electronic materials

Review

Polymer Composites Containing Ionic Liquids: A Study of Electrical Conductivity

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Abstract: Polymer composites are engineered materials that combine polymers with diverse fillers to enhance their physicochemical properties. The electrical conductivity of polymer composites is a vital characteristic that significantly broadens their use, particularly in electronic applications. The addition of ionic liquids into polymer composites represents a new method to enhance their functional properties, particularly in terms of electrical conductivity. In this brief review, several polymer matrices, conductive fillers, and ionic liquids utilized in polymer composites are categorized. Additionally, the effect of ionic liquids on the electrical conductivity of polymer composites is concisely explained. This review gives brief information that increases the understanding of electrical conductivity in polymer composites containing ionic liquids. In summary, most studies show that adding ionic liquids enhances the electrical conductivity of polymer composites regardless of the polymer matrix or conductive filler type. This enhancement is due to ionic liquids improving filler dispersion and promoting the creation of effective three-dimensional conductive networks within the matrix, thus boosting electron transport and mobility throughout the structure. This review provides new insights into the behavior of ionic liquids in composite systems, highlighting their role in improving properties for advanced applications. It encourages innovation in next-generation conductive materials and assists future research and development of more efficient materials for electronics.

Keywords: ionic liquid; polymer composite; electrical conductivity; thermoplastic; elastomer; thermoset

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1. Introduction

Polymer composites are concocted materials fabricated by combining polymers with various fillers or additives to improve their physicochemical properties. The integration of a polymer matrix with fillers or reinforcements like carbon, glass, aramid fibers, etc., characterizes these composites. This fabrication results in materials that display superior strength, robustness, and thermal stability compared to their polymer matrices [1]. Polymer composites are broadly applied in industries such as the construction, automotive, and aerospace fields and offer adaptability and high performance under required conditions. Their customized properties make them valuable for particular applications, transforming material design and creating new probabilities in engineering and manufacturing [2]. The electrical conductivity of polymer composites is a crucial property that substantially expands their applicability, specifically in electronic and electrochemical applications [3]. Conventionally, polymers are insulative, but the incorporation of conductive fillers alters these composites into electrically conductive materials [4]. Conductive fillers such as carbon nanotubes, graphene, carbon black, and metal particles construct pathways that assist electron flow when distributed within the polymer matrix [5]. The percolation threshold is the critical point at which the composite conversions from an insulator to a conductor remain vital for optimizing conductivity [6]. Beyond this threshold, a minor rise in filler content can lead to a significant improvement in conductivity [7].

Moreover, there is a firm reliance on the filler particle geometry, alignment, dispersion, aspect ratio, and filler–filler interactions [8]. This designed conductivity is essential for applications like electromagnetic shielding, anti-static packaging, and anti-corrosion technology [9]. Additionally, the mechanical flexibility combined with electrical functionality permits these composites to be worn in wearable electronics, where they can withstand bending and stretching while retaining their conductive properties [10]. Therefore, comprehending and tuning the electrical conductivity of polymer composites is critical for the development of advanced materials that converge exact technological requirements, making them valuable in innovative materials science and engineering. Adding ionic liquids into polymer composites signifies a novel approach to further improve their functional properties, especially in electrical and thermal conductivity [11,12]. Ionic liquids are salts that remain liquid at temperatures below 100 °C and are composed of organic cations and inorganic or organic anions [13,14]. Ionic liquids are also known for their non-volatile nature, high ionic conductivity, and high thermal stability [15,16]. Their exceptional properties, such as adjustable viscosity and conductivity, make them perfect for a broad range of applications, from green solvents [17,18] to components in advanced electrochemical systems [19]. These liquids facilitate good interactions when added to polymer matrices, considerably enhancing the electrical conductivity of the composites [20,21].

This addition not only increases conductivity but also offers toughness, thermal stability [22], and the capability to operate over a wide temperature range and conditions [23]. Such characteristics are specifically beneficial in high-performance applications like batteries, supercapacitors, actuators, membranes, etc. [24]. The changeability of ionic liquids provides for the adjustment of properties by modifying their chemical composition [25], thus allowing tailored solutions for specialized applications. The addition of ionic liquids into polymer composites unfolds new opportunities in the advancement of revolutionary materials that are both versatile and effective. Until now, there has been a lack of literature that particularly reviewed polymer matrices, conductive fillers, ionic liquids in such composites, and their electrical conductivity. This brief review aims to fill this gap by investigating the diverse types of polymer matrices used in composites containing ionic liquids, focusing on their regular usage. It also explores the various conductive fillers employed in these composites, detailing their common types. Furthermore, it highlights the different types of ionic liquids utilized in polymer composites, describing their unique selection. In addition, this review emphasizes how ionic liquids affect the electrical conductivity of polymer composites, underscoring their roles in the composites, as well as the significant importance of optimal conductive filler and ionic liquid contents.

2. Polymer Matrices in Composites Containing Ionic Liquids

Table 1 shows polymer matrices used in electrical conductivity studies of composites containing ionic liquids. It reveals that three main types of polymer matrices are employed: thermoplastics, thermosets, and elastomers. Thermoplastics are most frequently used, followed by elastomers, and thermosets are utilized with moderate regularity. Among thermoplastics, polyvinylidene fluoride (PVDF) is prominent due to its exceptional properties, such as high dielectric constant, chemical resistance, thermal stability, and notably its piezoelectric characteristics [1]. These properties make PVDF an excellent choice for high-performance applications requiring good environmental stability and favorable mechanical properties, which are essential in energy storage devices, sensors, actuators, and other electro-electronic devices [26,27]. Its ability to form stable composites that can integrate conductive fillers makes it a preferred polymer matrix for enhancing electrical conductivity [28]. Polymethyl methacrylate (PMMA) is another significant polymer used in conductivity studies. It stands out for its excellent optical clarity, outstanding mechanical performance, good weather resistance, and availability at a relatively low cost [29]. PMMA's ease of processing and adaptability to diverse manufacturing techniques, combined with its ability to be used in lightweight and durable composite forms, allows it to support a wide range of applications, including optical devices where conductivity is tailored through the inclusion of conductive nanofillers [30,31].

Another notable thermoplastic is polyaniline (PANI), which is unique due to its conductive properties. PANI is intrinsically conductive and can be processed in various ways to enhance its electrical properties. The inherent conductivity of PANI, coupled with its physicochemical stability and tunable electrical characteristics through doping [15], makes it an invaluable component in polymer composites for electronic applications, such as in anticorrosive coatings [15,19]. In the elastomers category, polychloroprene (CR) is used for its excellent chemical and heat resistance and low flammability [32]. These attributes are critical in applications requiring durable, flexible materials that maintain conductivity under stress and varying environmental conditions. CR's versatility in compounding with conductive fillers makes it suitable for use in flexible electronic circuits and robotic parts [7], where electrical conductivity is required alongside elasticity and durability. Styrene butadiene rubber (SBR) is widely used due to its good physical properties, cost-effectiveness, and easy processing. It offers good abrasion resistance and aging stability, which are beneficial in automotive applications such as tires, belts, hoses, seals, and rolls [20]. Figure 1 shows the chemical structures of PVDF, PMMA, PANI, CR, and SBR. Generally, each of these polymers has been chosen for specific traits that enhance the performance of composites in electrical conductivity applications.

Table 1. Polymer matrices used in electrical conductivity studies of composites containing ionic liquids.

Polymer Matrix	Abbreviation	References
Thermoplastic		
Cellulose acetate	CA	[33]
Polyaniline	PANI	[15,19,34]
Polybutylene terephthalate	PBT	[35]
Polyetherimide	PEI	[36]
Polymethyl methacrylate	PMMA	[29–31,37]
Polystyrene	PS	[3]
Polyvinyl alcohol	PVA	[38–40]
Polyvinylidene fluoride	PVDF	[1,23,24,26–28,41,42]
Thermoset		
Diglycidyl ester of aliphatic cyclo	DGEAC	[11]
Diglycidyl ether of bisphenol A	DGEBA	[6,21]
Phenolic formaldehyde	PF	[5,9]
Polyurethane	PU	[4,43]
Elastomer		
Ethylene acrylic elastomer	AEM	[8]
Natural rubber	NR	[44]
Polychloroprene	CR	[7,45–48]
Polydimethylsiloxane	PDMS	[10]
Poly(vinylidene fluoride-co-hexafluoropropylene)	FKM	[49]
Styrene butadiene rubber	SBR	[20,50–53]

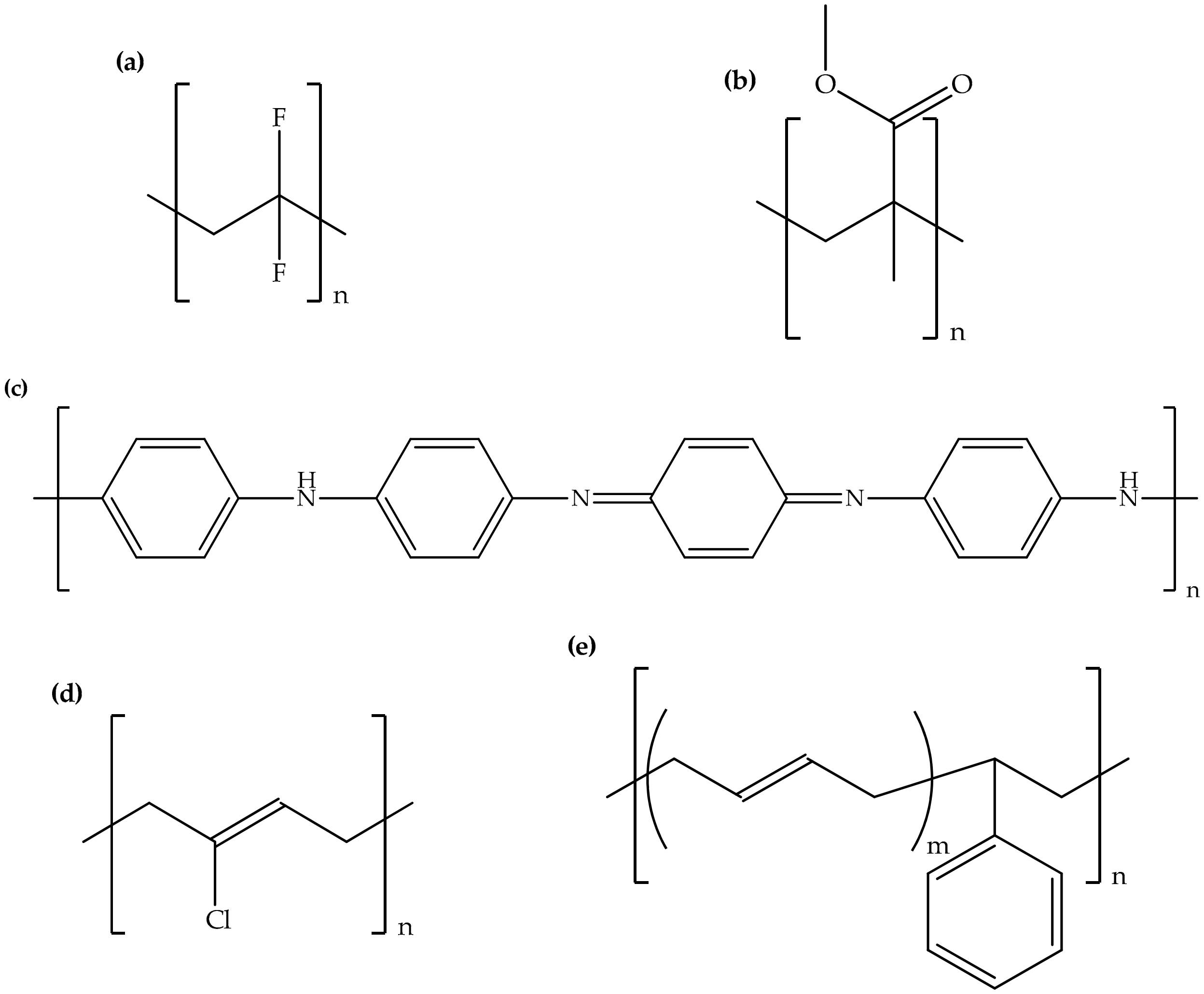


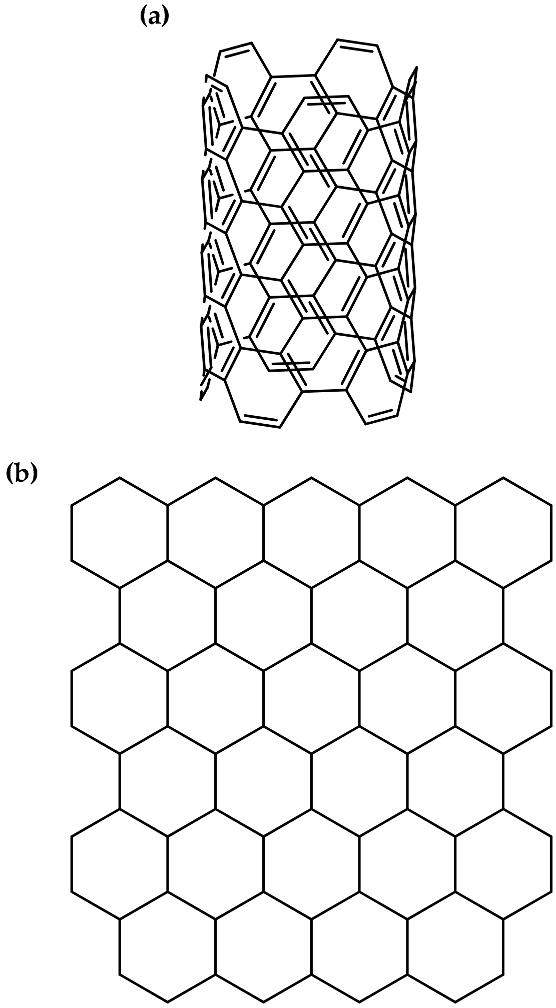
Figure 1. Chemical structures of (**a**) PVDF, (**b**) PMMA, (**c**) PANI, (**d**) CR, and (**e**) SBR.

3. Conductive Fillers in Polymer Composites Containing Ionic Liquids

Table 2 presents the conductive fillers employed in electrical conductivity studies of polymer composites containing ionic liquids. It demonstrates that various types of carbon-based materials are usually used. Multiwalled carbon nanotubes (MWCNTs) are the most prevalent conductive fillers in the field of conductive composites due to their exceptional electrical conductivity and mechanical strength. MWCNTs offer an excellent aspect ratio and network-forming capability [50,52], which enables efficient electron transport pathways within the polymer matrix. These characteristics make MWCNTs ideal for applications requiring robust conductive materials, such as electrostatic discharge dissipater, electromagnetic interference shielding materials, etc. [3]. Graphene (Gra), a single layer of carbon atoms arranged in a hexagonal lattice, follows closely due to its high surface area and outstanding thermal and electrical properties [31]. Its two-dimensional structure provides a high conductivity level, making Gra a potential choice for enhancing the performance of polymer composites in electronic communication and stealth technology [29]. Graphene oxide (GO), an oxidized form of Gra, is frequently utilized for its functional groups and its better interaction with polymers, which facilitate dispersion in polymer matrices [38]. GO is particularly valuable in applications where processability and compatibility with the polymer matrix are critical, such as in flexible electronics and smart electronic applications [33]. Figure 2 shows the chemical structures of MWCNTs, Gra, and GO. Reduced graphene oxide (rGO), derived from the reduction of GO, retains most of Gra's superior properties while being more processable [49]. rGO is used to improve the electrical conductivity of composites further, making it suitable for diverse applications and multifunctional materials [39,49]. Other conductive fillers, such as carbon black, carboxylated MWCNTs, exfoliated graphite, etc., also contribute to conductivity enhancement. These materials are typically selected for their ability to impart electrical properties effectively while being economically viable. In general, these fillers each provide unique development of polymer composites, tailoring and optimizing their conductive properties to meet specific technological demands, thus expanding the capabilities and applications of these advanced materials.

Table 2. Conductive fillers employed in electrical conductivity studies of polymer composites containing ionic liquids.

Conductive Filler	Abbreviation	References
Carbon black	CB	[28]
Carboxylated multiwalled carbon nanotubes	MWCNT-COOH	[35]
Exfoliated graphite	xGnP	[4]
Graphene	Gra	[9,20,23,29,31,42]
Graphene oxide	GO	[11,24,33,38,43]
Multiwalled carbon nanotubes	MWCNTs	[1,3,6–8,15,19,21,26,27,30,36,37,40,41,44–48,50–53]
Reduced graphene oxide	rGO	[34,39,49]
Silanized graphene	Gra-Si	[5]
Single-walled carbon nanotubes	SWCNTs	[10]



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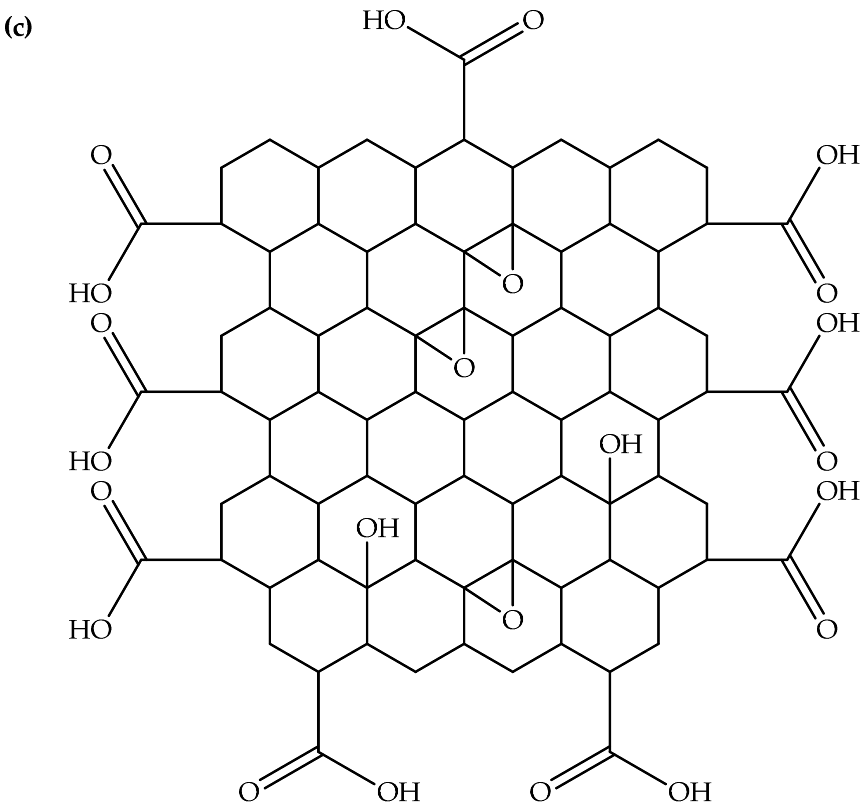


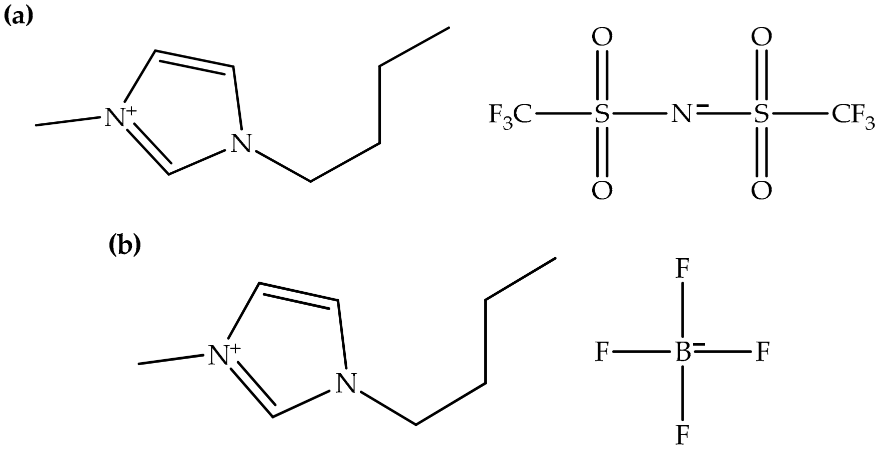
Figure 2. Chemical structures of (**a**) MWCNTs, (**b**) Gra, and (**c**) GO.

4. Ionic Liquids in Polymer Composites

Table 3 exhibits ionic liquids utilized in electrical conductivity studies of polymer composites. It exposes that imidazolium-based ionic liquids are mainly applied compared to other types of ionic liquids. 1-Butyl-3-methylimidazolium bis(trifluoromethylsulphonyl)imide ([C_4mim][NTf_2]) stands out as a preferred ionic liquid due to its exceptional thermal stability [29] and good plasticizing nature [51]. This ionic liquid is particularly valued for its ability to plasticize a wide range of polymers [10,44]. Its chemical structure allows for strong interactions with conductive fillers, facilitating better dispersion of fillers within the polymer matrix [46] and improving the electrical conductivity [7]. 1-Butyl-3-methylimidazolium tetrafluoroborate ([C_4mim][BF_4]) is another commonly used ionic liquid noted for its capability to act as a dispersing agent for the conductive fillers in polymer matrices [15,21]. The tetrafluoroborate anion contains fluorine, which induces compatibility with the fluorine-containing polymer matrix [42]. 1-Butyl-3-methylimidazolium hexafluorophosphate ([C_4mim][PF_6]) is selected for its ability to modify conductive fillers [30]. It enhances the electrical conductivity of polymer composites by creating a conductive filler network that provides an excellent electrical conductivity path [37]. Other types of ionic liquids, such as phosphonium-, pyridinium-, and ammonium-based ionic liquids, are increasingly being explored for their potential to enhance the electrical conductivity and performance of polymer composites. The phosphonium-based ionic liquid, such as trihexyl(tetradecyl)phosphonium bistriflimide ([$P_{6,6,6,14}$][NTf_2]), offers great non-covalent functionalization to conductive fillers, which promotes electrical conductivity within the composite structure [3]. The pyridinium-based ionic liquid, such as 1-butyl-4-methylpyridinium bromide ([C_4mpy][Br]), is also notable for its capability to interact with polymer matrix and conductive fillers, which facilitates improved polymer–filler interactions in composites [20]. Figure 3 shows the chemical structures of [C_4mim][NTf_2], [C_4mim][BF_4], [C_4mim][PF_6], [$P_{6,6,6,14}$][NTf_2], and [C_4mpy][Br]. These ionic liquids are mostly chosen based on their capacity to meet specific performance criteria in polymer composites, significantly impacting the materials' electrical conductivity and stability. Each brings unique benefits to the composites, making them indispensable in developing advanced conductive materials.

Table 3. Ionic liquids utilized in electrical conductivity studies of polymer composites.

Ionic Liquid	**Abbreviation**	**References**
Imidazolium-based		
1-Allyl-3-methylimidazolium chloride	[Amim][Cl]	[8,38]
1-(2-Aminoethyl)-3-methylimidazolium bromide	[Aemim][Br]	[24,41]
1-Benzyl-3-methylimidazolium chloride	[Bzmim][Cl]	[40,50]
1,6-Bis[3-(vinylbenzyl)imidazolium-1-yl]hexane chloride	[Bvbzimh][Cl]	[31]
1,10-Bis[1-vinylimidazolium-3-yl]decane bromide	[Bvimd][Br]	[5,9]
1-(3-Butoxy-2-hydroxypropyl)-3-methylimidazolium tetrafluoroborate	[Bhpmim][BF$_4$]	[11]
1-Butyl-3-methylimidazolium bis(trifluoromethylsulphonyl)imide	[C$_4$mim][NTf$_2$]	[7,10,44–48,51]
1-Butyl-3-methylimidazolium chloride	[C$_4$mim][Cl]	[33]
1-Butyl-3-methylimidazolium hexafluorophosphate	[C$_4$mim][PF$_6$]	[30,37,42]
1-Butyl-3-methylimidazolium tetrafluoroborate	[C$_4$mim][BF$_4$]	[15,19,21,36,42]
1-Decyl-3-methylimidazolium chloride	[C$_{10}$mim][Cl]	[52]
1-Ethyl-2,3-dimethylimidazolium bis(trifluoromethylsulphonyl)imide	[C$_2$dmim][NTf$_2$]	[53]
1-Ethyl-3-methylimidazolium bis(trifluoromethylsulphonyl)imide	[C$_2$mim][NTf$_2$]	[1]
1-Ethyl-3-methylimidazolium dicyanamide	[C$_2$mim][Dca]	[49]
1-Hexadecyl-3-methylimidazolium bromide	[C$_{16}$mim][Br]	[23]
1-Hexyl-3-methylimidazolium hexafluorophosphate	[C$_6$mim][PF$_6$]	[30]
1-Methylimidazolium chloride	[Cim][Cl]	[43]
1-Methyl-3-octylimidazolium chloride	[Coim][Cl]	[8]
1-Oxiranylmethyl-3-methylimidazolium tetrafluoroborate	[Oxmmim][BF$_4$]	[35]
1-(3-Sulfonato)-propyl-3-methylimidazolium hydrogen sulfate	[Supmim][HSO$_4$]	[34]
1-Vinyl-3-ethylimidazolium tetrafluoroborate	[Veim][BF$_4$]	[26,28]
Phosphonium-based		
Tributyl(ethyl)phosphonium diethyl phosphate	[P$_{4,4,4,2}$][Dep]	[27]
Trihexyl(tetradecyl)phosphonium bistriflimide	[P$_{6,6,6,14}$][NTf$_2$]	[3,27]
Trihexyl(tetradecyl)phosphonium bis(2,4,4-trimethylpentyl)phosphinate	[P$_{6,6,6,14}$][Tmpp]	[6]
Trihexyl(tetradecyl)phosphonium dicyanamide	[P$_{6,6,6,14}$][Dca]	[4]
Others		
1-Butyl-4-methylpyridinium bromide	[C$_4$mpy][Br]	[20]
1-Butylpyridinium bromide	[C$_4$py][Br]	[20]
N-Dodecyl-4-vinylpyridinium bis(trifluoromethylsulphonyl)imide	[C$_{12}$vpy][NTf$_2$]	[29]
Methacryloxyethyltrimethylammonium chloride	[Moetmam][Cl]	[39]



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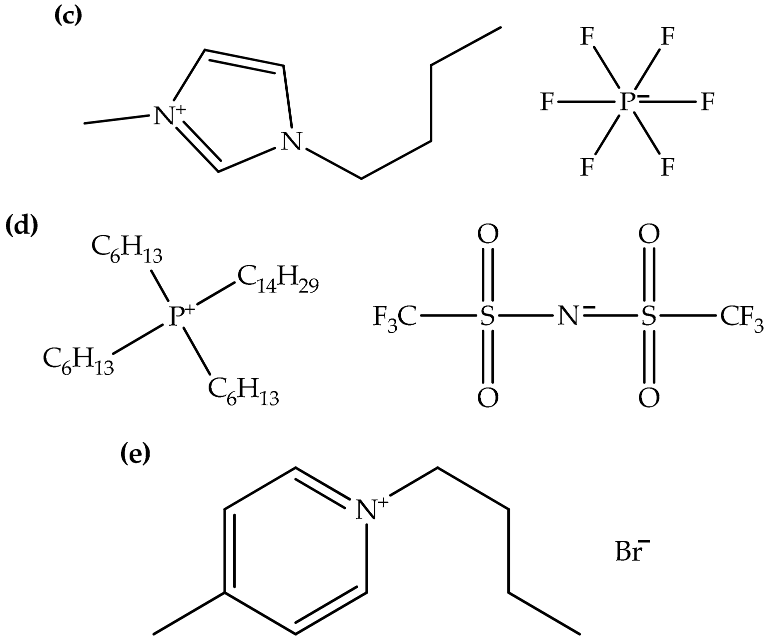

Figure 3. Chemical structures of (**a**) [C_4mim][NTf_2], (**b**) [C_4mim][BF_4], (**c**) [C_4mim][PF_6], (**d**) [$P_{6,6,6,14}$][NTf_2], and (**e**) [C_4mpy][Br].

5. Electrical Conductivity of Polymer Composites Containing Ionic Liquids

5.1. Electrical Conductivity of Thermoplastic Composites Containing Ionic Liquids

Table 4 shows the electrical conductivities of thermoplastic composites with optimal contents of conductive fillers and ionic liquids. Calheiros Souto et al. studied the electrical conductivity of PANI/MWCNTs/[C_4mim][BF_4] composites prepared via inverted emulsion polymerization [19]. They discovered that the electrical conductivity of the composite containing 12 wt.% MWCNTs and 5.0 wt.% [C_4mim][BF_4] was 4.5 S/cm, which is slightly higher compared to the composite without [C_4mim][BF_4] at 4.1 S/cm. This enhancement arises from the interactions between [C_4mim][BF_4] and the growing PANI chains, which improve chain orientation and subsequently increase conductivity [19].

Table 4. Thermoplastics, conductive fillers, ionic liquids, optimal contents, preparation methods, and electrical conductivities of thermoplastic composites.

Thermoplastic	Conductive Filler	Content (wt.%)	Ionic Liquid	Content (wt.%)	Preparation Method	Electrical Conductivity (S/cm)	References
PANI	MWCNTs	12	[C_4mim][BF_4]	5.0	IEP	4.5	[19]
PBT	MWCNT-COOH	0.7	[Oxmmim][BF_4]	2.0	MB	1×10^{-6}	[35]
PEI	MWCNTs	0.5	[C_4mim][BF_4]	1.0	ME	1.06×10^{-5}	[36]
PMMA	Gra	1.8	[C_{12}vpy][NTf_2]	8.9	SM	2×10^{-6}	[29]
PMMA	MWCNTs	2.0	[C_4mim][PF_6]	10	MB	1×10^{-2}	[37]
PS	MWCNTs	0.66	[$P_{6,6,6,14}$][NTf_2]	3.34	MB	1×10^{-3}	[3]
PVA	GO	2.89	[Amim][Cl]	0.67	SM	9.28×10^{-3}	[38]
PVDF	GO	3.0 *	[Aemim][Br]	-	SM	8.9×10^{-3}	[24]
PVDF	MWCNTs	2.0 *	[C_2mim][NTf_2]	-	MB	1×10^{-3}	[1]

* Ionic liquid-modified filler, IEP = inverted emulsion polymerization, MB = melt blending, ME = melt extrusion, and SM = solution mixing.

Lv et al. studied the electrical conductivity of PBT/MWCNT-COOH/[Oxmmim][BF_4] composites prepared through melt blending [35]. They found that the electrical conductivity of the composite containing 0.7 wt.% MWCNT-COOH and 2.0 wt.% [Oxmmim][BF_4] was 1×10^{-6} S/cm, which is significantly higher compared to the composite without [Oxmmim][BF_4] at 2.24×10^{-16} S/cm. This improvement stems from the formation of an ideal three-dimensional conducting network induced by [Oxmmim][BF_4] [35], which facilitates efficient electron mobility throughout the composite structure.

Ke et al. studied the electrical conductivity of PEI/MWCNTs/[C_4mim][BF_4] composites prepared via melt extrusion [36]. They discovered that the electrical conductivity of the composite containing 0.5 wt.% MWCNTs and 1.0 wt.% [C_4mim][BF_4] was 1.06×10^{-5} S/cm, which is significantly higher compared to the sample without MWCNTs/[C_4mim][BF_4] at 1.49×10^{-18} S/cm. This enhancement arises from the synergistic effect of MWCNTs and [C_4mim][BF_4], with [C_4mim][BF_4] also facilitating the dispersion of MWCNTs [36].

Caldas et al. studied the electrical conductivity of PMMA/Gra/[C_{12}vpy][NTf_2] composites prepared through solution mixing [29]. They found that the electrical conductivity of the composite containing 1.8 wt.% Gra and 8.9 wt.% [C_{12}vpy][NTf_2] was 2×10^{-6} S/cm, which is higher compared to the composite without [C_{12}vpy][NTf_2] at 2×10^{-9} S/cm. This improvement results from the role of [C_{12}vpy][NTf_2] in enhancing the dispersion of Gra and facilitating the formation of conductive pathways within the PMMA matrix [29].

Zhao et al. studied the electrical conductivity of PMMA/MWCNTs/[C_4mim][PF_6] composites prepared via melt blending [37]. They discovered that the electrical conductivity of the composite containing 2.0 wt.% MWCNTs and 10 wt.% [C_4mim][PF_6] was 1×10^{-2} S/cm, which is significantly higher compared to the composite without [C_4mim][PF_6] at 1.2×10^{-12} S/cm. This enhancement is due to [C_4mim][PF_6] improving the dispersion of the nanotubes, thereby enabling the individually dispersed MWCNTs to form an interconnected network structure readily [37].

Soares da Silva et al. studied the electrical conductivity of PS/MWCNTs/[$P_{6,6,6,14}$][NTf_2] composites prepared through melt blending [3]. They found that the electrical conductivity of the composite containing 0.66 wt.% MWCNTs and 3.34 wt.% [$P_{6,6,6,14}$][NTf_2] was 1×10^{-3} S/cm, which is higher compared to the composite without [$P_{6,6,6,14}$][NTf_2] at 1×10^{-7} S/cm. This improvement stems from [$P_{6,6,6,14}$][NTf_2] enhancing the dispersion of MWCNTs, which facilitates the formation of conductive pathways with a smaller quantity of the conductive filler [3].

Sahu et al. studied the electrical conductivity of PVA/GO/[Amim][Cl] composites prepared via solution mixing [38]. They discovered that the electrical conductivity of the composite containing 2.89 wt.% GO and 0.67 wt.% [Amim][Cl] was 9.28×10^{-3} S/cm, which is slightly higher compared to the composite without [Amim][Cl] at 7.38×10^{-3} S/cm. This enhancement originates from the formation of extensive interconnected three-dimensional conductive networks assisted by [Amim][Cl] [38], which facilitate improved electron mobility and boost the electrical conductivity of the composite.

Maity et al. studied the electrical conductivity of PVDF/GO/[Aemim][Br] composites prepared through solution mixing [24]. They found that the electrical conductivity of the composite containing 3.0 wt.% [Aemim][Br]-modified GO was 8.9×10^{-3} S/cm, which is significantly higher compared to the sample without [Aemim][Br]-GO at 4.6×10^{-13} S/cm. This improvement is due to the interfacial interaction between [Aemim][Br]-modified GO and the PVDF matrix, as shown in Figure 4, and the appropriate content of modified GO in the matrix, which facilitates the formation of a three-dimensional conductive network [24].

Sharma et al. studied the electrical conductivity of PVDF/MWCNTs/[C_2mim][NTf_2] composites prepared via melt blending [1]. They discovered that the electrical conductivity of the composite containing 2.0 wt.% [C_2mim][NTf_2]-modified MWCNTs was 1×10^{-3} S/cm, which is slightly higher compared to the composite without [C_2mim][NTf_2] at 1×10^{-4} S/cm. This enhancement results from [C_2mim][NTf_2] non-covalently wrapping MWCNTs, improving the dispersion of the nanotubes and facilitating the formation of an effective network within the PVDF matrix [1].

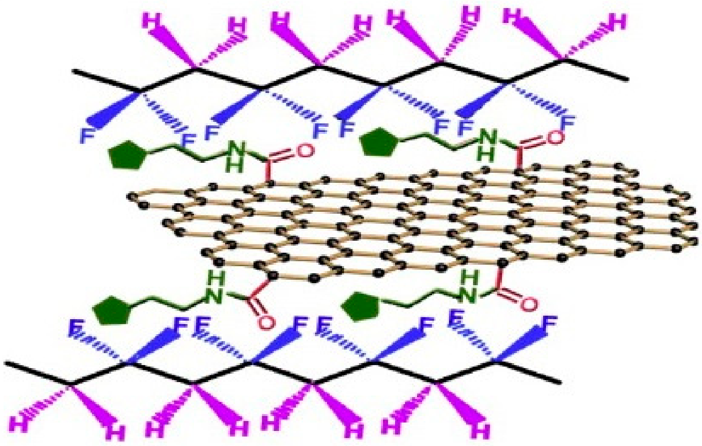

Figure 4. Schematic presentation showing the interaction between [Aemim][Br]-modified GO and the PVDF matrix. Reprinted from [24], with permission from Elsevier.

5.2. Electrical Conductivity of Thermoset Composites Containing Ionic Liquids

Table 5 shows the electrical conductivities of thermoset composites with optimal contents of conductive fillers and ionic liquids. Lopes Pereira and Soares studied the electrical conductivity of DGEBA/MWCNTs/[C_4mim][BF_4] composites prepared through solution mixing [21]. They found that the electrical conductivity of the composite containing 0.5 wt.% [C_4mim][BF_4]-modified MWCNTs was 1×10^{-3} S/cm, which is higher compared to the composite without [C_4mim][BF_4] at 1×10^{-5} S/cm. This improvement arises from enhanced interactions between [C_4mim][BF_4] and MWCNTs, leading to improved dispersion of the filler and a subsequent increase in electrical conductivity [21].

Table 5. Thermosets, conductive fillers, ionic liquids, optimal contents, preparation methods, and electrical conductivities of thermoset composites.

Thermoset	Conductive Filler	Content (wt.%/phr)	Ionic Liquid	Content (wt.%/phr)	Preparation Method	Electrical Conductivity (S/cm)	References
DGEBA	MWCNTs	0.5 *	[C_4mim][BF_4]	-	SM	1×10^{-3}	[21]
DGEBA	MWCNTs	1.0	[$P_{6,6,6,14}$][Tmpp]	10	SM	6×10^{-4}	[6]
PF	Gra	15	[Bvimd][Br]	5.0	HB	9.2×10^{-4}	[9]
PF	Gra-Si	15	[Bvimd][Br]	5.0	HB	1.45×10^{-3}	[5]
PU	xGnP	7.0 *	[$P_{6,6,6,14}$][Dca]	-	SB	2.47×10^{-2}	[4]

* Ionic liquid-modified filler, SM = solution mixing, HB = hot blending, and SB = solution blending.

Soares et al. studied the electrical conductivity of DGEBA/MWCNTs/[$P_{6,6,6,14}$][Tmpp] composites prepared via solution mixing [6]. They discovered that the electrical conductivity of the composite containing 1.0 phr MWCNTs and 10 phr [$P_{6,6,6,14}$][Tmpp] was 6×10^{-4} S/cm, which is significantly higher compared to the sample without MWCNTs/[$P_{6,6,6,14}$][Tmpp] at 1×10^{-13} S/cm. This enhancement stems from effective interactions between [$P_{6,6,6,14}$][Tmpp] and MWCNTs, facilitated by the non-covalent functionalization of MWCNTs, leading to better electrical conductivity [6].

Wang et al. studied the electrical conductivity of PF/Gra/[Bvimd][Br] composites prepared through hot blending [9]. They found that the electrical conductivity of the composite containing 15 wt.% Gra and 5.0 wt.% [Bvimd][Br] was 9.2×10^{-4} S/cm, which is higher compared to the composite without [Bvimd][Br] at 5.6×10^{-5} S/cm. This improvement originates from [Bvimd][Br] acting as a bridge for electron transfer between Gra layers, thereby facilitating the formation of a conductive network in the PF matrix [9].

Li et al. studied the electrical conductivity of PF/Gra-Si/[Bvimd][Br] composites prepared via hot blending [5]. They discovered that the electrical conductivity of the composite containing 15 wt.% Gra-Si and 5.0 wt.% [Bvimd][Br] was 1.45 × 10^{-3} S/cm, which is higher compared to the composite without [Bvimd][Br] at 4.14 × 10^{-5} S/cm. This enhancement is due to the imidazolium ring in [Bvimd][Br] improving the dispersion of Gra-Si within the PF matrix, which leads to the formation of more electrically conductive pathways [5].

Vargas et al. studied the electrical conductivity of PU/xGnP/[$P_{6,6,6,14}$][Dca] composites prepared through solution blending [4]. They found that the electrical conductivity of the composite containing 7.0 wt.% [$P_{6,6,6,14}$][Dca]-modified xGnP was 2.47 × 10^{-2} S/cm, which is significantly higher compared to the composite without [$P_{6,6,6,14}$][Dca] at 3.68 × 10^{-7} S/cm. This improvement results from the effective interaction of the dicyanamide anion in [$P_{6,6,6,14}$][Dca] with the PU matrix, promoting even xGnP dispersion and the development of a conductive network within the polymer [4].

5.3. Electrical Conductivity of Elastomer Composites Containing Ionic Liquids

Table 6 shows the electrical conductivities of elastomer composites with optimal contents of conductive fillers and ionic liquids. Subramaniam et al. studied the electrical conductivity of CR/MWCNTs/[C_4mim][NTf_2] composites prepared via mechanical mixing [45]. They discovered that the electrical conductivity of the composite containing 3.0 phr [C_4mim][NTf_2]-modified MWCNTs was 1 × 10^{-3} S/cm, which is higher compared to the composite without [C_4mim][NTf_2] at 1 × 10^{-6} S/cm. This enhancement stems from a combination of effective dispersion and improved networking between the nanotubes of [C_4mim][NTf_2]-MWCNTs, which together facilitate superior electrical connectivity throughout the composite [45].

Table 6. Elastomers, conductive fillers, ionic liquids, optimal contents, preparation methods, and electrical conductivities of elastomer composites.

Elastomer	**Conductive Filler**	**Content (phr/wt.%)**	**Ionic Liquid**	**Content (wt.%)**	**Preparation Method**	**Electrical Conductivity (S/cm)**	**References**
CR	MWCNTs	3.0 *	[C_4mim][NTf_2]	-	MM	1 × 10^{-3}	[45]
PDMS	SWCNTs	0.1	[C_4mim][NTf_2]	2.0	SM	4 × 10^{-5}	[10]
SBR	MWCNTs	5.0 *	[Bzmim][Cl]	-	MM	1 × 10^{-1}	[50]
SBR	MWCNTs	10 *	[C_4mim][NTf_2]	-	MM	1 × 10^{-2}	[51]

* Ionic liquid-modified filler, MM = mechanical mixing, and SM = solution mixing.

Zhao and Ounaies studied the electrical conductivity of PDMS/SWCNTs/[C_4mim][NTf_2] composites prepared through solution mixing [10]. They found that the electrical conductivity of the composite containing 0.1 wt.% SWCNTs and 2.0 wt.% [C_4mim][NTf_2] was 4 × 10^{-5} S/cm, which is significantly higher compared to the sample without SWCNTs/[C_4mim][NTf_2] at 1 × 10^{-15} S/cm. This improvement originates from enhanced dispersion of SWCNTs in the PDMS matrix and interactions between [C_4mim][NTf_2] and SWCNTs, which prevent SWCNTs aggregation and create an interconnected network structure [10].

Abraham et al. studied the electrical conductivity of SBR/MWCNTs/[Bzmim][Cl] composites prepared via mechanical mixing [50]. They discovered that the electrical conductivity of the composite containing 5.0 wt.% [Bzmim][Cl]-modified MWCNTs was 1 × 10^{-1} S/cm, which is significantly higher compared to the sample without [Bzmim][Cl]-MWCNTs at 1 × 10^{-7} S/cm. This enhancement is due to [Bzmim][Cl] interacting with SBR and MWCNTs through aromatic π–π and cation–π interactions, respectively (as shown in Figure 5), which improves the dispersion of MWCNTs and facilitates more efficient transport of electrons via three-dimensional continuous MWCNT networks [50].

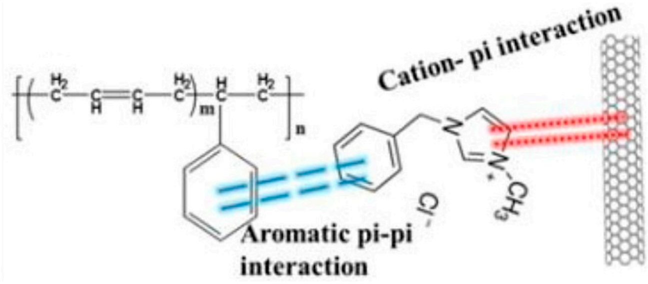

Figure 5. Schematic diagram showing the interactions between [Bzmim][Cl], SBR, and MWCNTs. Reprinted from [50], with permission from Elsevier.

Subramaniam et al. studied the electrical conductivity of SBR/MWCNTs/[C_4mim][NTf_2] composites prepared through mechanical mixing [51]. They found that the electrical conductivity of the composite containing 10 phr [C_4mim][NTf_2]-modified MWCNTs was 1×10^{-2} S/cm, which is significantly higher compared to the composite without [C_4mim][NTf_2] at 1×10^{-10} S/cm. This improvement results from [C_4mim][NTf_2] facilitating the creation of additional conductive pathways for electron transport due to enhanced networking between the fillers [51].

6. Conclusions

In this review, polymer matrices, conductive fillers, and ionic liquids utilized in polymer composites are briefly classified. In addition, the electrical conductivity of polymer composites containing ionic liquids is compared in this brief review. Polymer matrices such as thermoplastics (specifically PVDF, PMMA, and PANI), thermosets (explicitly DGEBA and PF), and elastomers (particularly CR and SBR) are predominantly used in these composites. Conductive fillers like carbon-based materials, especially MWCNTs, Gra, and GO, are usually employed in polymer composites. Imidazolium-based ionic liquids, notably [C_4mim][NTf_2], [C_4mim][BF_4], and [C_4mim][PF_6] are frequently applied in such composites. Most studies indicate that the addition of ionic liquids improves the electrical conductivity of polymer composites, irrespective of the type of polymer matrix and conductive filler used. This improvement is attributed to the interaction of the ionic liquids with the conductive fillers and polymer matrices. Ionic liquids not only enhance the dispersion of conductive fillers but also facilitate the formation of effective interconnected three-dimensional conductive pathways within the polymer matrix, thereby enhancing electron transport and mobility throughout the composite structure. This review offers novel insights into the consistent behavior of ionic liquids across diverse composite systems, providing a fresh perspective on their role in enhancing material properties for advanced technological applications. These insights pave the way for targeted innovations in the design and use of next-generation conductive materials. Furthermore, this review enriches the composite field's knowledge base and assists future research and development of more efficient conductive materials for electronic technologies.

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