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THERMAL CONDUCTIVITY OF SOOT FILLED COMPOSITION MATERIALS

Abstract: This article discusses the thermal conductivity of composite materials. Experimental results of studies of the dependences of the thermal conductivity of compositions based on polystyrene on the concentration of soot and on temperature are presented. The presence of reversible structural rearrangements in polymer soot filled compositions is revealed. The fact that the research of heat conductivity of the received polymeric soot filled compositions shows what various components and dispersion of filler has significant effect on heat conductivity of compositions that is caused by features to structure of education at their formation is given in article.

Key words: Composition materials, composite, polymer materials, polymer matrix, fillers, thermal conductivity, heat capacity, structuration, experimental results.

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Introduction

Theories explaining the thermal conductivity of polymers can be divided into two types. One of them starts from the theory of low state and considers the transfer of energy between repeating links through chemical bonds. In others, the concept of solid state theory is used, in which the collective motion of repeating units and the scattering of phonons that limit the region of the transport energy are considered.

At low temperatures, when oscillations are excited with a wavelength much greater than the distance between repeating links, naturally theories of the second type are used. In the case of amorphous polymers at high temperatures, when the mean free path -1 is of the same order as the distance between repeating units, the application of theories of the second type is practically equivalent to using the theory of low state. The criterion of applicability, the theory of the second type, and the questions of the thermal conductivity of polymers is the condition that the mean free path should be much greater than the distance between the particles.

Methods

The research was made by method of the electronic paramagnetic resonance (EPR) on a radio spectrometer of the E-4 model (Varian the minimum concentration of spin which was available to fixation by this device there was a size of 10¹¹ backs. Improvement of resolution often leads to deterioration by sensitivity to receive a prize in resolution and vice versa. That it was possible to determine the area under an absorption curve amplitude of modulation was selected as follows

$H_m - (2 - 4)H_{PP\max}$

where: $H_{PP \max}$ – the maximum width of a signal depending on field microwave oven power.

Such experimental conditions when neither the standard, nor a sample were sated were chosen and the studied and standard samples were at an identical temperature. Concentration of the paramagnetic centers I was determined by a graphic method integration by amplitudes and to the areas to I paid off by comparison with reference samples (α , $\dot{\alpha}$ the diphenyl, β pycrylgydrazil) (DPPG) on the following formula:



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$$N_{\chi} = \frac{N_a}{m_{\chi}} \frac{S_{\chi}}{S_a} \tag{1}$$

Where: N_a – concentration of spin of a standard (backs/g); m_{χ} – the mass of the studied substance (backs/g); S_{χ} , S_a – the area under a curve of a sample and a standard respectively.

Error of calculation of concentration of the paramagnetic centers makes 20-30%. The size of width of the H_{PP} line paid off directly according to the sheets calibrated by development scale.

Results

The study of the thermal conductivity of the resulting polymeric soot filled compositions shows

that the various components and dispersion of the filler have a significant effect on the thermal conductivity of the compositions, which is due to the peculiarities of the structure of formation during their formation.

Figure 1 shows the dependence of the thermal conductivity χ on v₁ soot filled compositions based on polystyrene (PS). From here it is visible, that with increase in a volume fraction of a filler the thermal conductivity of compositions grows. The most sensitive growth of thermal conductivity for soot filled compositions based on PS is observed at a filling volume of $0.4\div0.5$.



Figure 1. Concentration dependence of the thermal conductivity of polymer compositions based on polystyrene: 1, 2 – calculated by formulas (1), (2), respectively; 3 – experimental data for a soot filled polymer composition

Comparison of theoretical calculations with experimental data makes it possible to obtain an idea of the structure formation of the resulting polymer compositions. According to the Miesner theory for structures with cubic insulated inclusions, the thermal conductivity coefficient has the form:

$$\chi = \chi_2 \cdot \left[\left(1 - \sqrt[3]{v_1^I + \frac{1}{\sqrt{-1}}} \right) + 1 \right]$$
 (1)

Where
$$v = \frac{\chi_2}{\chi_1}$$
; χ , χ_1 , χ_2 – the thermal

conductivity of the filler and the binder, respectively, of the Aiken-Odelevski formula: on the basis of which lies the model of a simple chaotic system with isomer particles that fill the entire volume of material with: $v_1 < v_{\kappa n}$

$$\chi = \chi_2 \left[\frac{\frac{\nu_1^{2/3}}{\nu_1^{-1}}}{1 - (1 - \nu)\nu_1^{\frac{1}{3}}} + \left(1 - \nu_1^{2/3}\right) \right]$$
(2)

and for $v_1 < 0.5$

$$\chi = \chi_2 \left[c^2 + v \left(1 - c^2 \right) + 2vc \left(1 - c \right) \left(vc + 1 - c \right)^{-1} \right]$$
(2a)

where c – the positive root of the cubic equation $2c^3 - 2c^2 + 1 = v_2$.

To explain the experimental dependence of the thermal conductivity of amorphous polymers on temperature, a theoretical analysis of the structural model was carried out [7]. In calculating the intermolecular thermal conductivity, the mean free path of photons can be assumed to be temperature dependent and equal to the average intermolecular distance between two neighbouring molecules, and a potential corresponding to the calculation of the intermolecular interaction is used. The second assumption concerns the local anisotropy of thermal conductivity, which is a consequence of the chain structure of polymers and reduces to postulating the relationship between intra- and intermolecular thermal conductivities, it is assumed that the intramolecular thermal conductivity is much higher



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than the intermolecular one. With these assumptions considered, the thermal conductivity of the structural model (figure 2) [7] was considered, in which the atoms of the main chain form a cubic quasi lattice.

Each such atom is connected by two covalent bonds with neighbouring atoms and four Van der Waals bonds. The direction of covalent bonds in the lattice is static.



Figure 2. A model lattice for calculating the thermal conductivity of amorphous polymers (double lines correspond to covalent chemical bonds, single lines to Van der Waals bonds)

This, in this model, the amorphous polymer is modelled by a grid formed by two types of thermal bridges. Covalent bonds are characterized by high thermal conductivity $-\chi_{\beta}$ (low temperature resistance) and Van der Waals bonds – low thermal conductivity – χ_{ω} (high thermal resistance).

Calculation of the χ_{ω} basis on the Debye equation:

$$\chi = \frac{1}{2} \cdot \rho \cdot C_V \cdot \overline{C} \cdot \overline{l} \tag{3}$$

in which macroscopic quantities are replaced by molecular parameters:

$$C_{V} = \frac{3K}{a^{2}} \cdot b$$

$$\overline{C} = a \cdot \sqrt{\frac{f_{\omega}}{m}}$$
(4)

 f_{ω} – the elastic constants of the Van der Waals bond, m – is the mass of particles, $\bar{l} = a$. From equation (3) has:

$$\chi = K \left(\frac{3K_0}{b}\right) \cdot \sqrt{\frac{f_{\omega}}{m}}$$
(5)

the thermal conductivity of the polymer is proportional to the elementary thermal conductivity χ_{ω} . $\chi = K \cdot \chi_{\omega}$. The final equation will look like:

$$\Delta \left(\frac{1}{\chi} \cdot \frac{dx}{dT}\right) = -5,8\Delta a \tag{6}$$

As is known, the physical reason for the change in the thermal conductivity coefficient at the glass transition temperature is the difference in the coefficients of thermal expansion of amorphous polymers before and after verification.







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Figure 3 shows the course of the temperature dependence of the thermal conductivity of soot filled polymer compositions. As can be seen from the figure, with increasing temperature, the thermal conductivity of the composition increases. In the temperature dependence of the thermal conductivity, for all the samples studied, a jump in χ appears in the region of T_C, which is apparently associated with conformational changes in the structure of polymer compositions. It should be noted that as the concentration of soot increases, the thermal conductivity of the composition increases [8].

In the region $T > T_C$, the growth of $\chi(T)$ appears to be related to the linear increase in C_p(T), and in accordance with the expression:

$$\chi(T) = \frac{1}{3} \int C_V(\omega) \overline{V}_{Si\Delta} \cdot \overline{l}_i(\omega) d\omega \tag{7}$$

Here: $C_{Vi}(\omega)$ – is the heat capacity, \overline{V}_{Si} – is the average sound velocity, $\overline{l}_i(\omega)$ – is the mean free path of the i – phonon.

As can be seen from figure 3, two regions of the temperature coefficient change in $\chi(T)$, which indicates a change in the conditions of heat transfer through the sample. At high temperatures, a decrease of χ is observed for samples with a small filler concentration.

The study of $\chi(T)$ in a constant volume showed (figure 3, dotted lines) that a decrease in thermal conductivity in such a cell at high measurement temperatures is not observed. Apparently, this is due to the thermal expansion of the sample, i.e. with increasing temperature, the free volume increases and the average distance between neighbouring chains increases. As a result, the thermal resistance increases and the thermal conductivity decrease [9].

The obtained experimental data for PS-based compositions can apparently be interpreted, in that scattering of phonons in a more densely packed composition with strong intermolecular interaction occurs at greater lengths and, as a result, energy transfer is intense. The nature of the temperature dependence of the thermal conductivity, namely, the increase in temperature is determined both by the intensity of the thermal motion and by the change in the intermolecular interaction.

As is known, the thermal conductivity of the carbon black is greater than the thermal conductivity of the polymer, moreover, with the addition of the soot micro particles to the polymer composition, the degree of crystallinity of the polymer composition is increasing [10-12].

Therefore, with an increase in the concentration of soot, an increase in the thermal conductivity of the composition in the temperature range 200-300 K is more significant (figure 3).

Conclusion

A study of the concentration dependence of the thermal conductivity on the volume fraction of the filler and temperature shows that the most suitable formula for describing the dependence of $\chi(V)$ is the Eiken-Odelevski formula. In the temperature dependence of $\chi(T)$, a jump occurs at T_c of the composition, which is associated with conformational changes in the polymer.

According to the above, the composites studied in our work are dielectrics with a semiconductor mechanism of conduction. This fact, together with the EPR results of these materials, allows us to consider them as ferrimagnets and thus creates the basis for new developments in the field of magnetic materials such as composite materials in the future.

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