

Review article

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Antistatic polymer materials

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ABSTRACT: Introduction. In connection with the growing demand for living and working conditions in civil and industrial construction, there is an increasing need for high-quality building materials with the required set of performance properties. Polymer reinforced composite materials are promising materials in the construction industry due to their high strength, durability, reliability and economy. Polymers such as polyvinyl chloride, polyurethanes, polyacrylates, epoxy resins, polypropylene are used in construction for the manufacture of decorative elements, self-leveling floors, coatings for appliances and equipment. However, the use of polymeric materials creates risks of electric shock due to the generation of static charge. The use of electrically conductive nanomaterials as fillers makes it possible to reduce the resistivity of polymeric materials and slow down the flow of electric charges. **Main part.** This review article presents the benefits and drawbacks of antistatic additives for polymer materials used in industry and in the construction industry. **Conclusion.** An analysis of the literature has shown that over the past seven years, the largest number of relevant papers has been devoted to carbon materials as antistatic additives (8 articles), metal and metal oxide nanoparticles (7 articles), ionic liquids (7 articles), and polyaniline (7 articles). The most studied characteristics of antistatic polymer materials are the specific surface R_s and volume R_v resistances. According to the reviewed articles, metal and metal oxide nanoparticles are the most suitable antistatic additives to polymeric materials, since they are well dispersed in the polymer matrix. However, further research is needed to eliminate the negative effect of nanoparticles on the mechanical properties of polymeric materials.

KEYWORDS: nanoparticles, static electricity, carbon nanotubes, graphene oxide, ionic liquids, construction industry.

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INTRODUCTION

Static electricity is a serious problem for industrial production [1], industrial and civil construction, hospital wards [2]. This is explained by the fact that static charges create a fire and explosion hazard [3, 4]. Static electricity is caused by accumulation of positive or negative charges on the surface of polymer coatings and paintwork materials, for example, an excess of electrons due to surface friction upon contact with dust [5, 6]. Instruments, equipment and packaging used at industrial plants are made of polymeric materials with high surface resistivity, which is about $10^{11}–10^{14} \Omega/\text{cm}^2$ [7]. To eliminate static electricity at industrial plants and in the household, it is necessary to develop antistatic coatings for measuring instruments and equipment, panels of internal and external walls of buildings, which, unlike polymers, have high electrical conductivity and allow safe discharge of excess charge into

the ground [8]. To remove the charge, the coating must have an electrical resistivity in the range of $10^3–10^{10} \Omega/\text{cm}^2$ [9]. With a lower resistivity, the risk of electric shock to the human body increases [10].

Conductive ionic liquids [11], metal [12] and metal oxide [13] nanoparticles, carbon nanotubes [14], graphene oxide nanosheets [15], conductive polymers (polyaniline, polythiophene, polypyrrole) [16], and surfactants [17] are used as additives to polymer coatings that endow them with antistatic properties.

Nanomaterials are widely used in the construction industry. Nanoparticles are added to paints and varnishes to make them antistatic and scratch resistant and to protect them against dust and fog. The adhesives used for the manufacture of antistatic floors based on nanomaterials have a high electrical conductivity [18]. Coatings containing nanomaterials reduce dust adhesion on photovoltaic panels that are placed on the roofs of eco-buildings [19].

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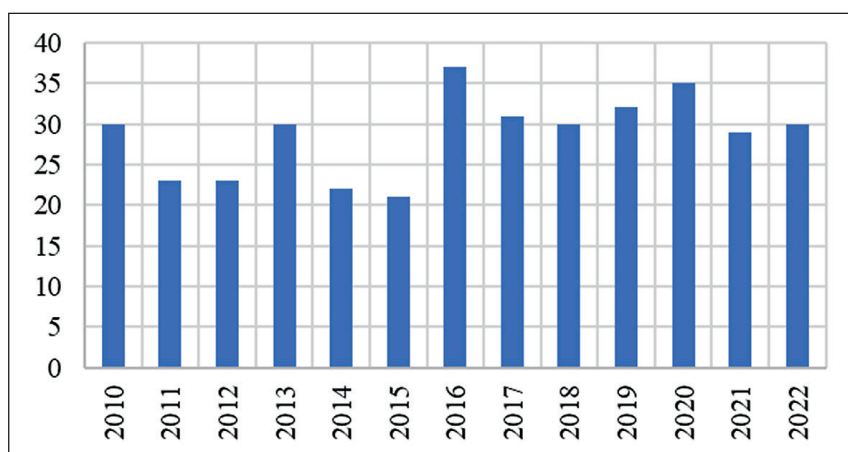


Fig. 1. Number of articles according to the Scopus database (search query: antistatic coatings)

The development of antistatic additives for polymer coatings is a promising area of research, as evidenced by a large number of articles published in the period of 2010–2022, according to the Scopus database (Figure 1). From the above data, it can be seen that the greatest interest in this topic was observed in 2016, while the smallest interest was expressed in 2015. In 2022, the number of publications has decreased with respect to 2016, but the interest in antistatic coatings is still high (30 articles per year).

CARBON-BASED ANTISTATIC ADDITIVES

Carbon-based conductive fillers include carbon black [20, 21], carbon fibers [1, 22], carbon nanotubes [13, 23–26], graphite [27], graphene [28, 29], and graphene oxide (GO) [30, 31].

Carbon nanotubes

Carbon nanotubes, particularly multi-walled carbon nanotubes (MWCNTs), are used most widely due to their low weight, large aspect ratio, good mechanical properties, chemical stability, and electrical conductivity comparable to that of metals [32].

The main disadvantage of using carbon nanotubes in antistatic polymer coatings is their low degree of dispersion in polymer matrices, especially in highly crystalline polymers, due to their entanglement and aggregation into bundles [33, 34]. This is caused mainly by high energy of van der Waals interactions and electrostatic forces in MWCNTs. To improve the dispersion of MWCNTs in the polymer, the nanotubes are treated with stabilizing compounds using covalent and non-covalent (or ultrasonic) modification [35, 36].

The non-covalent modification without the formation of chemical bonds between MWCNTs and the stabilizer is the method of choice, since the covalent modification may cause rupture into shorter segments [37]. For

example, Yan W. et al. used environmentally friendly and cheap tannic acid as a stabilizer. The authors attributed the stabilizing effect to the formation of non-covalent π - π bonds between the aromatic phenol rings of tannic acid and graphene structures of the nanotubes [38].

Liu L. et al. [39] used MWCNTs (Seoul, Korea, 10–20 nm in diameter, 20–100 μ m long) stabilized with environmentally friendly poly(tannin urethane) to produce an antistatic coating based on poly(vinyl chloride) (PVC).

When MWCNTs were added to the polymer, the surface resistance decreased from 10^{16} to $10^8 \Omega/\text{cm}^2$. Moreover, the content of the antistatic additive was low and amounted to 0.5 wt. %. Poly(tannin urethane) showed an excellent dispersing ability for MWCNTs. Considering the poly(tannin urethane) structure, the authors concluded that the aromatic rings of poly(tannin urethane) were attached to the MWCNT surface via π - π interactions, which promoted disaggregation [40].

The authors [39] compared the characteristics of the fabricated PVC coating with the coatings based on polyurethane-containing MWCNTs and found that the coating showed similar surface resistivity characteristics [39, 41].

Cyanuric acid and an ionomer based on the ethylene-co-acid-co-sodium acid copolymer were used to enhance the MWCNT dispersion in the polyoxymethylene matrix and to improve the electrical properties (electrical conductivity, surface resistance) of the polymer nanocomposite [42]. As a result, the surface resistance of the composites was reduced by an order of magnitude compared to pure polyoxymethylene (Figure 2). It was found that the most pronounced decrease in the surface resistance is achieved at an ionomer concentration of 3 wt. % (Figure 2 a) and cyanuric acid concentration of 0.5 wt. % (Figure 2 b). The authors explained the MWCNT stabilization by non-covalent π - π interaction with the external surface of the nanotube (wrapping of nanotubes) for cyanuric acid and cation- π interaction for the ionomer.

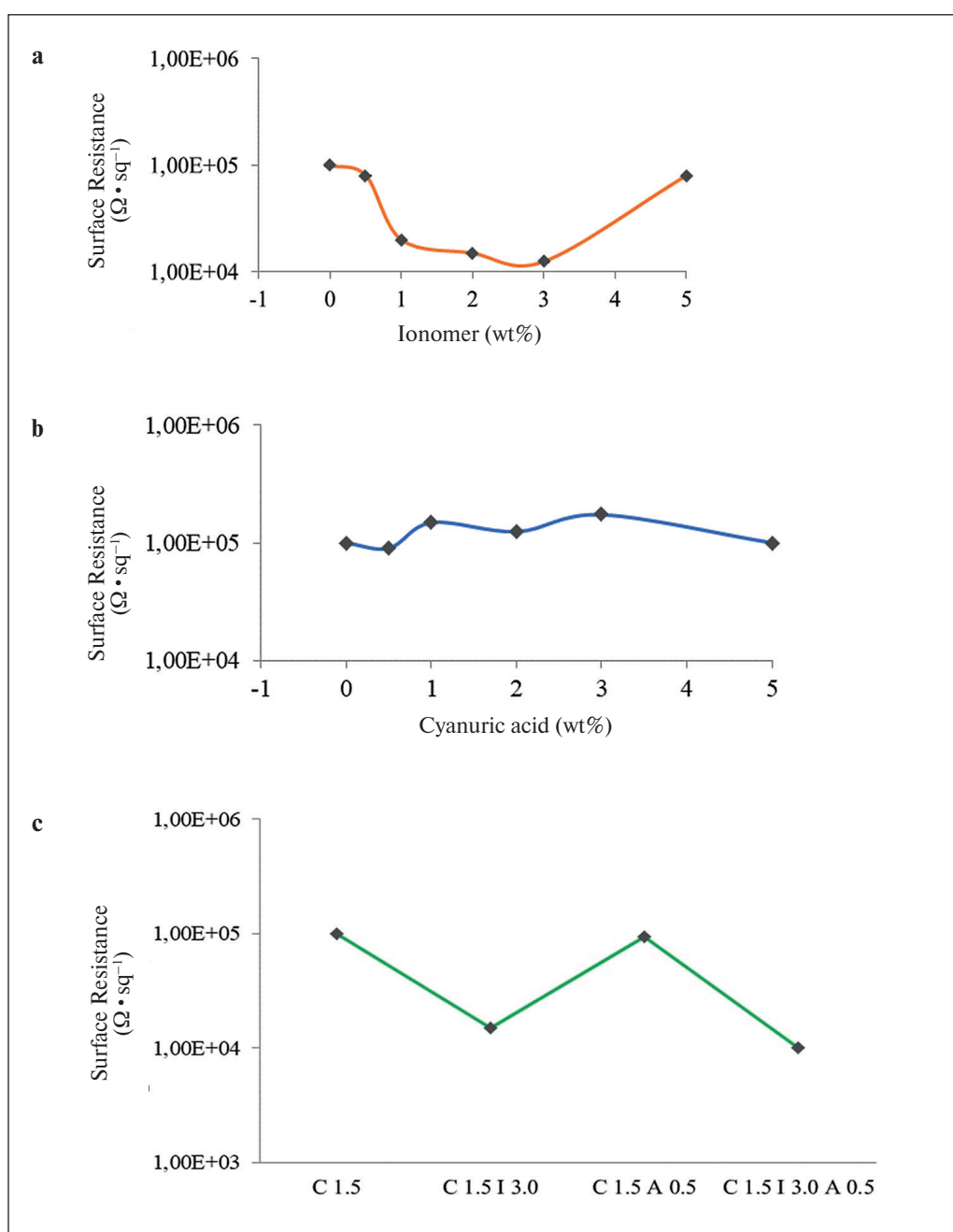


Fig. 2. Surface resistance of composites containing MWCNTs the surface of which is coated with ionomer (a), cyanuric acid (b), and ionomer and cyanuric acid (c) vs. concentration of the additive. C are nanotubes, I is ionomer, A is cyanuric acid [42]

However, the disadvantage of polymeric materials obtained in [39, 42] is weak adhesion, which leads to a short service life and environmental pollution, as well as to low tensile strength and impact strength.

Reduced graphene oxide as an antistatic additive

Reduced graphene oxide (rGO) nanosheets, like carbon nanotubes, have high mechanical strength, large specific

surface area, and high electrical conductivity, which makes them promising for use in polymer composites as antistatic agents [43–46]. However, the disadvantage of graphene oxide nanosheets, unlike nanoparticles of metals and metal oxides, is their poor dispersibility in the polymer matrix, which makes it difficult to create electron-conducting paths [47, 48]; one more drawback is low stability. To improve the dispersibility, graphene oxide is coated *in situ* with chemical modifiers, in particular surfactants [49, 50] or polymers

[51]. For example, polydopamine with catechol, amino, and imino functional groups covalently grafted to GO surface provides good dispersibility with polyurethane due to improved interfacial adhesion [52]. The poor dispersibility and tendency to aggregation and exfoliation of reduced graphene oxide nanosheets is due to van der Waals forces at the nanosheet interface [53].

The chemical modification of graphene oxide with dimethylformamide and carboxylic acids generated a positive charge on graphene sheets with a zeta potential of about 20 mV [54]. The addition of rGO to water-based polyurethane coatings made it possible to reduce the surface electrical resistance by five orders of magnitude. In the presence of 3 wt. % rGO, the surface electrical resistance was $5.4 \cdot 10^9 \Omega/\text{cm}^2$, which is sufficient for the manufacture of antistatic coatings. The authors attributed this fact to sp^2 hybridization in the rGO structure. The use of rGO nanohybrids with conductive polymers, in particular polyaniline, reduced the surface resistivity down to $10^6 \Omega/\text{cm}^2$ [54]. According to the authors [54], this was a result of the synergistic effect of rGO and polyaniline caused by the conjugated structure of polyaniline.

However, the authors [54] did not provide a detailed explanation of how the conducting paths are formed in the polyurethane matrix, which is a significant drawback of the study.

In another work [55], rGO nanosheets and copper nanoparticles were introduced into a polyurethane coating (the content of the additives was 3 wt. %). This resulted in a decrease in the surface resistivity of polyurethane from $10^{14} \Omega/\text{cm}^2$ to $10^9 \Omega/\text{cm}^2$. The authors suggested that the synergistic antistatic effect was due to the formation of conducting paths by copper nanoparticles, which acted as conductors between the rGO nanosheets (Figure 3).

Thus, small amounts of carbon-based additives reduce the surface resistivity of polymeric materials by several orders of magnitude. However, carbon-based fillers can deteriorate the mechanical properties and increase the water uptake of polymer coatings [54]. The main problem associated with the use of carbon materials in industry is the difficulty of their dispersion in polymers and the need to endow polymer fibers and textiles with hydrophilicity needed for their processing [56]. Carbon fillers deteriorate the film transparency of films, which requires the use of

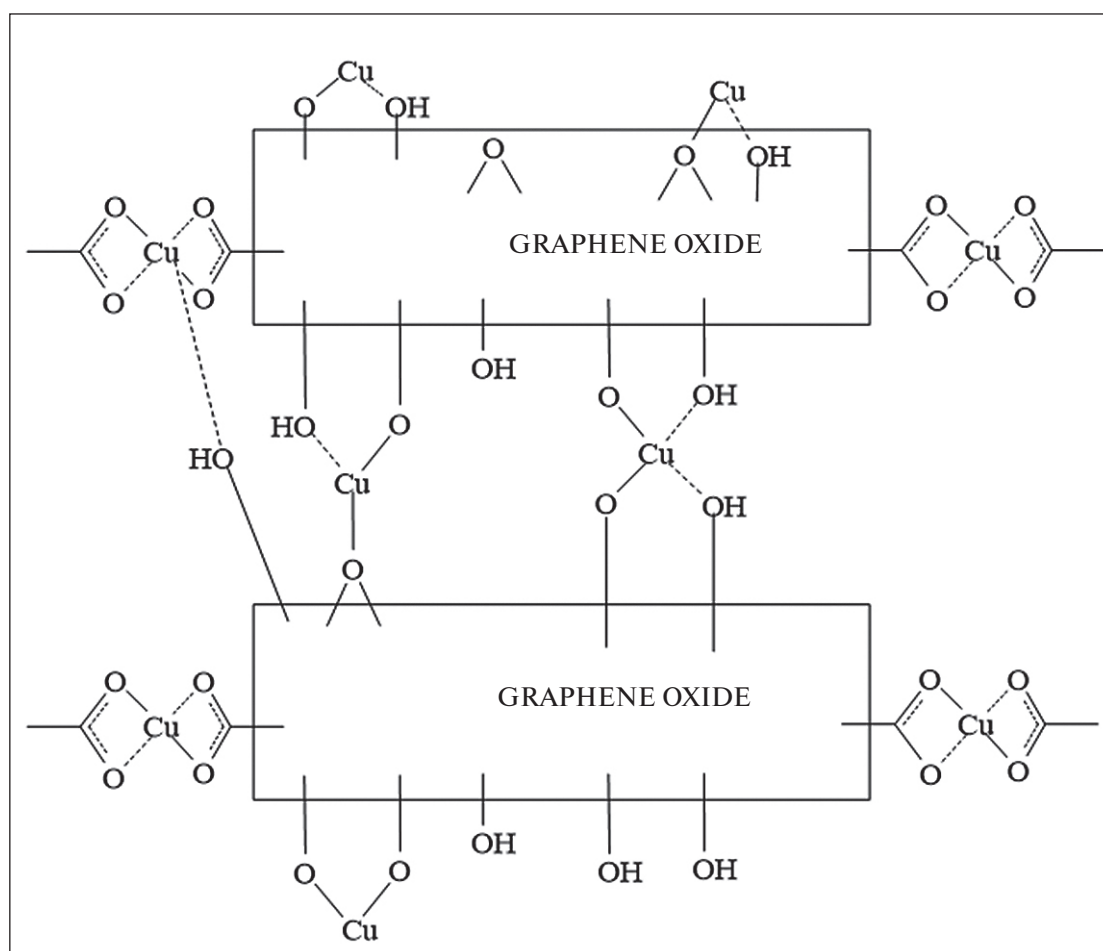


Fig. 3. Structure resulting from the reaction of rGO and copper(II) sulfate [55]

layer-by-layer assembly. For example, in order to maintain the film transparency [57], poly-L-lactide films were successively treated with two types of polyelectrolytes: positively charged chitosan or polyethyleneimine and negatively charged GO. This method allows deposition of multiple bilayers on the film.

Surfactants as antistatic additives

The surfactants applicable as antistatic additives include compounds containing hydrophilic groups and lipophilic hydrocarbon groups. This makes it easy to combine surfactants with epoxy resins [58], PVC [59], and textile fibers based on polyesters [60]. Cationic surfactants, that is, quaternary ammonium salts, are used most widely as antistatic agents, since they are readily soluble in epoxy resins and are easily adsorbed on negatively charged polyester fibers without damaging them [61]. For example, antistatic coatings based on epoxy resin (E-51), polyetheramine (hardener), and quaternary ammonium salts of poly(di-n-propylamine-co-epichlorohydrin) P(DPA-EPI) and poly(di-n-butylamine-co-epichlorohydrin) P(DBA-EDI). The coatings were deposited on a polypropylene substrate. The coatings retained their transparency when the surfactant was added. To achieve the antistatic effect, only 1 wt. % surfactant was required (Figure 4). As a result, the surface resistivity decreased by four orders of magnitude (from 10^{12} to $10^8 \Omega/\text{cm}^2$). According to the authors, this was due to the adsorption of air moisture by the surfactant and the formation of a conductive hygroscopic layer by the polar hydroxyl and quaternary ammonium groups of the surfactant.

In addition, the surface resistivity of the coatings with P(DBA-EDI) decreased to a higher extent than that of the coatings containing P(DPA-EPI). Thus, the length of the hydrocarbon chain at the nitrogen atom affects the antistatic properties of epoxy coatings.

However, a drawback of surfactants is their migration to the surface of the polymer material over time, which

leads to the loss of antistatic properties [62]. In addition, the hardness of epoxy coatings decreases upon the addition of surfactants, despite good adhesion [56]. Therefore, the interest in the use of surfactants in antistatic polymeric materials is declining.

Nanoparticles as antistatic additives

Nanoparticles of metals and metal oxides provide electrically conductive structures for polymer coatings [63]; unlike carbon particles, they are easily dispersed in the polymer matrix [64], generating a long-term antistatic effect; unlike surfactants, they do not migrate to the surface of the polymer material and are not leached [65, 66]; in addition, they improve the UV resistance of polymer fibers [64, 67–69].

Zhang, J. et al. [70] fabricated an antistatic coating based on polypropylene decorated with silver nanoparticles incorporated in the porous structure of coal gasification slag microbeads. Due to the small particle size, low density, and large specific surface area, silver nanoparticles were easily built into microbeads, and the filler was easily dispersed in the polypropylene matrix. Upon the addition of 10 wt. % of microbeads into the polypropylene matrix, the volume resistivity was $3.35 \cdot 10^3 \Omega \cdot \text{cm}$; this fits into the range required for antistatic properties. However, the filling deteriorated the mechanical properties of polypropylene. In particular, the mechanical strength of polypropylene decreased, which was explained by an increase in stress concentration at the polymer-filler interface. However, a shortcoming of this study is the lack of explanation why silver nanoparticles were used in microbeads rather than by themselves.

Nanoparticles of metal oxides, for example, aluminum(III) oxide, zinc(I) oxide, iron(III) oxide, and zirconium oxide (IV), are environmentally benign, chemically stable, and inexpensive antistatic additives for polymer coatings and fibers [63, 71, 72].

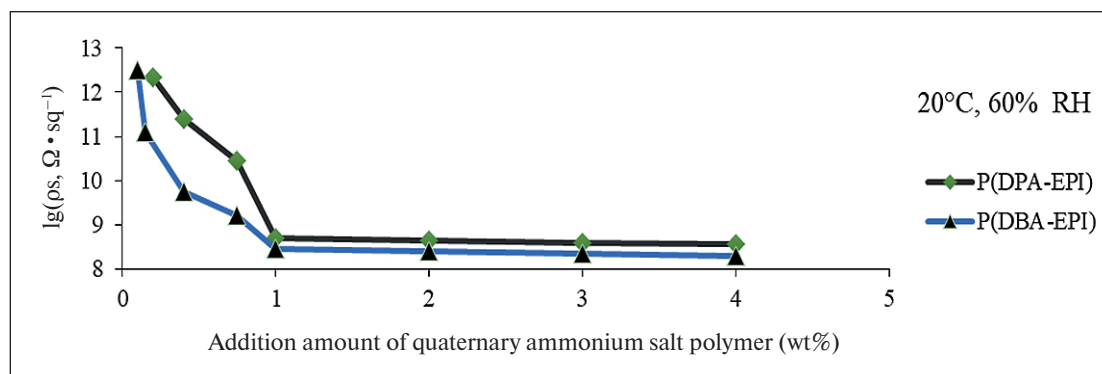


Fig. 4. Change in the surface resistivity of the epoxy coating with increasing surfactant content. RH is air humidity [58]

The introduction of aluminum oxide nanoparticles (particle size of 20–40 nm) into polypropylene was shown to increase the electrical conductivity of polypropylene [73]. The modified polypropylene was meant for the use as an antistatic coating for tools and instrumental equipment. It was reported that an increase in the nanoparticle concentration from 1 to 5 wt. % led to a decrease in the surface resistivity of the modified samples. When the concentration of nanoparticles in the composite was low (below 5 wt. %), the modification resulted in increasing tensile strength. However, when the concentration of nanoparticles in the polypropylene bulk was 30 wt. %, the mechanical and surface properties of polypropylene are deteriorated. Electron irradiation of modified polypropylene samples slightly increased the conductivity (on average, by 5–6%). The authors interpreted this fact by assuming that polypropylene chains break on exposure to radiation, resulting in an increase in the number of free radicals and unsaturated bonds and in the formation of conjugated double bonds.

The application of 6 wt. % zirconium dioxide nanoparticles with a size of 20–50 nm and a spherical shape in polyurethane coatings makes it possible to dissipate the electrostatic charges and reduce the surface resistivity to $10^9 \Omega/\text{cm}^2$. In addition, an advantage of zirconium dioxide nanoparticles over other nanoparticles is their ability to improve the adhesive strength of a polyurethane coating and form a rough structure on the coating surface, which prevents dust accumulation [74].

However, there are no data in the literature [63, 71–74] on the mechanism of the antistatic action of metal oxide nanoparticles.

Despite the fact that metal and metal oxide nanoparticles are better dispersed in the polymer matrix than carbon materials, higher concentrations of nanoparticles compared to the concentrations of carbon materials are required to attain the antistatic effect; this results in deterioration of mechanical properties. In addition, metal nanoparticles (copper, silver) are prone to oxidation, which necessitates their additional processing, in particular, alloying [75].

Ionic liquids as antistatic additives

Ionic liquids (ILs) are salts with high thermal stability and low combustibility [76]. They represent a promising alternative to nanosized powdered inorganic fillers of polymeric materials due to their environmental safety and the possibility of using renewable natural raw materials for their manufacture [77–83]. Ionic liquids can reduce the resistivity of polymer coatings in both wet and dry states. For example, Mudzakir A. et al. [84] tested an ionic liquid, *cis*-oleylimidazolium iodide, as an antistatic agent in ceramic and wooden floor coverings. The ionic liquid was mixed with polyurethane. The surface resistivity of the coating with IL (2–9 wt. %) was in the range from 10^6 to $10^9 \Omega/\text{cm}^2$, which allows slow and safe discharge of the electrical charge into the ground. According to the authors [84], the antistatic properties of IL were due to the electrical conductivity of iodide ions and to the ability to attract less dust. The resulting coating can be used in the construction industry for floor treatment and as a decorating element. A disadvantage of the reported coating and of the study as a whole is the yellow color of the resulting

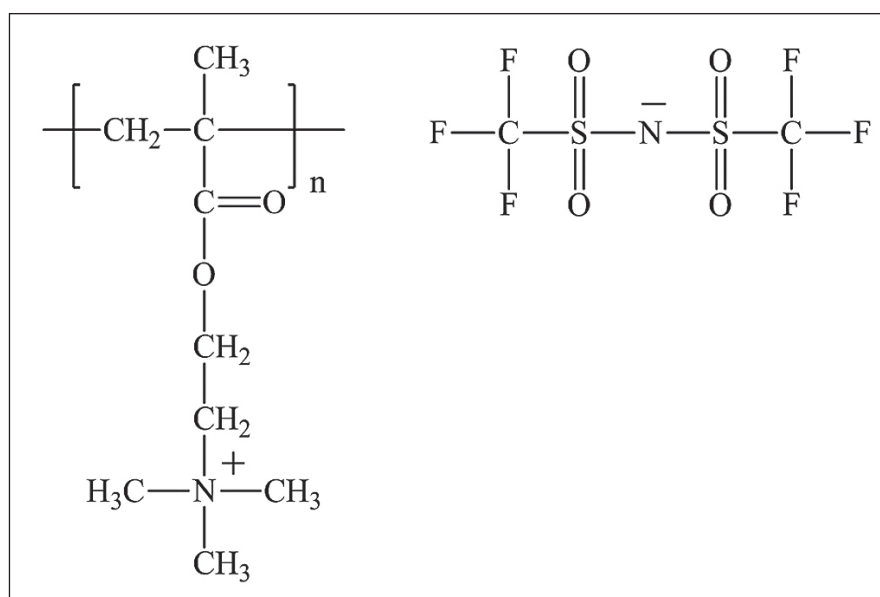


Fig. 5. Polymerized bis(trifluoromethanesulfonyl)ethyl trimethylammonium [85]

Table 1
Specific surface R_s and volumetric R_v electrical resistances of polyurethane films containing IL

IL content, ppm	$R_s, \Omega \cdot \text{cm}^{-2}$	$R_v, \Omega \cdot \text{cm}$
0	$2.1 \cdot 10^{12}$	$5.1 \cdot 10^9$
10	$3 \cdot 10^{11}$	$9 \cdot 10^9$
100	$6 \cdot 10^{10}$	$9.8 \cdot 10^8$
1000	$4.7 \cdot 10^9$	$5 \cdot 10^7$

coating caused by the presence of iodide anion in IL, which precludes the use of this coating on white surfaces.

To eliminate this drawback, a iodine-free IL, that is, polymerized bis(trifluoromethanesulfonylimide) [2-(methacryloyloxy)ethyl]trimethylammonium was utilized as a filler for polyurethane films [85] (Figure 5).

Owing to the use of IL, it was possible to reduce the surface and volume resistivity of polyurethane by two to three orders of magnitude at a low filler concentration of only 1000 ppm (Table 1). The film retained transparency upon addition of IL. The authors interpreted the mechanism of the antistatic action of IL by assuming that IL dissociates in the polyurethane matrix and $[\text{Tf}_2\text{N}]^-$ anions migrate along the fluctuations of polyurethane chains [84]. This IL was not washed away from the film in a methanol solution. However, a disadvantage of this IL is the fact that it is a solid, which complicates its dispersion in the polymer matrix.

The incorporation of ILs into poly(methyl methacrylate) and poly(butyl methacrylate) films and polyurea-based coatings made it possible to reduce their resistivity without loss of transparency [86–88]. Lower-viscosity ILs containing shorter alkyl chains have better antistatic properties [86, 88]. This is explained by the fact that an elonga-

tion of the IL alkyl chain leads to an increase in the van der Waals forces and a decrease in the ion mobility [89–91]. However, ILs based on imidazolium and ammonium cations are better compatible with the acrylate matrix than phosphonium cations, which is due to their better solubility [86]. Therefore, the compatibility of ILs with polymers is greatly affected by the hydrophobicity of the cations.

Ionic liquid anions also affect the conductivity. Tsurumaki A. et al. [86] compared the volume and surface resistivity of poly(butyl methacrylate) films (Blank) containing ionic liquids based on imidazolium cations $[\text{C}_4\text{mim}]^+$ and $[\text{BF}_4]^-$, $[\text{CF}_2\text{SO}_3]^-$, $[\text{PF}_6]^-$, $[\text{FSI}]^-$, and $[\text{Tf}_2\text{N}]^-$ anions. The most pronounced decrease in the surface resistivity was attained for the IL with the $[\text{BF}_4]^-$ anion, while the smallest decrease was observed for the IL with the $[\text{Tf}_2\text{N}]^-$ anion (Figure 6). The authors attributed the decrease in the resistivity to the plasticizing effect of IL and to the formation of a continuous conductive path in the polymer matrix. However, the influence of the type of anion on the resistivity was left without a proper explanation, which is a significant shortcoming of the work.

The addition of low-viscosity IL in low concentrations to polymer materials makes it possible to obtain antistatic polymer films and coatings. In particular, to reduce the resistivity of polyurea by five orders of magnitude, it is sufficient to introduce 2 wt. % potassium hexafluorophosphate [11, 87]. At the same time, the transparency of the coatings is preserved, unlike that upon the addition of carbon fillers. However, ionic liquids are more expensive antistatic additives than graphene, aluminum and copper powder. For example, the price of potassium hexafluorophosphate IL is 46 euro per kg; meanwhile, carbon black costs approximately \$1000 per ton. It is also possible that the mechanical properties of coatings based on polyurea can be deteriorated upon the addition of IL, in particular, the tensile strength may decrease due to IL migration to the surface [87].

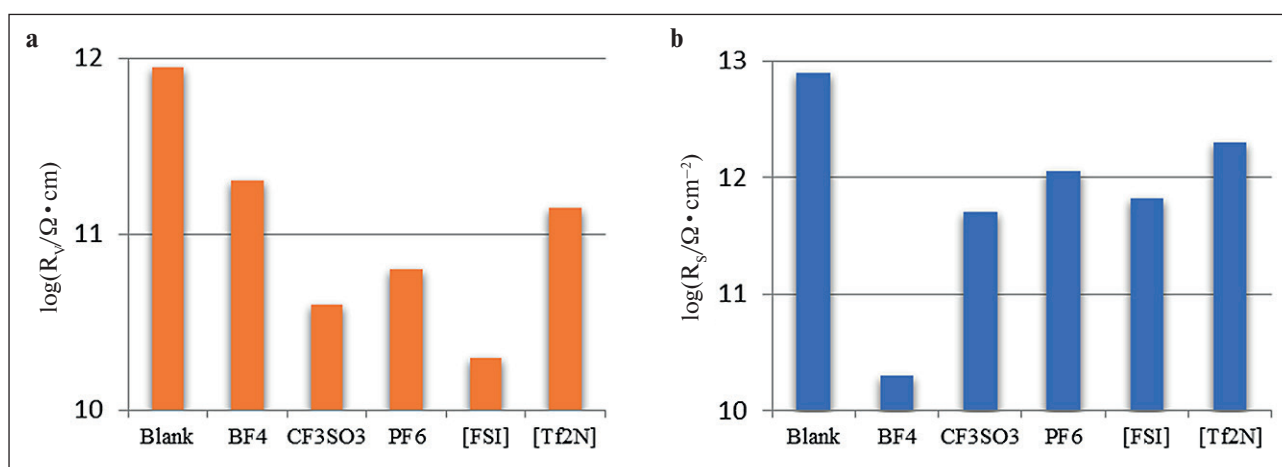


Fig. 6. R_v (orange) and R_s (blue) of poly(butyl methacrylate) films containing 10 wt. % IL [86]

Conductive polymers as antistatic additives

Conductive polymers, mainly polyaniline, are inexpensive and readily available alternatives to ILs. Conductive polyaniline was dispersed in methylpyrrolidone and added to polyurethane-acrylate latexes [92]. Polyaniline was added to improve the electrical insulation of damping coatings used in railway trains. As the content of polyaniline increased, the resistivity of the films decreased, while the electrical conductivity increased. When the content of polyaniline was 10 wt. %, the volume resistivity was $3.19 \cdot 10^5 \Omega \cdot \text{cm}$, and the best antistatic characteristics of the films were observed at a polyaniline dispersion content of 45 wt. %.

An advantage of polyaniline over nanoparticles is its controlled electrical conductivity due to the possibility of doping [93]. In particular, polyaniline was doped [93] with an acrylic ester grafting epoxy resin (A-g-EP) with carboxyl functional groups, which, in the opinion of the authors, provided conductive bridges between the polyaniline domains. As a result, the surface resistivity of the coatings decreased by 6 orders of magnitude compared to A-g-EP (from $1.6 \cdot 10^{11} \Omega/\text{cm}^2$ to $2.4 \cdot 10^5 \Omega/\text{cm}^2$). Gao X. et al. [94] showed that the addition of 3 wt. % polyaniline doped with dodecylbenzenesulfonic acid to polyurethane increases the electrical conductivity of the polyurethane film. The disadvantage of the works [92, 93] is the lack of data on the effect of polyaniline on the mechanical properties of polymer composites.

However, a disadvantage of polyaniline is the difficulty of its dispersion in the polymer matrix due to its aggregation, which this deteriorates the conductivity and complicates the formation of a conductive network in the coating; the lack of solubility in most organic solvents [95]; the need to use high concentrations to attain an antistatic effect (20 wt. % polyaniline was added to natural rubber in [96]); and lower electrical conductivity compared to titanium dioxide nanoparticles [97].

A high content of polyaniline in the matrix can lead to poor mechanical stability of the final product. To avoid deterioration of mechanical properties, polyaniline is mixed with nanoparticles [98]. Mirmohseni A. et al.

[98] inserted a nanohybrid of polyaniline, copper, and zinc oxide into the water base of a polyurethane dispersion. The resulting polyurethane coating had an increased adhesive strength to steel and scratch resistance compared to a coating without nanoparticles. The authors explained this fact by charged hydrophilic surface of the coating. Furthermore, the addition of the nanohybrid provides a better antistatic effect of polyurethane compared to single additives. The developed nanohybrid polyurethane coating is in demand in modern industrial construction for dedusting of soft steel and in hospital operating rooms.

CONCLUSION

Carbon nanotubes are promising antistatic additives to polymer coatings based on polyvinyl chloride for the manufacture of decorative finishing materials in construction and to composites based on polyoxymethylene intended for fixing devices for electronic equipment and gadgets (mobile phones, displays). However, further research is needed to improve the mechanical and adhesive properties of filled polymeric materials.

Graphene oxide nanosheets can only be used as additives in water-based polyurethane coatings as synergistic mixtures with metal nanoparticles or conductive polymers. The coatings can be used in civil engineering as decorative elements.

Metals and metal oxide nanoparticles, due to their good dispersibility in the polymer matrix, can be used as additives to polypropylene and polyurethane industrial coatings for devices and equipment.

Ionic liquids are promising additives in polyurethane coatings and polyacrylate films that can be used in civil and industrial construction and in electronics.

Surfactants are washed away from polymer coatings, and, therefore, they are not of interest to researchers.

Polyaniline is of interest as a cheap additive to water-based polyurethane coatings for the use in health care institutions (wall and floor coatings in hospitals, dental offices, etc.) only in combination with metal and metal oxide nanoparticles.

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