NOTATION

P, radiant flux; q, thermal flux; s, surface area; α, heat liberation coefficient; i, number of radiometer zone; T_i, temperature of i-th zone; T_0, temperature of surrounding medium; r and R, radii of inner and outer surfaces of radiometer; N, number of zones; U and E, thermo-emf of thermocouple during flux measurement and flux substitution; k, thermoelectric coefficient of thermocouple; M, electrical power of substitute heater; β, entrance orifice half-angle; ε, emissivity of cavity material; θ, angular coordinate in spherical coordinate system; $x = \cos\theta$, θ_0, and x_0 correspond to boundary of area irradiated by flux; $L_m(x)$, Legendre polynomial of first sort and m-th order; $W_m(x_0) = \int_{-1}^{1} L_m(x)\,dx$; λ, thermal conductivity coefficient of radiometer material; θ°, latitude at which curve $T(\theta)$ passes through zero; δ, cavity wall thickness; Q, thermal flux; μ and η, relative and absolute error in flux measurement; C, proportionality coefficient; σ, Stefan–Boltzmann constant.

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CONDUCTIVITY OF NONUNIFORM SYSTEMS

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UDC 536.24

Percolation theory and methods of generalized conduction theory are used to construct a model of a heterogeneous system and to determine the effective conductivity.

Statement of the Problem. We consider a very simple two-component heterogeneous system with random distribution of components, consisting of two kinds of identical isomeric particles, occupying the entire system volume without voids (Fig. 1a).

We need to find the effective conductivity Λ of the system (the thermal and electrical conductivity, the dielectric constant, the magnetic permeability, the diffusion, etc.) as a function of the conductivities Λ_i and the volume concentrations m_i of the i-th component when the latter do not interact, i.e., the quantity Λ_i does not depend on the concentration m_i.

The effective conductivity Λ of this system is determined from the equation

$$\langle \mathbf{j} \rangle = -\Lambda \langle \nabla\varphi \rangle, \tag{1}$$

where $\langle \mathbf{j} \rangle$ is the average flux over the volume V (heat, electricity, material, etc.) and $\langle \nabla\varphi \rangle$ is the average volume gradient of the potential due to the flux $\langle \mathbf{j} \rangle$:

$$\langle \mathbf{j} \rangle = \frac{1}{V}\int_V \mathbf{j}(\mathbf{r})\,dV, \quad \langle \nabla\varphi \rangle = \frac{1}{V}\int_V \nabla\varphi(\mathbf{r})\,dV. \tag{2}$$

Here for the local fluxes $\mathbf{j}(\mathbf{r})$ and the potential gradients $\nabla\varphi(\mathbf{r})$ we have the equations

$$\begin{gathered} \mathbf{j}(\mathbf{r}) = -\Lambda(\mathbf{r})\nabla\varphi(\mathbf{r}), \\ \operatorname{div}\mathbf{j}(\mathbf{r}) = 0, \\ \operatorname{curl}\nabla\varphi(\mathbf{r}) = 0. \end{gathered} \tag{3}$$

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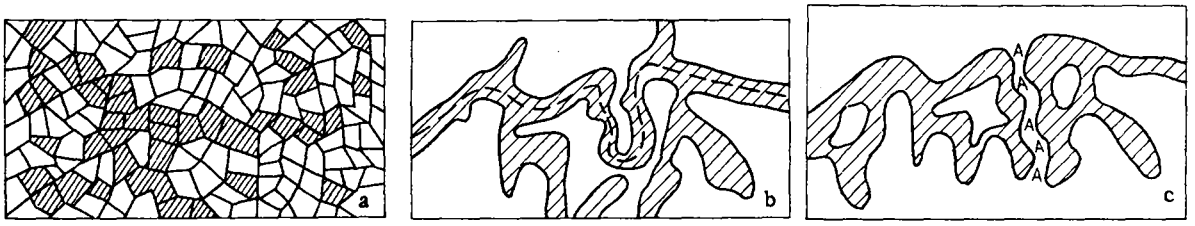

Fig. 1. Structure of the heterogeneous systems: a) mixture of two kinds of polyhedrons: b) planar representation of an infinite cluster near the percolation threshold $m_m \to m_c + 0$ (the shaded area); c) isolated cluster for $m_m < m_c$. The letter A denotes fine "conducting" seams of dielectric [10].

The expression for the average flux $\langle j \rangle$ and the potential gradient $\langle \nabla\varphi \rangle$ in a two-component system, according to Eqs. (2) and (3), can be written in the form

$$\langle j \rangle = -m_1\Lambda_m \langle \nabla\varphi_1 \rangle - m_2\Lambda_d \langle \nabla\varphi_2 \rangle, \tag{4}$$

$$\langle \nabla\varphi \rangle = m_1 \langle \nabla\varphi_1 \rangle + m_2 \langle \nabla\varphi_2 \rangle, \tag{5}$$

where

$$\langle \nabla\varphi_1 \rangle = \frac{1}{V_1}\int_{V_1} \nabla\varphi_1(\mathbf{r})\,dV_1; \quad \langle \nabla\varphi_2 \rangle = \frac{1}{V_2}\int_{V_2} \nabla\varphi_2(\mathbf{r})\,dV_2.$$

Here V_1, V_2, and $V = V_1 + V_2$ are the volumes of the first and second components and the entire system, respectively. Expressions (4) and (5), taking account of Eq. (1), can be rewritten in the form

$$N = m_1\Psi_1 + \nu m_2\Psi_2, \; N = \Lambda/\Lambda_m, \; \nu = \Lambda_d/\Lambda_m, \tag{6}$$

$$m_1\Psi_1 + m_2\Psi_2 = 1, \; \langle \nabla\varphi_i \rangle = \Psi_i \langle \nabla\varphi \rangle, \tag{7}$$

where Λ_m is the conductivity of the first component and Λ_d is the conductivity of the second component.

It can be seen from Eqs. (6) and (7) that to determine N we need additional information since the two equations contain three unknowns N, Ψ_1, Ψ_2. This information can be information on the structure (or the topology) of the heterogeneous system, which would allow us to close Eqs. (6) and (7). Usually the components are distributed either in an ordered fashion (the presence of a distant row) or randomly, in the model of the structures of the heterogeneous system. The first method is self-evident and allows us to obtain very exact results even when we replace the actual random position of the components by an ordered system [1]. However, it does not describe the probabilities of forming continuous bridges of conducting components, distributed in the nonconducting components. In addition, the diversity in structure leads to the appearance of a large number of formulas for N, which in certain situations gives rise to an indeterminacy in their choice.

The determination of conductivity in heterogeneous systems with a random distribution of components in a rigorous formulation of the problem encounters insurmountable mathematical difficulties, and one must use difference approximation schemes. A review of methods of this type and their capabilities has been given in [2].

Neither the ordered nor the random models can explain the appearance of a discontinuity in conductivity in poorly conducting systems $\Lambda_d = 0$ (d indicates dielectric), in which the good conducting components Λ_m are randomly distributed (m indicates metal). It has been established that in the conductivity of the components $\Lambda_m \neq 0$, $\Lambda_d = 0$ there is a discontinuity in the dependence of the effective conductivity Λ with a volume concentration of components $m_m = m_c$, which is designated the percolation threshold [3, 4].

The present paper puts forward a new model for a nonuniform two-component system which possesses the geometric clarity of the ordered models and simultaneously takes into account the probability of forming conducting bridges and accounts for the appearance of a discontinuity in the effective conductivity at the threshold value of concentration $m_m = m_c$. In the limiting cases this model converts to the various well-known models for ordered structures.

Some Results of the Percolation Theory. Recently, in determining the properties of heterogeneous systems, methods of percolation theory have been used (percolation processes) [3, 4] in which it is reported that with a volume concentration of the conducting component

$m_m = m_c$, a conducting infinite cluster (IC) is created, represented schematically in the plane of Fig. 1b. An expression for the effective conductivity of this medium, when the conductivity of the second component is $\Lambda_d = 0$ and $m_c \leqslant m_m < 0.5$ is represented in percolation theory in the form

$$N = A(m_m - m_c)^k, \; N = \Lambda/\Lambda_m, \; m_c \leqslant m_m < 0.5. \tag{8}$$

To determine the constants A and k one needs to know the topology of an infinite cluster. The topology of an IC is discussed in a number of references [5-10], and the computer modeling of a random distribution of components in heterogeneous systems and also experimental data has shown that $k = 1.8 \pm 0.2$ and $m_c = 0.15 \pm 0.03$.

Relatively, the values of A are not as definite. For example, in [3], on the basis of model experiments to determine the electrical resistance of a cube with cells of number 25 × 25 × 25 (the conductivity of each cell was assigned in a random fashion, and $\Lambda_m \neq 0$ and $\Lambda_d = 0$, respectively) the result was obtained that A = 1.6, k = 1.6.

It was shown in [10], using model experiments on a computer, that A = 1 and k = 1.6-2.

In [5, 6] attempts were made to generalize the results of theory for the case when $\Lambda_d \ll \Lambda_m$ but $\Lambda_d \neq 0$ while m_m varies throughout the whole range of concentrations $0 \leqslant m_m < 1$. For this the entire range of concentrations $m_m = 0$-1 was divided into three sections, in each of which a relation for N was recommended. For example, if $\nu_m \leqslant 5 \cdot 10^{-4}$ then N is

$$\begin{aligned} N &= \nu_m(1 - 5m_m)^{-1}, \; m_m < m_c, \\ N &= 1.6(m_m - m_c)^{1.6}, \; m_c \leqslant m_m < 0.5. \end{aligned} \tag{9}$$

If $m_m > 0.5$, then in [6] it was recommended that N be determined using a formula obtained on the "efficient medium" model in [11, 12]:

$$N = [(3m_1 - 1) + (3m_2 - 1)\nu_m]/4 + \sqrt{[(3m_1 - 1) + (3m_2 - 1)\nu_m]^2/16 + \frac{\nu_m}{2}}. \tag{10}$$

If $3 \cdot 10^{-2} \leqslant \nu_m < 1$, then Eq. (10) is recommended for the entire range of concentrations.

In our opinion, the use of the efficient medium model as a basis for generalizing the results of percolation theory and experimental data on the effective conductivity N for $\nu_m < 10^{-2}$ is formal in nature. The efficient medium model does not reflect the topology of an IC and does not account for its variation (the branching with variation of concentration m_m). A defect of the correlations made in [5, 6] is also that there are various formulas for each range of concentration and ratio of component conductivities, and the formulas suggested are not related directly with the topology of the IC.

Heterogeneous System Model. In constructing a model we shall start from the following experimentally and theoretically established fact, that for $\nu_m = 0$ N depends on m_m according to Eq. (8). It can be shown that, for the case $\nu_m = 0$, the volume concentration of the conducting component belonging to an infinite cluster is

$$m_{c1} = \frac{m_m - m_c}{1 - m_c}. \tag{11}$$

Then, putting $A = (1 - m_c)^{-1}$ in Eq. (8), we obtain an expression for N with $\nu_m = 0$ in the following form

$$N = \left(\frac{m_m - m_c}{1 - m_c}\right)^k, \; 1 \geqslant m_m \geqslant m_c. \tag{12}$$

We have established that the discrepancy between the value calculated from Eq. (12) and the experimental and theoretical data of percolation theory is less than 20% for k = 1.6. Thus, we can assert that the conductivity of an IC with $\nu_m = 0$ is

$$\Lambda = \Lambda_m\left(\frac{m_m - m_c}{1 - m_c}\right)^{1.6}, \; 1 \geqslant m_m \geqslant m_c. \tag{13}$$

We shall single out a macroscopic tube of size L within the volume of a heterogeneous system. Here L is the minimum distance in which the conductivity of the tube is equal to the

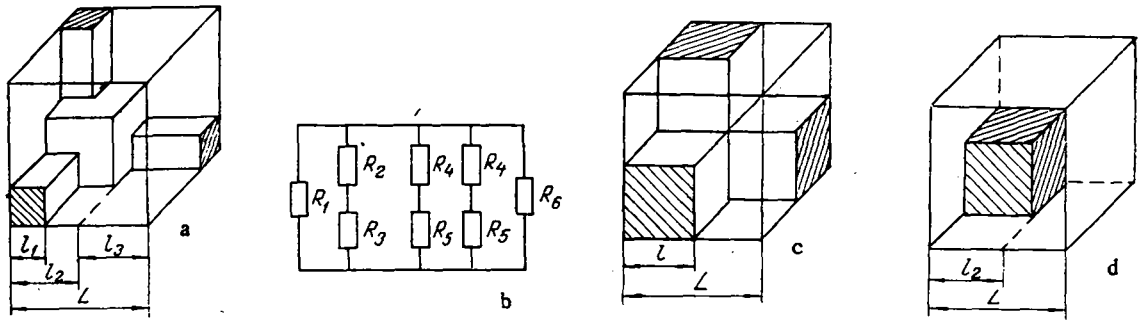

Fig. 2. Modeling of the structure of heterogeneous systems: a) the elementary cell of the model; b) the equivalent scheme for combining the resistances of the elementary cell when there is "adiabatic" partition; c) model with interpermeable components; d) the Eucken–Odelevskii model.

effective conductivity Λ of the nonuniform system, and in addition, the dimensions of the nonuniformities must be greater than the mean free path of the flux carrier (charge, energy, mass, etc.), i.e., the dimension L must not be less than the radius of correlation between the conducting particles. The resistance of the cube to current $\langle j \rangle$ flowing along the normal to one side, is

$$R = \frac{L}{\Lambda S}, \quad S = L \times L. \tag{14}$$

The resistance of the conducting IC, as follows from the definition, must be related to the mean length L_{IC} along which the flow passes and to the average area S_{IC} of the cross section of the IC by the relation

$$R_{IC} = \frac{L_{IC}}{\Lambda_m S_{IC}}. \tag{15}$$

If $\nu_m = 0$, then the cube resistance is equal to the resistance of the IC, i.e., $R = R_{IC}$ and it follows from Eqs. (14) and (15) that

$$\Lambda = \Lambda_m \frac{S_{IC}}{S} \frac{L}{L_{IC}}. \tag{16}$$

We shall denote the complex of unknown quantities S_{IC} and L_{IC} appearing in Eq. (16) by

$$S^*_{IC} = S_{IC} \frac{L}{L_{IC}}$$

and represent Eq. (16) in the form

$$\Lambda = \Lambda_m \bar{S}_1, \quad \bar{S}_1 = \frac{S^*_{IC}}{S}. \tag{17}$$

By comparing Eqs. (13) and (17), we obtain a law for the variation of the effective cross section S^*_{IC} of the conducting IC

$$\bar{S}_1 = \left(\frac{m_m - m_c}{1 - m_c} \right)^{1,6}, \tag{18}$$

which takes into account the most important features of this parameter.

We shall construct a model of the heterogeneous system structure, in which the following special features must be reflected: transition of an IC to an isolated cluster (IsC) for $m_m \leqslant m_c$; and the variation in the cross section of the conducting IC according to Eq. (18).

To do this we represent the topology of the IC in a cube with side L in the form of a three-dimensional figure shown in Fig. 2a. Here the isolated clusters (IsC) are individual inclusions of cubic shape with side l_2 and volume concentration $(l_2/L)^3 = m_c$, are located at distances l_3 apart, and joined by conducting links whose cross section is $S_1 = l_1^2$.

We now divide the cube with infinitely thin "adiabatic" planes parallel to the flow, and write* the average flux $\langle j \rangle$, passing through the cube (Fig. 2) along the normal to one of the sides (Fig. 2a), in the form

$$\langle j \rangle = \overline{S}_1 \langle j_1 \rangle + \overline{\Delta S} \langle j_{12} \rangle + 2\overline{S}_3 \langle j'_{12} \rangle + \overline{S}_4 \langle j_2 \rangle . \tag{19}$$

In Eq. (19) $\langle j_1 \rangle$ is the flux passing only through the first component (IC) with conductivity Λ_m; $\langle j_{12} \rangle$ is the flux passing sequentially through the second component (in the length $L - l_2$) and the first component in the cube length l_2; and $\langle j'_{12} \rangle$ is the flux passing sequentially through the second component (in the length $L - l_1$) and the first component in the cube length l_1; and $\langle j_2 \rangle$ is the flux passing only through the second component. The transverse sections of the individual parts of the cube are

$$\overline{S}_1 = \left(\frac{l_1}{L}\right)^2; \quad \overline{S}_2 = \left(\frac{l_2}{L}\right)^2; \quad \overline{S}_3 = \frac{(L - l_2) l_1}{L^2}, \quad \overline{S}_4 = 1 - \overline{S}_2 - 2\overline{S}_3.$$

Here it is assumed that

$$\overline{\Delta S} = \overline{S}_2 - \overline{S}_1, \quad \overline{l}_2 = m_c^{\frac{1}{3}}, \quad \text{if} \quad \overline{S}_2 > \overline{S}_1,$$
$$\overline{\Delta S} = 0, \quad \overline{l}_2 = \overline{l}_1, \quad \text{if} \quad \overline{S}_2 \leqslant \overline{S}_1.$$

The value $\overline{l}_1$ we find from Eq. (18):

$$\overline{l}_1 = \left(\frac{m_m - m_c}{1 - m_c}\right)^{0,8}, \quad \overline{l}_1 = \frac{l_1}{L}, \quad \overline{S}_i = \frac{S_i}{S}. \tag{20}$$

The equivalent scheme for combining the resistances to the passing flux $\langle j \rangle$ of individual sections of the cube is shown in Fig. 2b, where the resistances R_i have the following structure:

$$R_1 = \frac{L}{S\Lambda_m} \overline{S}_1^{-1}, \quad R_2 + R_3 = \frac{L}{S\Lambda_m} \frac{1 - (1 - \nu_m)\overline{l}_2}{\overline{\Delta S}\,\nu_m}, \tag{21}$$

$$R_4 + R_5 = \frac{L}{S\Lambda_m} \frac{1 - (1 - \nu_m)\overline{l}_1}{\overline{S}_3 \nu_m}, \quad R_6 = \frac{L}{S\Lambda_d} \overline{S}_4^{-1}.$$

The total cube resistance is

$$R^{-1} = R_1^{-1} + (R_2 + R_3)^{-1} + 2(R_4 + R_5)^{-1} + R_6^{-1}. \tag{22}$$

Taking into account that R may be written in the form of Eqs. (14) and (22), and using the resistance values from Eq. (21), we obtain an expression for the effective conduction of the heterogeneous system

$$N = \overline{S}_1 + \nu_m \left[\frac{\overline{\Delta S}}{1 - (1 - \nu_m)\overline{l}_2} + 2 \frac{\overline{S}_3}{1 - (1 - \nu_m)\overline{l}_1} + \overline{S}_4 \right]. \tag{23}$$

We shall consider special cases of Eq. (23).

a) If $\overline{\Delta S} = 0$, i.e., $S_2 = S_1$ (this condition holds for $m_m > 0.5$), then Eq. (23) takes the form

$$N = \overline{S}_1 + 2\nu_m \frac{\overline{S}_3}{1 - (1 - \nu_m)\overline{l}_1} + \overline{S}_4. \tag{24}$$

We choose the notation $C = l_1/L$ and substitute the values $\overline{S}_i$, and then Eq. (24) takes the form of the Dul'nev formula, which is valid for a model with interpermeable components, whose beams have the same thickness (Fig. 2c) [1]:

$$N = C^2 + 2\nu_m \frac{(1 - C)C}{1 - C + \nu_m C} + \nu_m (1 - C)^2. \tag{25}$$

b) If $\overline{S}_1 = 0$, i.e., $m_m \leqslant m_c$, then $\overline{S}_3 = 0$ and the model (Fig. 2a) converts to the Eucken–Odelevskii model with isolated subdivisions (Fig. 2d). For the range of concentrations $m_m \leqslant m_c$

*The justification for the method of "adiabatic subdivision" is given in [1].

$$\bar{S}_2 = m_m^{\frac{2}{3}}, \quad \bar{l}_2 = m_m^{\frac{1}{3}}, \quad \bar{S}_4 = 1 - m_m^{\frac{2}{3}}, \quad \bar{S}_1 = 0$$

and the expression for Λ, according to Eq. (23), takes the form

$$N = \left[\frac{m_m^{\frac{2}{3}}}{1 - (1 - \nu_m) m_m^{\frac{1}{3}}} + 1 - m_m^{\frac{2}{3}} \right] \nu_m. \qquad (26)$$

Thus, for concentrations $m_m \geqslant 0.5$, Eq. (23) converts to Eq. (25) for a model with interpermeable components, and for $m_m \leqslant m_c$, it converts to Eq. (26) for a model with isolated inclusions. If it is known that the material consists of continuous fibers or is a system with closed inclusions, then one can at once use Eqs. (25) and (26), respectively, for analysis over the entire range of concentration variation.

Comparison of Calculated Results with Experimental Data. Comparison of the calculated N from Eq. (23) with experimental data shows that the error in computation is comparable with the measurement error: if $\nu_m < 5 \cdot 10^{-4}$, then $m_c = 0.16 \pm 0.1$; if $\nu_m \geqslant 5 \cdot 10^{-4}$, then $m_c = 0.09$.

The variation in the model parameter m_c with increase of ν_m results from the fact that if ν_m is small but not equal to zero ($\nu_m > 5 \cdot 10^{-4}$), then when IC undergoes transition to IsC($m_m \to m_c$) thin layers of dielectric are formed between the conducting isolated clusters (CIsC),and their conductivity in these conditions is comparable with that of the CIsC itself [10] (see Fig. 1c). The presence of "conducting" thin dielectric layers between IsC, leads to a shift in m_c toward smaller m_m, i.e., transition of conducting IC to IsC, between which conducting bonds are formed, and this occurs for $m_m < m_c = 0.15 \pm 0.03$.

Figure 3 compares the calculated N using Eq. (23) with experimental data on the electrical conductivity of sintered copper powder with addition of alumina and sintered iron powder with addition of alumina [13]. The ratio of conductivity of the original components in these mixtures is $\nu_m = 0$. The comparison between the calculated and experimental data is satisfactory. Figure 4 compares the theoretical N from Eq. (23) with the experimental data on electrical conductivity of tungsten bronze Na_xWO_3 at T = 300°C [5, 6]. The ratio of conductivities in this system is $\nu_m = 10^{-4}$. Figure 4 also shows data of model experiments on electrical conductivity with $\nu_m = 10^{-4}$ in the range $m_m = 0.3$-0.5 [5, 6]. Comparison of the calculated N and the experimental data for $\nu_m = 10^{-4}$ shows good agreement (error of ∿20%).

Figure 4 also compares the calculated N and the experimental data on electrical conductivity of a NH_3–Li solution, in which the ratio of the component conductivities is $\nu_m = 1.2 \cdot 10^{-3}$ [5, 6].

It should be noted that N as calculated from Eq. (23) with $\nu_m > 10^{-2}$ is close to N from the Dul'nev formula [1] over the entire range of concentrations.

Recommended Formula for Calculating the Effective Conductivity of Heterogeneous Systems. As was pointed out, to close Eqs. (6) and (7) and determine the effective conductivity of heterogeneous systems Λ, one needs information on their structure. Analysis of the structures of heterogeneous systems shows that, in general, they can be divided into three basic types: I) structures with isolated impregnations; II) structures with interpermeating components; and III) structures where there is transition from structures with isolated impregnations to structures with interpermeating components, as the concentration varies.

Heterogeneous systems with structures of type I and II have been described quite well in the literature using the Eucken–Odelevskii and the Fre–Dul'nev models [1, 12].

For type III structures the model constructed (Fig. 2a) and the formula obtained for the effective conductivity N, Eq. (23), are in good agreement with experimental data over a wide range of the parameters $0 \leqslant \nu_m \leqslant 1$ and $0 \leqslant m_m \leqslant 1$. Here the geometric parameters of the model (Fig. 2a) were chosen from experimental and theoretical data of percolation theory. We note that the model proposed encompasses all three types of structures — I-III.

It should be noted that the more complete and reliable the information which is used on the structure of heterogeneous systems, in choosing geometric model parameters, the better is the agreement between the calculated and experimental data on the properties of heterogeneous systems. If the structure of a heterogeneous system is unknown, we can recommend the following investigative procedure.

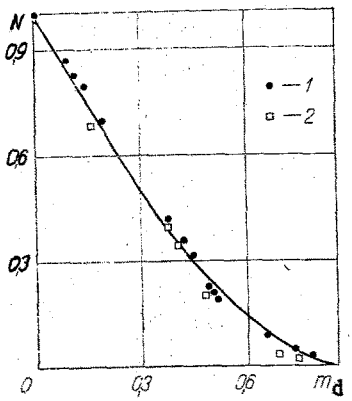

Fig. 3

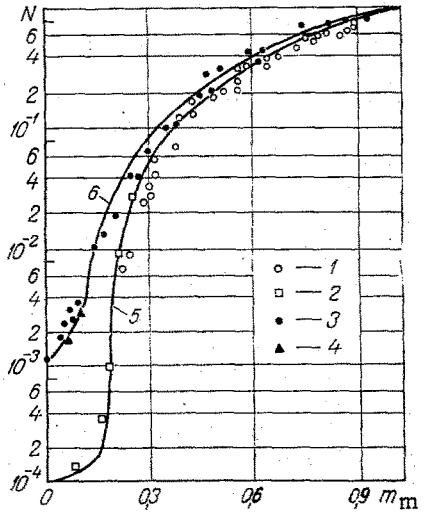

Fig. 4

Fig. 3. The effective electrical conductivity of sintered powders with addition of alumina ($\nu_m = 0$). The points are the experimental data of [13]: 1) copper powder; 2) iron powder. The curve is the calculated value from Eq. (23).

Fig. 4. The effective electrical conductivity of heterogeneous systems with random structure. The points are the experimental data of [5, 6]: 1) tungsten bronze at T = 300°K ($\nu_m = 10^{-4}$); 2) data of model experiments ($\nu_m = 10^{-4}$); 3) a solution NH_3—Li ($\nu_m = 1.2 \cdot 10^{-3}$); 4) model experimental data ($\nu_m = 1.2 \cdot 10^{-3}$). The curves are calculations from Eq. (23): 5) with $\nu_m = 10^{-4}$, $m_c = 0.17$; 6) with $\nu_m = 1.2 \cdot 10^{-3}$, $m_c = 0.09$.

With the chosen model — I, II, or III — we solve the inverse problem, i.e., we compare the calculated values of N for different geometrical model parameters with a limited number of experimental data for N, and thus we can determine the structure of the given heterogeneous system. For the chosen structure we subsequently determine the transfer coefficients analytically.

NOTATION

Λ, effective conductivity; m_i, volume concentration of the i-th component; Λ_i, effective conductivity of the i-th component; $\langle j \rangle$, mean flux of material; $\langle \nabla\varphi \rangle$, mean potential gradient; $j(r)$, local flux of material; $\nabla\varphi(r)$, local potential gradient; m_c, percolation threshold; $\nu_m = \Lambda_d/\Lambda_m$; $N = \Lambda/\Lambda_m$.

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