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THERMAL CONDUCTIVITY OF FILLED POLYMER COMPOSITIONS

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An equation for calculating the thermal conductivity of filled polymer compositions is derived and confirmed experimentally.

In what follows, we examine the thermophysical properties of concentrated polymer solutions in hydrocarbon solvents, filled with metallic particles. In view of the high viscosity of a gel, sedimentation proceeds extremely slowly and the composition can be viewed as homogeneous and isotropic. If the volume concentration of the metal is high, the metallic particles form a branching network, encompassing practically the entire volume of the composition. In this case, the dispersed system must be viewed as consisting of two interpenetrating components: the gel and the metal. The properties of such systems are examined in detail in [1].

For relatively small volume concentrations of metallic powder, the composition can be viewed as consisting of a binding medium (gel) with randomly positioned metallic inclusions [1]. The difference between the situations can be easily observed experimentally. In the first case, the coefficient of effective thermal conductivity of the composition depends on the thermal conductivity of the metal. In the second, in view of the fact that the coefficients of thermal conductivity of metals are several orders of magnitude greater than the coefficient of thermal conductivity of a gel, the effective thermal conductivity depends on the nature of the packing, i.e., on the volume content, dispersion, etc., but not on the nature of the metal.

Quite often, the powder particles are covered by an oxide film, whose thermal conductivity in most cases exceeds the thermal conductivity of the gel by 1.5-2 orders of magnitude. As calculations of the thermal conductivity of a dispersed system with coated spherical particles have shown [2], in this case, the screening action of the coating is not great and it can be neglected. In fluidlike polymer compositions, separate particles as well as floccules, consisting of several particles, can form isolated inclusions. This is due to aggregation processes, which occur for sufficiently high specific surface of the powder. Usually, chains consisting of several particles are formed with aggregation.

In a number of works [3-5], the formation of elongated aggregates is explained by the fact that the potential barrier, which must be overcome by the particles in order to connect to an ellipsoid, is lower along the long axis of the ellipsoid than along the short axis. As is well known, with the formation of aggregates, aside from the change in the polydispersity of the inclusions, one other factor appears: the volume fraction of particles in the aggregate is greater than the average volume fraction of the dispersed phase. The effect of this factor on the dielectric permeability of the composition with spherical aggregates was analyzed in [6], where the aggregate was viewed as a system with a higher concentration of the dispersed phase. As shown in [6], this effect is significant only when the volume concentration of the dispersed phase exceeds 25%. For lower concentrations of the dispersed phase, the aggregate can be viewed approximately as a particle with a thermal conductivity of the order of the thermal conductivity of a metal.

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Thus, in order to calculate the effective thermal conductivity of a fluidlike polymer composition with metallic packing, it is necessary to examine a homogeneous system with thermal conductivity λ_b, containing randomly oriented polydisperse inclusions with thermal conductivity λ_d; in addition $\lambda_d \gg \lambda_b$. Calculations of the effective thermal conductivity of such systems for a wide range of volume concentrations (taking into account the possibility for the formation of a continuous network) were carried out in [7-9]. In these calculations, the real random system is replaced by an ordered system that models it, and the effective thermal conductivity of an elementary cell of the model system is calculated. There also exists a statistical approach to calculating the transport coefficients in dispersed systems starting from the properties of the particles and the medium. The theoretical and experimental studies of the effect of polydispersity on the effective dielectric permeability and electrical conductivity of compositions are reviewed in [6], where a comprehensive bibliography on this problem is presented.

In the statistical approach, the effective coefficient of thermal conductivity of the composition is determined from the relations

$$\begin{gathered}\lambda_e \langle \nabla T \rangle = \lambda_d \langle \nabla T_d \rangle + \lambda_b \langle \nabla T_b \rangle, \\ \langle \nabla T \rangle = \langle \nabla T_d \rangle + \langle \nabla T_b \rangle .\end{gathered} \tag{1}$$

Here, T_b and T_d are the temperature fields in the binding medium and in the inclusions; the brackets indicate averaging with respect to volume. The temperature fields T_b and T_d are found from a solution of the stationary heat conduction equations (Δ is the Laplacian operator):

$$\Delta T_d = \Delta T_b = 0 \tag{2}$$

with the conditions that the temperature and the heat flux are continuous on particle surfaces Γ

$$T_b|_\Gamma = T_d|_\Gamma;\ \lambda_b \nabla T_b \cdot \mathbf{n} = \lambda_d \nabla T_d \cdot \mathbf{n}, \tag{3}$$

and with boundary conditions on the external surface of the composition. In order to transform relations (1) into a more convenient form, we will use the following equations for averaging the temperature gradients:

$$\begin{gathered}\langle \nabla T_d \rangle = \frac{1}{V} \sum_i \int_{V_i} \nabla T_d dv = \frac{1}{V} \sum_i \oint_{S_i} T_d d\mathbf{s}, \\ \langle \nabla T_b \rangle = \frac{1}{V} \left[\sum_i \left(\oint_{S_{0_i}} T_b d\mathbf{s} - \oint_{S} T_b d\mathbf{s} \right) + \int_{V - \sum_i V_{0_i}} \nabla T_b dv \right],\end{gathered}$$

where V is the volume of the composition; V_{oi}, volume containing only a single particle i; S_{oi}, surface enclosing this volume; V_i and S_i, volume and surface of the i-th particle. The summation is carried out over all particles. With the help of these expressions, we obtain from (1)-(3)

$$\begin{gathered}\oint_{S_{0_i}} [\mathbf{r}(\nabla T_b \cdot d\mathbf{s}) - T_b d\mathbf{s}] = \frac{\lambda_d - \lambda_b}{\lambda_b} \oint_{S_i} T_d d\mathbf{s}, \\ (\lambda_e - \lambda_b) \left(\int_{V - \sum_i V_{0_i}} \nabla T_b dv + \sum_i \oint_{S_{0_i}} T_b d\mathbf{s} \right) = -\lambda_b \sum_i \oint_{S_{0_i}} [T_b d\mathbf{s} - (\nabla T_b d\mathbf{s}) \cdot \mathbf{r}].\end{gathered} \tag{4}$$

If the volume content of particles is not large, then in calculating the integrals in (4) over the surfaces S_{oi}, sufficiently far removed from the particles, it is possible to consider only the dipole expansion of the temperature field

$$T_b = -\mathbf{G} \cdot \mathbf{r} + \frac{\mathbf{P}_i \cdot \mathbf{r}}{r^3} . \tag{5}$$

Here $\mathbf{P}_i$ is the dipole moment of the i-th inclusion, determined from Eqs. (2) and (3) with the boundary condition

$$\nabla T_b|_{r \to \infty} \to -\mathbf{G}.$$

In view of the linearity of the heat problem

$$\mathbf{P}_i = v_i \sum_{k,n=1}^{3} \alpha_{kn}^{(i)} G_n \mathbf{l}_k, \tag{6}$$

where $\alpha_{kn}^{(i)}$ is the symmetrical particle polarizability tensor [10] and v_i is its volume. Substituting (5) and (6) into (4) and taking into account the random particle orientation, we obtain

$$(\lambda_e - \lambda_b)/(\lambda_e + 2\lambda_b) = \sum_{j=1}^{N} (4\pi/9) I_j C_j, \tag{7}$$

where the summation is carried out over the N types of inclusions; C_j is the volume concentration of the inclusions; $I_j = \sum_{k=1}^{3} \alpha_{kk}^{(j)}$ is the first invariant of the tensor polarizability. As is well known, $I_j > 0$ for $\lambda_d > \lambda_b$ and $I_j < 0$ for $\lambda_d < \lambda_b$. In the case that $\lambda_d \gg \lambda_b$ and $\lambda_d \ll \lambda_b$, the quantity I_j does not depend on λ_d, but is determined only by the shape of the inclusion. For one form of inclusions, expression (7) reduces to a Maxwell type equation

$$(\lambda_e - \lambda_b)/(\lambda_e + 2\lambda_b) = kC, \ k = 4\pi I/9, \tag{8}$$

where the coefficient k takes into account the polydispersity of the particles. Equation (8) is valid also for systems containing inclusions of different shape, but with identical coefficients of thermal conductivity. However, in this case, the I-averaged (over the particles) invariant of the polarizability tensor is $I = \sum_{j=1}^{N} P_j I_j$, where P_j is the fraction of particles with different shapes. For an elliposid, the polarizability tensor is presented in [10]. Calculating its first invariant, we obtain

$$k = \frac{1}{9} \sum_{n=1}^{3} (\lambda_d - \lambda_b)/(\lambda_b + (\lambda_d - \lambda_b) A_n), \tag{9}$$

where A_n is the depolarization factor. For spherical inclusions,

$$A_n = 1/3, \ k = (\lambda_d - \lambda_b)/(\lambda_d + 2\lambda_b)$$

and expression (8) goes over into a well-known Maxwell equation for a medium with spherical particles. For $(\lambda_d/\lambda_b - 1)\min A_n \gg 1$ and $\min(1/A_n - 1) \gg \lambda_d/\lambda_b$, Eq. (9) is simplified:

$$k = \frac{1}{9} \sum_{n=1}^{3} \frac{1}{A_n}, \ \left(\frac{\lambda_d}{\lambda_b} - 1\right) \min A_n \gg 1, \tag{10}$$

$$k = -\frac{1}{9} \sum_{n=1}^{3} \frac{1}{1 - A_n}, \ \min\left(\frac{1}{A_n} - 1\right) \gg \frac{\lambda_d}{\lambda_b}. \tag{10a}$$

For ellipsoidal particles, Eq. (8) with coefficient (9) coincides with a similar formula for the dielectric permeability derived by Stepin [11]. In [11], the mutual effect of particles is taken into account with the help of the Lorentz equation for interaction of pointlike induced dipoles, as was done by Odelevskii [12]. The derivation of Eq. (8), presented above, permits avoiding the contrast method used for calculating dispersed systems with ellipsoidal particles by Pol'der, Van-Santen, and Fricke (these works and the inadequacies of the contrast method are analyzed in [6]). In deriving Eq. (8), it is assumed that the centers of the inclusions are distributed uniformly over the composition and for this reason the form of the correlation function of the mutual positioning of the particles is not taken into account. Methods for calculating the effective characteristics of compositions (viscosity, thermal conductivity, etc.) taking into account the form of the binary distribution function for inclusions (structure formation changes this function) were developed by Yu. A. Buevich and co-workers (in application to thermal conductivity see [13]). As numerical calculations carried out for spherical particles in [14] have shown, the effective thermal conductivity of the composition depends weakly on the form of the binary distribution function. At the same time, for the case that we are interested in, $\lambda_d \gg \lambda_b$ for $C \leqslant 0.15$-0.20, the results of the

TABLE 1. Values of the Coefficients k for Axisymmetric Ellipsoids (b = c)

b/a	A_1	k			
		$\lambda_d/\lambda_c=0$	$\lambda_d/\lambda_c=0{,}1$	$\lambda_d/\lambda_c=10$	$\lambda_d/\lambda_c=\infty$
0,1000	0,0203	—0,5490	—0,4800	1,2153	5,9272
0,1111	0,0238	—0,5480	—0,4589	1,1944	5,1238
0,1250	0,0285	—0,5465	—0,4580	1,1681	4,3561
0,1428	0,0346	—0,5447	—0,4568	1,1368	3,6717
0,1667	0,0432	—0,5422	—0,4552	1,0970	3,0365
0,2000	0,0558	—0,5386	—0,4530	1,0467	2,4619
0,2500	0,0754	—0,5334	—0,4498	0,9833	1,9543
0,3333	0,1087	—0,5256	—0,4448	0,9046	1,5208
0,5000	0,1735	—0,5131	—0,4369	0,8142	1,1782
1,00	0,3333	—0,5000	—0,4285	0,7500	1,0000
1,50	0,4459	—0,5079	—0,4334	0,7720	1,0514
2,00	0,5272	—0,5260	—0,4443	0,8135	1,1508
2,50	0,5881	—0,5496	—0,4579	0,8599	1,2682
3,00	0,6354	—0,5765	—0,4728	0,9062	1,3938
3,50	0,6730	—0,6059	—0,4884	0,9465	1,5119
4,00	0,7036	—0,6358	—0,5034	0,9933	1,6574
4,50	0,7290	—0,6670	—0,5186	1,0334	1,7924
5,00	0,7505	—0,6992	—0,5334	1,0713	1,9301
5,50	0,7688	—0,7318	—0,5478	1,1065	2,0669
6,00	0,7846	—0,7649	—0,5816	1,1396	2,2050
8,00	0,8308	—0,8994	—0,6128	1,2540	2,7636
10,00	0,8608	—1,0370	—0,6572	1,3440	3,3219
20,00	0,9262	—1,7363	—0,8078	1,6085	6,1422

calculations for different distribution functions (even neglecting the impenetrability of inclusions) practically coincide.

In [15], starting from Fricke's equation [16] for the dispersed medium with ellipsoidal inclusions

$$\lambda_e-\lambda_b=\frac{C}{3}\sum_{n=1}^{3}\frac{\lambda_d-\lambda_b}{\lambda_b+(\lambda_d-\lambda_b)A_n}[\lambda_b(1-A_n)+\lambda_e A_n],$$

an empirical equation is proposed for taking into account the polydispersity of inclusions

$$\frac{\lambda_e-\lambda_c}{\lambda_e+(n-1)\lambda_b}=\frac{\lambda_d-\lambda_b}{\lambda_d+(n-1)\lambda_b}C, \tag{11}$$

where n is an empirical coefficient that accounts for the polydispersity of the inclusions. The expression $n = 3/\Psi$, $\Psi \leqslant 1$, is usually used; n = 3 for a sphere. This equation has been quite widely used in the literature on the thermophysical properties of filled polymer media. An equation similar to (11) is also used in the literature on the electrical conductivity of liquid dispersed systems [17]. The value of n (or Ψ) for a given composition is calculated from Eq. (11) from the experimental data. For $\lambda_d >> \lambda_b$ and $\lambda_d << \lambda_b$, it follows from (11) that

$$\frac{\lambda_e-\lambda_b}{\lambda_e+(n-1)\lambda_e}=\begin{cases} C, & \lambda_d\gg\lambda_b,\\ -\dfrac{C}{n-1}, & \lambda_d\ll\lambda_b.\end{cases} \tag{12}$$

Let us compare expressions (8) and (12) with Maxwell's equation for $\lambda_d >> \lambda_b$ and $\lambda_d << \lambda_b$:

$$\frac{\lambda_e-\lambda_b}{\lambda_e+2\lambda_b}=\begin{cases} C, & \lambda_d\gg\lambda_b,\\ -\dfrac{C}{2}, & \lambda_d\ll\lambda_b.\end{cases} \tag{13}$$

As is evident from (8) and (13), polydispersity of inclusions increases the effective volume fraction of polydisperse particles by a factor k for $\lambda_d << \lambda_b$ and by a factor $2|k|$ for $\lambda_d << \lambda_b$, compared with the same volume content of spherical particles. This result has a clear physical interpretation. In contrast to (8), Eqs. (12) relate the increase in the effective thermal conductivity of the composition with polydisperse particles to the thermal

TABLE 2. Results of Analysis of Experimental Data on the Thermal Conductivity of Polymer Compositions

Filler	λ_f, W/m·K	Shape of inclusions	Size, μm	c, %	λ_g, W/m·K; T, K	$\frac{\lambda_e}{\lambda_g}$	k	$A_1^{(*)}$	$b/a^{(*)}$
[21], Copper powder	394,3 [24]	Nonspherical	50 and 15	10,0 20,0	0,08 20	1,750 2,625	1,8052	0,0842 0,7311	3,68 4,55
				10,0 20,0	0,22 3000	1,9545 4,545	2,1256	0,0673 0,7758	4,35 5,71
[22], Copper powder	394,3 [24]	Same	13	13,0 24,5	0,076 20	1,974 2,632	1,5353	0,1071 0,6758	3,03 3,54
[23], Industrial diamond	629 [25]	Nonspherical, arbitrary	0.5— —1.0	4,8 15,2	0,21 300	1,429 2,381	2,0971	0,0686 0,7724	4,29 5,60
[23], Crystalline quartz	10,42 [25]	Irregular	11	10,3 21,4	0,21 300	1,429 1,952	1,1433	0,1522 0,5768	2,25 2,40
[23], Corundum	28,5 [25]	Plates	6,9	8,0 17,0		1,619 2,095	2,0915	0,0688 0,7718	4,29 5,59
			1,8	7,2 11,6 21,3		1,476 1,905 2,857	1,8463	0,0734 0,7535	4,06 5,09
[16], Balsa wood	0,045 [25]	Discs	800× 7200	14,0 25,0	0,22 300	0,756 0,677	0,5182	0,8995	14,3
[16], Aluminum	207 [25]	Parallelepipeds	1600× 1600× 400	15,5		1,986	1,5887	0,1016 0,6886	3,17 3,75
		Cylinders	2700× 270	15,5		2,165	2,3750	0,0584 0,8013	4,85 6,60
			5000× 1000	15,5		1,872	1,4524	0,1171 0,6534	2,82 3,23
Magnesium	165 [25]	Milled	Data in the present work				1,2935	0,1436 0,5989	2,36 2,60
Aluminum powder	207 [25]	Chains					5,2683	0,0232 0,9136	9,17 16,9

Note. Upper values correspond to an elongated ellipsoid and lower values to a compressed ellipsoid.

conductivity with the same content of inclusions, but with the effective thermal conductivity of the medium increased by a factor (n — 1)/2, while for $\lambda_d \ll \lambda_b$, with an additional decrease by a factor (n — 1)/2 of the effective filler volume. These results are physically incorrect.

In the case of randomly oriented inclusions, their shape can be modeled by an effective axisymmetric ellipsoid. Determining the coefficient k in Eq. (8) from experimental data, it is possible to calculate the ratio of the semiaxes of this ellipsoid with the help of expression (9). The depolarization factors for the ellipsoid are calculated from the equations in [10]. Table 1 presents the values of the coefficient k for elongated and compressed elliposids of revolution in two limiting cases: $\lambda_d \gg \lambda_b$ and $\lambda_d \ll \lambda_b$. In [18], it was proposed that the ratio of the semiaxes of the equivalent axisymmetric ellipsoid be determined from the measured dielectric permeability of the dispersed system. A special investigation of the effect of the particle shape on the value of the polarizability coefficients is presented in [17]. The relative increase in the resistance of a parallelepiped filled with an electrolyte with nonconducting axisymmetric particles immersed in it was measured. This permits calculating the quantity α along the particle axis perpendicular the to electrolyte. The experiments carried out showed that α is determined mainly not by the shape, but only by the ratio of the lengths of the particle axes. The results of the measurements agree well with the values of the polarizability coefficients calculated for the corresponding ellipsoids of revolution.

We measured the thermal conductivity of hydrocarbon gels, thickened by polyisobutylene with different metallic powders, using a constant intensity probe method [19]. The experimental technique, taking into account the properties of polymer compositions (high viscosity, poor wettibility, slow structural formation processes, etc.), was perfected for pure gels with different content of polyisobutylene. After this was done, we carried out measurements for filled gels. A relative variant of this method, in which the heating rates of two identical probes placed in the medium being studied and in a comparison-medium [20] are compared, was used. The following powders were used: milled magnesium; two fractions of a chipped form of magnesium and aluminum; four fractions of spherical aluminum and aluminum powder in flaked form.

Analysis of the experimental data leads to the relation

$$(\lambda_f/\lambda_g - 1)/(\lambda_f/\lambda_g + 2) = kC, \tag{14}$$

where λ_f/λ_g is the ratio of the coefficients of thermal conductivity of the filled and pure gel; C is the volume content of the metal. The values of the coefficients for magnesium and aluminum powders are given in Table 2. For the rest of the powders, the particles are nearly spherical and independent of the granulometric composition of the powders, $k \approx 1$.

The experimental data in [16, 21-23], where the effective thermal conductivities of rubbers filled with powders with nonspherically shaped particles were measured, were analyzed with the help of Eq. (8). The values of the thermal conductivities of the filler materials are taken from [24, 25]. The basic experimental data in [16, 21-23] and the values of k computed from them are presented in Table 2. As is evident from this table, for coarsely dispersed powders, $k \approx 2$. Starting from the values of the coefficients k, the parameters of the effective ellipsoid are calculated. The results of the calculations for model fillers [12] show that the ratio of the semiaxes gives the correct information concerning the degree of polydispersity of the particles.

Thus, the experimental investigation carried out confirms the validity of Eq. (8), presented above, for the thermal conductivity of compositions with polydisperse particles for relatively low filler volume concentrations. The ratio of the semiaxes of the effective ellipsoid of revolution calculated from the experimental data gives a good qualitative characterization of the degree of polydispersity.

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THERMAL CONDUCTIVITY OF HYDROCARBONS IN THE NAPHTHENE GROUP UNDER HIGH PRESSURES

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The thermal conductivity of hydrocarbons in the naphthene group has been experimentally determined. An equation is now proposed for calculating the thermal conductivity over the given temperature and pressure ranges.

The thermal conductivity of seven hydrocarbons in the naphthene group (cyclopentane, cyclohexane, ethyl cyclohexane, trans-1,2-dimethyl cyclohexane, cis-1,3-dimethyl cyclohexane, trans-1,4-dimethyl cyclohexane, and cyclohexene) was measured by the method of coaxial cylinders at temperature from 20 to 180°C and under pressures from 0.1 to 150 MPa. The construction of the test equipment and the measuring procedure have already been described [1]. The maximum error of a λ determination was ±1.5%.

The experimentally determined values of the thermal conductivity are given in Table 1.

Only a few published data are available on the thermal conductivity of hydrocarbons in the naphthene group. For cyclopentane, e.g., only one value of λ and the magnitude of its derivative $(\partial\lambda/\partial t)_p$ are given. A comparison of the data in [2] with the results of this study indicates that there the value of the thermal conductivity at 20°C is 2.5% higher and the value of the temperature coefficient at this temperature $\alpha + (1/\lambda_{20})(d\lambda/dt)_p$ is 11.7% lower.

The thermal conductivity of cyclohexane was studied by several authors, the results having been surveyed and analyzed in another report [3]. Its thermal conductivity under atmospheric pressure was studied most thoroughly by Filippov [4] and Mukhamedzyanov [5]. The value of λ_{30} in [4] is 4.7% higher and the value of α is 25% lower than according to the data of this study.

The values in [5] are consistently higher than those obtained in this study: 2.5% at 20°C, 5.1% on the average, and 7.0% maximum. The temperature coefficient α for cyclohexane in [5] is 30% higher.

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