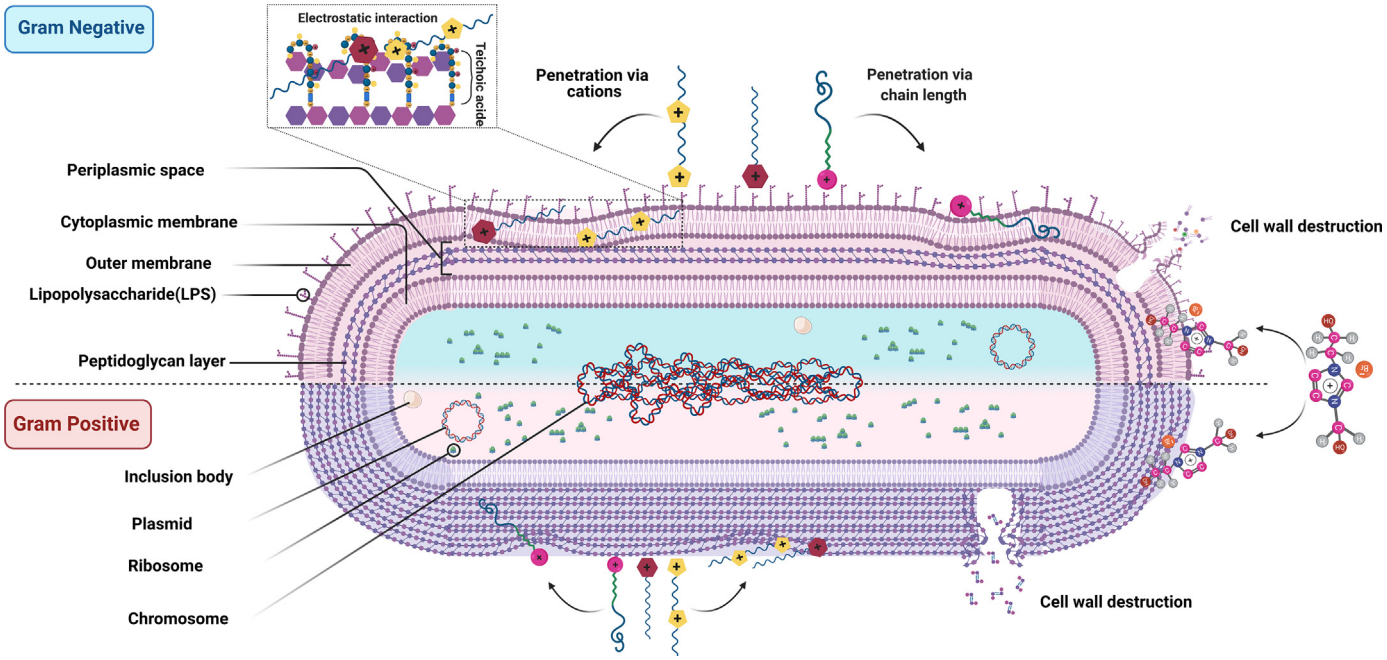


Fig. 2. Mechanism of the breakdown of cell walls of Gram-negative bacteria and Gram-positive bacteria by ionic liquids. Bacteria cell death is attributed to the penetration of ionic liquids, electrostatic interaction between the phospholipid bilayer functional groups, and ionic liquids approaching the membrane.

7.1. Water treatment

In the 21st-century, water pollution appeared as one of the most notable concerns owing to the rapid rise in population, urbanization, and industrialization. As announced by the world health organization, around 6 billion world's population will face the water scarcity problem by the year 2050 [100]. Giving due consideration to emerging water scarcity and depleting the access to freshwater with every passing day, several methods have been used in the treatment of wastewater as the main source of access to fresh water. The focus in water treatment is being diverted towards the implementation of an efficient and cost-effective method enabling resource recovery [101]. Hence, the development of specific approaches and technologies for the decontamination of contaminated water is a pressing environmental challenge. Several types of adsorbents, such as natural/synthetic polymers, and biobased materials have been utilized for the removal of pollutants from wastewaters and aqueous media; use of adsorbents with the ability to decontaminate contaminated water is an enticing topic in water treatment. Consequently, the construction of innovative adsorbents with selective adsorption and effective adsorption/desorption behavior under mild and greener conditions is essential.

Ionic liquids play a significant role in water treatment due to their high potential for the removal of a wide range of pollutants from aqueous solutions [9]. Ionic liquid-based antimicrobial materials are attractive candidates as adsorbents with dual functions of adsorbing the organic/inorganic contaminants and removing the microbial pollutants from aqueous solutions. The following examples represent the impact of antimicrobial ionic liquids as chemical and biological adsorbents in water treatment processes.

The aminothiazolyl-functionalized phosphonium (HATPS) ionic liquid was fabricated and used as an effective adsorbent for the removal of heavy metal ions (Fe^{3+}, Zn^{2+}, Cu^{2+}, and Ag^{+}) from aqueous media (Fig. 3a and b) [102]. The new HATPS ionic liquid displayed an excellent scavenging efficiency for heavy metal ions (Ag^{+}: up to 96%, adsorbate initial concentration; $C_o = 0.03$ M and adsorbent dose; m = 0.03 M), and could be simply regenerated and reused. The scavenging process occurred through complexation of heavy metal ions with deprotonated HATPS (ATPS) chelate or with HATPS as monoprotic bidentate NO^{-} ligand (Fig. 3c). Also, HATPS was applied as a spectrophotometric probe for Fe^{3+} determination in an aqueous solution with a lower limit of detection for $Fe^{3+} = 0.322$ mg L^{-1}. In addition, the scavenging outputs have been assigned as new candidates for antibiotics. Further, the antimicrobial studies exhibited high antibacterial activity for the scavenging outputs with preferable potency against *Escherichia coli* than *Staphylococcus aureus* [minimum inhibitory concentration (μg mL^{-1}): *Escherichia coli* = 25.5–128.0, and *Staphylococcus aureus* = 32.5–250.0], but with lower fungicidal activity (Fig. 3d). The significant improvement in the bioactivity of metal-ATPS in comparison to the HATPS could be related to the merger of diverse bioactive pharmacophores into the metal-ATPS structure performing synergistic antimicrobial effects (Fig. 3e).

In another study, the aliquat 336-based methyltrioctylammonium naphthylacetate, [A336][NAA] ionic liquid was synthesized and applied as an extractor with antimicrobial activity for the removal of hexavalent chromium (Cr^{6+}) [103]. The extraction of Cr^{6+} by using [A336][NAA] dissolved in toluene provided the removal efficiency of 99.5% at pH = 3. The Cr^{6+} extraction in [A336][NAA]/toluene could be related to the complexation of both $[A336]^{+}$ and $[NAA]^{-}$ with Cr^{6+} ions, and the charge balance of nitrate (NO_3^{-}) ion in the complex. The antibacterial effects of [A336][NAA] (concentration = 1000 μg mL^{-1}) were examined via inhibition zone diameter (IZD) assay against *Bacillus subtilis* (IZD = 12 mm), *Escherichia coli* (IZD = 10 mm), *Pseudomonas aeruginosa* (IZD = 13 mm), *Salmonella typhi* (IZD = 10 mm), and *Staphylococcus aureus* (IZD = 10 mm). Moreover, the ionic liquid antifungal activity was tested towards *Aspergillus niger* (IZD = 16 mm), *Candida* sp. (IZD = 9 mm), *Penicillium* sp. (IZD = 15 mm), *Microsporum gypseum* (IZD = 12 mm), and *Trichophyton* sp. (IZD = 17 mm). The bacterial and fungal inhibition results showed that the growth of all the tested organisms was significantly inhibited by [A336][NAA].

The generation of magnetic property in adsorbents for water treatment is economically more desirable because of their easier recycling. Consequently, the magnetic polyoxometalate supported-ionic liquid phase (POM-SILP: specific surface area = 100 m^2 g^{-1} and pore volume = 0.70 cm^3 g^{-1}) was prepared for the removal of organic, inorganic, microbial, and microplastics pollutants from water (Fig. 4) [104]. The composite was prepared based on the creation of polyoxometalate ionic

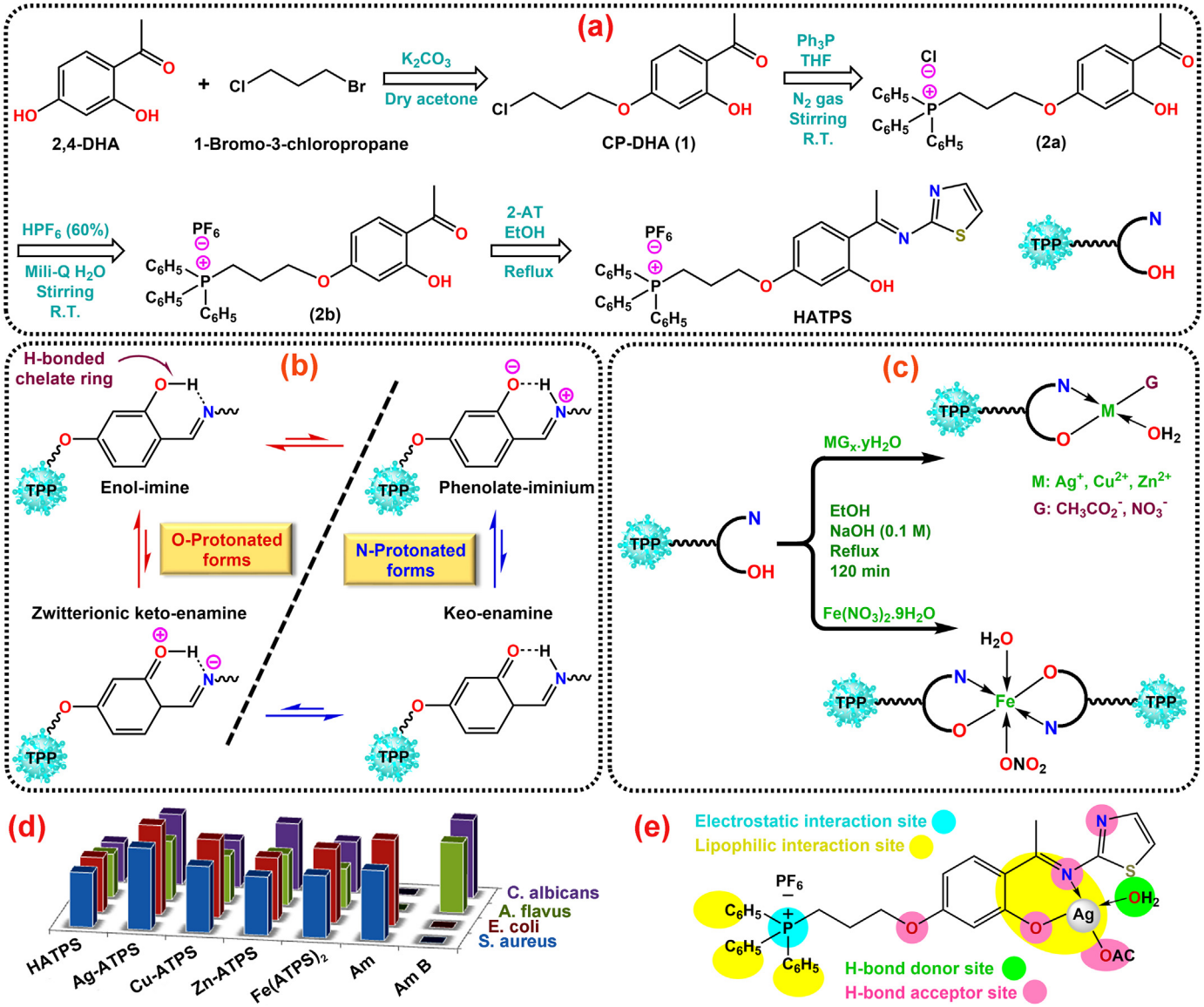


Fig. 3. (a) Preparation of 2,4-dihydroxyacetophenone (DHA)-based triphenyl-phosphonium (TPP) ionic liquids (2a, b: DHA-TPPILs), and aminothiazolyl-functionalized phosphonium ionic liquid (HATPS). Procedure: I) The synthesis of 1-[4-(3-chloropropoxy)-2-hydroxyphenyl]-1-ethanone (1; CP-DHA) through a reaction of 2,4-dihydroxyacetophenone (2,4-DHA), 1-bromo-3-chloropropane, and anhydrous potassium carbonate in acetone at reflux condition for 16 h. II) The preparation of 3-(4-acetyl-3-hydroxyphenoxy)propyl) triphenylphosphonium chloride (2a) via the addition of solution 1 in dry tetrahydrofuran (THF) to a triphenylphosphine (Ph_3P or TPP) solution in dry THF at room temperature (R.T.) under nitrogen atmosphere (N_2 gas) during 30 min, and stirring under N_2 gas and reflux condition for 3 h. III) The creation of 3-(4-acetyl-3-hydroxyphenoxy)propyl)triphenyl phosphonium hexafluorophosphate (2b) by adding the hexafluorophosphoric acid solution (HPF_6, 60 w%) to solution 2a in milli-Q water under cooling in an ice bath over 1 h, and stirring at R.T. for 24 h. IV) The formation of HATPS from adding of 2b ethanolic solution to a solution of 2-amino-thiazole (2-AT) in ethanol and glacial acetic acid under reflux for 3–5 h. (b) Tautomeric modes and forms of hydrogen bonding in HATPS. (c) The scavenging of heavy metal ions by HATPS. (d) The inhibition zone for scavenger and scavenging outputs (M-ATPS complexes) against microbial species. (e) The pharmacophores belonged to the most potent antimicrobial scavenging product [Ag-ATPS: $Ag(ATPS)(H_2O)(CH_3COO)$]. Reproduced with modification and permission from ref. [102].

liquid onto the microporous magnetic Fe_2O_3@SiO_2 core/shell (Fig. 4a). The magnetic POM-SILP displayed high adsorption efficiency for heavy metal ions (Pb^{2+}, Ni^{2+}, Cu^{2+}, and Co^{2+}) removal from aqueous solutions. The Lacunary Keggin tungstate anions combined with POM-ILs featured heavy metal ions binding sites. In addition, when magnetic POM-SILP was used to adsorb the patent blue V, the results indicated a removal efficiency of more than 99% due to the presence of water-immiscible POM-ILs bearing large and hydrophobic Q7 cations in magnetic POM-SILP structure with binding capability towards organic contaminants (Fig. 4b). Furthermore, the spherical polystyrene beads (as a model of environmentally persistent microplastic) removal was assessed by magnetic POM-SILP (Fig. 4c). The polystyrene removal investigation exhibited that the viscous POM-IL coating with long-chain quaternary organo-ammonium cation onto the magnetic core surface could attach the magnetic POM-SILP onto the polystyrene beads, and thus enabling their magnetic retrieval from the water. Besides, the antibacterial water purification effects of magnetic POM-SILPs were assayed against *Bacillus subtilis* and *Escherichia coli* bacteria. The antibacterial activity of magnetic POM-SILP was confirmed using scanning electron microscopy and transmission electron microscopy images that illustrated the bacterial morphology was affected by the presence of magnetic POM-SILP; bacterial removal was 100% for both bacterial strains at the magnetic POM-SILP concentration of 10 mg mL^{-1}. The antibacterial activity of magnetic POM-SILP against *Bacillus subtilis* remained unchanged after consecutive third cycles, while the particle influences on *Escherichia coli* decreased after two cycles.

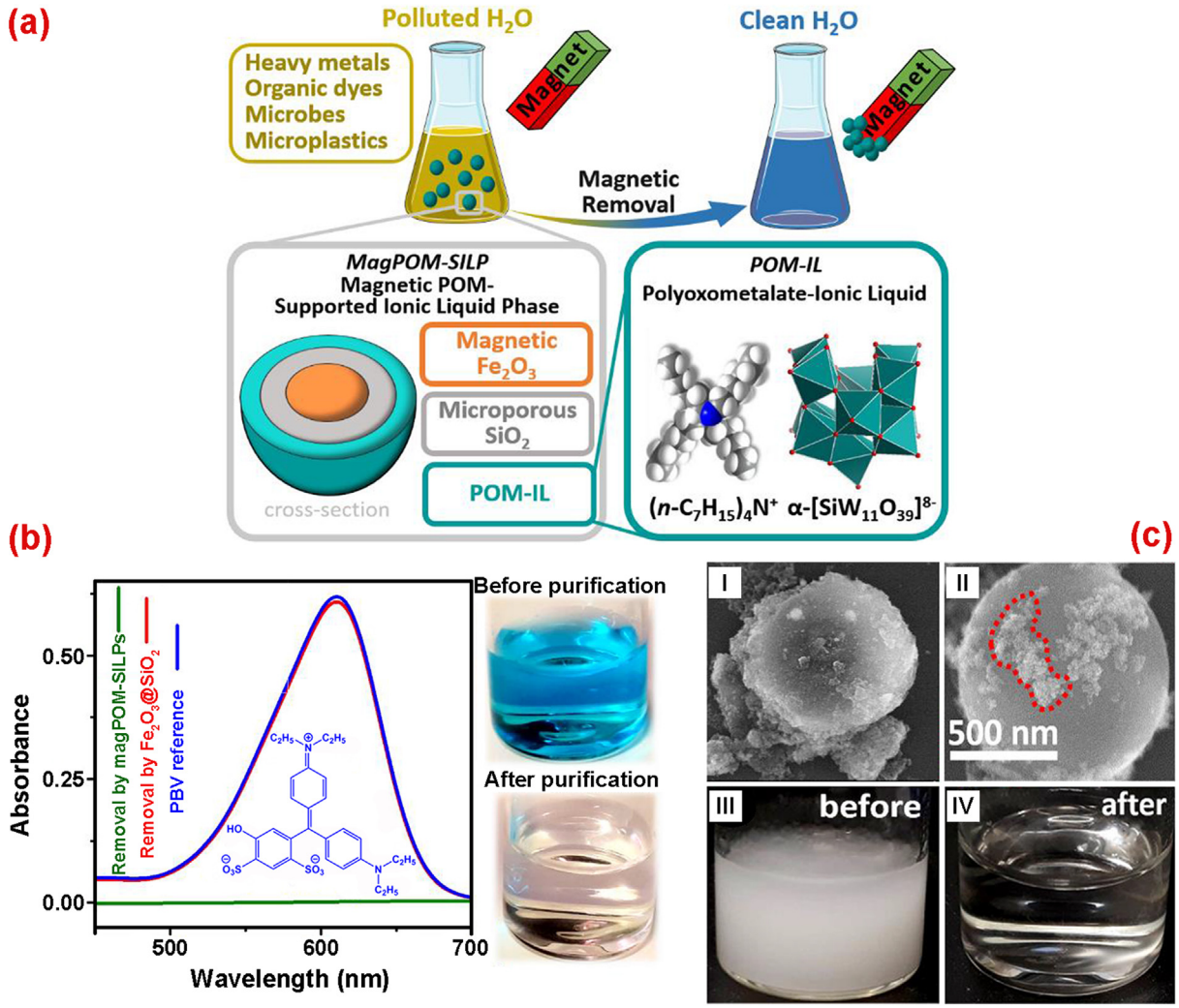


Fig. 4. (a) Preparation of magnetic polyoxometalate supported-ionic liquid phases (magPOM-SILPs): I) The formation of magnetic iron oxide/silica (Fe_2O_3@SiO_2) core-shell particles from the reaction of surfactant Brij 56 with aqueous Fe(III) solution and subsequently treating with $Si(OEt)_4$ via reverse water-in-oil microemulsion method in cyclohexane organic phase, II) The polyoxometalate ionic liquid (POM-IL) synthesis by a combination of the antimicrobial tetra-*n*-heptyl ammonium cation [Q7: $(n\text{-}C_7H_{15})_4N^+$] and the lacunary-Keggin cluster anion [$(\alpha\text{-}SiW_{11}O_{39})^{8-}$], and III) The magPOM-SILPs (20 wt% POM-IL loading) preparation by dispersing Fe_2O_3@SiO_2 particles in the POM-IL solution in acetone, followed by vacuum drying. (b) Patent Blue V (PBV) removal by adsorbents from water; Left: Ultraviolet-visible spectra of PBV before adsorption (blue line), after adsorption onto the Fe_2O_3@SiO_2 (red line), and after adsorption onto the magPOMSILP (green line). Right: The images of PBV solutions before and after removal. (c) Microplastics removal by magPOM-SILP; Up: the scanning electron microscopy (SEM) images of polystyrene beads covered with magPOM-SILPs with the magnification of 1 μm and 500 nm in images I and II, respectively. Below: photographs of microplastic solutions before and after polystyrene adsorption. Reprinted with modification and permission from ref. [104].

Photocatalysis is one of the effective and widely used methods for the degradation of contaminants and elimination of microbial species in water. In this context, the poly(methyl methacrylate)/ionic liquid-TiO_2 (PMMA/IL-TiO_2) nanocomposite film was prepared from 1-butyl-3-methylimidazolium tetrafluoroborate ionic liquid and methyl methacrylate in presence of benzoyl peroxide initiator and triton X-100 surfactant via micro-emulsion technique [105]. The PMMA/IL-TiO_2 film showed a photodecomposition effect on the trypan blue dye as well as antibacterial activity against *Bacillus* sp. The PMMA/IL-TiO_2 exhibited a high photocatalytic effect under visible light due to the ionic liquid role in improving the transportation of photo-induced electrons and holes. The highest photocatalytic efficiency along with antibacterial activity was obtained for PMMA/IL-TiO_2 film with 40 μm thickness and 0.01 wt% of TiO_2.

Benzimidazole-based ionic liquid coated on ZnO nanoparticles (ZnO@IL) was synthesized and applied as a photocatalyst for the 4-nitroaniline oxidation under ultraviolet light (Fig. 5a) [106]. The 4-nitroaniline oxidation by ZnO@IL revealed that a significant degradation occurred according to the first-order kinetic (Fig. 5b), and a higher oxidation rate ($k = 2.01 \times 10^{-4}$ mMs^{-1}) was observed at pH = 4 compared with that at pH = 10 ($k = 0.26 \times 10^{-4}$ mMs^{-1}). The ZnO@IL (150 μL, 5 mM) exhibited antibacterial activity against bacteria (2.5×10^5 CFU/mL) with microbial inhibition of 43% and 40% against *S. aureus* (Gram-positive), and *E. coli* (Gram-negative), respectively (Fig. 5c). The cell growth inhibition of ZnO@IL was associated with the imidazolium ring and the carboxylate group as essential reactive center that could generate the reactive oxygen species, interact with proteins (amide, carboxyl, phosphate, hydroxyl tails, and carbohydrate-related moieties) or intercalate between the purine and pyrimidine components in DNA of both types' bacteria (Fig. 5d). The ZnO@IL could easily enter into the cell, affect metabolic functions, prevent the respiration system, break the cell wall, leach out the cell materials, and eventually leading to cell death (Fig. 5e).

Adhering to the goals of green chemistry, biopolymers from natural and renewable sources in view of their abundance, and biocompatibility, have garnered much attention for the preparation of natural polymer-based ionic liquids with antimicrobial activity for water treatment [107]. In this respect, spherical microgel comprising gum Arabic-

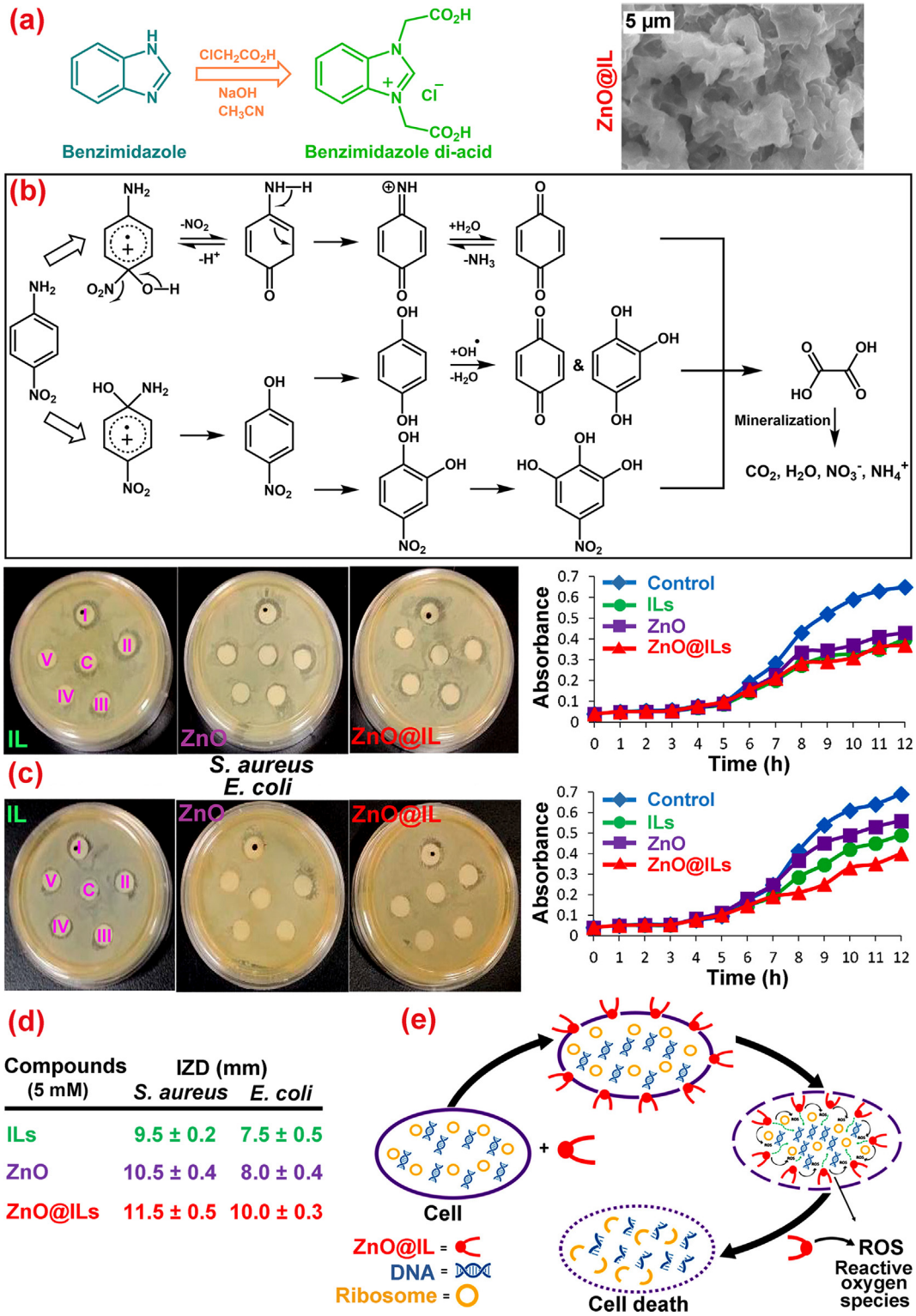
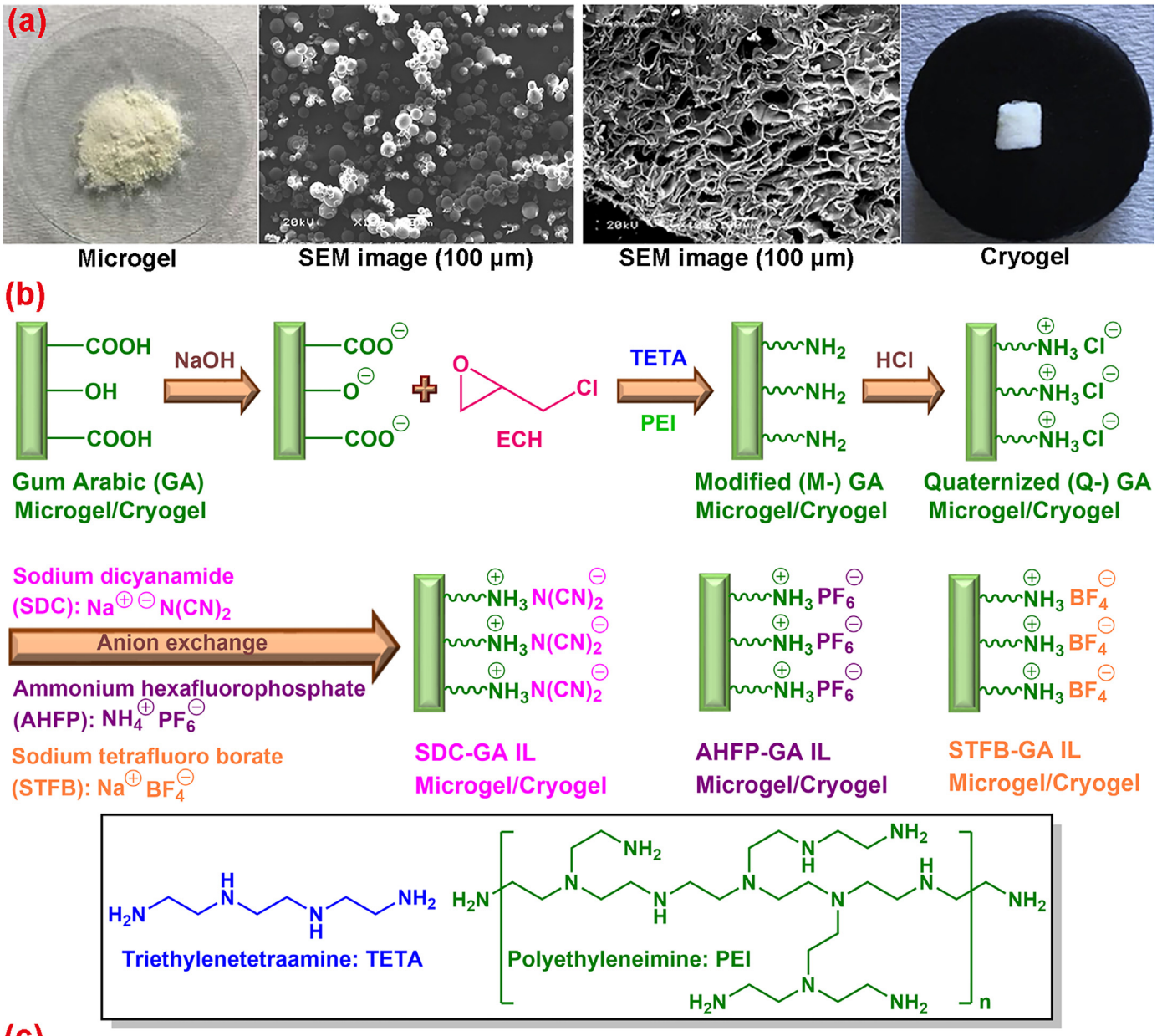


Fig. 5. (a) Preparation of benzimidazolium di-acid ionic liquid and benzimidazolium di-acid@ZnO (ZnO@IL). Experimental procedure: (I) Synthesis of white solid benzimidazole-based ionic liquid through the dissolution of benzimidazole and chloroacetic acid in dry acetonitrile at pH = 8, reflux for 5 h, and solution pH adjustment to 2.0–3.0. (II) The synthesis of ZnO@IL by adding benzimidazolium di-acid ionic liquid to zinc acetate and NaOH solution in methanol, reflux for 2.0 h, filtration, and washing with methanol and water. (b) A plausible mechanism for 4-nitroaniline oxidation via OH radical produced by ZnO@IL under ultraviolet light. (c) Kirby-Bauer disk diffusion analysis and the minimum inhibition concentration data. [Concentrations of samples in ethanol (µL); $I = 150$, II = 125, III = 100, IV = 75, V = 50, and C: control]. (d) Inhibition zone diameter (IZD) of compounds in an antibacterial test. (e) The proposed interactions between bacterial cells and ZnO@ILs. Reprinted with modification and permission from ref. [106].

based ionic liquid (GA-IL), was prepared from the gum Arabic in a single step, and super porous gum Arabic cryogel was synthesized under cryogenic conditions (Fig. 6a). Then, the chemical modification of gum Arabic microgel and cryogel was performed with polyethyleneimine and triethylenetetraamine to produce the modified gum Arabic microgels/cryogels. Finally, the GA-IL-based microgels/cryogels were

formed by the anion exchange reaction of protonated modified gum Arabic microgels/cryogels with ammonium hexafluoroborate, sodium dicyanamide, and sodium tetrafluoroborate (Fig. 6b). Bare, modified, and GA-IL cryogels have been deployed for the adsorption of drug (sodium diclofenac) and pesticide (paraquat) from aqueous solutions because of their superior porous nature (Fig. 6c). Adsorption studies



revealed that the modified and GA-IL cryogel could remove the higher amount of sodium diclofenac and paraquat in comparison to bare cryogel due to their tunable chemical and physical properties. Furthermore, the antimicrobial effect of gum Arabic microgels was assayed by disc diffusion method, and the quaternized polyethyleneimine microgel was found to be an effective antibacterial agent for the growth inhibition of Gram-positive (*Staphylococcus aureus*) and Gram-negative (*Escherichia coli*) bacteria with 13 ± 1 mm inhibition zones.

Membrane separation is an imperative technology employed worldwide to produce clean and pure water with low energy consumption and a small energy footprint [108]. One of the emerging and intriguing candidates for membrane fabrication in the water treatment arena are polymeric ionic liquids in which functional polymers bear the moieties of antimicrobial ionic liquids. The structural synergy between the macromolecular architecture and antimicrobial ionic liquid component extends the window of physicochemical characteristics of membranes, thus, enriching their application and functional range by linking the advantageous attributes of ionic liquids [109].

Polyamide ionic liquid membranes (PAILMs) have been prepared through surface modification of polyamide with 1-aminoethyl-3-methylimidazolium bromide ionic liquid (AMIB IL) [110]. The PAILMs were synthesized by 'piperzine ~ trimesoyl chloride' (PIP-TMC) interfacial polymerization. The AMIB IL was attached to the membrane surface through 'acyl chloride ~ amine' amidation reaction. The thin film of polyamide composite membranes exhibited antibacterial activity, antibiotics separation, high water permeability, and salt rejection. The modified membrane (PIP-TMC-AMIB) displayed water flux and salt rejection of 135 ± 5 L m^{-2} h^{-1} and 95 ± 1% (Na_2SO_4) at 6 bar pressure, indicating a four-fold improvement compared to the pristine PIP-TMC membrane. The selective and rapid transportation of water from inside the PIP-TMC-AMIB could be related to the higher surface energy, hydrophilicity, and loose structures after membrane modification with AMIB. The salt rejection of PIP-TMC-AMIB could be consistent with the negative charge of the modified membrane and a strong electrostatic repulsion between SO_4^{2-} and negatively charged membranes. Also, the PIP-TMC-AMIB was used for the filtration of antibiotics solutions containing ciprofloxacin, erythrocin, levofloxacin, oxytetracycline, and tetracycline. In the antibiotics filtration, the PIP-TMC-AMIB showed higher water flux (150 ± 5 L m^{-2} h^{-1}) in comparison with Na_2SO_4 filtration because of the osmosis pressure effect and exhibited high antibiotics rejection (93.5 ± 3%). In addition to the high performance for salt and antibiotics rejection, the PIP-TMC-AMIB displayed efficient separation of sodium chloride/antibiotics mixtures, as well as promising stability and antibacterial activity with an inhibition effect of 99% against *Escherichia coli* growth.

Recently, an ionic liquid grafted polyamide 6 (IL-g-PA6) was synthesized by grafting the 1-vinyl-3-butylimidazolium chloride ionic liquid ([VBIM][Cl]) onto the polyamide 6 main chains via radiation-induced grafting methodology (Fig. 7a) [111]. The bulky cations of grafted ionic liquid as spacers could enlarge the distance between polyamide 6 chains, and decrease the hydrogen bonding strength in the polyamide 6 matrix. Thus, the ion-containing polyamide 6 could be easily melt-spun into fibers due to the good melt processability. The fibers derived from ion-containing polyamide 6 showed antibacterial activities particularly against *Escherichia coli*, as well as, antistatic performances and good mechanical properties (Fig. 7b). The IL-g-PA6 with a unique ion structure was applied to design a porous membrane through the immersion precipitation phase inversion method [112]. The adsorption capability of IL-g-PA6 membranes was assayed for the chromate removal, and results revealed that the observed excellent adsorption efficacy was associated with the special interaction between grafted ionic liquid and chromate ions (Fig. 7c). These researches offered a new strategy to prepare the interesting scavenger membranes by using the IL-g-PA6 for the removal of heavy metal ions and microbial contaminations from aqueous solutions.

Supported ionic liquid membranes (SILMs) have been prepared by using imidazolium-based ionic liquids and hydrophobic polytetrafluoroethylene, and were deployed for the π electron-induced separation of aromatic compounds (toluene, styrene, divinylbenzene, congo red, eriochrome black T, and ramazol brilliant blue R) from non-aqueous (hexane) and aqueous media [113]. The separation efficacy of SILMs was established with the trend, toluene < styrene < divinylbenzene from non-aqueous solutions, and in the case of dyes, the congo red and ramazol brilliant blue R were preferentially removed from aqueous solutions. The ionic liquid affinity towards π electron cloud is the driving force for this separation trend, while the highest selectivity for congo red was related to the complicated electronic configuration. The antibacterial effects of SILMs were assayed against *Staphylococcus aureus* and *Pseudomonas aeruginosa*. The SILMs exhibited satisfactory antibacterial activity against Gram-positive bacteria, and the colony-forming unit value (2.82E+09) was significantly reduced to 1.10E+03, 1.2E+04, and 4.4E+06 after 5 h of exposure to SILMs with bromohexane, chlorobutane, and allylbromide moieties, respectively. The trend of antibacterial activities of ionic liquids (bromohexane > chloro butane > allylbromide) could be associated with greater number of carbon atoms on the side chain of imidazolium ring.

In another study, a polyethersulfone grafted ionic liquid porous membrane (PES-g-IL [BF_4]) was fabricated by chemical bonding of 1-vinyl-3-butylimidazolium tetrafluoroborate ionic liquid ([VBIm][BF_4] or IL (BF_4)) on the main chains of polyethersulfone (PES) through radiation-induced grafting technique (Fig. 8a) [114]. The PES-g-IL (BF_4) membranes revealed extremely improved water flux, antifouling, and antibacterial properties in comparison to the neat polyethersulfone porous membranes (Fig. 8b).

Antimicrobial poly(ionic liquid)s, synthesized by polymerizable antimicrobial ionic liquids, encompass the combination of antimicrobial ionic liquids' unique properties together with intrinsic polymer properties. The antimicrobial poly(ionic liquid)s seem to be the promising materials to design the antimicrobial membranes with improved durability and processability, enhanced mechanical and thermal stability, along with increased permeation rates and separation capability [115].

In this context, antibacterial poly(ionic liquid)s membranes (PILMs) have been fabricated through a two-step synthetic method: I) preparation of ionic liquid monomers (1-vinyl-3-butylimidazolium chloride, 1-vinyl-3-hexylimidazolium bromide, and 1-vinyl-3-allylimidazolium bromide), and II) the synthesis of PILMs by the reaction of ionic liquid monomers and co-monomers (acrylonitrile and styrene) with the 1,4-divinylbenzene cross-linker [116]. These PILMs showed considerable stability in apolar-aprotic, polar-aprotic, and polar-protic solvents, demonstrating their wide applications in various solvents. Pre-compaction of PILMs led to stable water flux, and membranes derived from ionic liquids with chlorobutane, bromohexane, and allybromide moieties showed water flux of 315 L m^{-2} h^{-1} at 1.5 psi, 948 and 982 L m^{-2} h^{-1} at 0.5 psi, respectively. The PILMs were applied for protein (bovine serum albumin and dextran) rejection, and

Fig. 6. (a) Scanning electron microscopy (SEM) images of gum Arabic (GA) microgel and cryogel. (b) Preparation of GA based ionic liquids (GA-ILs) via post-modification reactions of GA microgels/cryogels; I) NaOH treatment of GA microgel/cryogel to generate negative charges, II) The reaction of deprotonated GA microgel/cryogels with epichlorohydrine (ECH) as a binder for interaction with modifying agents (polyethyleneimine: PEI and triethylenetetraamine: TETA) to create modified (M-) GA microgel/cryogels containing amine groups, III) The protonation of amine groups onto M-GA microgel/cryogels to construct quaternized (Q-) GA microgel/cryogels, and IV) The GA-ILs formation through anion exchange of Q-GA microgel/cryogels. (c) Representation of adsorption efficiency of bare, modified, and GA-ILs cryogels for the drug (sodium diclofenac) and pesticide (paraquat) removal. Reprinted with modification and permission from ref. [107].

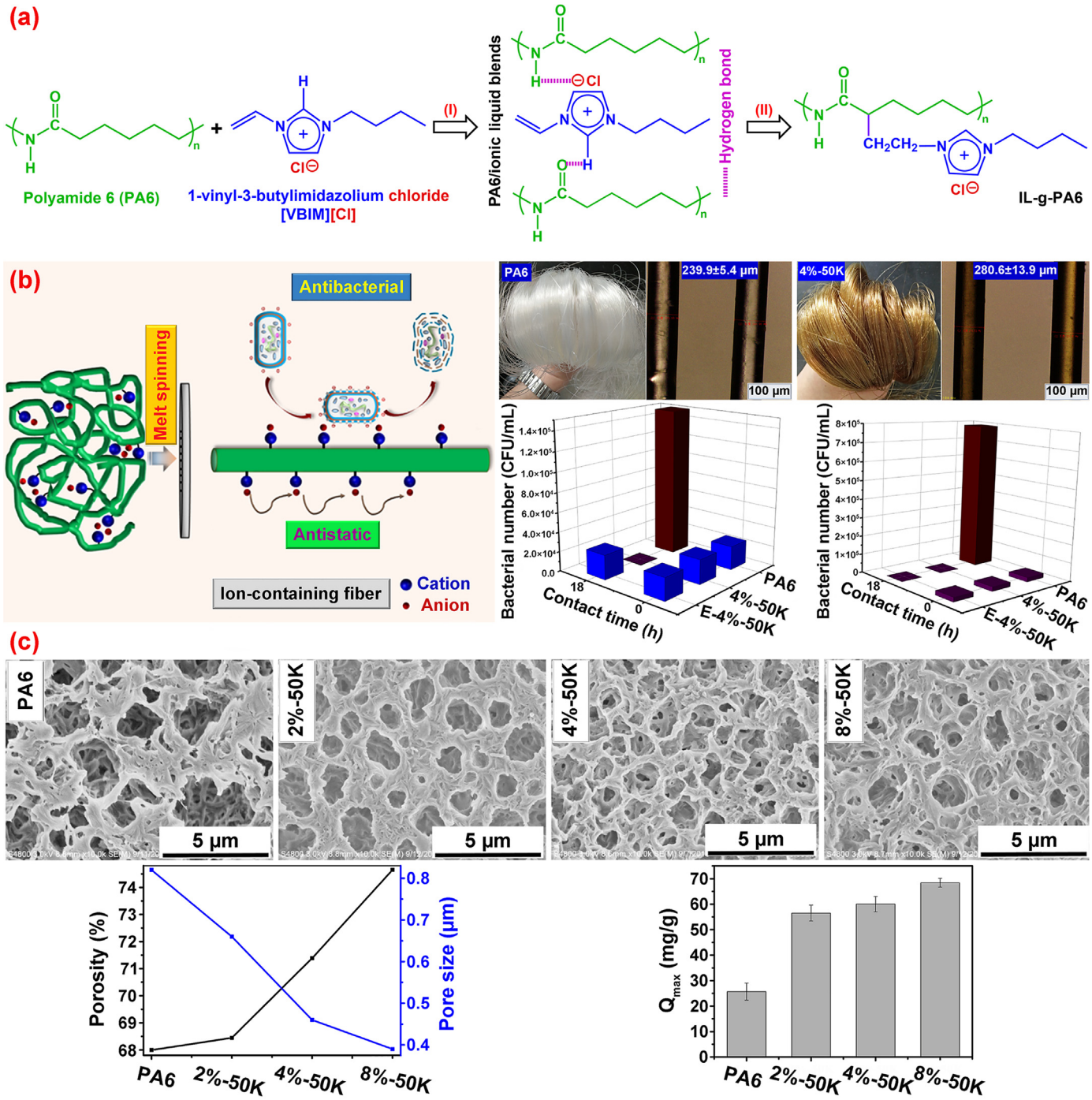


Fig. 7. (a) Preparation of ionic liquid-grafted polyamide 6 (IL-g-PA6) during 2 steps: (I) the creation of PA6/ionic liquid blends by melt mixing of polyamide 6 (PA6) and 1-vinyl-3-butyl imidazole chloride ionic liquid at 220 °C for 2 min at a rotation speed of 20 rpm, followed by 5 min rotation at 50 rpm. (II) the IL-g-PA6 preparation via graft polymerization by γ-ray irradiation at 50 kGy dosages of blends with different ionic liquid loadings under room temperature for 17 h. (X%-50 K represents a PA6 matrix bearing X wt% of the ionic liquid that was exposed to a 50 kGy absorption dosage of γ-ray irradiation, and *E*-X%-50 K represents the extracted IL-g-PA6 for removal of non-grafted ionic liquids). (b) The formation of ion-containing PA6 fibers by the melt-spinning process of IL-g-PA6 films, and investigation of their antibacterial properties. (c) The preparation of IL-g-PA6 membranes via simple immersion-precipitation phase inversion technique, and evaluation of their porosity and adsorption capability for chromate ions removal. Reprinted with modification and permission from ref. [111, 112].

results confirmed that the tenability of membrane's rejection related to the polymerization time and imidazolium ring substitution. The *Staphylococcus aureus* and *Pseudomonas aeruginosa* growth were inhibited by PILMs, and the antibacterial effect was dependent on the imidazolium ring substituents.

Recently, antibacterial poly(amino acid ionic liquids) membranes (PAAILMs) were synthesized by blending the poly(amino acid ionic liquids) with polyvinylidene fluoride and were applied to separate the amino acids viz. L-aspartic acid and *L*-phenylalanine. The PAAILM with L-lysine anion showed 65% selectivity for the mixed amino acids through one-step separation based on repulsion between positively charged membrane surface and electronegative amino acids. These PAAILMs displayed high mechanical strength and thermostability along with efficient antifouling and antibacterial effects [117].

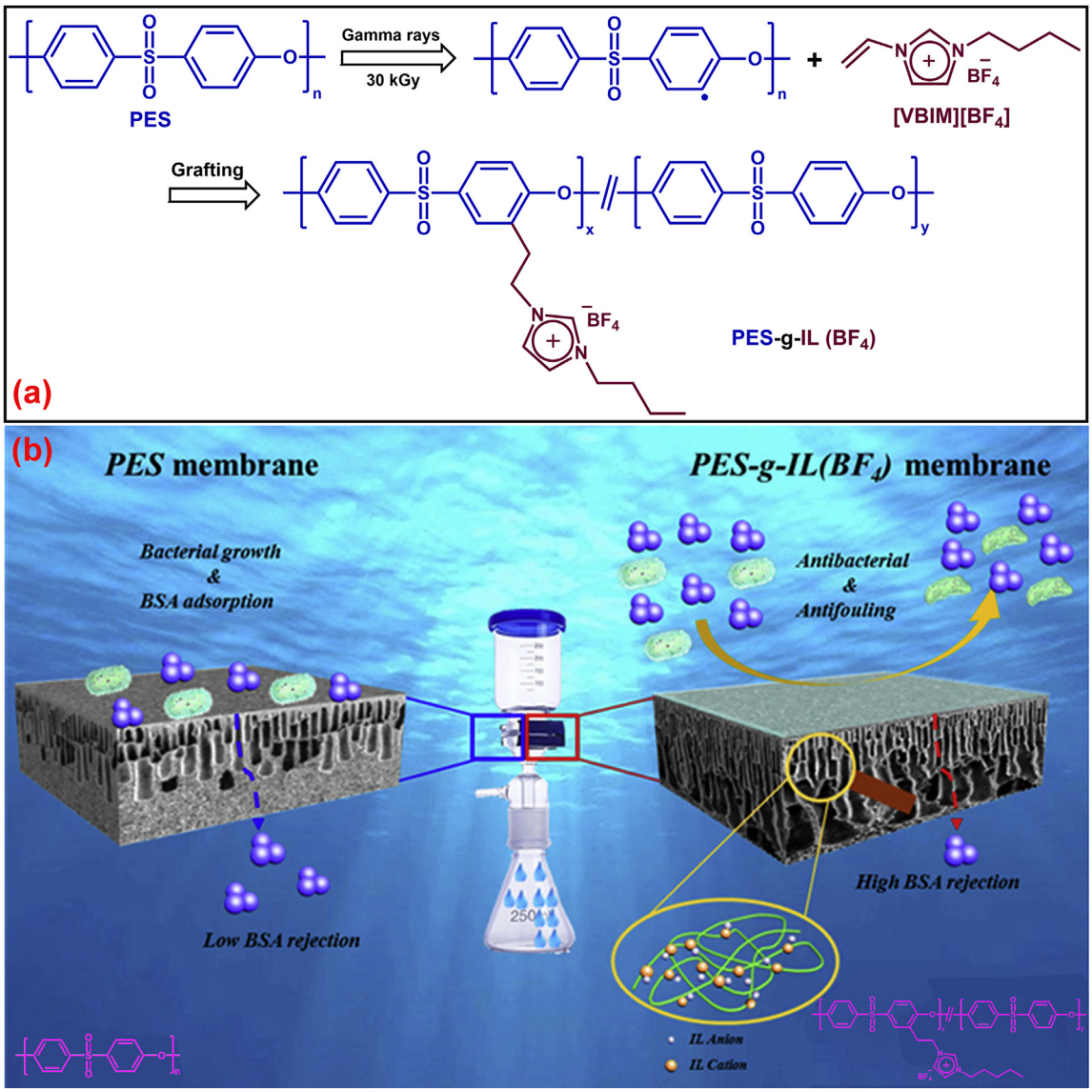
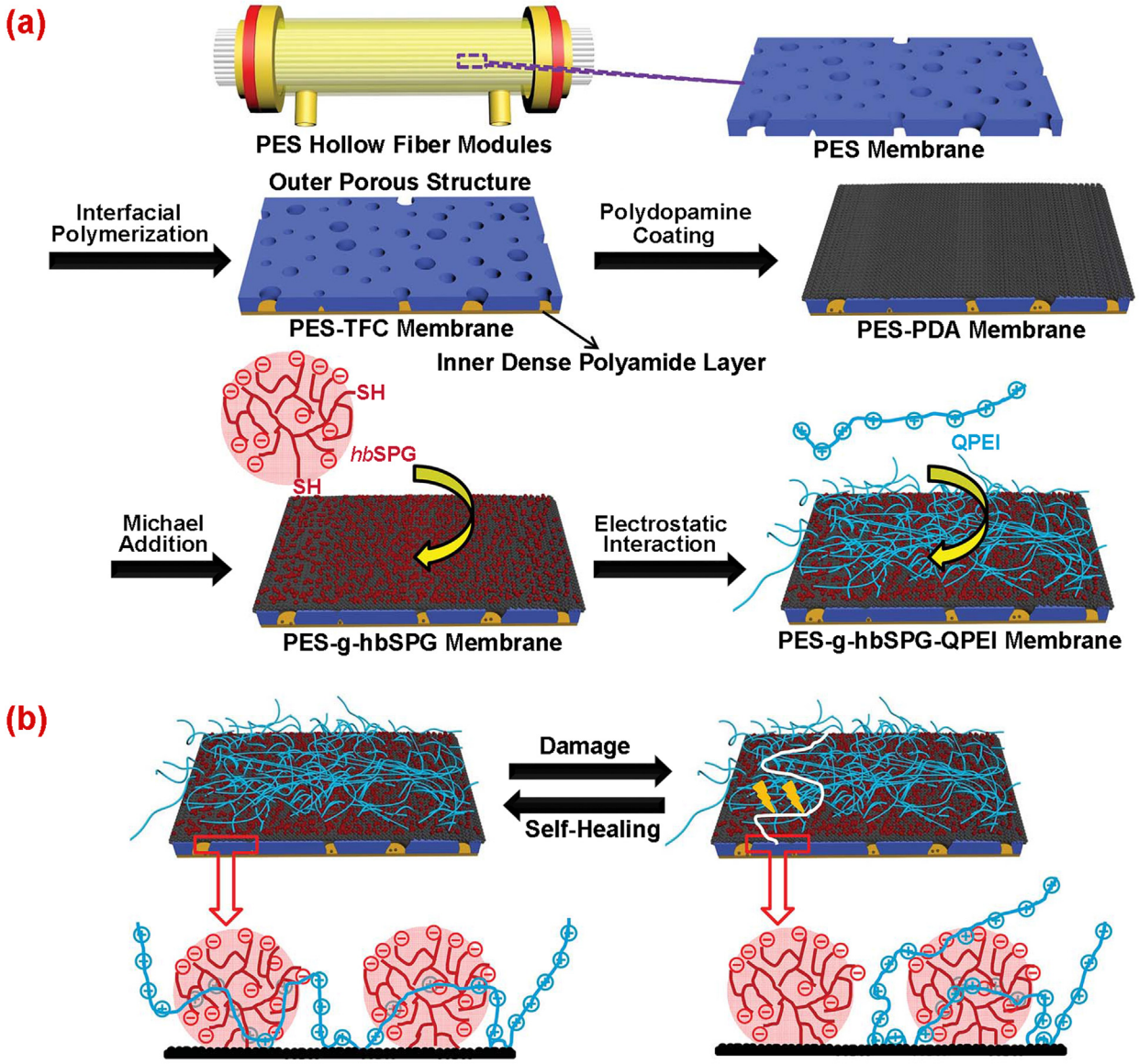


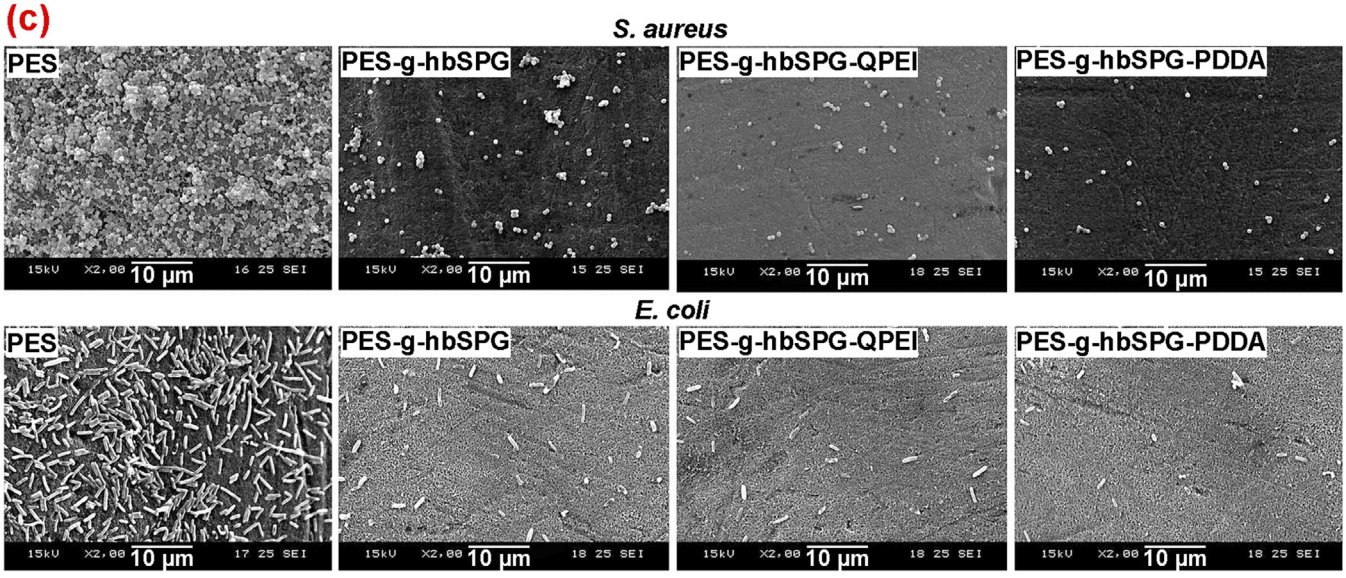
Fig. 8. (a) Procedure for preparing of polyethersulfone grafted ionic liquid porous membrane. (I) Formation of PES/IL (BF_4) binary blend: desolvation of polyethersulfone (PES) and 1-vinyl-3-butyl imidazolium tetrafluoroborate ([VBIm][BF_4]) or IL (BF_4)) in dimethylformamide, and drying of obtained homogenous solution. (II) Synthesis of PES-g-IL (BF_4): irradiation of PES/IL (BF_4) blends with γ-ray at 30 kGy dosages during 17 h at room temperature. (III) Preparation of PES-g-IL (BF_4) membrane: fabrication of homogenous dopes by dissolving of PES-g-IL (BF_4) in dimethylformamide, liquid membrane formation from casting of homogenous dopes on glass, and porous membrane creation via liquid membrane immersing into the coagulation bath, then freeze-drying. (b) Antibacterial, antifouling, bovine serum albumin (BSA) rejection behavior of PES-g-IL (BF_4) membrane, and its comparison with PES membrane. Reprinted with modification and permission from ref. [114].

Antibacterial hyperbranched poly(ionic liquid) functionalized poly (ether sulfone) membrane (PES-g-hbSPG-QPEI) was fabricated via sequential immobilization of hyperbranched polyglycerol (hbSPG) and quaternized polyethylenimine (QPEI) or poly(diallyldimethylammonium chloride) solution (PDDA) on the poly(ether sulfone) (PES) membranes pretreated with polydopamine (PDA) and utilized for osmotic power generation as superior antifouling agents with self-healing property (Fig. 9a). The healable antifouling characteristics of PES-g-hbSPG-QPEI hollow fiber membranes were confirmed by the excellent anti-protein sorption performances and improved antibacterial activities on both, the aged and non-aged samples soaked in real wastewater for two months. Under pressure, retarded osmosis analysis, the poly(ether sulfone) membranes were seriously polluted under high-pressure operation leading to a 60% flux drop. After back-flushing and hydraulic pressure impulsion, the non-aged membrane was obtained at a recovery rate of 98% and a flux reduction of 30%, while the aged membranes were obtained in a flux reduction of 40%. The polyionic liquid coating layers with intrinsically antifouling and healable behaviors demonstrated the excellent potential for the construction of high-performance pressure retarded osmosis membranes towards osmotic power generation. The higher chain mobility of polyionic liquid network awards the self-healing properties to membranes, in which the reversible ion-pair exchange could readily compensate damaged interface edges, leading to repeated healing function, prolonged membrane lifespan, and preserved membrane performance (Fig. 9b). The PES-g-hbSPG-QPEI with hbSPG containing abundant sulfonate groups on the surface-displayed less prone to a bacterial attachment (Fig. 9c) [118].

7.2. Air filtration

The rapid urbanization and industrialization for improving the human life have brought unwanted air pollution issues. Air pollutants, especially carbon dioxide and particulate materials are considered the most serious environmental problems causing intense damage to


(a)
PES Hollow Fiber Modules
PES Membrane
Outer Porous Structure
Interfacial Polymerization
Polydopamine Coating
PES-TFC Membrane
PES-PDA Membrane
Inner Dense Polyamide Layer
SH
hbSPG
SH
QPEI
Michael Addition
Electrostatic Interaction
PES-g-hbSPG Membrane
PES-g-hbSPG-QPEI Membrane
(b)
Damage
Self-Healing


(c)
S. aureus
PES
PES-g-hbSPG
PES-g-hbSPG-QPEI
PES-g-hbSPG-PDDA
E. coli
PES
PES-g-hbSPG
PES-g-hbSPG-QPEI
PES-g-hbSPG-PDDA
10 μm

human health [121]. Air pollution is not only harmful to the air passages and lungs but can also damages other organs of the human body [122]. Hence, there have been growing concerns about reducing or eliminating the risks emanating from the air pollutants. Air filtration techniques can eradicate air pollutants and effectively improve the respiratory air quality [123]. The filtration compounds play a key role in the air filtration process, and the design of innovative air filters has received major attention in recent years [123]. However, conventional filtration materials are ineffective for removing extremely small size particles (with particulate material 2.5 nm). Consequently, air filtration by using nanofibrous filters and nanofibrous membranes has garnered considerable popularity; especially, electrospun nanofibrous membranes have gained significance in the filtration arena due to their prominent properties [124].

Tiny particulate materials such bioaerosols may carry various viruses, bacteria, and fungi which can cause severe allergic, respiratory, and infectious diseases through airborne transmission, thus necessitating the fabrication of antimicrobial and antiviral air filters [125,126]. Today, ionic liquids-based materials are among the most potential candidates for air filter fabrication because of their exceptional gas separation, photoresists, anticorrosion, and antimicrobial properties thus enabling the development of a fairly new approach in the last two decades. Various types of ionic liquids-based membranes have been applied for air treatment including supported ionic liquid membranes, polymer/ionic liquid composite, polymer/ionic liquid/inorganic composite, and ionic liquid-gelled membranes [127].

In this context, an antimicrobial fibrous mat (PLA/PHB-IL) was prepared from polylactide/polyhydroxybutyrate (PLA/PHB) nanofibers that are functionalized with quaternary ammonium-based ionic liquid and utilized for aerosol particle filtration [10]. The capability of the electrospun nanofibrous mat was considered by assessing the filter penetration of sodium chloride aerosol particles, and the results revealed that the quality factors were dependent on the electrospinning time to attain a fiber diameter of ~500 nm with the distribution of 350 fibers in filter mat. The PLA/PHB-IL biodegradable filters displayed fungicide activity against *Aspergillus niger* and *Chaetomium globosum*, and the antifungal effects could be improved with increasing ionic liquid concentration. Consequently, the ionic liquid functionalized nanofibers not only could be used for antimicrobial air filtration but also can create "eco-friendly" compounds due to their biodegradable behavior.

The cellulose acetate/poly(ionic liquid) composite was prepared as an antimicrobial air filter for the elimination of inhalable particulate materials via macromolecular design and electrostatic spinning technique (Fig. 10a) [128]. The diameter of fibers was efficiently reduced by an introduction and increased content of poly(ionic liquid)s, and so the nanofibrous filters were obtained with a removal rate of 99.65% for particulate materials 10 nm (Fig. 10b). The nano-filters exhibited antibacterial activities against *Escherichia coli* and *Staphylococcus aureus*, without obvious cytotoxic effects on cultured cells (Figs. 10c and d). The nanofilters preserved the fibrous morphology and outstanding antimicrobial effects after multiple recycling, owing to the sturdy covalent bonds of poly(ionic liquid)s with cellulose acetate (Fig. 10d). In contrast, the filters containing ionic liquids moieties with excellent hydrophilicity and solubility are prone to be lost during application making it hard to recycle. As a result, the need of the hour for progress in this field is to prepare filters with low air resistance, high recyclability as well as good biocompatibility, and environmentally friendly behavior.

7.3. Food packaging

Food packaging is an integral part of the food industry to improve the food protection and safety. The development of novel innovative packaging materials with antimicrobial properties can be achieved by adding agents such as antimicrobial ionic liquids to inhibit microbial growth by reducing live counts of microbes [129]. Green antimicrobial composite films have been prepared using 1-allyl-3-mthylimidazolium chloride and biopolymers such as cellulose, starch, and lignin. as suitable candidates for fresh food packaging [130].

A bioactive ionic liquid i.e. choline salicylate was utilized for the production of gelatin-based ionogel films (Figs. 11a) with enhanced antimicrobial and antioxidant features [11]. The ionic liquid acted as a plasticizer in the structure of the film and did not experience any chemical changes during the synthesis process. The ionic liquid/gelatin-based films exhibited excellent bacterial growth inhibition compared to the films without ionic liquid. This may be due to the good antibacterial effect of cholinium salicylate against *Bacillus subtilis* (Fig. 11b). The ionic liquid-loaded films also demonstrated the ability to improve the shelf life of *Malus pumila* (red apple) by preventing air oxidation (Fig. 11c); films displayed exceptional ultraviolet shielding and antioxidant properties with high mechanical strength. The results of antioxidant activity induced by free radical scavenging assay revealed that the radical scavenging effect did not occur in the film without ionic liquid whereas the ionic liquid loaded films with increased ionic liquid concentration displayed increased antioxidant capacity (Fig. 11d).

Likewise, cholinium citrate and cholinium hexanoate ionic liquids were used in combination with pullulan and chitosan for the preparation of biocompatible and antibacterial transparent films. The films prepared with chitosan indicated antibacterial activity against *Klebsiella pneumoniae* and *Staphylococcus aureus*, whereas for pullulan the film is bioactive when prepared with cholinium citrate [131].

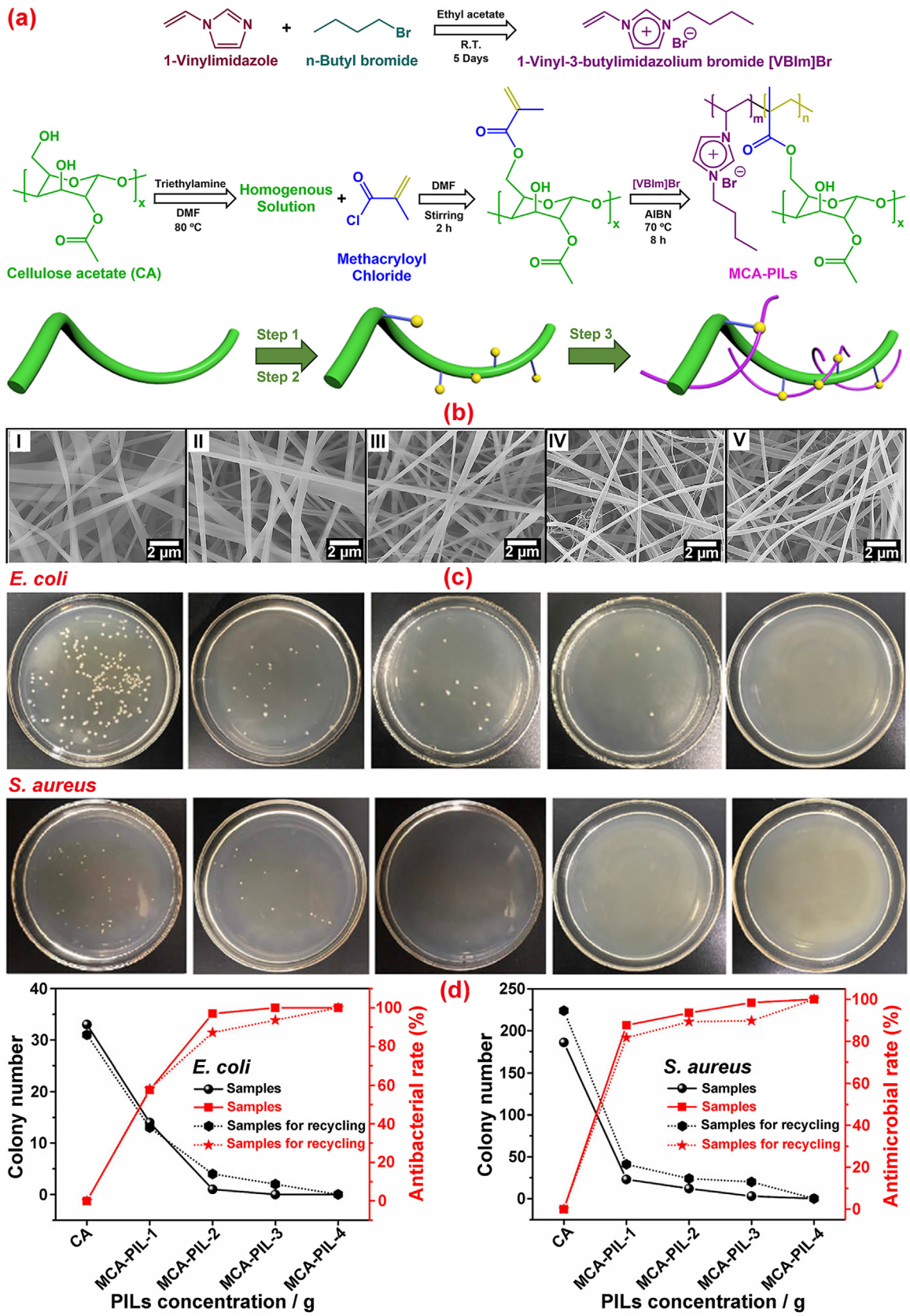
Overall, the application of ionic liquids for the preparation of food packaging films is very limited compared to bioactive or biopolymer-based films. In the future, research needs to be preferentially focused on the development of active, intelligent, and stable films as well as efforts to enhance safety by preventing the associated toxicity of film which could be encountered when they are exposed to the natural environment. These materials packaging films can be optimized further and commercialized for deployment as active and intelligent packaging for visual quality evaluation of fresh food products [132].

7.4. Coating

A topical issue for materials sciences is the protection of materials surfaces by coating, thus an inclusive diversity of coating techniques are accessible with a common purpose of surface protection from chemical or physical damages, and biodegradation [13]. Although the coating strategy affords advantages, there are still disadvantages restricting their usage. According to the rules governing human health and environmental protection, the use of hazardous compounds is severely forbidden in coating technology. These restrictions along with the absence of alternate coating techniques to fulfill all the technological requirements prompted the creation of a new coating strategy. In this regard, scientists are paying significant attention to designing and fabricating smart and green coating materials. The manufacture of

Fig. 9. (a) Procedure for hyperbranched poly(ionic liquid) functionalized poly(ether sulfone) membranes. The poly(ether sulfone) (PES) membrane creation from PES hollow fiber formed by spinning via a dry-jet wet-spinning method [119]. The poly(ether sulfone)-thin-film composite (PES-TFC) membrane formation via interfacial polymerization to form thin-film polyamide layers on the inner surfaces of hollow fibers [120]. The pretreated poly(ether sulfone)-polydopamine (PES-PDA) membranes fabrication via formation of a polydopamine coating on the PES-TFC dual-layer by using a tris buffer solution containing dopamine hydrochloride (DA), $CuSO_4$, and H_2O_2. The preparation of poly(ether sulfone) grafted hyperbranched sulfonated polyglycerol (PES-g-hbSPG) intermediate membranes by replacing the polydopamine coating with hbSPG solution containing triethylamine (TEA) through Michael addition reaction during 12 h. The synthesis of PES-g-hbSPG-QPEI or PES-g-hbSPG-PDDA membranes by forming the second layer of quaternized polyethylenimine (QPEI) or poly(diallyldimethylammonium chloride) (PDDA) polyelectrolytes on the PES-g-hbSPG through electrostatic interactions. (b) Self-healing behavior of PES-g-hbSPG-QPEI or PES-g-hbSPG-PDDA membranes. (c) Antibacterial effects of pristine PES, PES-g-hbSPG, PES-g-hbSPG-QPEI, and PES-g-hbSPG-PDDA against *S. aureus* and *E. coli* at an initial cell concentration of 10^8 CFU/mL at 37 °C for 6 h. Reprinted with modification and permission from ref. [118].

non-toxic smart coatings has gained widespread attention through the developments of low-hazardous air/water pollutants and low-volatile organic compounds, endowed with super-hydrophobic, anti-wearing, antimicrobial, anti-corrosion, self-cleaning, and self-healing behaviors [133]. The smart coatings, fabricated from responsive materials that respond actively to environmental changes, have special tasks including sensing, protecting, and healing [134]. The protection quality of coatings was improved via the development of advanced smart



coating techniques by using ionic liquid-based materials with unique properties.

The antibacterial stainless steel has been prepared through ionic liquids grafting onto the stainless steel surface via silane modification followed by the thiol-ene click reaction [135]. The antibacterial effect of ionic liquids grafted stainless steel (IL-g-SS) was investigated against *Escherichia coli*, and the results indicated the significant antibacterial activity of IL-g-SS which could be due to the lipophilicity of imidazolium ring with long alkyl chains. The antibacterial activity of IL-g-SS could also be related to the counter anion of imidazolium cation. It has been reported that the ionic liquids with bromide anion exhibited better antibacterial activity than ionic liquids with hexafluorophosphate and tetrafluoroborate anions. The stability of IL-g-SS surface was assayed, and the obtained results established that the *Escherichia coli* growth was efficiently inhibited by IL-g-SS after 5 cycles, representing its considerable durability and stability under operating conditions.

In another study, the ionic liquid-based block copolymers, alternating copolymers, and statistical copolymers were prepared from ionic liquid homopolymers containing a polycation/polyanion structure fabricated with ionic liquid monomers substituted with a polymerizable group [136]. The prepared polyionic liquids, via free radical polymerization and group transfer polymerization mechanisms, were applied as binders in coating processes due to their admirable film formation behavior; their antimicrobial activities were examined against *Escherichia coli* and *Micrococcus luteus*. These studies demonstrated that the antibacterial effect of hydrophilic homopolymers containing imidazolium or pyrrolidinium cation and 4-vinylbenzene moiety, and hydrophilic copolymers with alternating positive and negative sections can lead to the formation of antimicrobial polyionic liquids surfaces.

Ionic liquid bearing 3-(12-mercaptododecyl)-1-methyl-1H-imidazol-3-ium bromide (IL(Br)) moiety was prepared with the minimum inhibitory concentration against *Staphylococcus aureus* (4 μg mL^{-1}) and *Escherichia coli* (8 μg mL^{-1}). The molecular brushes decorated ion-responsive surface IL(Br)/PDA@PES was fabricated from the thiol-terminated IL(Br) clicked onto the poly(ethersulfone) (PES) membrane using polydopamine (PDA) coating as a post-functionalization platform [137]. The antibacterial IL(Br)/PDA@PES surface was assayed for bacteria-killing, and the antibacterial results showed the high bactericide efficiency of 94% and 99% against *Escherichia coli* and *Staphylococcus aureus*, respectively. The IL(Br)/PDA@PES surface could release the dead bacteria by an anion-exchange of Br^- with Tf_2N^- because of the ionic liquid ion sensitivity, and the releasing efficacies were 95% and 97% for *Escherichia coli* and *Staphylococcus aureus*, respectively. So, this intelligent antibacterial coating fabricated via a facile mussel-inspired method could achieve long-term sterilization through a simple counter anions exchange.

The neutral and ionic hybrid compounds, containing diaminothiadiazole Schiff-bases or 1,2,4-triazole and ionic liquid terminal sections, have been synthesized by a combination of quaternization, heterocyclization, and Schiff-base strategies [138]. The triazole derivative was prepared to form 4-*tert*-butylphenol through subsequent alkylation, hydrazinolysis, nucleophilic addition, and heterocyclization reactions. The in vitro bacterial growth inhibition of ionic liquid-based hybrids exhibited broad bactericide efficiency and modest to excellent antibacterial effects compared with DCMU®, as a standard antifoulant, against *Staphylococcus aureus*, *Escherichia coli*, *Aeromonas hydrophila*, and *Vibrio* sp. as common parts of marine bacteria with biofilm-inducing ability. The ionic liquid-based hybrids were combined with the inert commercial paint matrix to create the anti-biofouling coatings. The anti-biofouling ionic liquid-based hybrids could successfully prevent the creation of slime films, the adhesion and invasion of marine biofouling organisms including asidian, bryozoan, macroalgae, tube worms, and zooids onto the coated surfaces. The most effective antifouling behavior was dedicated to 2,5-bis-(3-methoxysalicylideneimino)-1,3,4-thiadiazole with astonishing resistance for colonization of zooids (0% settlement during 147 days), red algae (0% settlement during 147 days), and tube worms (12% settlement after 147 days). Thus, the blending of ionic liquid-based hybrid can offer promising antifouling composites for coating purposes to fight against marine biofoulings.

Ionic liquids encompassing benzylidene-bis-iminothiazole and salicylidene-iminothiazole Schiff bases have been prepared and explored for their antifouling and antibacterial properties [139]. The synthesized ionic liquids-based Schiff bases inhibited the growth of broad-spectrum bacteria, against *Escherichia coli*, *Aeromonas hydrophila*, *Vibrio* sp., and *Staphylococcus aureus* as the most common marine biofilm-inducing bacteria, with moderate to excellent bactericide actions. Inhibition zone examination demonstrated that the synthesized ionic liquids-based Schiff bases displayed higher efficacy against Gram-negative bacteria, *Escherichia coli*, and *Aeromonas hydrophila*. The integration of bactericidal ionic liquid-based Schiff bases into the matrix of inert commercial paints for coating formulations meaningfully improved the antifouling property of coatings applied in polyacrylic panels.

Biofouling assessment of new ionic liquid-based Schiff bases indicated that the majority of tested coatings showed the considerable anti-biofouling effect relative to the Diuron® antifoulant-containing coating under strict marine conditions. The extraordinary usefulness of ionic liquid-based Schiff bases for inhibition of biofilm formation, fouling with bryozoans, colonization of barnacle and macro-algal, tube-worms settlement, and growth of ascidians and zooids can provide the next generation of outstanding antifouling additives.

The thermostable and hydrophobic epoxy-amine networks have been fabricated from imidazolium ionic liquid monomers containing a resembling structure of bisphenol A diglycidyl ether epoxy prepolymer, with no usage of toxic epichlorohydrin and bisphenol A via a one-step polyaddition reaction-based polymerization [140]. These cross-linked epoxy networks represented strong inhibition against *Escherichia coli* biofilm formation (~95%), and their antibacterial activity is primarily associated with the hydrophobicity of polymeric networks. Thus, the development of epoxy coatings was considered a promising strategy for the construction of an antibacterial active surfaces.

7.5. Anticorrosion

Microorganisms in non-sterile solutions can significantly affect the materials/solutions interface structure and can form biofilms onto the surface of materials. The microbial colonization and the biofilms accumulation onto materials surface can cause localized gradients of dissolved oxygen, corrosive anions, and pH, resulting in an accelerated corrosion rate [141]. Therefore, the microbial corrosion which is initiated by microbes must be mitigated by killing the microorganisms or inhibiting their growth rate not only in the solutions but also on the surfaces of the materials in the environment. For this purpose, the design of new compounds with biostatic or biocidal properties was considered

Fig. 10. (a) Synthesis of cellulose acetate/poly (ionic liquid) composite. Experimental procedure: (I) Preparation of 1-vinyl-3-butylimidazolium bromide ionic liquid ([VBIm]Br IL) from the reaction of 1-vinylimidazole with n-butyl bromide at room temperature for 5 days. (II) The formation of cellulose acetate/poly (ionic liquid) composite in 3 steps; Step 1: the homogenous solution creation from cellulose acetate (CA) in dimethylformamide (DMF) in the presence of trimethylamine at 80 °C. Step 2: dropwise addition of methacryloyl chloride with DMF into the CA homogenous solution under stirring, and their reaction during 2 h. Step 3: addition of [VBIm]Br IL and 2,2′-azobis(isobutyronitrile) into the reaction solution under stirring at 70 °C for 8 h, then dropwise addition of reaction mixture into the water with vigorous stirring to obtain a white solid, and finally the filtration and freeze-drying of obtained white solid to form the modified cellulose acetate by poly (ionic liquids) (MCA-PILs). (b) Scanning electron microscopy (SEM) images of fibrous filters; I: CA, II: MCA-PILs-1, III: MCA-PILs-2, IV: MCA-PILs-3, and V: MCA-PILs-4 (MCA-PILs ingredients: 4.0 g CA, 0.4 g methacryloyl chloride, with 1.0, 2.0, 3.0 and 4.0 g [VBIm]Br, respectively). (c) The growth inhibition of bacteria by CA and MCA-PILs. (d) The antibacterial activity of CA, MCA-PILs, and recycling samples. Reprinted with modification and permission from ref. [128].

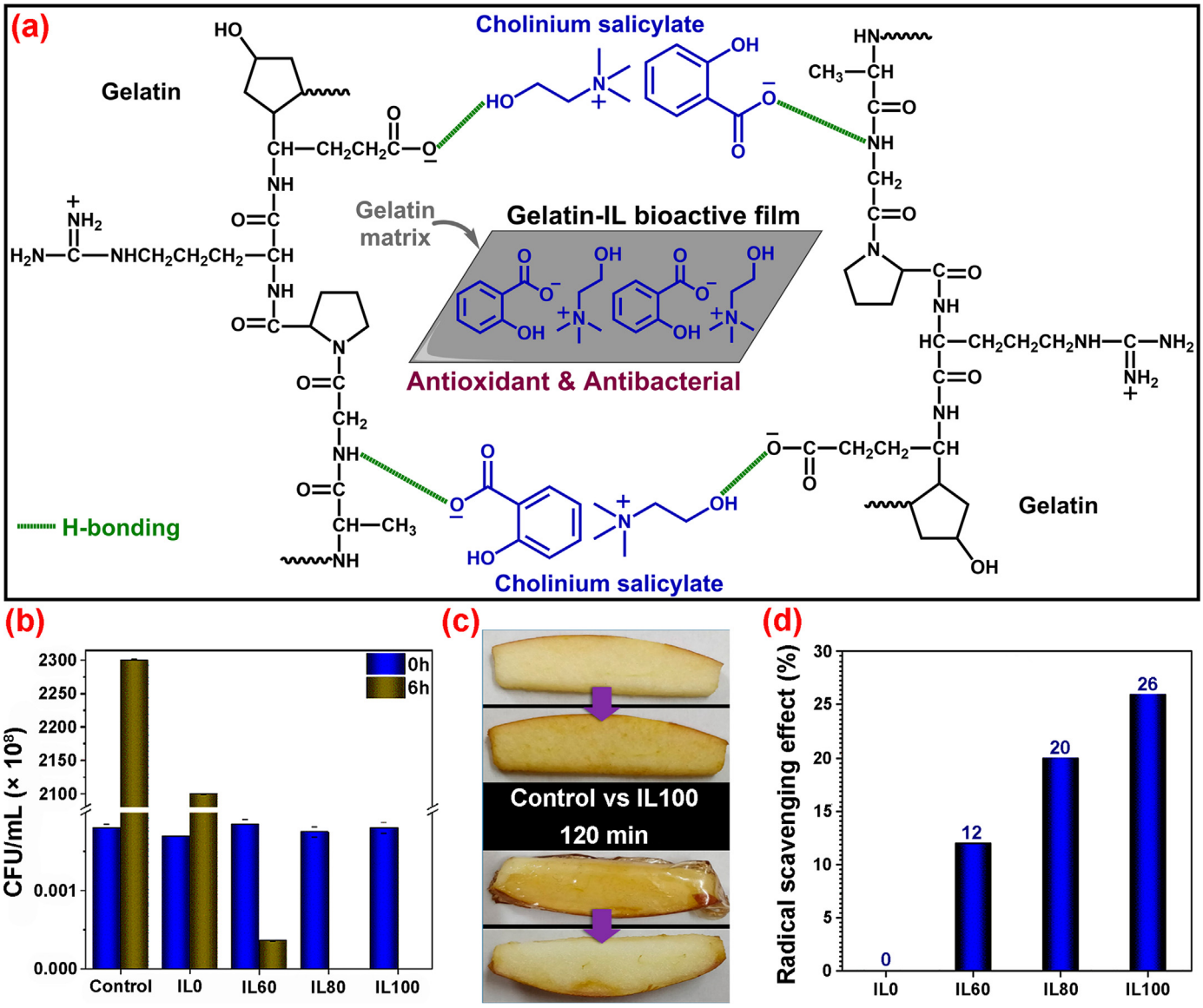


Fig. 11. (a) Ionic liquid/gelatin-based films and the H-bonding interactions between ionic liquid and gelatin in its structure. Film Preparation: I) the gelatin solution (10% *w*/*v*) formation in hot water, II) the addition of choline salicylate ionic liquid (IL) to gelatin (with 60%, 80%, and 100% *w*/w proportions), III) the creation of gelatin-IL homogeneous solutions via mechanical stirring, and IV) the preparation of different films (IL60, IL80, and IL100) from casting and drying of gelatin-IL solutions. (b) Antibacterial effect of synthesized films against *Bacillus subtilis*. (c) Test of peeled apple pieces' preservation. (d) Free radical scavenging effect of prepared gelatin films with different ionic liquid content. Reprinted with modification and permission from ref. [11].

which avoids the environmental microbial corrosion. In this context, the role of ionic liquid-based antimicrobial materials with anticorrosion impacts is reviewed here.

Metal and alloy microbial corrosion is an environmental issue that has been a serious industrial problem because it constitutes about one-fifth of the overall damages caused by corrosion. Hence, the anticorrosion effects of ionic liquid-based antimicrobial materials were investigated onto metals and metal-based materials. Recently, the antibacterial and anti-corrosion effects of 1-butyl-1-methyl pyrrolidinium trifluoromethyl sulfonate ([Py1,4]TfO) ionic liquid were studied on the planktonic and sessile bacterial growth, and mild steel corrosion [12]. The obtained results revealed that the sessile and planktonic bacterial growth was efficiently inhibited by using the [Py1,4]TfO, and the inhibition efficacy was dependent on the [Py1,4]TfO concentration. The growth inhibition of sessile bacteria required a higher concentration of [Py1,4]TfO in comparison to the planktonic bacteria. The anticorrosive efficiency of [Py1,4]TfO on the mild steel has also been evaluated in 3.5% NaCl solutions via potentiodynamic polarization and impedance spectroscopy analyses. The anticorrosive effect of more than 80% was attained by using 100 mg L^{-1} of [Py1,4]TfO on the surface of mild steel. The anticorrosion influence of [Py1,4]TfO is related to the adsorption of ionic liquid onto the mild steel surface and the formation of a protective layer against the corrosion attack.

The anticorrosion and antibacterial activities of ionic liquids-based on benzalkonium chloride derivatives (BCILs; *N*-benzyl-*N*,*N*-dimethyldodecan-1-aminium chloride: ILa, *N*,*N*-dimethyl-*N*-(4-methylbenzyl)dodecan-1-aminium chloride: ILb, *N*-(4-chlorobenzyl)-*N*,*N*-dimethyldodecan-1-aminium chloride: ILc, and *N*,*N*-dimethyl-*N*-(4-nitrobenzyl)dodecan-1-aminium chloride: ILd) were examined for the carbon steel immersed in the contaminated solution by sulfate-reducing bacteria [142]. The BCILs displayed high anticorrosive efficacy in contaminated solution by sulfate-reducing bacteria even at low concentration, due to the capability of BCILs to adsorb on the surface of carbon steel. The order of BCILs biocidal activity was determined to be ILc < ILd < ILa < ILb, and the minimum inhibitory concentration against planktonic sulfate-reducing bacteria mixed consortium was reported

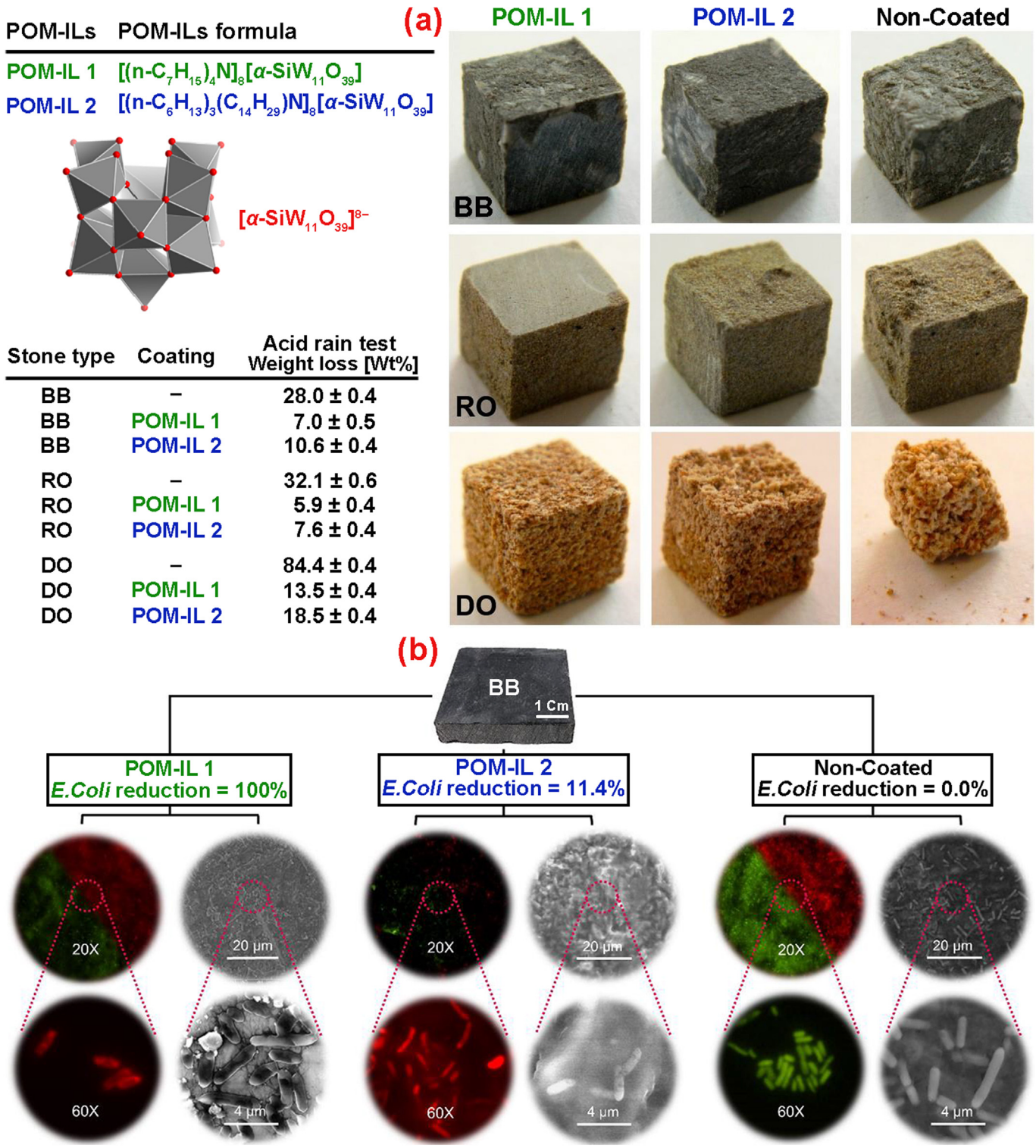


Fig. 12. (a) Anticorrosion effects of polyoxometalate-ionic liquids (POM-ILs) coated onto the stone (Belgian Blue: BB, Romery: RO, and Dom: DO) samples under acid rain exposure. Conditions: simulated acid rain: aqueous acetic acid (20 wt%), flow rate 60 mL min^{-1} per sample, and exposure time: 3 h. (b) Antibacterial activity assay of POMILs onto BB stone for prevention of *E. coli* biofilm formation via fluorescence microscopy and environmental scanning transmission electron microscopy analyses. Reprinted with modification and permission from ref. [144].

to be 200, 100, 50, and 25 μg mL^{-1} for ILc, ILd, ILa, and ILb, respectively. The BCILs biocidal effects against sulfate-reducing bacteria were controlled by an electron transfer mechanism. The sulfate-reducing bacteria tend to respond to the electron donors ILa and ILb with higher biocidal efficacy compared to the electron withdrawers ILc and ILd with lower biocidal activities. The results confirmed that the BCILs performance as corrosion inhibitors depended on the structure and concentration of ionic liquids cation. Therefore, the BCILs are considered promising protective additives in the petroleum industry to protect metal structures from corrosion and bio-corrosion.

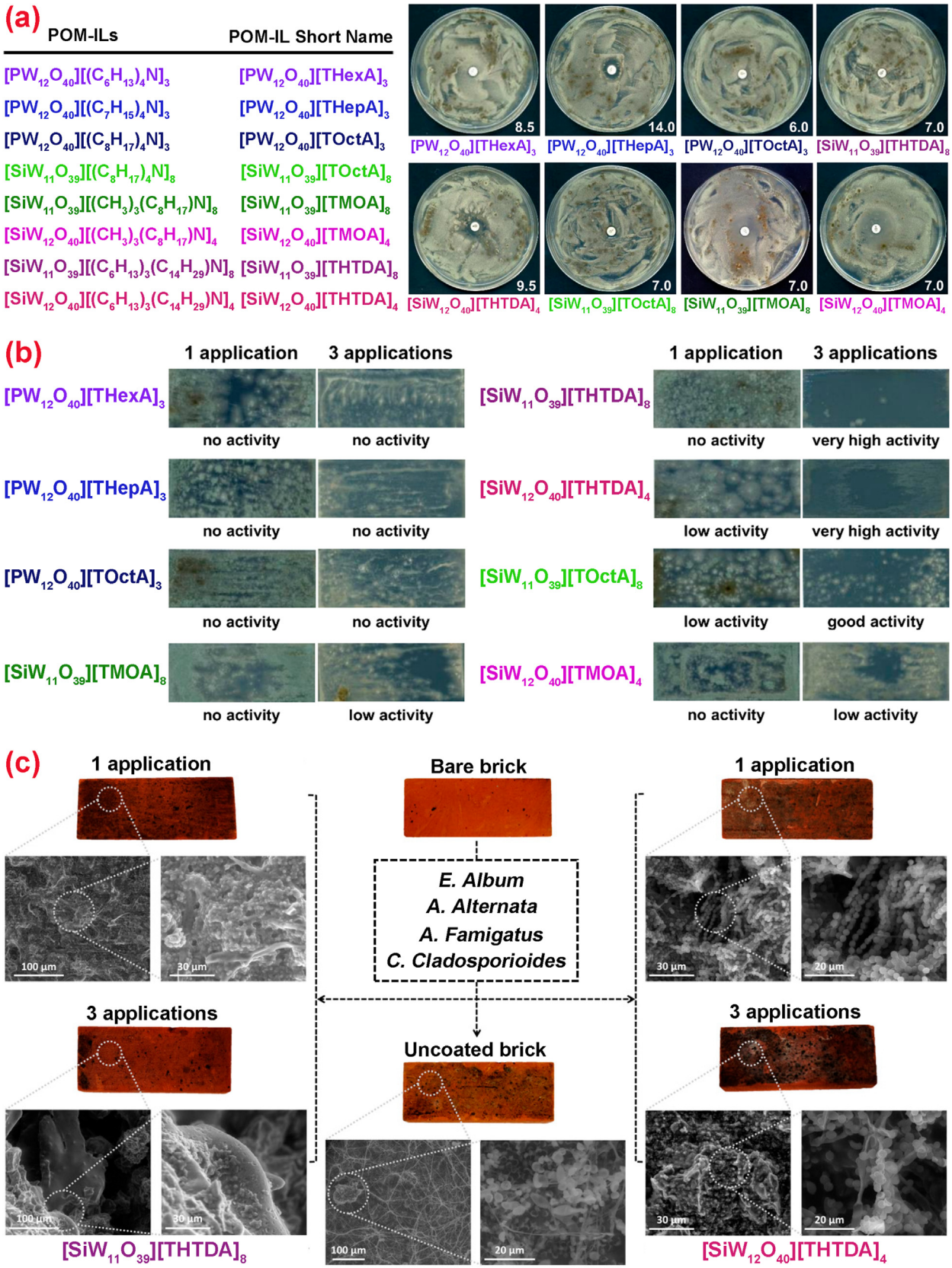


Fig. 13. (a) Inhibition of mold growth by polyoxometalate-ionic liquids (POM-ILs). (Zone's diameter unite = mm). (b) POM-ILs antifungal activity onto historical brick samples inhabited by mixed culture of molds. (c) Environmental scanning transmission electron microscopy images of antifungal effects of POM-ILs onto brick samples. Reprinted with modification and permission from [145].

The antibacterial and anticorrosion activities of 1-(2-hydroxyethyl)-3-methylimidazolinium chloride ([OH-EMIm]Cl) and 1-ethyl-3-methyleimidazolinium chloride ([EMIm]Cl) ILs were evaluated on the planktonic bacterial growth inhibition and the mild steel corrosion inhibition in 3.5% NaCl solutions [143]. The employed ionic liquids demonstrated dually function as effective biocides to mitigate the bacterial growth, and as acceptable inhibitors for controlling the microbially induced corrosion. The results indicated that the [OH-EMIm]Cl displayed a better biocidal effect in comparison to the [EMIm]Cl. The enhanced antibacterial and anticorrosive activities of [OH-EMIm]Cl were related to the incorporation of hydroxyl group into the side chain of imidazolium cation leading to the synergistic effects of O and N heteroatoms. Also, the dual effects of ionic liquids were improved with increasing the concentration of ionic liquid; inhibitory effects were obtained ~100% in the presence of 50 mg L^{-1} of ionic liquids during one hour.

Polyoxometalate-ionic liquids {POM-ILs; POM-IL 1: $[(n\text{-}C_7H_{15})_4N]_8[\alpha\text{-}SiW_{11}O_{39}]$ and POM-IL 2: $[(n\text{-}C_6H_{13})_3(C_{14}H_{29})N]_8[\alpha\text{-}SiW_{11}O_{39}]$}, as anticorrosion and antibacterial coatings, were assayed for protection of natural stones (Belgian Blue, Romery, and Dom) from weathering (corrosion) and biodeterioration (Fig. 12) [144]. Stone samples were coated with a transparent thin film of hydrophobic, acid-resistant POM-ILs with antibacterial activity. The POM-IL coated stones showed negligible corrosion compared to the unprotected stones in exposure to simulated acid rain (Fig. 12a). The surface POM-ILs coatings were mechanically stable and could not be removed even under harsh chemical and mechanical conditions. The biocidal effects of POM-ILs prevented the biofilms formation onto the coated stones by forming an antibacterial surface layer (Fig. 12b). The anticorrosive and antibacterial results confirmed that the modification of cation in the POM-ILs structure affected the anticorrosion effects as well as the antibacterial effects; POM-IL 1 displayed higher anti-corrosive and antibacterial properties in comparison to POM-IL 2.

The in vitro antifungal activity of POM-ILs ($[PW_{12}O_{40}][(C_6H_{13})_4N]_3$ or $[PW_{12}O_{40}][THexA]_3$, $[PW_{12}O_{40}][(C_7H_{15})_4N]_3$ or $[PW_{12}O_{40}][THepA]_3$, $[PW_{12}O_{40}][(C_8H_{17})_4N]_3$ or $[PW_{12}O_{40}][TOctA]_3$, $[SiW_{11}O_{39}][(C_8H_{17})_4N]_8$ or $[SiW_{11}O_{39}][TOctA]_8$, $[SiW_{11}O_{39}][(CH_3)_3(C_8H_{17})N]_8$ or $[SiW_{11}O_{39}][TMOA]_8$, $[SiW_{11}O_{39}][(C_6H_{13})_3(C_{14}H_{29})N]_8$ or $[SiW_{11}O_{39}][THTDA]_8$, $[SiW_{12}O_{40}][(CH_3)_3(C_8H_{17})N]_4$ or $[SiW_{12}O_{40}][TMOA]_4$, $[SiW_{12}O_{40}][(C_6H_{13})_3(C_{14}H_{29})N]_4$ or $[SiW_{12}O_{40}][THTDA]_4$), and their efficiency in eradication of molds from historical bricks has been evaluated (Fig. 13) [145]. The antifungal effects of POM-ILs were assayed against a mixed culture of molds involving *Alternaria alternata*, *Aspergillus fumigatus*, *Cladosporium cladosporioides*, and *Engyodontium album* isolated from the surfaces of historical brick barracks. POM-ILs displayed high antifungal activity against molds because of the mold growth inhibition through toxic effects on fungal conidia, and restriction on access to oxygen and nutrients by coating the brick surface. The obtained results revealed that the POM-ILs antimicrobial effects were mostly related to cations, and their key role emanated from the amphiphilic property allowing their insertion into the phospholipid bilayer of microbes' plasma membranes. Also, the combination of two active components in the POM-ILs structure generated the water immiscibility property that reduces the microbially induced biodeterioration. These desired characteristics provide the opportunity to design new compounds as protective coatings for reducing the microbial biodeterioration of mineral-based materials.

The amino acid bearing ionic liquids have been deployed as anticorrosive and water-based lubricating additives in water. For example, tetrabutylphosphonium amino acid ionic liquids (P4444-Cys, P4444-His, P4444-Lys, P4444-Met, P4444-Phe, P4444-Ser, and P4444-Trp) bearing tetrabutylphosphonium cation were prepared by using cysteine, histidine, lysine, methionine, phenylalanine, serine, and tryptophan amino acids as counter anions [146]. The results uncovered that the wear and friction coefficient was significantly diminished after the amino acid ionic liquids addition. The amino acid ionic liquids could intensely adsorb onto the metal surface to generate a physical adsorption film thus improving the anticorrosion effect. Besides, it could react with metal friction pairs to form chemical reaction films enriching the tribological properties of the additive. The tribological behavior of amino acid ionic liquids lubricant additives was associated with the synergy of N and P polar elements of boundary lubricant film on the steel-steel friction pair surface. Based on the biotoxicity and soil phytotoxicity analyses, the synthesized amino acid ionic liquids not only presented tribological effects but also displayed antibacterial activity and low toxicity to plants. Among the prepared amino acid ionic liquids, the P4444-Trp demonstrated remarkable tribological and bactericide effects and was less toxic to the plant. So, the environmental influence of water-based lubricating additives can be considerably improved by the amino acid ionic liquids.

8. Conclusion

Herein, the recent advances related to the environmental application of ionic liquids-based antimicrobial materials are presented. Ionic liquids have been utilized in various domains including materials science, resulting in the production of antimicrobial film membranes, nanoparticles, coatings, and surfaces and mainly demonstrating their dual-functional properties. The finding of the studies summarized throughout this review illustrated that ionic liquids-based materials are promising antimicrobial materials with an ability to combat against a wide spectrum of fungi and bacterial strains. This further highlights the potential in developing additional ionic liquids-based materials with different architectures i.e., ionic liquids macromolecular and monomer structures using hyperbranched, branched, or linear structures in order to prepare more effective antimicrobial ionic liquids-based materials. It has been observed that cationic ionic liquids-based materials were studied extensively due to the better interaction of cationic ionic liquids moieties with the negatively charged microbial cells membrane. In terms of molecular structure, the flexibility of ionic liquids is related to their capability to modulate anions and cations as salt and benefit the properties of both. Additionally, low-cost production methods for ILs, will facilitate particularly their use as antimicrobial agents against antibacterial resistance strains [92,147,148].

However, in many cases, the studies also indicated the superior activity of anionic ionic liquids-based materials with active countercations which is associated with the mobility of the cationic moieties. Therefore, in future efforts emphasis should be on investigating the potential of anionic ionic liquids antimicrobial materials and understanding of synergistic effects between counter-ion and ionic liquids. Another challenge towards the application of ionic liquids-based materials is their low selectivity values due to associated toxicity which need to be addressed in the future.

In order to have more sustainable systems, it is essential to strive for obtaining ionic liquids from renewable resources based on natural carboxylic acids, which has benefits for other applications as well. The utilization of renewable resources will also reduce the toxicity compared to synthetic ionic liquids-based materials. To obtain effective, strong, and non-harmful antimicrobial ionic liquids materials, all the features need to be considered to overcome the global health concerns of pollution, antibiotic resistance, and all existing infection threats. Additionally, the advantages encompassing bioavailability, activity, and multifunctionality are feasible via ionic liquids chemistry thus providing an encouraging novel platform for future strategies. Despite many efforts to make antimicrobials-based materials, there have been no reports about the fabrication of medical gowns, gloves, and antimicrobial coatings on the walls and floors of hospitals based on ionic liquids-based antimicrobial materials.

In our view, the key features needing further development in both, the existing and newer applications is a better understanding of ionic liquids mixtures with other ionic liquids and with molecular species. The investigations need to focus more on the structural influence on physical and chemical properties at the nanometer level that may

persevere after dilution into solvents and on the economic and environmental impacts of using antimicrobial ionic liquids in small- and large-scale industrial processes. The cost appears to be a critical barrier in the exploitation of the advantageous features of ionic liquids and identifying pathways for large-scale applications. A better understanding of both the theoretical and practical application of antimicrobial ionic liquids would be of tremendous assistance for progress in this field. Hopefully, the research community continues these innovative investigations in hitherto unexplored areas.

Declaration of Competing Interest

The authors deny any conflict of interest.

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