Investigation of dielectric parameters of structural nanocomposites based on polyethylene

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> **Abstract.** The dependence of the dielectric constant and specific volume resistance of a structural composite based on (80 wt%) synthetic isoprene rubber and (20 wt%) low density polyethylene on the content of nanosized particles of aluminum fillers, Nalchikin and carbon black in small quantities is shown graphically. Using an electron probe method for studying the microstructure of a structural material, a hydrostatic weighing method based on determining the density of a composite by its double weighing and an electron shadow microscope, this paper presents models and physical mechanisms that lead to extrema in the permittivity and conductivity of composite materials.

1 Introduction

One of the most important tasks of this work is the process of ordered reorientation of the molecules of a structural composite, in which aluminum powder, soot and Nalchikin are present in various concentrations, under the action of an external electric field. As a result of polarization, their electrically charged particles are redistributed along with the molecules. Of the variety of layered silicates that exist in nature, the most promising is montmorillonite clay or Nalchikin, which, under certain conditions, can delaminate into separate plates with a thickness of about 1 nm and a diameter of 20–250 nm. The relevance of the work lies in monitoring changes in the dielectric properties of a structural composite from nanoadditives. In practice and for scientific purposes, a high-strength composite is increasingly being used. The strength of the composite is increased with the introduction of reinforcing components. The behavior of the permittivity for microsystems is usually considered according to the Maxwell-Wagner theory of polarization. Polarization comes from the relaxation of polarization at an interface between two materials with different dielectric properties. It is known [1] that in dielectrics with nanofillers, additives must have known values of conductivities and permittivities.

2 The experimental part

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The aim of this work is to study the dielectric permittivity and resistivity of structural polymer composites based on synthetic isoprene elastomer and low density polyethylene containing carbon black, aluminum, and Nalchikin nanofillers in various amounts.

The amounts of soot, Nalchikin and aluminum were calculated using the formula:

$$
c = 0.1e^n,\tag{1}
$$

where c is the filler content, $wt.\%$. n – integers from 0 to 4 inclusive, e is the base of natural logarithms, equal to approximately 2.71

3 Research results and their discussion

Figure 1 graphically shows how the impurity content affects the dielectric constant. Observing the curve of the dependence of the dielectric constant on the concentration of Nalchikin nanoadditives, we found that with a small amount (0.3% Nalchikin), the dielectric properties of the structural material fall, then, with an increase to 5.5%, they increase to 4.4 (which is higher than the dielectric permeability of the composite without Nalchikin impurities). We continue to observe changes in the dielectric properties of the structural material by changing the impurities. By adding aluminum in an amount of 0.1%, we find a drop in the dielectric constant, and extremes are also observed in the composite with additions of 0.56%; 1.02% and with the addition of aluminum powder up to 5.5%, we observe an increase in dielectric properties up to 4.4. When soot nanoadditives are added to the structural material, the dielectric constant decreases, at a soot concentration of $0.27% = 2.52$, when the pure composite is 4.

Fig. 1. Concentration dependences of the real part of the dielectric constant: $1 - 80\%$ SKI-3 + 20% LDPE + carbon black; 2 - 80% SKI-3 + 20% LDPE + Al2O3; 3 -80% SKI-3 + 20% LDPE + nalchikin.

From Figure 2 (curve 3) it can be seen that with an increase in the soot content to 0.27 wt%, the specific volumetric electrical resistance P увеличивается increases and, with a further increase in the content of soot nanoadditives, leads to saturation. A comparison of the data on the dependencies $p_y = p_y(c)$ for a structural composite of 80% SKI-3+20% LDPE containing Al nanoparticles (Fig.2, curve 1), nalchikin (Fig.2, curve 2) and soot (Fig.2, curve 3) shows that the course of these curves also varies greatly. Such a paradoxical drop

or increase in various physical parameters of the composite under study with small additions of Al, soot and nalchikin nanoparticles can be called a nanoeffect and is observed in the study of the mechanical strength of the composite, in the study of the density of the structural composite and the number of aluminum, nalchikin and soot particles on the surface of the composite. This is due to the effect of small additives, when nanoscale particles, filling the pores and voids of the matrix, lead it to hardening. Based on the above, it is not possible to explain the obtained concentration dependences pv(c) and εˊ(c) for the studied structural composite within the framework of the Maxwell-Wagner polarization model.

Fig. 2. Concentration dependences of resistivity for a structural composite of 80% SKI-3 + 20% LDPE + Al2O3 (curve -1), nalchikin (curve-2), soot (curve-3).

These results, as noted above, cannot be explained either by the Maxwell-Wagner polarization of dielectrics with conductive inclusions, or by the Khokhlov-Grossberg reptation model [1]. In order to explain the experimental data obtained-dielectric parameters and specific electrical conductivity-we conducted studies of the structure and morphology, the change in the density of the structural composite SKI-380% + LDPE 20%, with nano additives of Al, nalchikin and soot by electron microscopy, electron shadow microscope and hydrostatic weighing. These data are presented in Fig. 3 and 4, respectively. The data of elemental scanning of the surface obtained by an electron microscope show (Fig.3a) that aluminum particles with small additives are evenly distributed in the matrix of the structural composite. However, when the aluminum content increases to 5.37 wt%, a certain part of the nanoparticles lines up in the 2nd order, possibly located at the boundaries or delineating the shapes of supramolecular formations in the composite. This arrangement of Al nanoparticles in the structural composite is apparently due to the fact that they occupy the most energetically advantageous positions when mixing the ingredients of the composite in the melt.

Al $K\alpha1$

Fig. 3. Distribution of aluminum on the surface of SKI-3 containing: a) 0.1 wt. %; b) 5,37 wt. % of filler particles

The authors of the work [5] observed a similar distribution of carbon black particles for some heterogeneous polymer mixtures, which led to a superadditive electrical resistance. It was shown in [6] that such a redistribution of filler particles is due to the thermodynamic benefits of this process and in the vast majority of cases, the filler (above 5% by mass) is localized at the phase interface. Unlike inert aluminum (due to the fact that the particles have an oxide film (Al2O3)), soot is quite active and its nanoparticles form agglomerates ranging in size from 100 nm to 10 microns. (Fig. 4).

Fig. 4. An image obtained by an electron shadow microscope for a structural composite of 80 wt. % $SKI-3 + 20$ wt. % LDPE + 0.1 wt. % soot.

However, during the preparation of the composite, the carbon black associates are destroyed to nanometers [7]. The authors of this work have shown that soot agglomerates are crushed into parts under the action of stresses arising during mixing. Another approach is that these agglomerates experience "corrosion", in which small pieces break out of their surfaces[8].

The experimental data presented in Figures 1 (curve 1), Figure 2 (curve 3) for the dependencies $\varepsilon = \varepsilon(c)$ and $p_y = p_y(c)$, as noted above, do not fit into the framework of known models and cannot be explained by structural studies. But we can assume the existence of a special effect - fractal polarization for a structural composite of 80% SKI-3 + 20% LDPE + carbon black. The paper[10] shows the fact that the microscopic properties of the composite depend on the fractal structure. Figures 5 a and b show our proposed structure of fractal conductive structures in a structural composite of 80% SKI-3+20% LDPE+ carbon black.

Fig. 5. The assumed schematic structure of fractal conductive formations in a structural composite of 80% SKI-3+20% LDPE+ carbon black.

a) The percolation threshold has not occurred, contains only inclusions of the type 1.

b) The percolation threshold has occurred, inclusions of the type (2) appear, but inclusions of the type (1) remain.

The schematic structure shown in Figure 5 is a sample of the investigated structural composite-dielectric, which contains fractal clusters-conductive inclusions of carbon black nanoparticles.

It is known that when introducing the concept of dielectric permittivity, the concept of isotropy of a substance [11] and possible small deviations are specified. However, when considering composites, especially those containing conductive inclusions, the isotropy conditions are significantly violated.

4 Conclusions

The dependences of the dielectric permittivity and specific volume resistance of a composite of 80 wt% SKI-3+20 wt% LDPE on the concentration of nanoscale particles of aluminum fillers, nalchikin and soot were experimentally investigated. The features of these curves are considered.

It is shown that at low concentrations of Al, nalchikin and soot nanoparticles in the composite, significant changes are observed - extremes - on the curves of dependence ε = ε (C), P_v= p_v(C), which do not fit into the framework of the Maxwell-Wagner polarization model.

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