WEB OF SCIENTIST: INTERNATIONAL
SCIENTIFIC RESEARCH JOURNAL
ISSN: 2776-0979 (Volume 2, Issue 5, May, 2021

STUDY OF THERMAL CONDUCTIVITY OF SOFT-FILLED COMPOSITIONS BASED ON POLYSTYRENE AND POLYVINYLDENEFLUORIDE

Kamalova Dilnavoz Ikhtiyorovna
PhD in Technical Sciences, Associate Professor of the Department of Methods of Teaching Physics and Astronomy, Navoi State Pedagogical Institute
e-mail: kamalova.di@mail.ru

Annotation

In this article, a study of the thermal conductivity of soot-filled compositions based on polystyrene and polyvinylidene fluoride. Also presented are the experimental results of studies of the dependences of the thermal conductivity of compositions based on polystyrene on the concentration of soot and on temperature. The presence of reversible structural rearrangements in polymer soot-filled compositions was found.

Keywords: Composite, polymer matrices, fillers, thermal conductivity, heat capacity, structure formation.

Introduction

Theories explaining the thermal conductivity of polymers can be divided into two types. In one of them, they proceed from the theory of the low state and consider the transfer of energy between repeating links through chemical bonds. In others, the concept of the theory of a rigid body is used, in which the collective motion of repeating units and the phenomenon of phonon scattering are considered, which limit the region of transfer energy. At low temperatures, when an oscillation with a wavelength much greater than the distance between the repeating links is excited, theories of the second type are naturally used. In the case of amorphous polymers at high temperatures, when the mean free path is of the same order of magnitude as the distance between repeating units, the application of the second type theories is practically equivalent to the use of the low state theory. The criterion of applicability of the theory of the second type and the issues of thermal conductivity of polymers is the condition that the average free path should be much greater than the distance between particles.

Experiment and Discussion

The study of the thermal conductivity of the obtained polymer soot-filled composites shows that various components and dispersion of the filler have a significant effect on the thermal conductivity of the composites, which is due to the peculiarities of the structure of formation during their formation. Fig.1 shows the dependence of the thermal conductivity coefficient on of soot-filled composites based on polystyrene. Hence it can be seen that with an increase in the volume fraction of the filler, the thermal conductivity of the compositions increases.

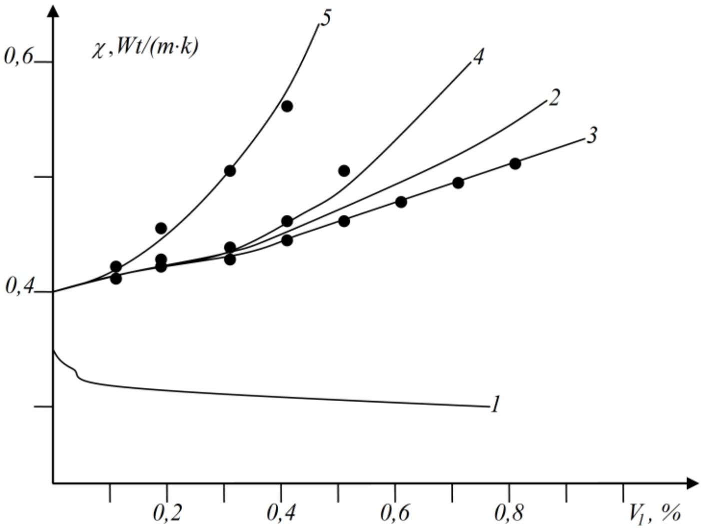

Fig. 1. The concentration dependence of the thermal conductivity of the polymer composition based on polystyrene: 1, 2 – calculated by formulas (1), (2), respectively; 3 – experimental data for a soot-filled polymer composition

The most sensitive increase in thermal conductivity for soot-filled compositions based on PS is observed with a filling volume of 0,4÷0,5. Comparison of theoretical calculations with experimental data makes it possible to get an idea of the structure formation of the obtained polymer compositions. According to Misner's theory, for structures with isolated cubic inclusions, the thermal conductivity coefficient has the form:

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

Where $v = \frac{\chi_2}{\chi_1}$; χ, χ_1, χ_2 – thermal conductivity of the composition, filler and binder, respectively, Formula Aiken-Odelevsky, which is based on the model of a simple chaotic system with isomeric particles filling the entire volume of the material at $v_1 < v_{кр}$ [2]:

$$\chi = \chi_2 \left[\frac{v_1^{2/3}}{1-(1-v)v_1^{\frac{1}{3}}} + \left(1 - v_1^{2/3}\right) \right] \qquad (2)$$

and for $v_1 < 0{,}5$ (3)

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

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

To explain the experimental temperature dependence of the thermal conductivity of amorphous polymers, a theoretical analysis of the structural model was carried out. When calculating the intermolecular thermal conductivity, the value of the average free path of photons can be considered at a temperature-dependent and equal to the average intermolecular distance between two neighboring molecules and the potential can be used to calculate the intermolecular interaction. The second assumption concerns the local anisotropy of thermal conductivity, which is a consequence of the chain structure of polymers and is reduced to postulating the relationship between intramolecular and intermolecular thermal conductivities; it is assumed that intramolecular thermal conductivity is much higher than intermolecular.

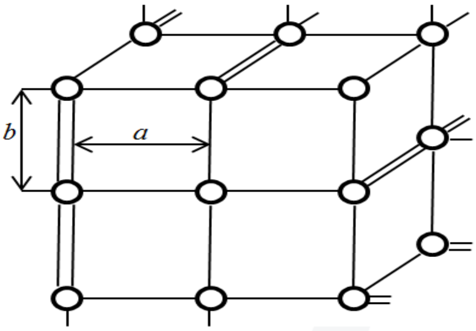

Fig. 2. model lattice for calculating the thermal conductivity of amorphous polymers (double lines correspond to covalent chemical bonds, single lines correspond to van der Waals bonds)

Taking these assumptions into account, the thermal conductivity of the structural model was considered (fig.2), in which the atoms of the main chain form a cubic quasilattice, each such atom is connected by two covalent bonds with neighboring atoms and four van der Waals bonds. The direction of covalent bonds in the lattice is static. Thus, in this model, an amorphous polymer is modeled by a network formed by two types of thermal bridges. Covalent bonds are characterized by high thermal conductivity – χ_β (low thermal resistance), and van der Waals bonds – by low thermal conductivity – χ_ω (high thermal resistance). Calculation of the χ_ω basis on the Debye equation:

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

in which macroscopic quantities are replaced by molecular parameters:

$$\left.\begin{aligned} C_V &= \frac{3K}{a^2}b \\ C &= a\sqrt{\frac{f_\omega}{m}} \end{aligned}\right\} \tag{4}$$

f_ω – Van der Waals coupling elasticity constants, m – particle mass. e=a equation (3) has:

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

The thermal conductivity of the polymer is proportional to the elemental thermal conductivity χ_ω, $\chi = K\cdot\chi_\omega$. The final equation will be:

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

As is known, the physical reason for the change in the temperature coefficient of thermal conductivity at the glass transition temperature is the difference in the coefficients of thermal expansion of amorphous polymers before and after glass transition. Figure 3 shows the course of the temperature dependence of the thermal conductivity of soot-filled polymer composites.

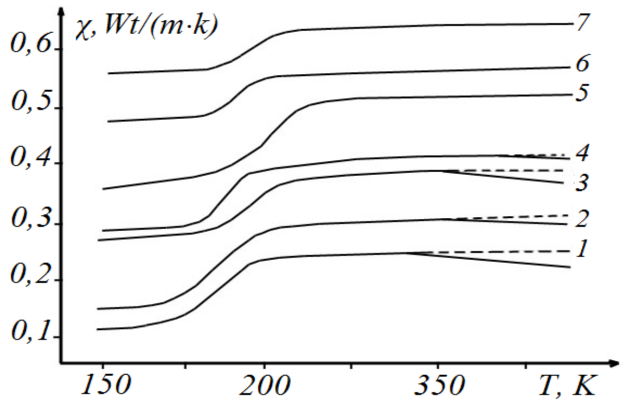

Fig. 3. Temperature dependence of the thermal conductivity of polymer compositions based on polystyrene (PS) with the magnitudes of soot particles: 1 – 0,2; 2 – 0,3; 4 – 0,5; 5 – 0,6.

As can be seen from Figure 3, with increasing temperature, the thermal conductivity of the composition increases. A jump is observed in the temperature dependence of thermal conductivity for all studied samples in the region T_c, apparently associated with conformational changes χ in the structure of polymer composites. It should be noted that with an increase in the concentration of soot, the thermal conductivity of the composition increases.

In the area $T>T_c$, growth seems χ(T) to be associated with a linear increase $C_P(T)$, and in accordance with the expression:

$$\chi(T)=\frac{1}{3}\int C_V(\omega)V_{Si\Delta}\cdot l_i(\omega)d\omega \tag{7}$$

Here: $c_{Vi}(\omega)$ – heat capacity, V_{si} – average speed of sound, $e_i(\omega)$ – mean free path i of phonon. As can be seen from fig.3, two regions of variation of the temperature coefficient χ(T) are distinguished, which indicates a change in the conditions for heat transfer through the sample. At high temperatures, for samples with a low concentration of filler, χ a decline is observed. A study in a constant χ(T) volume showed (fig.3, dashed 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., as the temperature increases, the free volume increases and the average distance between adjacent chains increases. As a result, thermal resistance increases and thermal conductivity decreases.

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

As is known, the thermal conductivity of soot is greater than the thermal conductivity of the polymer; moreover, with the introduction of soot micro particles into the polymer composition, the degree of crystallinity of the polymer composition increases. Therefore, with an increase in the concentration of soot, the increase in the thermal conductivity of the composition in the temperature range of 200-300 K is more significant (fig.3).

Conclusions

Studies of the concentration dependence of thermal conductivity on the volume fraction of the filler and temperature show that the most appropriate formula for describing the dependence is the Aiken-Odelevsky formula χ(V). In the temperature dependence χ(T), a jump is observed at T_c of the composition, which is associated with conformational changes in the polymer.

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