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Review

Ionic liquid-based antioxidant materials: a bird's eye view on the most recent literature

Salvatore Marullo and Francesca D'Anna

Dipartimento STEBICEF, Università degli Studi di Palermo, Palermo 90128, Italy

FD, 0000-0001-6171-8620

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Author for correspondence:
Francesca D'Anna
e-mail: francesca.danna@unipa.it

The unwanted effects of radicals formed in some oxidation processes have propelled the obtainment of protecting antioxidant materials for packaging or coatings. A recent development in this regard is constituted by ionic liquid (IL)-based antioxidant materials. This brief perspective surveys the literature and discusses works that appeared in the last decade on such topics. The discussion is divided into two sections: polymer composites and gel-based materials. The latter section also includes gels made by polymerized ILs. Throughout this perspective, the performance of the materials and the role exerted by ILs are discussed. This is one of the few perspectives on this topic, aiming also to discuss the trends of the research currently being conducted and possible suggestions for future research.

This article is part of the discussion meeting issue 'Ionic liquids and the future of soft materials'.

1. Introduction

Oxidation processes are the primary cause of unwanted events like food spoilage or oxidative stress and damage to cells caused by radicals, such as reactive oxygen species and reactive nitrogen species. Preventing oxidation is also primarily important to preserve the properties of materials, like polypropylene and polyethylene. This urges the search for efficient antioxidant materials, and since many of such applications involve contact of these materials with food, or human tissues, it is paramount to consider their biocompatibility and safety [1].

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The most commonly used approaches to obtain antioxidant materials to be used as protective coatings involve integration of natural antioxidants in polymer blends or surface grafting of antioxidant species on nanomaterials [2].

In many instances, the same materials can serve as protective packaging or coatings by acting as a barrier towards atmospheric oxygen.

Sustainability and safety considerations have motivated, in the last years, an increased use of biopolymers in antioxidant materials, owing to their suitable mechanical properties, robustness, low cost, in some cases, and inherent radical scavenging ability [3], which drives the development of fully natural antioxidant materials [4].

In such a context, non-conventional solvents like ionic liquids (ILs) have recently gained sizeable interest.

ILs are organic salts with a low melting point, conventionally lower than 100°C, often liquid at room temperature. Therefore, they constitute a unique class of solvents, entirely composed of ions, with low flammability and volatility, and with a remarkable structural organization. The reader is addressed to comprehensive reviews on these aspects [5–8]. In the context of materials science, an attractive characteristic of ILs is the possibility to significantly tailor their properties for a given application in a straightforward way, by changing the nature of the constituting ions. In particular, a judicious choice of the anion has been shown to yield inherently antioxidant ILs [9]. In addition, some ILs display a high solubilizing power towards biopolymer, owing to disruption of intra- and inter-chain hydrogen bonding, thus overcoming solubility issues involving biopolymers in conventional molecular solvents [10].

Based on all these considerations, this brief perspective aims to present a bird's eye view on the works that appeared in the literature from 2015 onwards, concerning IL-based antioxidant materials. It is important to note that, in this work, the term 'IL based' refers to materials in which ILs are a significant and permanent component of their formulation. Thus, it does not mean that the antioxidant action is necessarily due to the ILs. As a guiding criterion, the papers herein selected are limited to works describing antioxidant materials in which ILs are permanently retained. Materials in which ILs are only used as an aid in processing or synthesis and then removed have been excluded. Examination of the selected literature clearly shows that the IL-antioxidant materials described fall into two categories: polymeric composite materials and IL gels (ILGs), which, in this perspective, are presented and discussed separately in the following sections. In particular, the IL gels section also includes those formed by polymerized ILs. Though polymerized ILs cannot be considered ILs in their own right, given their very different properties, their solid state instead of liquid, to name one, they could nevertheless be included in this perspective, as IL-derived polymers.

As will be discussed in the following discussion, in the majority of cases, the IL itself provides antioxidant properties to the materials, deriving from inherently antioxidant cations or anions. In other cases, the presence of IL is nonetheless crucial to facilitate component solubilization, blending and dispersion. Thus, in such cases, the presence of ILs is still relevant, because they endow the materials with other properties that are essential for their very synthesis or to exert their functions, including the antioxidant one.

(a) Composite materials

A viable strategy to improve the properties of materials is constituted by the use of composite materials, in which a synergistic effect induced by the concomitant presence of the mixture often results in improved mechanical strength, stability and robustness. In this context, Gomes *et al*. prepared beads of the nanocomposite formed by two polysaccharides, namely alginate and acemannan [11], loaded with the IL cholinium caffeate [Ch][Caff] (figure 1a).

The materials used in this work were prepared by Ca(II)-induced gelation of alginic acid, in the presence of acemannan and variable amounts of [Ch][Caff], ranging from 0.5 to 3 wt%. The IL considered is composed of non-toxic ions, and the IL anion is a polyphenolic compound,

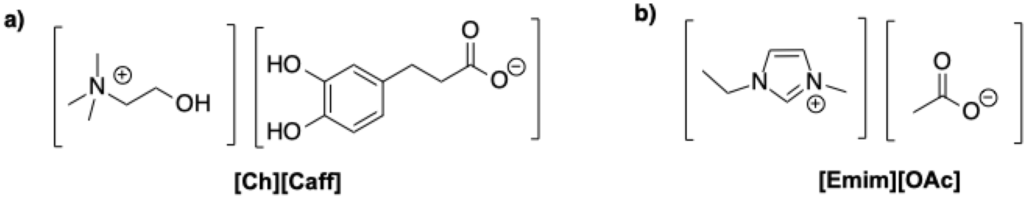

Figure 1. Structure of (a) cholinium caffeate and (b) [Emim][OAc].

therefore endowing the materials with antioxidant ability. Thus, the IL imparts its antioxidant properties to the composite materials prepared. Radical scavenging efficiency, evaluated against 2,2-diphenyl-1-picrylhydrazyl radicals (DPPH) [12,13], increased by 95% in the presence of the IL, compared with the pristine composite. This material achieved high antioxidant ability coupled with anti-inflammatory effect, and the solid matrix could be used for the sustained release of [Ch][Caff], showing potential as a therapeutic tool for osteoarthritis.

A commonly sought-after application of composite materials with antioxidant activity is food packaging. In this context, it is important that the materials employed are not pollutant agents. Following this approach, a composite material was prepared, blending the hydrophobic polymer poly(caprolactone) with the hydrophilic cellulose (CE), in the presence of the IL [Emim][OAc] (figure 1b) [14]. In the absence of the IL, the material could not be prepared. In particular, the monomer ε-caprolactone was polymerized in a CE solution in [Emim][OAc], in the presence of ZnO nanoparticles as catalyst. The ensuing materials showed uniformly dispersed nanoparticles and smooth surfaces. Notably, the composite material showed the same crystallinity as the parent CE, showing superior antioxidant ability and UV–visible barrier properties compared with the parent polymers. Furthermore, the IL further contributes to providing antioxidant ability, given that imidazolium acetate ILs have been reported in the literature to possess appreciable radical scavenging ability [15].

In a related example, doping of a biopolymer nanocomposite with a cholinium-based IL was shown to be essential to endow the material with antioxidant ability [16]. In particular, IL cholinium indole-3-acetate (figure 2a) was added in high amounts, from 25 to 100 wt% of biopolymers, to composites of sodium alginate and k-carrageenan, to obtain transparent colourless films (figure 2b). The authors determined the radical scavenging efficiency of the IL, finding that it showed inherent antioxidant ability, and for this reason, added it to the formulation of materials to confer further antioxidant properties. More specifically, the presence of the IL enhanced the radical scavenging ability of the nanocomposite, which increased from 28 to 65% as assessed by the DPPH assay and imparted antibacterial and partial UV-blocking properties. Adding a further 0.9 wt% of lignin (LG) produced coloured transparent films with higher blocking of UV-A radiation, leaving the mechanical and antioxidant properties practically unchanged (figure 2c).

In a different approach, ILs are not directly employed to provide radical scavenging ability to the material, but rather, to ensure uniform dispersion of the antioxidant agent. Such a strategy was employed by Amini *et al.*, using the IL [Emim][OAc] to prepare LG nanoparticles by sonication as the antioxidant agent [17]. These dispersions were then added with CE to produce the all-CE composite material. The resulting materials showed increasing antioxidant ability, as evaluated by the ABTS assay [18], on increasing LG nanoparticle content up to 7 wt%. As can be seen from the pictures reported in figure 3, the inclusion of LG nanoparticle maintained the transparency of the CE films, although a lower crystallinity degree was observed.

As previously mentioned, a notable characteristic of ILs is the possibility to endow them with specific properties by a judicious choice of the ions. Therefore, in the context of IL-based antioxidant materials, this property can be conferred by using an IL with an antioxidant anion or cation. This strategy was employed in fabricating films from a composite of gelatin

Figure 2. Structure of (a) cholinium indole-3-acetate, (b) films of sodium alginate/k-carrageenan composite with IL and (c) the same films after adding 0.9 wt% of LG. Adapted with permission from *ACS Sustainable Chem. Eng*. 2024, *12*, 4314. Copyright 2024 American Chemical Society.

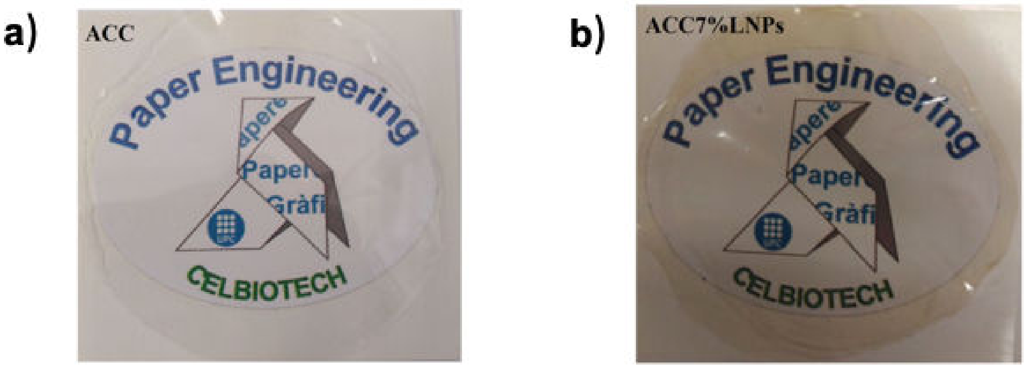

Figure 3. Pictures of all-CE films (a) without LG and (b) with 7 wt% LG nanoparticles. Adapted from Amini *et al.* *Wood Sci. Technol*. 2023, *57*, 1319–1344, licensed under a Creative Commons Attribution 4.0 International License, https://creativecommons.org/licenses/by/4.0/

and carboxymethylcellulose, which employed the IL 1-methyl-3-octyl imidazolium salicylate, [omim][Sal] [19], depicted in figure 4a.

The IL was used in the formulation of the film in varying amounts, ranging from 5 to 20 wt%. The results obtained showed that adding the IL confers radical scavenging ability to the ensuing film, owing to the presence of the salicylate anion, having well-known antioxidant ability. Furthermore, the addition of the IL also endowed the material with antibacterial ability, tested against *Escherichia coli* and *Staphylococcus aureus* strains, and improved tensile strength. Finally, the material prepared with 20 wt% of IL could be used to develop a package for apples, which resulted in significant protection from oxidation compared with untreated control.

A similar approach was pursued by Lu *et al.*, developing films based on different types of collagen peptides by mixing them with the IL arginine lactobionate, [Arg][LAA], reported in figure 4b [20]. Once again, the IL anion has well-established antioxidant properties, also retained in the collagen-based films, prepared at concentrations of biopolymer ranging from 0.5 to 2.0 wt%. In such a medium, collagen peptides undergo self-assembly, giving rise to a transparent film, enabling a significant release and penetration of collagen peptides when in contact with skin. Finally, compatibility tests on human skin cells demonstrated the potential of such films in the cosmetic field.

Alkaloid-derived ILs based on matrinium cation and long-chain carboxylates, [Mat][Coc], depicted in figure 5, were employed in mixtures with gelatin to obtain transparent composite films [21].

Films obtained at IL concentrations ranging from 5 to 10 wt% exhibited radical scavenging efficiencies higher than 90% as assessed by both DPPH and ABTS assays. In addition, these materials exhibited strong thermostability, with decomposition temperatures equal to or higher than 200°C, and when applied as coating on apples, significantly reduced browning and weight loss, compared with the untreated control. Also in this case, IL with antioxidant ions endows the final material with radical scavenging efficiency.

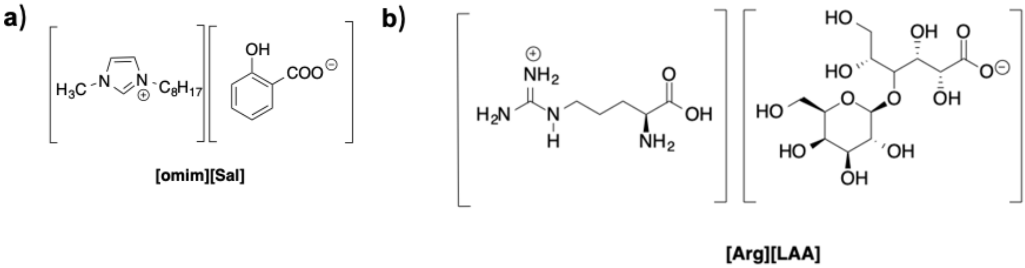

Figure 4. Structure of (a) 1-methyl-3-octyl imidazolium salicylate and (b) arginine lactobionate ILs.

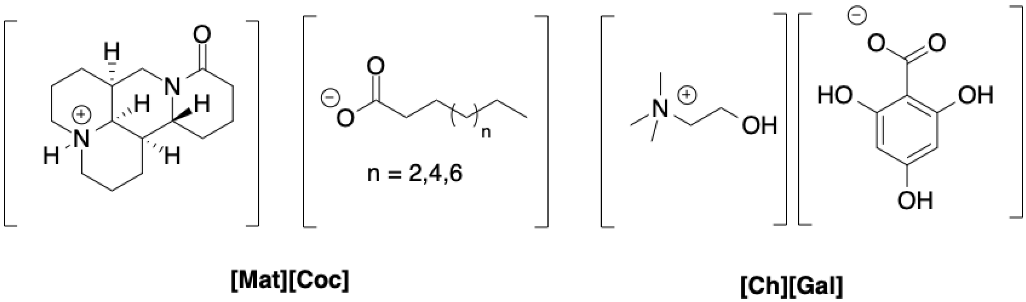

Figure 5. Structure of matrinium-based ILs.

As a further example, Gomes *et al.* [22] described the preparation of antioxidant sponges of silk fibroin in the presence of the IL cholinium gallate, [Ch][Gal], reported in figure 5b, whose anion has antioxidant properties. Solid-state nuclear magnetic resonance and Fourier transform infrared analyses pointed out that the structure of fibroin is preserved in the IL-doped sponges, which also showed increased radical scavenging ability compared with the control sample, without IL. The materials were able to support adipose stem cell growth and proliferation, proving blood compatibility as evaluated by haemolysis tests. In addition, they showed anti-inflammatory effects owing to inhibition of tumor necrosis factor-α, a protein involved in chronic conditions such as Crohn's disease or rheumatoid arthritis. However, a disadvantage of these materials lies in the rather long and elaborate preparation procedure.

In a different example, Tyagi *et al.* reported starch/zein composite films, in which the IL [bmim][Cl] was used as plasticizer, enhancing the tensile strength of the film [23]. In this case, antioxidant ability was imparted by adding sorghum bran extract, which also conferred UV-shielding ability to the material. Remarkably, the presence of the IL was crucial to the preparation of the material, owing to a substantial enhancement of solubilization of sorghum bran extract and of its compatibility with the other biopolymers.

The dual role of plasticizer and inducer of radical scavenging ability by an IL is demonstrated by the preparation of collagen-polyvinyl alcohol (PVA) composite films added with cholinium acetate, [Ch][OAc], depicted in figure 6a [24].

Seeking application of such materials as therapeutic agents, the composite films were doped with the antibiotic ciprofloxacin. Preparation of the films, schematically depicted in figure 6b, involved mixing PVA with aqueous acidic solutions of collagen extracted from sliced fish skin, followed by addition of ciprofloxacin and [Ch][OAc] in amounts ranging from 25 to 50% with respect to the dry collagen/PVA composite mass. Subsequently, the gelatinous solution obtained was cast into a Petri dish, and the film was obtained after drying under reduced pressure.

The films prepared showed excellent radical scavenging efficiency and UV-blocking ability. Notably, when compared with the relevant films in which a commonly used plasticizer, glycerol, was employed, the IL-based film showed increased surface roughness and

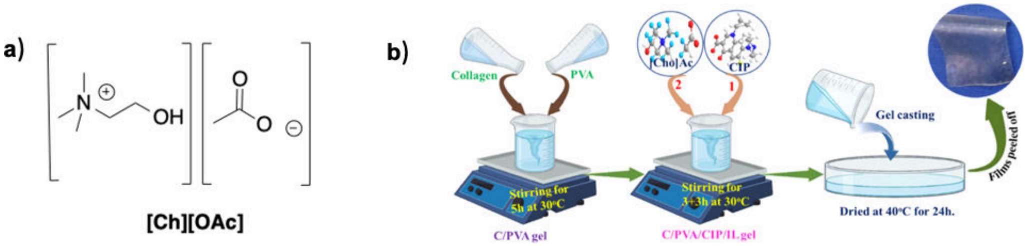

Figure 6. Structure of (a) cholinium acetate and (b) schematic of the preparation of collagen/PVA films doped with ciprofloxacin. Adapted with permission from *ACS Appl. Bio Mater*. 2023, *6*, 663. Copyright 2024 American Chemical Society.

hydrophobicity. Finally, while the glycerol-based film doped with ciprofloxacin exhibited burst release of the drug, for the IL-based types, a sustained release profile was observed.

For a different application, a poly(*p*-phenylenediamine)/Fe_3O_4 doped with the IL pyridinium hydrogenosulfate showed antioxidant ability, brought about by both the polymer and iron oxide. This composite was used as a heterogeneous catalyst for the one-pot synthesis of polyhydroquinoline derivatives from aromatic aldehydes, dimedone, benzyl acetoacetate and ammonium acetate, as shown in figure 7 [25].

In this case, IL endows the catalyst with Brønsted acidity, enhancing catalytic efficiency.

More recently, Lu *et al*. reported on the preparation of liposomes formed by the IL obtained from betaine and the carotenoid pigment astaxanthin (ASTA-Bet; figure 8) in the presence of soybean lecithin and polysorbate as stabilizing agents [26].

Astaxanthin is a well-known potent antioxidant, but it is sparingly soluble in water. Therefore, conversion into an IL significantly increased its solubility in aqueous medium, allowing it to self-assemble into liposomes. These materials showed good stability, skin permeability and *in vivo* antioxidant ability, in this case entirely conferred by the ASTA-Bet antioxidant IL anion.

(b) Gel-based materials

Among the various classes of antioxidant materials, gels have gained increasing interest owing to their convenient properties. In particular, gels are viscoelastic materials formed by dispersing a solid (gelator) in a liquid phase (solvent). Despite being predominantly composed of liquid, their rheological behaviour is solid-like, since the gelator forms a sample-spanning network, able to entrap significant amounts of solvent by capillary forces, which in turn prevents it from flowing [27]. Gels are usually classified as covalent or supramolecular. In covalent gels, the gelator is a cross-linked polymer that forms a gelatinous network underpinned by covalent bonds. On the other hand, supramolecular gels originate from the self-assembly of mostly low-molecular-weight molecules, but also polymers, and are underpinned by non-covalent interactions [28].

Another common classification subdivides gels based on the solvent in which they are prepared, thus distinguishing them in hydrogels (water) or organogels (organic solvents). A recent development in this field is represented by gels in non-conventional solvents, namely eutectogels (deep eutectic solvents) and ILGs [29–31]. In particular, ILGs allow joining the convenient properties of ILs with those of solid-like nature of gels and have found application in different fields, such as catalysis [32,33], non-conventional reaction media [34], antibacterial [35] and antioxidant materials [15]. A notable advantage of ILGs over those based on molecular solvents is the minimization of solvent leaching from the gel by evaporation, owing to the very low volatility of ILs.

Regarding antioxidant ILGs, Luque *et al.* described the preparation of ILGs of PVA, in cholinium-based ILs bearing polyphenol anions [36], namely salicylate [Ch][Sal], gallate [Ch][Gal], caffeate [Ch][Caff] and vanillate [Ch][Van], whose structures are reported in figure 9.

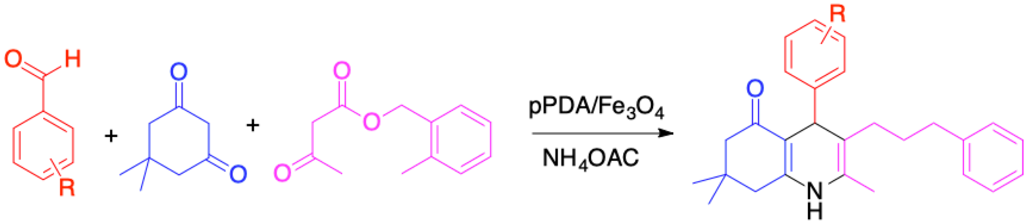

Figure 7. One-pot synthesis of polyhydroquinoline derivatives.

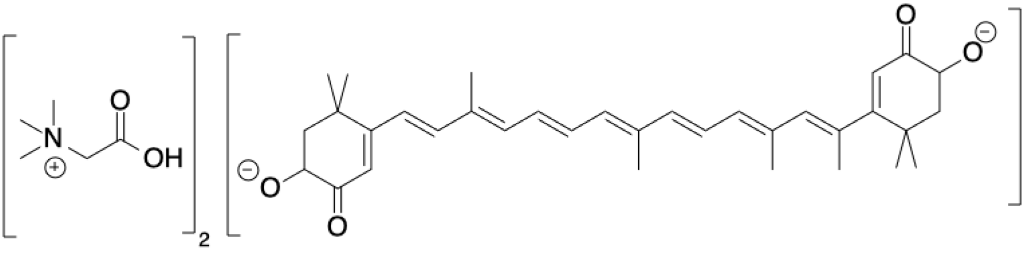

Figure 8. Structure of ASTA-Bet IL.

The phenolic moieties in the IL anions not only endow the gel with antioxidant ability but also play the role of cross-linker for the PVA chains. All the gels obtained proved thermoreversible, flexible and elastic. In addition, they showed significant antioxidant and antibacterial activity, with the PVA-[Ch][Van] gel exhibiting the highest radical scavenging efficiency, while the PVA-[Ch][Sal] displayed the best antibacterial activity, tested against the *E. coli* strain. Finally, all these ILGs proved blood compatible, showing promise for application in wound healing patches.

Antioxidant polymer ILGs, once again used for wound dressing purposes, were reported by Zhi *et al.* [37]. The preparation of these gels follows a quite complex route, as anthocyanidin is first immobilized in a dual cross-linked ILG formed by pectin, acrylamide and the IL 1-hydroxyethyl-2-methylimidazolium chloride, in the presence of $Ti_3C_2T_x$ MXene, acting as cross-linker (figure 10). This suspension was then added to ethylene diacrylate and photopolymerized.

Sterilized ILGs were then applied to wounds on mice, showing significant healing compared with the control sample, which was attributed to the radical scavenging ability provided by anthocyanidin and pectin. It is, however, important to note that, in this work, the antioxidant ability was not quantitatively determined.

As already mentioned, ILs are particularly suited to obtain biopolymer-based gels. Along this line, Marullo *et al.* obtained ILGs from binary mixtures of biopolymers such as CE, chitosan (CS), chitin (CT) and LG (figure 11a) [15]. The gels were formed in imidazolium and pyrrolidinium-based ILs, reported in figure 11b, with a simple heat-cool cycle.

Gelation tests showed that most of the gels formed in the presence of CS-containing mixtures, whereas no gel formed from the CT : LG mixture. An important advantage of these gels is that they are supramolecular in nature and their preparation is straightforward, without any need for cross-linkers or radical initiators. Notably, the IL anion had a marked influence on the properties of these gels. In particular, rheological measurements showed that gels obtained in chloride-based gels were the stiffer ones, while those obtained in acetate-based ILs proved more flexible. All the gels showed high scavenging efficiency for DPPH radicals, always higher than 90% and always significantly higher than the scavenging efficiency of the individual components. Once again, the IL anion exerted a major influence, with gels formed in acetate-based IL showing faster radical removal. Finally, the gels also exhibited bactericidal effects against the *E. coli* strain.

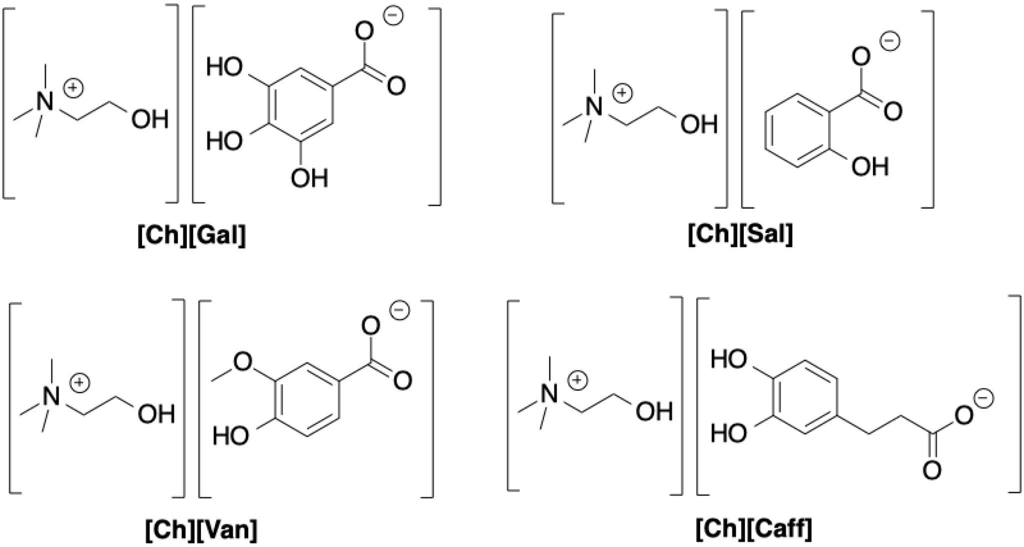

Figure 9. Cholinium-based ILs with polyphenol anions used to obtain ILGs from PVA.

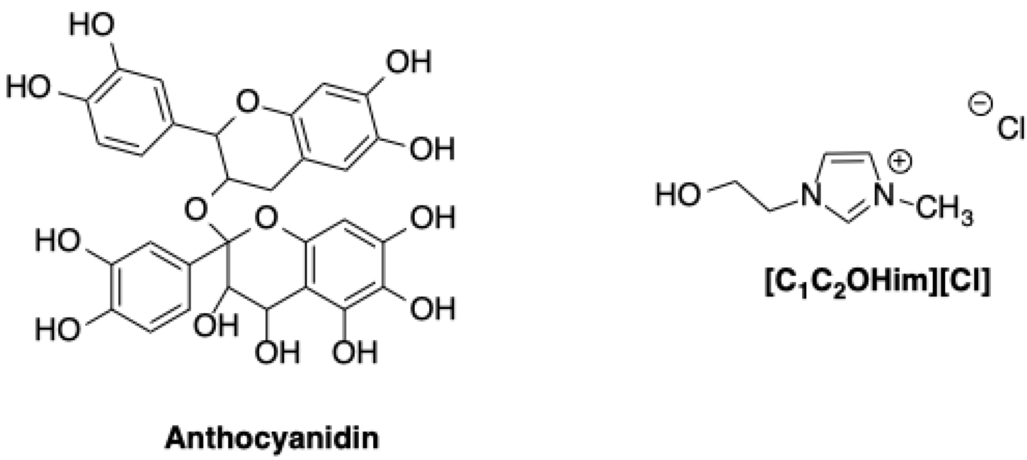

Figure 10. Structures of anthocyanidin and ILs used.

Incorporation of natural antioxidants in hybrid ILGs has been shown to enhance their radical efficiency. This is the case of hybrid supramolecular gels of some diimidazolium polycarboxylate salts, depicted in figure 12a, which harden several imidazolium and pyrrolidinium ILs, reported in figure 12b [38]. These gels were further doped with 1-carboxypropyl carbon nanotubes, non-covalently modified with natural antioxidants, including α-tocopherol and quercetin (figure 11c).

The presence of the carbon nanomaterial enhances the gelation propensity of the diimidazolium salts, exerting a template effect on the gelation process. Despite the very low amount of antioxidants, 0.0002 wt%, the gels achieved almost complete scavenging of DPPH after 1 h, and the radical scavenging efficiency of the gel was found to be higher than those of the individual gel components.

No leaching of carbon material was detected from the gel, which could be potentially spread as a protective coating. Interestingly, the gel could be dispersed within a poly(methylmethacrylate) film, maintaining its antioxidant properties.

In another example, antioxidant supramolecular ILGs were obtained by doping gel formed by diimidazolium dicarboxylate salts as gelator, in imidazolium ILs, incorporating nitrogen-doped carbon nanodots (NDCNs; figure 13) [39].

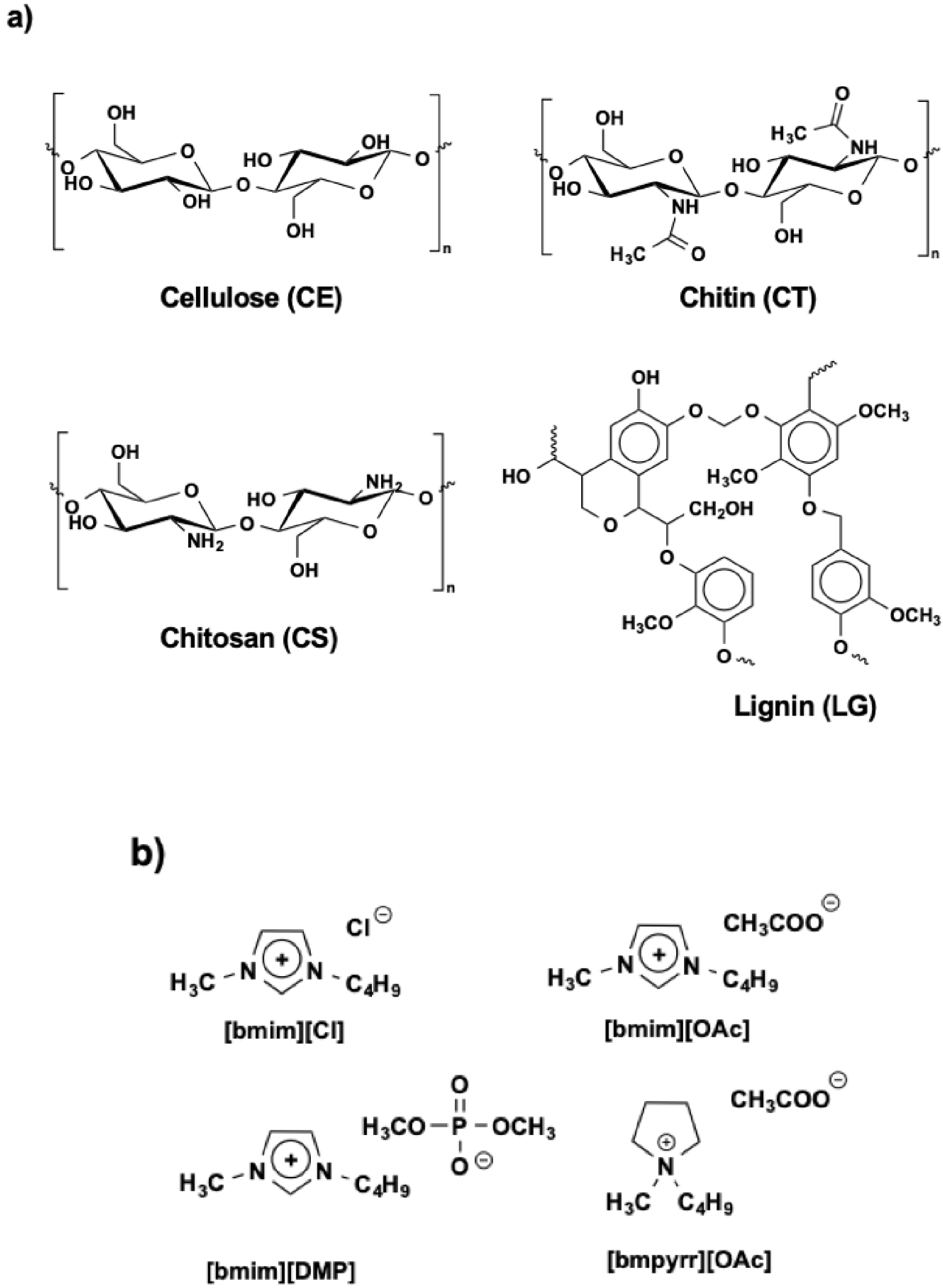

Figure 11. Structures of (a) biopolymer and (b) ILs.

The inclusion of NDCN into the gel matrix exerted a stabilizing effect on the gel owing to H-bonding and π-stacking interactions. In addition, NDCNs endowed the ensuing gel with emissive ability and with high radical scavenging efficiency, evaluated against DPPH radicals. Notably, the antioxidant efficiency of NDCNs within the gelatinous network was higher than the one displayed in the solution of the ILs considered, evidencing a favourable effect of the confinement in the gel phase.

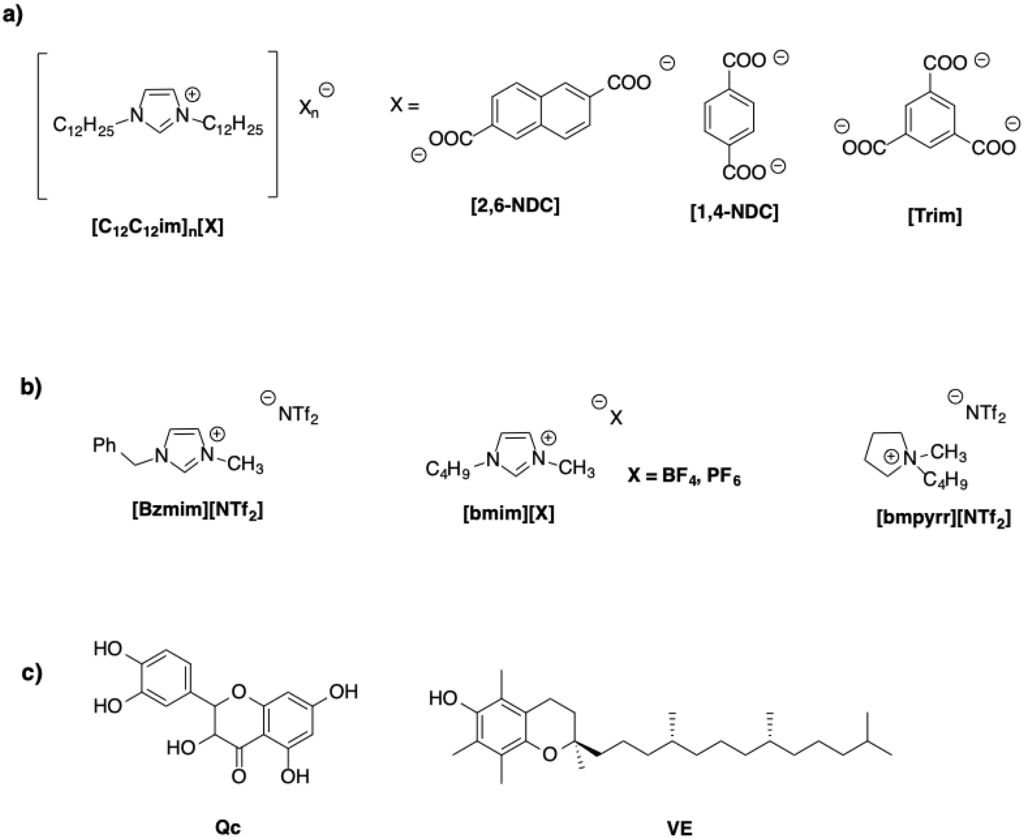

Figure 12. Structures of (a) gelators, (b) ILs and (c) natural antioxidants.

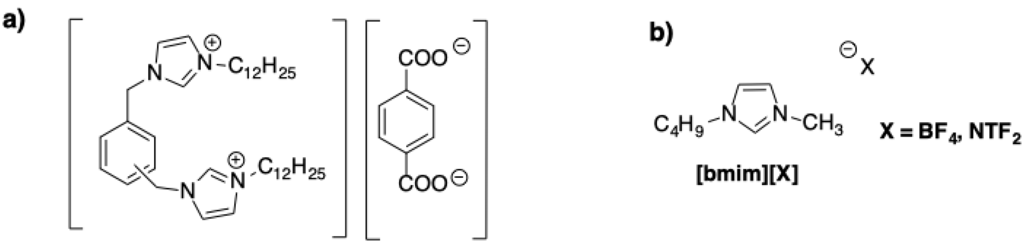

Figure 13. Structures of (a) gelators and (b) ILs.

Mehta & Kumar reported on antioxidant films obtained from ILGs of gelatin in the IL cholinium salicylate, [Ch][Sal] [40]. The gels and the relevant films, obtained in pure ILs (IL-100; figure 14a), were transparent with a tensile strength of 100 MPa. The IL imparted radical scavenging ability to the films, although a reduction of efficiency assessed by DPPH assay was detected on going from neat IL to the gel phase. These films were tested as protective packaging for *Malus pumila,* resulting in significant preservation effects compared with the untreated control, as shown in figure 14b.

Other works reported in the literature deal with antioxidant gels obtained in water upon polymerization of vinylimidazolium ILs. As previously mentioned in the introduction section, although these hydrogels formed by poly(ILs) cannot be strictly considered as IL-based, this section also covers these works, since in our view they are consistent with the focus of this perspective. As an example, a hydrogel was obtained by photopolymerization of a vinylimida-

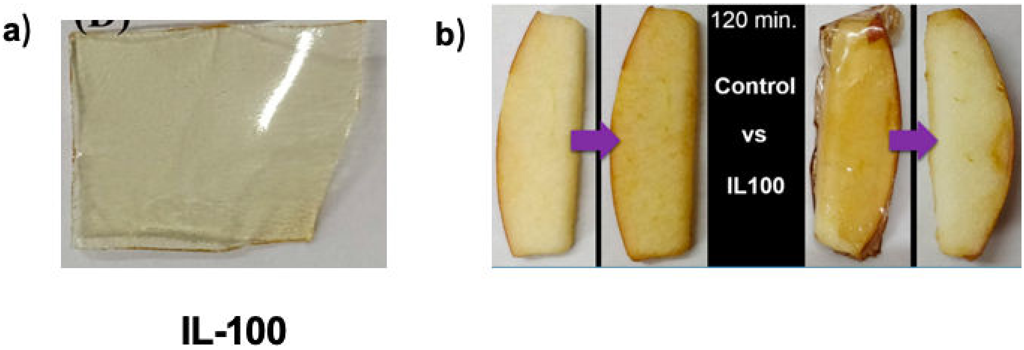

Figure 14. Pictures of (a) gelatin/[Ch][Sal] film and (b) treatment of *Malus pumila* with film. Adapted with permission from *ACS Sustainable Chem. Eng*. 2019, *9*, 8631. Copyright 2019 American Chemical Society.

zolium IL bearing a boronic acid functionality (figure 15) within a methacrylate gelatin matrix [41].

The IL not only initially allowed solubilization of the polymer but also acted as a cross-linker during photopolymerization. The hydrogels showed antioxidant activity evaluated both against DPPH and PTIO, a water-soluble radical. The extremely rapid gelation time of 3 s made these gels particularly suitable for application as wound dressing. Accordingly, successful wound healing was observed upon application by *in situ* photopolymerization on wounds of diabetic mice. It is, however, worth noting that the authors did not investigate in this case which gel component provides antioxidant ability.

Using a similar approach, Zhang *et al.* reported a hydrogel obtained by copolymerization of 1-butyl-3-vinyl imidazolium bromide with methacrylate-functionalized LG and hydroxyethyl-methacrylate as shown in figure 16 [42].

The gels obtained showed high tensile strength and long-term antibacterial activity. In addition, high antioxidant ability was detected against DPPH radicals and proved efficient dressing when applied on skin wounds. In this material, antioxidant ability was ascribed mainly to the polyphenol backbone of LG.

In another example, a hydrogel was obtained by a poly(IL) obtained by polymerizing 1-(3-aminopropyl)-3-(4-vinylbenzyl) imidazolium chloride (figure 17a) in the presence of oxidized sodium alginate and a mixture of polysaccharides known as dendrobium officinale polysaccharides (DOP) [43].

The material obtained showed antioxidant and antimicrobial properties, proving efficient in promoting *in vitro* and *in vivo* diabetic wound healing. However, the antioxidant properties were due only to the presence of the DOP components, while the IL provided antimicrobial properties.

Similar comments can be drawn from the work by Zaki-Germi *et al*., describing the synthesis of a composite based on functionalized polyphenolic two-dimensional nanosheets and gelatin methacrylate, forming a hydrogel [44]. In particular, the polyphenolic nanosheet, obtained from pyrolysis of beehives residues, was first functionalized with a diimidazolium IL (figure 17b) and then photopolymerized in the presence of gelatin methacrylate to form a hydrogel. This material has good mechanical, antioxidant and antimicrobial properties. Once again, the antioxidant activity was brought about by the polyphenolic nanosheet, whereas the IL only provided antimicrobial properties against different pathogenic bacterial strains. A limitation of this protocol is the rather long and laborious route required to obtain such material.

In all the examples seen so far, the antioxidant ability is provided to the gel by one of its components. In a variation to this theme, the radical scavenger species can be generated *in situ*, within the gel. In a notable example, Xu *et al*. constructed a poly(IL) based hydrogel capable of releasing the antioxidant species, i.e. hydrogen [45].

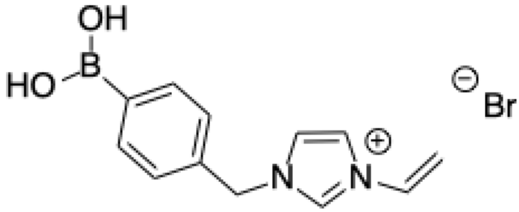

Figure 15. Structure of boronic acid-functionalized monomer.

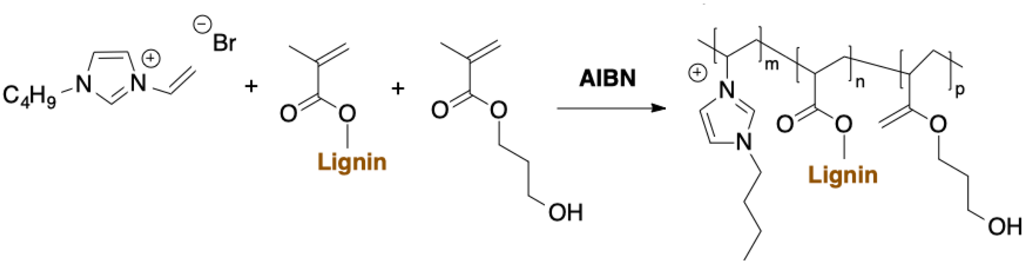

Figure 16. Schematic of copolymerization to obtain hydrogels.

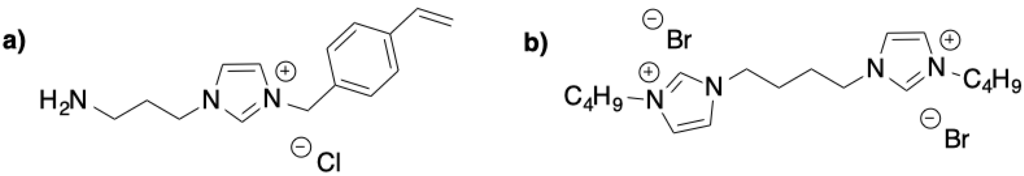

Figure 17. (a) Structures of polymerizable IL and of (b) diimidazolium IL.

In particular, the gel component which releases H_2 is Chlorella, a kind of green algae that, when present in agglomerated state, in a hypoxic environment, release hydrogen. In the light of this, Chlorella agglomerates were formed by flocculation in water, induced by the poly(IL) previously obtained by 1-hydroxymethyl-3-vinyl chloride. This mixture is further photopolymerized in the presence of methacrylate-functionalized hyaluronic acid.

The hydrogels showed high radical scavenging efficiency, as evaluated in solution against DPPH and peroxy radicals. The antioxidant ability was also investigated against reactive oxygen species in a cellular environment by confocal microscopy. The release of H_2 also endowed the hydrogels with antibacterial properties, making them suitable for application on to diabetic wounds.

2. Conclusion

The aim of this brief perspective is to provide a bird's eye view on recent application of ILs to obtain antioxidant material. The works, covering the last decade, clearly show that most of the efforts have been geared to composite materials films of IL-based gels. In addition, the vast majority of applications are focused on two kinds of applications, namely protective food packaging and wound dressing. A further look at the works covered in this perspective allows us to draw the following comments on the research currently done on this topic:

— ILs appear particularly advantageous over molecular solvents for designing antioxidant materials for three main reasons. First, their low volatility provides more robust antioxidant materials, owing to minimized leaching of solvent by evaporation. The second is the high solubilizing power towards different types of biopolymers, enabling preparation of biocompatible antioxidant materials. Third, unlike conventional organic solvents, ILs are often active components of such materials providing radical scavenging efficiency to the ensuing materials. It is worth noting that such ability can be imparted in a straightforward way to ILs, simply by a judicious choice of the constituting ions.

— The overwhelming majority of the works covered here, either composite or gel-based materials, employ imidazolium-based ILs. Since antioxidant materials are expected to be used in contact with food or human skin, it is important that the ILs used are as low toxic as possible, biocompatible and also beneficial in terms of sustainability. For these reasons, future research on this topic would significantly benefit from shifting to safer and less toxic ILs, like those based on aliphatic cations [46] and also to bio-based ILs [47].

As a final note, herein, the aim was to give a clear picture of the work done in the last decade, evidencing the significant progress and providing possible suggestions for future work in this area, hoping that this could incite the search for novel applications of these materials.

Data accessibility. This article has no additional data.

Declaration of AI use. We have not used AI-assisted technologies in creating this article.

Authors' contributions. S.M.: funding acquisition, writing—original draft; F.D.: conceptualization, funding acquisition, supervision, writing—review and editing.

Both authors gave final approval for publication and agreed to be held accountable for the work performed therein.

Conflict of interest declaration. We declare we have no competing interests.

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