ISSN 1027-5495. Functional Materials, 30, No.3 (2023), p. 398-402
doi:http://dx.doi.org/10.15407/fm30.03.398

Electrical conductivity of epoxy composites with silicon carbide powder filler

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Received May 10, 2023

Specific electrical conductivity of ED-20 epoxy-diane resin with silicon carbide powder filler of different dispersions was investigated. The obtained increase in the specific electrical conductivity of the epoxy resin with the increasing content of the silicon carbide powder filler is explained by the higher value of its particular electrical conductivity than that of the epoxy matrix. It was established that the specific electrical conductivity of investigated epoxy composites also depends on the size of the filler grains and their shape. Samples of epoxy composites and silicon carbide powder with the grains size of 60 to 150 microns have the highest specific electrical conductivity. From the analysis of the theoretical calculations carried out within the model of spherical conductive inclusions and the obtained photographs of the shape of the grains of silicon carbide powder, it follows that such grains have a form close to spherical. An increase of the degree of deviation from the sphericity of the shape of the grains with dimensions $d > 150$ μm is the reason decreasing the specific electrical conductivity of both the silicon carbide powder and the obtained epoxy composite samples. The obtained results will have practical applications for the development based on the epoxy-diane resin with silicon carbide powder filler of theconductive antistatic coatings, protective screens against the aggressive action of electromagnetic radiation and radiation, and elements of electronic equipment .

Keywords: epoxy-diane resin, silicon carbide powder filler, specific electrical conductivity, dispersion, non-uniform material.

Електропровідність епоксикомпозитів з наповнювачем порошку карбіду кремнію. *С.В. Луньов, М.В. Хвищун, Ю.В. Коваль, А.І. Цизь*

Досліджено питому електропровідність епоксидно-діанової смоли марки ЕД-20 з наповнювачем порошку карбіду кремнію різної дисперсності. Одержане зростання питомої електропровідності епоксидної смоли зі збільшенням вмісту наповнювача порошку карбіду кремнію пояснюється більшим значенням його питомої електропровідності, ніж епоксидної матриці. Встановлено, що питома електропровідність досліджуваних епоксикомпозитів залежить також від розміру зерен наповнювача та їх форми. Найбільшу питому електропровідність мають зразки епоксикомпозитів та порошок карбіду кремнію з розміром зерен від 60 до 150 мкм. З аналізу проведених теоретичних розрахунків в рамках моделі сферичних провідних включень та одержаних фотографій форми зерен порошку карбіду кремнію слідує, що такі зерена мають форму близьку до сферичної. Збільшення ступеня відхилення від сферичності форми зерен розмірами $d > 150$ мкм є причиною зменшеня питомої електропровідності як порошку карбіду кремнію, так і одержаних зразків епоксикомпозитів. Одержані результати матимуть практичне застосування для розробки на основі епоксидно-діанової смоли з наповнювачем порошку карбіду кремнію провідних антистатичних покриттів, захисних екранів від агресивної дії електромагнітного випромінювання та радіації, елементів електронної техніки.

1. Introduction

Obtaining polymer composite materials with special electrophysical properties largely depends on the nature of the filler, the shape, size, and nature of the distribution of its particles, as well as on the degree of interaction between the components [1-4]. Characteristics of particle aggregation, crystallization conditions, and a number of other factors change the morphology of the polymer matrix, and as a result, the obtained on their basis composite materials acquire unique properties, which leads to the expansion of the possibilities of their practical application. For example, polymers used to protect against electromagnetic radiation and radiation, as well as in static charge removal systems, must have a certain level of electrical conductivity [5-9]. So, in order to remove a static charge, materials with a resistivity of 10^5-10^7 Om·cm are suitable, and for protection against electromagnetic radiation, the resistivity should be of the order of 10^1–10^3 Om·cm. The synthesis of polymers with the intrinsic (natural) electrical conductivity is an expensive but effective solution for the described applications. Conductive polymer systems in which a conductive filler is added to a relatively non-conducting (dielectric) matrix are more promising from a commercial point of view. In this regard, electroactive polymer materials based on epoxy resins with semiconductor fillers, which are widely used in electric microphones, dosimeters, pressure sensors, air filters, and electromechanical converters, are of particular interest to many researchers [10-15]. One of the promising such semiconductor fillers can be silicon carbide powder, which in many characteristics, such as thermal conductivity, breakdown voltage, microhardness, relatively low cost, and high thermal and radiation resistance, exceeds the silicon, germanium and gallium arsenide [16-18]. Therefore, it is interesting from both a scientific and a practical point of view study of the impact of content, shape, and dispersion of silicon carbide powder on the electrical properties of epoxy composites.

2. Samples of epoxy composites and the experimental method

Cardboard forms in the form of rectangular parallelepipeds, 1×1×3 cm in size were initially produced to conduct research on the electrical conductivity of ED-20 epoxy-diane resin with polyethylene polyamine (PEPA) hardener, without fillers and with fillers of silicon carbide powders of different grain sizes. Epoxy resin or a ready-made solution of epoxy resin with silicon carbide powder was poured into such molds, which were thoroughly mixed to achieve maximum homogeneity. 12% PEPA hardener was added for quick and high-quality hardening of the epoxy-diane resin. As it was previously established by other authors [19-21], this content of hardener is the most optimal to ensure homogeneity of hardening of epoxy resin and its high physico-mechanical and operational properties. Conductors 1-4 were inserted into the surface area of the parallelepiped to a depth of approximately 2 mm, to which the probes of the E6-13A teraohmmeter were connected (Fig. 1). Identical homogeneity in the distribution of silicon carbide powder over the volume of the investigated sample reaffirmed by the reproducibility of the electrical conductivity values of a batch of samples (3-5 samples) filled epoxy composites. Studies of the specific electrical conductivity of the epoxy composite along the length of the sample were conducted for controlling the uniformity of the distribution of silicon carbide in the volume of the sample and its possible shrinkage. For this, the resistance between contacts 1-3, 1-4, 2-3, 2-4 and 1-2 was measured. Taking into account the distance between these contacts along the length of the sample, the specific electrical conductivity of the epoxy composite was determined. The same values of specific electrical conductivity (with an accuracy of 10%) for different lengths of the sample indicated the uniformity of the distribution of the filler in the volume of the epoxy composite and the absence of shrinkage of the silicon carbide powder.

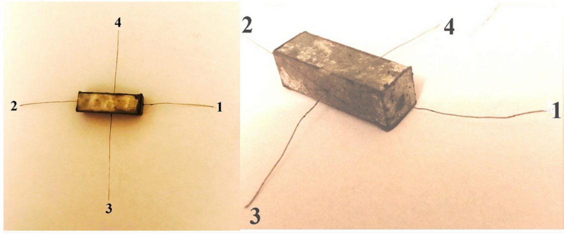

Fig. 1. Photographs of the epoxy composite sample for conducting electrical conductivity measurements.

A special cylindrical capsule was created for measuring the electrical conductivity of silicon carbide powder. Silicon carbide powder of a given volume was pouredIt into this capsule consisting of an insulating case and two movable electrodes, the ends of the cylinder, between which the resistance was measured. The electrical conductivity of such a capsule will be determined by the resistance of the powder, since the resistance of the insulating case is much greater. The research of the shape of silicon carbide powder grains was carried out with the help of an optical microscope MMP-14C.

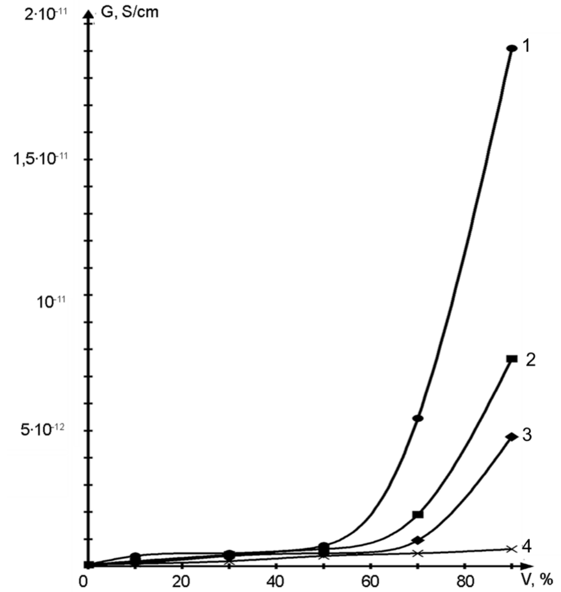

Fig. 2. Dependencies of specific electrical conductivity of epoxy resin on the content of silicon carbide powder filler with different grain sizes d, μm: *1* – 80; *2* – 60; *3* – 150; *4* – 320.

3. Experimental results and their discussion

The resistance of the sample of the correct geometric shape

$$R = \rho \frac{L}{S}, \qquad (1)$$

where ρ is the resistivity of the material, L is the length of the sample, which in our case is equal to 3 cm, and S is the cross-sectional area (for the epoxy composite sample, $S=1\ cm^2$).

Specific electrical conductivity

$$\sigma = \frac{1}{\rho} = \frac{L}{RS}. \qquad (2)$$

Dependencies of the specific electrical conductivity of the epoxy resin with the silicon carbide powder filler of different content and dispersion were obtained on the basis of expression (2) and experimental values of the resistance of the epoxy composite, the (Fig. 2).

As follows from Fig. 2, a significant (by about an order of magnitude) increase in specific electrical conductivity is observed for epoxy resin samples, filled by the silicon carbide powder with the content in the volume of the sample of more than 50%. This is explained by the fact that the specific electrical conductivity of the powder grains is greater than that of the epoxy resin. The dependence of the specific electrical conductivity of the investigated samples on the grain size of the silicon carbide powder is also quite significant. According to Fig. 2 (curve 1), the epoxy resin sample with grain size d=80 μm has the highest specific electrical conductivity.

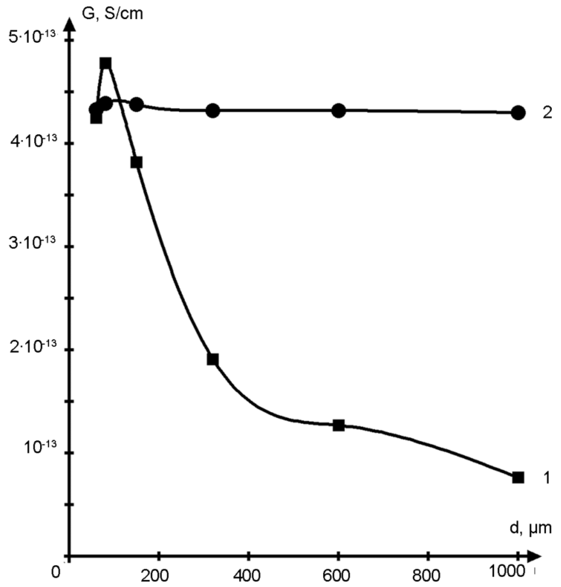

Fig. 3. Dependencies of the specific electrical conductivity of the epoxy resin on the grain size of the silicon carbide powder filler with the content of 30% in the sample: *1* – experimental results; *2* – results of theoretical calculations.

Dependencies of the specific electrical conductivity of epoxy resin and silicon carbide powder on the size of its grains were investigated for a more detailed study of this effect (Fig. 3 and Fig. 4).

According to Fig. 3, the specific electrical conductivity reaches a maximum at $d = 80$ μm and then decreasing as the grain size increases. This may be due to the fact that when increasing the size of the grain, it is also necessary to take into account its shape, which is obviously different from the spherical one. A qualitatively similar dependence of specific electrical conductivity on the size of the filler grain was obtained for silicon carbide powder (Fig. 4). To assess the impact of the non-spherical shape of silicon carbide powder grains on the electrical conductivity of the obtained epoxy composites, calculations of the specific electrical conductivity of epoxy resin with a filler of this powder of different dispersion were also carried out based on the theory of electrical conductivity of heterogeneous material with conductive inclusions of spherical shape [22]:

$$\sigma = \sigma_2 \left[1 + \frac{(\sigma_1 + \sigma_2) f}{\sigma_2 + \frac{1}{3}(\sigma_1 - \sigma_2)(1 - f)} \right], \qquad (3)$$

where σ_1 and σ_2 are the specific electrical conductivities of the inclusions and the conductive matrix, respectively, and f is the fraction of the sample volume occupied by the inclusions. For our case, σ_1 is the specific electrical conduc-

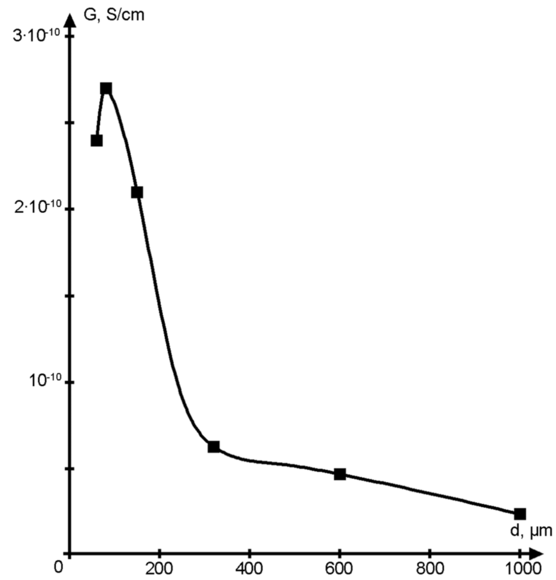

Fig. 4. Dependence of the specific electrical conductivity of the silicon carbide powder on its grain size.

tivity of silicon carbide powder, and σ_2 is the specific electrical conductivity of epoxy resin. Taking into account the obtained experimental results of the specific electrical conductivities σ_1 (Fig. 4) and σ_2, calculations (for the case $f = 0.3$) of the specific electrical conductivity of epoxy composites with a filler of silicon carbide powder of different dispersion were carried out on the basis of expression (3) (Fig. 3, curve 2). As follows from the comparison of the curves 1 and 2 of Fig. 3, the best agreement between the experimental results and the corresponding theoretical calculations is for the samples of epoxy composites, filled by the silicon carbide powder with the grain size of 60 to 150 μm. It is obvious that the shape of such grains is close to spherical. Increasing the degree of non-sphericity of the shape of grains with the size of $d > 150$ μm leads to a significant decrease in the specific electrical conductivity of the obtained epoxy composites. This statement is also confirmed by the photographs of grains of silicon carbide powder, obtained at the 20-fold magnification (Fig. 5).

As we can be seen from these photos, the shape of the grains of silicon carbide powder is really different from spherical. Grains ranging in size from 60 to 150 μm have a more convex shape, close to spherical.

4. Conclusions

Growing specific electrical conductivity of the epoxy resin with the increasing content of the silicon carbide powder filler is explained by the higher value of its particular electrical conductivity in relation to the value of the specific

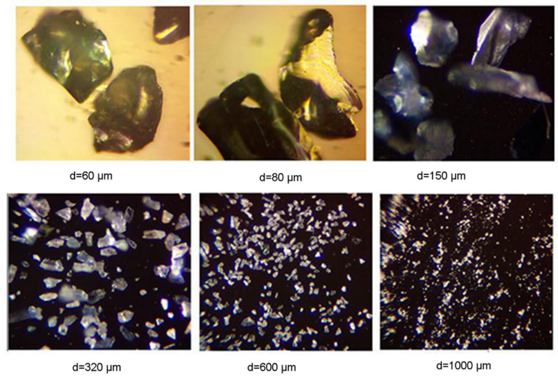

Fig. 5. Photos of grains of silicon carbide powder of different dispersion.

electrical conductivity of the epoxy matrix. It was established that the specific electrical conductivity of epoxy resin significantly depends on the size of the grains of silicon carbide powder filler, as well as their shape. Samples of epoxy composites and silicon carbide powder with the grain size of 60 to 150 microns have the highest specific electrical conductivity. The shape of such grains is closest to spherical. The deviation from the sphericity of the shape of the grains is the reason for the decrease in the specific electrical conductivity of both the silicon carbide powder and the epoxy resin filled with this powder with the grains size of $d > 150$ μm. Therefore, the shape criterion is more noticeable for large grain sizes and, accordingly, has a greater effect on the specific electrical conductivity of the studied epoxy resin samples. This is also confirmed by the calculations of the specific electrical conductivity of epoxy resin in the model of spherical inclusions of silicon carbide powder.

The obtained electrically conductive composite materials based on epoxy-diane resin with silicon carbide powder filler will be able to find their practical use as a raw material for or manufacturing the pressure and temperature sensors, conductive antistatic coatings, protective screens for the electronic equipment elements against electromagnetic radiation and ionizing radiation.

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