



Article

Aryloxyphosphazene-Modified and Graphite-Filled Epoxy Compositions with Reduced Flammability and Electrically Conductive Properties

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Abstract: A method has been developed for producing an epoxy composition based on a low-viscosity epoxy-resorcinol resin, a phosphazene-containing curing agent, isophoronediamine, and thermally expanded graphite as a filler. The degree of cure and the absence of side reactions during the curing process were confirmed using IR spectroscopy. The influence of the content of phosphazene-containing curing agent and filler on the physico-mechanical properties of the composition, its fire resistance, and antistatic properties were studied. Using the UL-94 HB horizontal burning test, it was found that the addition of 10 and 20 wt. % phosphazene-containing curing agent (relative to isophoronediamine) reduces the burning speed by 10 times compared to a sample without phosphazene. The addition of a filler to a composition containing phosphazene reduces the burning speed by 25 times compared to a composition without phosphazene and imparts antistatic properties to the epoxy composition, as evidenced by the specific volume electrical resistance of the order of 10^1 Ohm·m. Phosphazene-containing curing agent had no statistically significant effect on specific volume electrical resistivity ($p > 0.05$). Tests of physico-mechanical and adhesive properties (tensile strength, compressive strength, water absorption, water solubility, abrasion resistance, and adhesive strength) of filled epoxy compositions with 10 and 20 wt. % phosphazene-containing curing agent demonstrated that these properties met the requirements for floor coverings in construction and parts of electrical devices.

Keywords: phosphazene; fire resistance; coatings; abrasion resistance; electrical conductivity; antistatic material



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1. Introduction

To ensure fire safety in production, equipment, floors, and walls must be covered or made of materials that simultaneously have resistance to static discharge, low resistivity, low electrostatic attraction for charged dust particles, and increased fire resistance [1]. This is explained by the fact that the spark discharge has sufficient energy to ignite powders of organic materials, solvent vapors, polymeric fibrous materials, etc., which is the cause of fires and explosions. In addition, static electricity creates very high voltages (several thousand volts) and damages electronic devices.

The occurrence of static electricity is explained by the Helmholtz electric double layer theory. According to this theory, when two materials are in close contact with each other and then separated, a two-layer charge is formed at the material interface. As a result, the surface with a smaller work of function is positively charged, while one with a larger work of function is negatively charged [2].

There are several methods to reduce the generation of static electricity. These include electrical grounding, an increase in air conductivity, an increase in air humidity, the appli-

cation of antistatic coatings on the surface of materials, and the method of internal mixing of an antistatic agent with a material [3].

Carbon materials [4–6], nanofibers [7,8], nanotubes [9–12], nanoparticles of metal and metal oxides [13,14], graphene [15–18], and graphene oxide [19], as well as electrically conductive polymers (polyaniline, polypyrrole, and PEDOT:PSS) [20] and environmentally friendly ionic liquids, such as tributyl(octyl) phosphonium bis(trifluoromethanesulfonyl) imide [21], can be used as conductive or antistatic agents to reduce the electrical resistance of materials. Carbon nanotubes and graphene have a conjugated structure in which carbon atoms are in the state of sp^2 hybridization, which makes it possible to transfer an electron along the entire carbon chain and conduct an electric current. Carbon nanotubes in the amount of only 0.1 wt. % reduced the specific surface resistance of the photocurable 3,4-epoxycyclohexylmethyl 3,4-epoxycyclohexanecarboxylate epoxy resin by five orders of magnitude (from 10^{11} Ohm/cm² to 10^6 Ohm/cm²), which makes it possible to obtain the resulting composite to a dissipative material, a material that dissipates static electricity [22]. Carbon nanotubes do not impair mechanical properties of cured epoxy resin. On the contrary, there is an increase in the modulation of elasticity and the glass transition temperature of the cured epoxy resin. By adding 0.5 wt. % of graphene filler to epoxy resin, specific volume resistance decreased from 10^{14} Ohm m to 10^8 Ohm m, which met the requirements of the resistance of antistatic material as per the IEC 61340 standard [23]. The disadvantage of carbon nanotubes and graphene is their high cost, tendency for aggregation, and a significant increase in the viscosity of the composition in which they are added, which limits their use as antistatic additives.

In turn, for the manufacture of fire-resistant materials, phosphorus- and nitrogen-containing fire retardants are used, which slow down the spread of flame during combustion by isolating the ignition source. Unlike halogen-containing flame retardants, halogen-free flame retardants are non-toxic to the human body, since they do not form toxic gases during combustion, such as dibenzodioxins and dibenzofurans. In addition, bromine- and chlorine-containing compounds are effective as flame retardants only when used together with a synergist, i.e., antimony trioxide, which is toxic and carcinogenic to humans [24].

Among halogen-free flame retardants for polymeric materials, compounds of various classes are known. They are inorganic–organic hybrid agents, for example, phosphorylated Zn-based metal–organic framework [25], ammonium polyphosphate and aluminum hydroxide [26], condensation product of 2-[(5-oxo-6H-phosphanthridin-5-yl) methyl]-butanedioic acid and melamine [27], bis (2-acrylamidoethyl) phenyl phosphate [28], phosphorus-containing liquid epoxidate polybutadiene [29], and phosphazenes.

The disadvantage of inorganic flame retardants is that they are not able to react with the polymer matrix (in particular, ammonium polyphosphate) and have poor compatibility with the polymer matrix due to the different polarities of the matrix and the flame retardant. This causes phase separation and deterioration of the physico-mechanical properties of the composition [30]. To improve the compatibility of ammonium polyphosphate with epoxy resin, additional chemical modification with amines is required [31].

Poly- and cyclophosphazenes contain alternating phosphorus and nitrogen atoms, which allows them to provide a cooperative flame-retardant effect [32]. Phosphazenes in the gas phase form a significant number of free radicals, $PO_2\cdot$ and $PO\cdot$, and non-combustible pyrolysis gases. The resulting radicals interact with the $H\cdot$ and $\cdot OH$ radicals and thereby create a quenching effect. In turn, non-combustible gases (nitrogen and ammonia) reduce the concentration of oxygen and produce a diluting effect. In the condensed phase, pyrolysis fragments rich in phosphorus with high molecular weights are formed, which improve the thermal stability of the char layers and prevent the combustion of the polymer matrix in the condensed phase [33].

The addition of 1 wt. % aryloxycyclotriphosphazene-containing hydrazine fragments into epoxy resin increases the limiting oxygen index by 17% compared to pure resin [34]. Epoxy resins cured with hexacyclohexylaminocyclotriphosphazene and diaminotetracy-

clohexylaminocyclotriphosphazene have a lower total heat release compared to epoxy resins cured with ethylenediamine [35]. Benzimidazolyl-substituted cyclotriphosphazene reduced total smoke and peak smoke of benzoxazine based on bisphenol A and aniline by 36.5% and 39.35%, respectively. The authors of [36] explain this fact by the formation of an intumescent and compact char layer. The advantage of phosphazenes over other inorganic–organic flame retardants is the ease of their functionalization, which makes it possible to obtain compounds capable of reacting with polymers, in particular epoxy resins. This property of phosphazenes ensures their good dispersibility and compatibility with the polymer matrix without deteriorating the mechanical properties of the latter [37,38].

The present study is devoted to the preparation of an epoxy composition based on a low-viscosity epoxy-resorcinol resin, a phosphazene-containing curing agent, and graphite. The phosphazene-containing curing agent was used to provide a flame-retardant epoxy composition. Thermally expanded graphite was used to provide this composition with antistatic properties, as it has high electrical conductivity, as well as has lower cost and exhibits aggregability in the polymer matrix compared to other carbon fillers.

2. Materials and Methods

2.1. Materials

The epoxy-resorcinol resin (UP-637, EEW = 260 gE) was purchased from OOO Doros (Yaroslavl, Russia). Thermally expanded graphite (TEG), Expanded EX05, was purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Isophoronediamine, 4-hydroxybenzaldehyde (98%), ethanol, anhydrous ($\geq 99.5\%$), chloroform, anhydrous ($\geq 99\%$), sodium (99.9%), tetrahydrofuran, anhydrous ($\geq 99.5\%$), and magnesium sulfate, anhydrous ($\geq 99.5\%$), were purchased from Sigma-Aldrich (Saint Louis, MO, USA). Hexachlorocyclotriphosphazene (99%) was purchased from Fushimi Pharmaceutical Co., Ltd. (Marugame, Kagawa Prefecture, Japan).

2.2. Methods

IR spectra were recorded using a Nicolet 380 spectrometer (Thermo Fisher Scientific, Waltham, MA, USA) in the spectral range $4000\text{--}500\text{ cm}^{-1}$ with a wavenumber accuracy of 0.01 cm^{-1} .

The glass transition temperature and the development of the storage modulus, loss modulus, and $\tan \delta$ mechanical losses were determined according to ASTM D7028-07(2015) using dynamic mechanical analysis with a Netzsch DMA 242E Artemis device (Erich NETZSCH GmbH and Co. Holding KG, Selb, Germany). Measurements were performed in a three-point bending mode within the temperature range of $30\text{--}200\text{ }^{\circ}\text{C}$ at a standard heating rate of 5 K/min in a nitrogen atmosphere at a flow rate of 100 mL/min . The oscillation frequency was 1 Hz , and the load amplitude was 12 N during all the performed tests.

The rheology properties of the obtained compounds were determined at $25\text{ }^{\circ}\text{C}$ according to ISO 3219-1993 [39] using a rotational viscometer on a Brookfield CAP 2000+ viscometer (AMETEK Brookfield, Middleboro, MA, USA) with a cone-plate geometry with CAP-1. All tests were performed with a constant shear rate depending on viscosity.

The tensile strength and tensile modulus were determined according to ISO 527-2:2012 [40] on a 50ST Tinius Olsen universal testing machine (Tinius Olsen TMC, Horsham, PA, USA) with a traverse movement speed of 1 mm/min . The strains were measured using a video extensometer VEM208 (Tinius Olsen TMC, Horsham, PA, USA).

The compressive strength was estimated according to ISO 604:2002 [41] on a 50ST Tinius Olsen universal testing machine (Tinius Olsen TMC, Horsham, PA, USA) with a punch movement speed of 1 mm/min .

The shear strength of the adhesive bond was determined according to ISO 4587-79 [42] on a 50ST Tinius Olsen universal testing machine (Tinius Olsen TMC, Horsham, PA, USA) with a traverse speed of 10 mm/min . Steel plates were used as the bonded material (steel grade St3, analogue: A57036 (USA), SS330 (Japan), and DC03 (Germany)).

The water absorption and water solubility content of the test compositions were determined according to ISO 62:2008 [43] methods 2 and 3, respectively.

Horizontal (UL 94 HB) burning tests were conducted to investigate the flammability properties of composites according to ASTM D635. Specimens are conditioned for at least 48 h at 23 ± 2 °C and $50 \pm 5\%$ relative humidity prior to testing. Specimens, 125×12.5 mm, are each marked at 25 mm and 100 mm from one end. Each specimen is clamped horizontally at the end nearest the 100 mm mark, with its transverse axis inclined at $45 \pm 2^\circ$ to the horizontal. A 20 mm high blue flame from a burner is applied to the end of the specimen for a period of 30 s, or whenever the flame front reaches the 25 mm mark, whichever comes first. The times to reach the 25 mm mark and the 100 mm mark, or when burning ceases are recorded, and the extent of burning are measured. The behavior of specimens was classified as HB (HB = Horizontal Burning) under the following conditions: the flame front does not pass the 25 mm reference mark, or the flame front passes the 25 mm reference mark but does not reach the 100 mm reference mark, or the flame front reaches the 100 mm reference mark and the linear burning rate does not exceed 40 mm/min for specimens having a thickness between 3 and 13 mm or 75 mm/min for specimens having a thickness less than 3 mm.

The specific volume electrical resistance of the materials was determined using a GOM-802 digital programmable milliohmmeter in accordance with ISO 3915:2022 [44]. The test specimens were thin strips of a composite of fixed dimensions (thickness up to 1 mm, width up to 5 mm, and length 40 mm) deposited on degreased glass.

The resistance R , in ohms, was calculated corresponding to each position of the potentiometric electrodes, using the relation,

$$R = \frac{\Delta U}{I}$$

where ΔU is the voltage drop, in volts, between the potentiometric electrodes, and I is the current, in amperes, passing through the specimen.

The specific volume electrical resistance ρ , expressed in ohm meters, was calculated by using the relation,

$$\rho = \frac{R \cdot w \cdot h}{L}$$

where R is the measured resistance, in ohms, L is the distance between electrodes, in meters, w is the width of the specimen, in meters, and h is the thickness of the sample, in meters. Four specimens were tested for each experimental condition. An average bulk resistivity and standard deviation for each sample were calculated for each sample.

Abrasion resistance was determined in accordance with ISO 7784-1:2023 [45].

The specimen is placed on the ground glass of the platform with the coated side up and firmly attached so that the specimen does not move during the test. Then, the light source is turned on, and at least 2 kg of sand is poured into the funnel.

From the funnel, the sand is poured into the guide tube and falls in a jet and then onto the sample. Sand is poured from the sample into the tank. If abrasion of the coating to the substrate has not occurred, the test is continued. In this case, the sand collected in the tank is returned to the funnel, substituting an empty tank instead of a filled tank. Abrasion is stopped as soon as damage to the coating of the substrate is detected. At this moment, the jet of sand in the guide tube is closed with a damper. The sand collected in the tank is weighed, and the total amount of sand used for abrasion from the start of the test is recorded.

Abrasion resistance X was calculated by the formula:

$$X = \frac{m}{h}$$

where m is the mass of sand used for abrasion, expressed in kilograms, and h is the thickness of the coating, millimeters. The test results are the arithmetic means of the results of at least three determinations.

The average values of the performance characteristics of various samples were compared using two-way ANOVA followed by Tukey's special analysis for multiple comparisons. The tables in parentheses present the maximum values of the average deviations of the parameters. The significance level was set up at $p < 0.05$. Five samples were used for each test.

2.3. Synthesis of Hexakis-[(4-formyl)phenoxy]cyclotriphosphazene (FPP)

4-Hydroxybenzaldehyde (7.32 g, 0.06 mol) was dissolved in ethanol (30 mL) in a three-necked flask equipped with a stirrer and a reflux condenser. After the complete dissolution of 4-hydroxybenzaldehyde, the alcoholic solution of sodium ethylate, which was obtained via the dissolution of sodium (1.15 g, 0.05 mol) in ethanol (20 mL), was added in the flask. The reaction time was 10 min; then, ethanol was distilled off on a rotary evaporator in vacuum. The residue was dried in vacuum up to a constant weight. The yield of the product was quantitative. 4-Hydroxybenzaldehyde phenolate (8.64 g, 0.06 mol) was loaded in a three-necked flask equipped with a stirrer and a reflux condenser, and tetrahydrofuran (40 mL) was added. A solution of hexachlorocyclotriphosphazene (2.61 g, 0.0075 mol) in tetrahydrofuran (20 mL) was added to the dispersion formed during stirring. The time of reaction was 9 h during solvent boiling. When the process was complete, the reaction mixture was filtered off, and the mother liquor was evaporated on a rotary evaporator. The product was recrystallized from the ethanol–chloroform mixture. Yield: 4.52 g (70%).

2.4. Synthesis of the Phosphazene-Containing Curing Agent

FPP was dissolved in 40 g of isophoronediamine at 100 °C in a 100 mL round-bottom flask equipped with a magnetic stirrer and a reflux condenser as per the formulations provided in Table 1. Then, magnesium sulfate was added to the resulting mixture, and synthesis was carried out at the same temperature for 24 h. Lastly, the precipitate was separated in a centrifuge. The resulting liquid transparent mass was used without further purification.

Table 1. Curing agent composition formulations.

Ingredients	Weight of FPP Added to IPDA, wt. %		
	10	20	30
FPP, g	4	8	12
MgSO ₄ , g	0.65	1.32	1.98

2.5. Preparation of a Composition Based on Epoxy-Resorcinol Resin and Phosphazene-Containing Curing Agent

Epoxy resin was pretreated under vacuum at 50 °C for 1 h. The resin was cooled to room temperature, and 1.5 g of the curing agent was added and mixed for 10 min, then the filler (or without it) was introduced into the composition in the amounts indicated in Table 2, and the mixture was stirred until a homogeneous mass was formed. The finished compound was poured into molds, stored for one day at 25 °C, and samples gradually cured for 5 h at 130 °C.

Table 2. The amount of resin dosed with the curing agent vs. the modifier content.

Epoxy-Resorcinol Resin Weight, g	Content of Phosphazene Modifier in the Curing Agent, wt. %	Filler Weight, g
4.54	0	1.2
4.02	10	1.1
3.51	20	1.0
2.99	30	-

3. Discussion

Epoxy-resorcinol resin was chosen as the base of the binder. The choice of this resin was due to its relatively low viscosity (0.7 mPa·s), which offsets the high viscosity of the phosphazene-containing curing agent ($1.32 \div 4.65$ Pa·s depending on the concentration). As a curing agent, pure IPDA was used, as well as IPDA containing 10, 20, and 30 wt. % PCA (Figure 1).

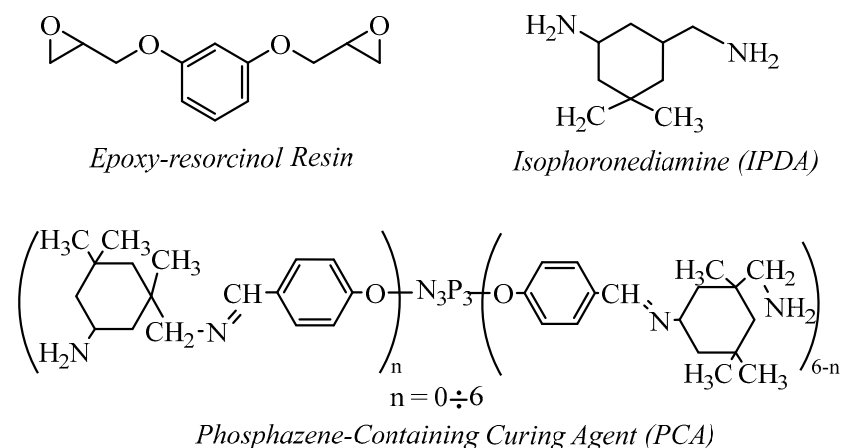


Figure 1. Formulas of compounds used to obtain epoxy compositions.

Thermally expanded graphite was used as a filler to impart antistatic properties to the developed material. The choice of graphite is due to its relatively high electrical conductivity and chemical resistance, good dispersibility in binders, and relative cheapness. The amount of filler introduced was determined experimentally and amounted to 20% by weight of the binder. At a lower concentration of graphite, the conductivity was unsatisfactory, and at a higher concentration, problems arose in obtaining a homogeneous composition.

Since the main objective of the study was the development of a material with reduced flammability, the corresponding samples of the cured compositions were initially prepared. The degree of curing of the resin was evaluated using IR spectroscopy. Comparing the spectra of the curing agent, as well as the original and cured resins (Figure 2), we can conclude that the conversion of epoxy groups occurs completely when using a curing agent containing up to 20 wt. % phosphazene (Figure 2D). This is based on the fact that, in the cured composition, there are no band characteristics of symmetric and asymmetric vibrations of oxirane rings ($C-O-C_{sym}$ and $C-O-C_{asym}$), which are observed in the original resin. However, when the content of phosphazene in the curing agent is 30 wt. %, bands of residual epoxy groups are visible in the spectrum of the sample. This can be explained by the high viscosity of the initial composition, which leads to poor homogenization of the mixture and, as a result, incomplete curing of the resin. A more thorough homogenization of the composition due to the increase in time was not possible due to the beginning of the curing process and an even greater increase in viscosity.

In addition, from the analysis of the IR spectra, it can be concluded that the azomethine groups and the phosphazene ring are not destroyed during the curing process.

The fire resistance rating was evaluated according to the UL-94 HB test with horizontal fixing of the cured samples. As can be seen from Table 3, when the content in the curing agent is 10 and 20 wt. % PCA, the burning rate of unfilled samples is reduced by a factor of 10 compared to a sample not containing phosphazene. However, at a concentration of 30 wt. % PCA in IPDA, the burning rate increases again. This fact can be explained by a significant increase in the viscosity of the compositions with an increase in their content of phosphazene, which prevents the uniform distribution of the components in each other and reduces their flame-retardant properties. It should also be noted that for all samples containing phosphazene, no burning droplets were formed during combustion, which were observed in the case of the unmodified composition.

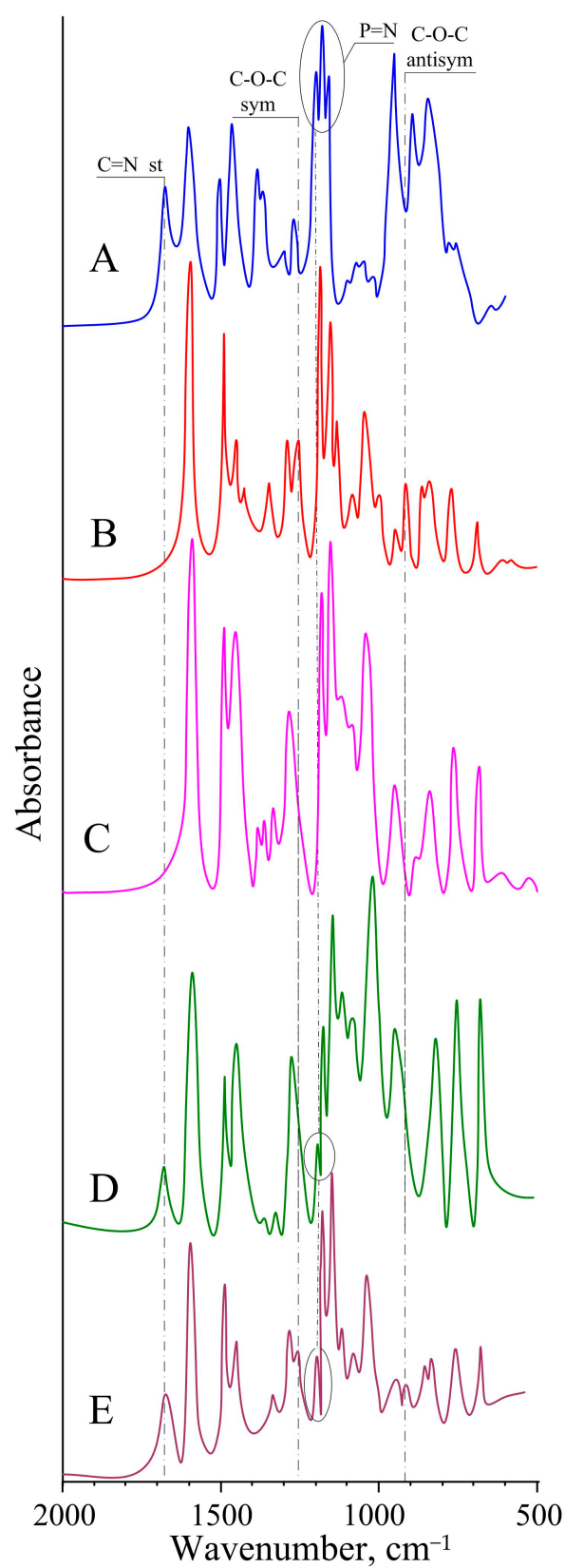


Figure 2. IR spectra of IPDA containing PCA (A); epoxy resin (B); epoxy resin cured IPDA (C); epoxy resin cured IPDA containing 20 wt. % PCA (D); and epoxy resin cured IPDA containing 30 wt. % PCA (E).

Table 3. Characteristics necessary for the selection of optimal composition formulations.

Parameter	Content of PCA in IPDA, wt. %						
	Without Filler				With Filler		
	0	10	20	30	0	10	20
Phosphorus content in cured compositions, wt. %	0	0.14	0.31	0.52	0	0.12	0.25
Burning rate of cured compositions, (± 0.5), mm/min	50	5	5	25	5	2	2
p	$p < 0.05$				$p < 0.05$		
Initial viscosity of uncured compositions, (± 0.1), Pa·s	0.63	1.23	2.12	3.7	7.61	8.5	10.2
Specific volume electrical resistance of cured compositions, (± 3), Ohm·m	-	-	-	-	20.9	16.55	18.5
p	$p > 0.05$						

In the case of filling compositions with graphite, the effect of viscosity becomes even more significant. Thus, when introducing a filler into compositions containing 30% PCA in IPDA, it was not at all possible to achieve a uniform structure of the material. Therefore, further studies were carried out using a curing agent containing no more than 20% PCA.

However, the filler had a significant positive effect on fire resistance. The burning rate of graphite-filled compositions that do not contain phosphazene decreased by 10 times, and with a content of 10 and 20% PCA in the curing agent, by 25 times. In this case, the formation of burning droplets was not observed for all samples. The burning rate of the composites did not exceed 40 mm/min (the wall thickness was more than 3 mm) at a phosphorus content of less than 1 wt. % (Table 3), which allowed them to be assigned to the HB flammability rating and indicated their reduced flammability.

As for the specific volume electrical resistance, PCA had almost no effect on this parameter ($p > 0.05$). The resistance values for all filled samples lie within the same numerical order, sufficient to use the developed compositions as antistatic materials. The obtained resistivity values were within the required range established by the ESD protection standard. In turn, at lower resistance, the risk of electric shock to the human body increases [46]. Specific volume electrical resistance was lowered by five orders of magnitude compared to the previously obtained compositions containing graphite [47]. The low value of specific volume electrical resistance of filled compositions and, consequently, high electrical conductivity are due to screening-oriented electron transfer through thermally expanded graphite due to hopping and tunneling mechanisms [48]. In the case of unfilled samples, the cured compositions are dielectrics. Their resistivity also does not depend on the content of phosphazene and lies in the region of 10^{14} Ohm·m.

To assess the potential areas of application of the obtained compositions, tests of their mechanical characteristics were carried out. The test results are presented in Table 4.

From Table 4, it can be observed that increasing the content of PCA contributes to an increase in tensile strength in unfilled compositions. This is due to the fact that phosphazene has a rigid polyconjugated structure, which, when distributed in a polymer matrix, acts as a rigid reinforcing material. The tensile strength values of the resulting filled compositions are of the same order as epoxy compositions reinforced with carbon nanotubes [49]. This is also consistent with the values of ultimate tensile strain, which also increases with increasing phosphazene content, but only slightly. Most likely, this fact is due to the orientation of the organic conjugated fragments of phosphazene along the tension axis.

In the case of filled composites, the opposite effect is observed: the values of tensile strength and ultimate tensile strain decrease with increasing PCA content in the composite. Presumably, this effect can be explained by a significant increase in the viscosity of the

filled compositions with an increase in the content of phosphazene (Table 3), due to which the system becomes less homogeneous, and the destruction of the material is easier.

Table 4. Characteristics necessary for the selection of optimal composition formulations.

Parameter	Content of PCA in IPDA, wt. %					
	Without Filler			With Filler		
	0	10	20	0	10	20
Tensile strength, (± 0.5), MPa	46.6	53.39	70.38	45.96	40.34	38.57
<i>p</i>	$(p < 0.05)$			$(p < 0.05)$		
Ultimate tensile strain, (± 0.05), %	2.2	2.26	2.56	1.3	1.12	1.00
<i>p</i>	$(p < 0.05)$			$(p < 0.05)$		
Compressive strength, (± 1), MPa	132	135	142	129	131	135
<i>p</i>	$(p < 0.05)$			$(p < 0.05)$		
Ultimate compression strain, (± 0.1), %	12	12.52	12.68	13.1	12.4	12.3
<i>p</i>	$(p < 0.05)$			$(p < 0.05)$		

When testing the developed materials for compression, in the case of unfilled samples, as in tensile testing, an increase in strength and ultimate strain was observed with increasing PCA content. At the same time, for filled compositions, an increase in compressive strength with an increase in the content of phosphazene is also observed, but the values of this characteristic are somewhat lower than those for unfilled characteristics. The increase in the strength of the filled compositions is explained by the fact that, during compression, the main load is assumed by the filler. The lower compression characteristics of filled compositions compared to unfilled compositions can also be explained by the effect of viscosity on the uniformity of the system, which is also confirmed by a decrease in ultimate compression strain with increasing phosphazene content. Nevertheless, the decrease in the values of the strength characteristics when filling the compositions with graphite is not critical; therefore, additional studies were carried out to more accurately assess the prospects for the use of the developed materials.

Initially, the water absorption and water solubility of the material were evaluated. As can be seen from Table 5, neither the content of PCA in IPDA nor the presence of a filler in the composition have any effect on these characteristics, which are extremely insignificant and lie within the experimental error. Accordingly, it can be concluded that the developed material can be used in wet conditions or subjected to wet cleaning.

It was also found that the glass transition temperature of the obtained materials practically does not depend on either the presence of the filler or the amount of phosphazene in it, and varies within the range of 116–124 °C. This parameter characterizes the preservation of the physical and mechanical characteristics of the material up to the specified temperatures.

It was found that the content of PCA in IPDA did not affect the abrasion resistance of filled and unfilled compositions ($p > 0.05$). However, this characteristic was significantly affected by the presence of a filler in the material, and the resistance increased by almost 1.5 times. This can be explained by the fact that the filler acts as an abrasive.

In turn, the content of phosphazene affected the adhesion strength of filled and unfilled compositions to aluminum. With an increase in the content of phosphazene in the material, the adhesion increased, which was probably due to the presence of azomethine groups in phosphazene that are capable of interacting with the aluminum surface. At the same time, in the case of filled compositions, the adhesive strength values turned out to be lower than those for unfilled compositions, which was quite explicable by a decrease in the contact points between the binder and the metal surface. For all samples during testing, an adhesive type of destruction of the adhesive joint was observed.

Table 5. Performance characteristics of the developed compositions.

Parameter	Content of PCA in IPDA, wt. %					
	Without Filler			With Filler		
	0	10	20	0	10	20
Water absorption, (± 0.2), %	0.3	0.3	0.4	0.2	0.3	0.2
p	$p > 0.05$			$p > 0.05$		
Water solubility, (± 0.2), %	0.2	0.3	0.3	0.4	0.3	0.3
p	$p > 0.05$			$p > 0.05$		
Glass transition temperature, (± 3), °C	116	119	119	116	116	124
p	$p > 0.05$			$p < 0.05$		
Abrasion resistance, (± 1), kg/mm	35	34	36	48	51	51
p	$p > 0.05$			$p > 0.05$		
Adhesion strength, (± 0.05), MPa	3.1	3.35	3.41	2.4	2.87	3.2
	$p < 0.05$			$p < 0.05$		
Type of destruction of the glue line	Adhesive					

4. Conclusions

The aim of the work was to develop a composite material based on epoxy resins, which simultaneously has antistatic properties, fire resistance, and stable physical and mechanical properties during operation. To obtain such a material, thermally expanded graphite was used as a filler, and a phosphazene-containing curing agent was used as a fire-retardant agent.

Low specific volume electrical resistance ($18.5 \pm 3 \text{ Ohm m}$), low burning speed ($2 \pm 0.5 \text{ mm/min}$), abrasion resistance ($51 \pm 1 \text{ kg/mm}$), low water absorption ($0.2 \pm 0.2\%$), and water solubility ($0.3 \pm 0.2\%$) of the developed epoxy composition will allow the material to be used for the manufacture of composite self-leveling floors at facilities where fire hazardous, explosive, and flammable substances are stored, in areas of high humidity and with increased requirements for the slightest accumulation of dust, and places with high burst loads. This is due to the complete distribution of the electrical static discharge over the floor surface and its successful removal to the grounding system.

Another area of application of the developed material is the cast insulation of measuring current and voltage transformers and power transformers, during prolonged operation in which the windings heat up, which can lead to fire. The use of an epoxy compound will allow the use of transformers in industrial premises where high fire safety is required. To use the developed material, a method of vacuum injection/impregnation of the transformer windings with an epoxide compound, followed by heat treatment, is proposed.

Adhesion strength to aluminum, equal to $3.2 \pm 0.05 \text{ MPa}$, and low specific volume electrical resistance of the developed filled epoxy composition will allow its use as a conductive adhesive-sealant for gluing electrical products, assemblies and parts of radio engineering and electronic devices, metal parts of measuring equipment, and fixing and sealing in the production of electronic equipment. The adhesive will form joints with sufficient strength to hold two surfaces together and have the ability to provide a secure connection without removing old adhesive residue. Conductive adhesive will create fire and explosion-safe conditions and service life of electrical products and increase their maintainability.

To improve the dispersibility of thermally expanded graphite in epoxy resin, which is important for the fire resistance and mechanical properties of the finished product, it is further proposed to use the following: (1) the method of ultrasonic treatment of epoxy resin and thermally expanded graphite before starting the curing process, (2) pre-dispersion of the filler in a volatile organic solvent (for example, acetone), mixed it with an epoxy resin, and followed by the removal of the solvent under vacuum, or (3) prolonged mechanical

stirring of the components when heated. Resin transfer molding or vacuum-assisted resin transfer molding methods can be used to produce multi-scale epoxy composites.

Due to its layered lattice structure, graphite has a low coefficient of friction and is used as a solid lubricant. Of great importance for the industrial application of the obtained epoxy compositions is the establishment of the relationship between tribological and mechanical characteristics, which is planned in our future research.

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