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EFFECTIVE CONDUCTIVITY OF MATRIX COMPOSITES

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The effective-field method is generalized to the problem of the conductivity of microhomogeneous media having a random structure, with allowance for the binary interaction of inclusions. The calculations of the effective conductivity by various methods are compared with experiments on the electrical and moisture conductivity of composites.

Determining the relation between the macroscopic properties of a material and its microscopic structure is a very important problem of physics and mechanics. This pertains to the transport properties of microinhomogeneous media, corresponding to processes of heat and mass transfer, the electric conductivity and permeance, and filtration of a Newtonian liquid in undeformable cracked-porous media [1-4]. The equations describing the steady-state conditions of these processes are mathematically equivalent. If the linear scale of the field of the average motive force of the transfer process in a heterogeneous medium consisting of a homogeneous matrix with randomly distributed inclusions is substantially greater than the characteristic size of the inclusions, it is natural to describe the transfer process within the framework of the continuum approximation. It is then sufficient to use the effective conductivity coefficients (such as the coefficients of thermal conductivity and diffusion, electrical conductivity, dielectric constant, permeance, Darcy's constant, etc.) for the medium as a whole.

Four groups of methods for determining the effective coefficients are known. The first group is that of model treatments, replacing the real stochastic structure of composites by a regular structure [5] or some particular cases of random structures [6]; percolation models, in particular, belong to this group [7]. The perturbation method [3, 4] gives correct results when the differences in the conduction coefficients of the ocmponents of a composite are small. The variational method [8, 9] is invariant under randomly oriented inclusions and gives too wide a spread of estimates of the effective properties for highly inhomogeneous materials. The fourth group consists of methods based on expressing the solution of the steady-state transfer equation with random rapidly oscillating coefficients in terms of the Green's function of the analogous equation for a homogeneous medium. Depending

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on the numerous ways of closing the integral equations obtained, the methods in this case are classified as the method of the effective medium [10-12], the singular approximation method [4], the method of arbitrary moments [13], and the Mori–Tanaka–Eshelby method of the average field [14-16]. These are virtually one-particle methods, in contrast to the perturbation [4] and multiparticle methods [17], which are correct only for small differences between the properties of the components and low inclusion densities. Below we develop an effective field method, which makes allowance for the multiparticle interaction of inclusions and was proposed earlier for solving a broad class of problems of microinhomogeneous media: elasticity, thermoelasticity, ideal plasticity, and viscosity of suspensions [18-21]. The method is based on the solution of problems for one inclusion and a finite number of inclusions in an unbounded matrix and a self-consistent procedure for closing the corresponding equations averaged over the ensemble of inclusions.

1. Let us consider an infinite homogeneous matrix with conductivity tensor k_0, which contains the random set $X = (V_k, x_k, \omega_k)$ of ellipsoids v_k with characteristic functions V_k and centers x_k forming a Poisson set, semiaxes $a_k^i (a_k^1 \geqslant a_k^2 \geqslant a_k^3)$, a set of Euler angles $\omega_k{}^i$, and conductivity tensors $k_0+k_1(x)=k_0+k_1^{(k)}(x)$ for $x \in v_k$; generally speaking, the tensor $k_1{}^{(k)}(x)$ is not uniform over the volume of the inclusion and $k_1{}^{(k)} = 0$ for $x|v_k$. The steady-state transfer equation (Fourier, Ficks, Ohm, Darcy, etc.) in a microinhomogeneous medium has the form

$$\nabla k_0 \nabla u = -\nabla k_1 \nabla u, \tag{1}$$

where ∇ is the gradient operator; u is a potential (e.g., temperature); a uniform field $\varepsilon(\partial w) = \varepsilon_0$, $\varepsilon \equiv \nabla u$ is prescribed at the boundary ∂w of region w containing a statistically large number of inclusions. Here and below we use the standard notion of tensor analysis: k_0 and k_1 are tensors of the second rank $k_0 = k_{0i}\delta_{ij}$, $k_1 = k_{1i}\delta_{ij}$) (no summation over i); the components of isotropic tensors are labeled with an upper index 0, e.g., $k_0{}^0 = k_{0i}$ (i = 1, 2, 3). The solution of (1) can be expressed in terms of the convolution of the fundamental solution G with a nonzero right side. Then differentiating and centering the resulting integral equation with the assumption that the distance from x to the boundaries of the region is substantially larger than the characteristic size of the inclusions, we obtain

$$\varepsilon(x) = \langle \varepsilon \rangle + \int U(x-y)(k_1(y)\,\varepsilon(y)\,V(y) - \langle k_1 \varepsilon V \rangle)\,dy, \tag{2}$$

where $V(y) = \sum_{i=1} V_i(y)$, $U(x-y) \equiv \nabla\nabla G(x-y)$; for an isotropic medium $G(x-y) = (4\pi k_0{}^0|x-y|)^{-1}$. In (2) and below $\langle\cdot\rangle$ and $\langle\cdot|x_1\rangle$ denote the mean and arbitrary mean over the ensemble of the ergodic field $X(\cdot|x_1)$ (or over the macroregion w) with the condition that an inclusion exists at point x_1. The arbitrary means are calculated by using a binary function of the distribution $\varphi(v_m|v_n)$ is the probability of the m-th inclusion being located in region v_m when the n-th inclusion is fixed. Since the inclusions do not intersect each other, $\varphi(v_m|v_n) \equiv 0$ inside the "correlation well" and $v_{mn}{}^0$ is an ellipsoid with orientation ω_m and axes $a_m^i + a_n^3$ and characteristic function $V_{mn}{}^0$. When the inclusions are uniformly distributed we have

$$\varphi(v_m|v_n) = \psi(\omega_m)(1 - V_{mn}^0)\, f_{mn}(|r|)(\operatorname{mes} w)^{-1}, \tag{3}$$

where from the normalization condition $\langle\psi(\omega_m)\rangle = 1$. The results of the Kirkwood, hypervalued, Percus-Yerwick, and other approximations [22] can be used directly for macroisotropic structures once the fucntions $f_{mn}(|r|)$ have been found; $f_{mn}(|r|)$ is assumed to be known within the framework of this study. Use is often made of a simpler approximation (which will be used here to obtain an analytical result)

$$f_{mn}(|r|) = n_\nu \text{ for } v_m \in X_\nu, \tag{4}$$

where n_ν is the calculated concentration of inclusions ν of component X_ν and is related to the bulk concentration by $c_\nu = 4/3\, a_\nu^1 a_\nu^2 a_\nu^3 n_\nu$.

To find the effective conductivity of the medium we average the local $F = -k\nabla u$, relating the flux and the potential gradient, over the region w: $\langle F \rangle = k_0\langle\nabla u\rangle + \langle k_1 \nabla u V\rangle$. Then

$$k_* = k_0 + \langle R_* \rangle, \quad \langle k_1 \nabla u V \rangle \equiv R_* \langle \nabla u \rangle. \tag{5}$$

Thus, in order to determine k_* we must find the tensor R_* from (2) and (5).

We fix an arbitrary realization of the field X and consider the field $\bar{\varepsilon}_k(x)$, $x \in v_k$ (henceforth called the effective field), in which the inclusion v_k is lcoated:

$$\bar{\varepsilon}_k(x) = \langle \varepsilon \rangle + \int U(x-y)\{V(y;\ x)\, k_1(y)\, \varepsilon(y) - \langle k_1 \varepsilon V \rangle\}\, dy, \tag{6}$$

where $V(y;\ x) = V(y) \backslash V_k(x)$.

2. To close the system (6) and then find its approximation solution we adopt the hypothesies of the effective-field methods [18-21]: 1) every ellipsoidal inclusion v is in a uniform field $\overline{\varepsilon_i}$; 2) each pair of inclusions v_i, v_j is in its own uniform field $\hat{\varepsilon}_{ij}$, which does not depend on the properties of these inclusions. The physical meaning of hypotheses 1 and 2 is explained in [18, 20].

For the ellipsoidal shape of the inclusion v we obtain from (2) and (6) the algebraic equation

$$\langle \varepsilon(x) \rangle_i - \langle U(x) \rangle_i \langle k_1(x)\, \varepsilon(x)\, V_i(x) \rangle_i = \langle \bar{\varepsilon}_i \rangle, \tag{7}$$

where we have adopted the notation $\langle(\cdot)\rangle_i = \int(\cdot)(x)V_i(x)dx$ and used the Eshelby theorem $\langle U(x)\rangle_i = \int U(x-y)V_i(y)dy = -P_i \equiv -Sk_0^{-1} = \text{const}$ for $x \in v_i$. The tensor S of rank two is analogous to the Eshelby tensor in the theory of elasticity and depends on the shape but not on the ellisoid size v. For example, for an isotropic matrix

$$S_{ii} = \frac{a^1 a^2 a^3}{2} \int_0^\infty \frac{ds}{(a^i + s)^2 \Delta(s)}, \quad \Delta(s) = \sqrt{((a^1)^2 + s)((a^2)^2 + s)((a^3)^2 + s)} \tag{8}$$

(no summation over i), $S_{ij} = 0$ at $i \neq j$. Analytical expressions for the tensor S for a sphere, elliptical cylinder, and oblate and prolate ellipsoids are given in [12, 14]; in particular, for a sphere $S_{11} = S_{22} = S_{33} = 1/3$.

The dependence $\langle k_1(x)\varepsilon(x)V_i(x)\rangle_i$ on $\langle\overline{\varepsilon_i}\rangle$ must be known in order to estimate the effective tensor in (5). Since problem (7) is linear, there exists a constant tensor A of rank two such that

$$\langle \varepsilon(x) \rangle_i = A_i \langle \bar{\varepsilon}_i \rangle, \tag{9}$$

$$\bar{v}_i \langle k_1(x)\, \varepsilon(x) \rangle_i = R_i \langle \bar{\varepsilon}_i \rangle, \tag{10}$$

where $R_i = -P_i(A_i - I)\bar{v}_i$, I is a unit tensor of rank two and the bar above the region denotes its measure: $\bar{v}_i = \text{mes}\, v_i$. When deriving (10) we assumed that $k_1(x)$, generally speaking, is not uniform over the volume of the inclusion; for a homogeneous inclusion $k_1(x) = k_1^{(i)} = \text{const}$ and from (7) we have

$$A_i = (I + P_i k_1^{(i)})^{-1}, \quad R_i = k_1^{(i)} A_i \bar{v}_i \tag{11}$$

(no summation over i). The value of A_i can be found numerically in the general case of an inhomogeneous inclusion. Analytical solutions of Eq. (1) are known at present for a two-layer inclusion consisting of isotropic ellipsoids in an isotropic matrix. In [15] the problem for a two-layer spheroid was solved in spheroidal coordinates, using Legendre polynomials. The problem of a layered ellipsoid in a uniform coordinate-linear field $\bar{\varepsilon}(x)$ was solved by using the Ferrers potential [23]. As an example let us consider the limiting case of the results of [15, 23], the existence of a resistance on the surface of a homogeneous ellipsoid, i.e., when the boundary conditions on the surface of the inclusion have the form

$$k_0 \frac{\partial u^-}{\partial n} = (k_0 + k_1) \frac{\partial u^+}{\partial n} = \beta (u^- - u^+), \tag{12}$$

where n is the outer normal of the ellipsoid; u^+ and u^- are the limiting values of the potential inside and outside the ellipsoid, near the surface of the inclusion. The case $\beta \to 0$ corresponds to "adiabatic" boundary conditions and $\beta \to \infty$ corresponds to "isothermal" boundary conditions. The boundary condition (12) is mathematically equivalent to the surface of a homogeneous ellipsoid having an infinitesimally thin layer with thickness $l \ll a^3$ and conduction coefficient $k = \beta/l$. Transforming the results of [24] for a spherical inclusion, we obtain

$$R_i^0 = 3k_0^0\,[k_1^0 + (k_1^0 + k_0^0)\,k_0^0/\beta a][k_1^0 + 3k_0^0 + (2k_1^0 + k_0^0)\,k_0^0/\beta a]^{-1}. \tag{13}$$

The error that arises when the layer of finite thickness on the surface of the inclusion is replaced by the boundary condition (12) was evaluated in [15].

When estimating k_* below we use the averaged tensor R_i over the possible orientations ω_i of the inclusion: $\langle R_i\rangle_\omega$. In particular, for an infinitesimal concentration of identical inclusions we have

$$k_* = k_0 + \langle R \rangle_\omega n \tag{14}$$

and $\langle R\rangle_\omega n$ has the connotation of the main term in the power expansion of k_* in the inclusion concentration $\langle V\rangle$. Formula (14) is the limiting formula for the effective medium [10-12], Mori–Tanaka–Eshelby [14-16], and other [25] methods. For example, for ideally conducting ($k_1{}^0 = \infty$) and nonconducting ($k_1{}^0 = -k_1{}^0$) spheroids with an equiprobable orientation in an isotropic matrix we obtain

$$\langle R \rangle_\omega^0 = k_0^0\,\frac{S_{11} + 1/3}{S_{11}(1 - S_{11})}\bar{v},\quad \langle R \rangle_\omega^0 = k_0^0\,\frac{5 - 3S_{11}}{1 - S_{11}}\bar{v}, \tag{15}$$

where the I axis of the local coordinate system, made to coincide with the major axes of the ellipsoid, is the symmetry axis of the spheroid.

3. The solution of the problem of two inclusions in an unbounded matrix is simplified by using hypotheses 1 and 2. Averaging Eq. (6) over the inclusion volume v_i we obtain

$$\langle \bar{\varepsilon}_i \rangle - \bar{v}_i^{-1} \iint U(x - y)\,V_j(y)\,V_i(x)\,\langle k_1(y)\,\varepsilon(y)|x \rangle_j\,dxdy = \langle \hat{\varepsilon}_{ij} \rangle . \tag{16}$$

Using the solution (10) for a single inclusion in a field $\langle\overline{\varepsilon_j}\rangle$, we rewrite (16) as

$$\langle \bar{\varepsilon}_i \rangle = T(x_i - x_j)\,R_j\,\langle \bar{\varepsilon}_j \rangle + \langle \hat{\varepsilon}_{ij} \rangle ,$$
$$T(x_i - x_j) = (\bar{v}_i\bar{v}_j)^{-1} \iint U(x - y)\,V_i(x)\,V_j(y)\,dxdy, \tag{17}$$

whereupon

$$R_i \langle \bar{\varepsilon}_i \rangle = \sum_{j=1}^{2} Z_{ij} R_j \langle \hat{\varepsilon}_{ij} \rangle ,$$

where the matrix $Z = (Z_{ij})$ (i, j = 1, 2) has the inverse Z^{-1} with elements $Z_{mk}{}^{-1}$ (m, k = 1, 2) in the form of submatrices

$$Z_{mk}^{-1} = I\delta_{mk} - (1 - \delta_{mk})\,T(x_m - x_k)\,R_k\ \ (m,\ k = 1,\ 2).$$

The solution (17) can also be constructed by iteration methods

$$R_i \langle \bar{\varepsilon}_i \rangle = \sum_{k=0}^{\infty} \sum_{m=0}^{1} (TR_jTR_i)^k (TR_j)^m \langle \hat{\varepsilon}_{ij} \rangle . \tag{18}$$

As an approximation of the tensor T we can use the point approximation of the inclusion size, which is asymptotically exact when the inclusions are infinitely apart:

$$T(x_i - x_j) = U(x_i - x_j). \tag{19}$$

Equation (19) is exact for spherical inclusions with possibly different radii in an isotropic medium.

An estimate by means of the expression

$$L_{ij} = \int T(x_i - x_j)\,\varphi(v_j|v_i) \sum_{j=1}^{2} Z_{ij} R_j dx_j \tag{20}$$

will be needed below. From (18) and (20) we obtain a representation for the components of the isotropic tensor L_{ij} as a rapidly divergent series

$$L_{ij} = c_j \sum_{p=1}^{\infty} \sum_{q=0}^{1} \frac{8\pi\,(\langle R_j \rangle_\omega \langle R_i \rangle_\omega)^p\,(\langle R_j \rangle_\omega)^q\,L_{2p+q}}{9\,(4\pi k_0)^{2p+q}\,\bar{v}_j^{p+q+1}\,\bar{v}_i^p\,(2p+q)} \int_{a_1^1+a_2^3}^{\infty} \frac{r^2\varphi_j(r)\,dr}{r^{3(2p+q)}n_j} \tag{21}$$

where the coefficients L_n are determined from the recurrence formulas $L_n = 2L_{n-1} + 3(-1)^{n-1}$, $L_1 = 3$. The value of L_{ij} can be determined by an equivalent method from Eqs. (16), (17), and (20):

$$\langle \varepsilon \rangle_i - \sum_j L_{ij} \langle \hat{\varepsilon}_{ij} \rangle = \langle \varepsilon \rangle - P_i(\langle k_1\varepsilon \rangle_i - \langle k_1\varepsilon \rangle). \tag{22}$$

In [17] the problem of two identical spheres in a uniform field $\hat{\varepsilon}_{ij}$ prescribed at infinity was solved exactly only by using bispherical coordinates and Legendre polynomials; the solution was obtained in the form of a slowly convergent series, whose first terms for L_{ij} (22) have the form

$$L_{ij}^0 = c\left(\frac{\gamma^2}{4} + \frac{3}{16}\gamma^2\frac{k_1^0 + k_0^0}{2k_1^0 + 3k_0^0} + \frac{\gamma^3}{64} + \ldots\right),$$

$$\gamma \equiv \frac{R^0}{k_0^0 v} = \frac{k_0^1}{k_0^0 + 3k_1^0}. \tag{23}$$

The graphs of $(L_{ij}{}^0/c) \sim \log(k_1{}^0 + k_0{}^0)/k_0{}^0$, calculated from the approximate (21) and exact (23) formulas are shown in Fig. 1. Formula (21) can be assumed to be satisfactorily accurate. The problem for two spherical isotropic inclusions with different parameters and properties was solved in [26].

4. The use of hypotheses 1 and 2 and the solutions (10) and (17) of the problems for one and two inclusions makes it possible to transform Eq. (6), averaged over the ensemble $X(\cdot|x_1)$, into a system of linear algebraic equations in $\langle\bar{\varepsilon}_i\rangle$ (for details see [18-20]):

$$\langle \bar{\varepsilon}_i \rangle = \langle \varepsilon \rangle + \sum_{\nu=1}^{N} P(V_{i\nu}^0) R_\nu n_\nu \langle \bar{\varepsilon}_\nu \rangle + \sum_{\nu=1}^{N} \int T(x_i - x_\nu) R_\nu (Z_{\nu\nu} - I)(1 - V_{i\nu}^0) n_\nu dx_\nu \langle \bar{\varepsilon}_\nu \rangle + \tag{24}$$
$$+ \sum_{\nu=1}^{N} \int T(x_i - x_\nu) R_\nu Z_{\nu i} n_\nu (1 - V_{i\nu}^0) dx_\nu \langle \bar{\varepsilon}_i \rangle,$$

where we have assumed that $\hat{\varepsilon}_{ij}(x) = \bar{\varepsilon}_j$ at $x \in v_i$, N is the number of components in the filler. From (24) we find

$$\langle \bar{\varepsilon}_i \rangle = D_i \langle \varepsilon \rangle, \quad D_i = \sum_{j=1}^{N} Y_{ij}, \quad i = 1, \ldots, N, \tag{25}$$

where matrix Y with elements in the form of submatrices Y_{ij} (i, j = 1, ..., N) has the inverse $Y^{-1} = (Y^{-1})_{mk}$(m, k = 1, .., N) with element-submatrices

$$Y_{mk}^{-1} = \delta_{mk}\left(I - \sum_{\nu=1}^{N} \int T(x_m - x_\nu) R_\nu Z_{\nu m} n_\nu (1 - V_{m\nu}^0) dx_\nu\right) - P(V_{mk}^0) R_k n_k - \tag{26}$$
$$- \int T(x_m - x_k) R_k [Z_{mk} - I] n_k (1 - V_{mk}^0) dx_k.$$

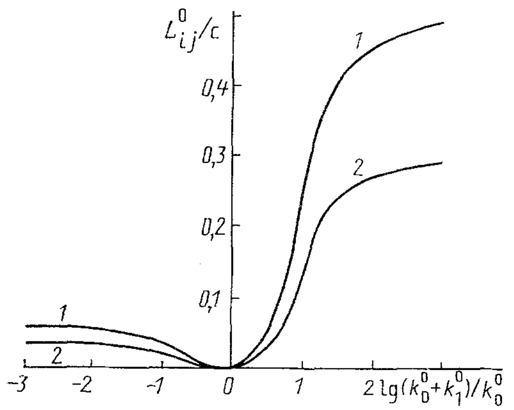

Fig. 1. Results of calculation from formulas (23) (curve 1) and 21 (curve 2) in dimensionless coordinates.

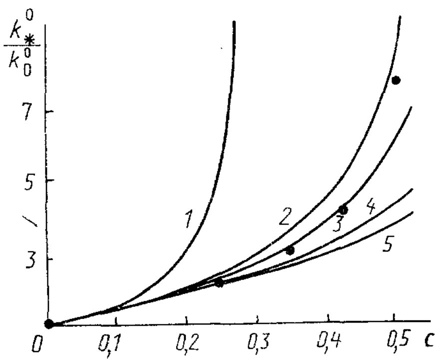

Fig. 2

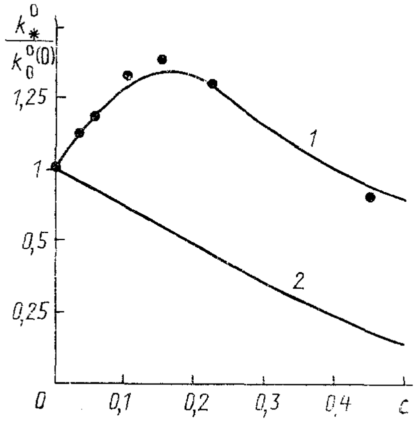

Fig. 3

Fig. 2. Experimental data (points) and calculated curves of the relative changes in the electrical conductivity: 1, 2, 3, 4, 5) calculation from formulas of [12] and formulas (30), (29), (31), and the formula of [13].

Fig. 3. Experimental data and calculated curves of the relative change in the moisture diffusion coefficient in a composite: curves 1 and 2 were calculated for constant and variable moisture diffusion coefficients in the matrix.

We simplify the operations of averaging over the orientations ω in (18) and (26) by replacing the tensors with their means $\langle R_j \rangle_\omega$. Expressions (25) are simplified for a one-component filler:

$$\langle \bar{\varepsilon}_1 \rangle = D_1 \langle \varepsilon \rangle, \quad D_1 = (I - P(V_{11}^0) \langle R_1 n_1 \rangle - L_{11})^{-1}. \tag{27}$$

From (5) and (25) we find an expression for the effective conductivity

$$k_* = k_0 + \sum_{i=1}^{N} \langle R_i n_i \rangle D_i. \tag{28}$$

We compare the experimental data of [27] and various methods of calculation of k_* on the example of the electrical conductivity of a composite with an isotropic matrix and an ideally conducting spherical inclusion of one size. Then from formulas (23), (27), (28) and (21), (27), (28) we find the relative change in the components of the isotropic tensors,

$$k_*^0/k_0^0 = 1 + 3c/(1 - 1{,}304c), \tag{29}$$

$$k_*^0/k_0^0 = 1 + 3c/(1 - 1{,}503c); \tag{30}$$

in similar fashion formula

$$k_*^0/k_0^0 = 1 + 3c/(1 - c) \tag{31}$$

was obtained by the Mori–Tanaka–Eshelby [14, 16] and effective medium [11] methods and accord with the lower boundary of the Hashin–Strickman form [4]; the difference between the last formulas and (29), (30) is attributed to the disregard of the binary interaction of inclusions and inclusion of the term L_{11} in (27). Estimates were also obtained by the method of [17], taking the binary interaction of inclusions into account, on the assumption that $\bar{\varepsilon} = \varepsilon_0$, $k_*^0/k_0^0 = 1 + 3c + 4.51\, c^2$; according to the method of arbitrary moments $k_*^0/k_0^0 = 1 + 3c(1 - c/3)/(1 - c)$ [13] the formula $k_*^0/k_0^0 = 1/(1 - 3c)$ [12] accords with the formula of Landauer [2]. We see from Fig. 2 that calculation from the proposed relations (29), (30) agree better with the experimental data [27] than does calculation from the formulas of [2, 4, 11, 12, 14, 7].

The above theory of the estimation of effective properties makes it possible to obtain estimates of the changes in the properties and structure of the components during the production of a composite. For example, Fig. 3 shows experimental data on the effective moisture diffusion coefficient at 60°C in a composite medium. The composite, based on ÉD-20 epoxy resin and a filler [KP-3 ground pulverized quartz, with ($a = (5\text{-}10)\cdot 10^{-5}$ m)], was hardened with polyethylene polyamine at room temperature. For the studies we prepared samples in the form of wafers with a thickness of $3\cdot 10^{-3}$ m by hardening the compositions in polyethylene molds. The diffusion coefficients were determined by the method of steady-state permeability. Curve 1, $k_*^0(c)/k_0^0$ calculated from formulas (23), (27) and (28) with $k_1^0 = -k_0^0$, for spherical inclusions of one size is not in agreement with the experimental data; the explanation for this is that the matrix is cross-linked to a lesser degree when the filler is introduced. This has been determined experimentally from electron-microscopic data. We then assume that the diffusion coefficient of the matrix increases with the surface area of the filler according to the formula $k_0^0(c)/k_0^0(0) = (1 - k_0^\infty)\exp(-\alpha c) + k_0^\infty$. The empirical parameters $\alpha = 5.1$ and $k_0^\infty = 2.2$ are found from the condition of the best rms fit of the experimental to the calculated curve 2 of the relative change in the diffusion coefficient, $k_*^0(c)/k_0^0(0)$.

NOTATION

k_0, conductivity tensor; $k_1^{(k)}$, jump in the conductivity tensor; u, potential; V_k, characteristic fucntion; v_k, ellipsoidal region; $\varepsilon = \nabla u$, potential gradient; $\langle(\cdot)\rangle$, operation of averaging over the ensemble of inclusions; G, fundamental solution; $U = \nabla\nabla G$; $\varphi(v_m|v_n)$, arbitrary inclusion distribution density; ψ, density of orientational distribution of inclusions; φ_{mn}, numerical concentration n of inclusions at a fixed inclusion m; c_ν, bulk concentration of inclusions of component ν; k_*, effective conductivity; A_i, potential conductivity tensor at i-th inclusion; and Z, Y, D, matrices describing the inclusion interaction.

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