Journal of Engineering Physics and Thermophysics, Vol. 78, No. 6, 2005

EFFECTIVE CONDUCTANCE OF A HIGH-CONTRAST, RANDOM-STRUCTURE COMPOSITE. NUMERICAL SIMULATION

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This paper considers a high-contrast, two-component composite of random structure, for whose simulation a two-dimensional network model is used. The dependence of the medium conductance on the volume content and composition of the filler that is characteristic of percolation theory has been obtained: up to some volume content, the effective conductance is small and then it grows rapidly. The results are based on statistical modeling (solving a large number of problems at various random distributions of inclusions and with subsequent statistical processing).

In the present paper, the effective properties of composites are calculated with the example of the problem of calculating the effective permittivity of polymers filled with ceramic particles. Polymers are filled with ceramic powder to increase the composite permittivity (ceramics can have a permittivity up to 10,000, polymers — from 2 to 5 [1]). Another example — polymers filled with ceramic heat-conducting particles increasing the heat transfer (the heat-conductivity coefficients of the polymer and the ceramics differ by a factor of 100–1000 [1]). Such materials are used in the electrical industry. The above-mentioned media are characterized by a random distribution of components and their high contrast.

The available theoretical results on the averaging of random media [2, 3] have not yet been realized in practice. Valid formulas for calculating the effective properties of random media have been obtained only for a small number of particular cases [4, 5]. The empirical formulas of [6] are applicable in the range of values typical of them, but for high-contrast composites they predict considerably overestimated values of effective characteristics. Numerical simulation of a random high-contrast medium on the basis of the finite-difference or finite-element methods is theoretically possible, but the problem dimension thereby turns out to be very large (since, if the representative element of the medium contains tens or even hundreds of inclusions, upon discretization of inclusions and the matrix by the network its size turns out to be considerable). Moreover, the contrast of coefficients makes it difficult to use the numerical methods that are oriented (without modifications) to noncontact media. It has been noted [7] that in contrast media the fluxes are concentrated in zones between adjacent particles. This means that the application of homogeneous networks will lead to a count by "empty place" (regions where fluxes are immaterial) and, having made no appreciable contribution to the calculation of the field where it is concentrated, will lead to unnecessary time expenditures. The remark on the concentration of fluxes permits passing from a continuous problem to a discrete one (the so-called "network" model [7, 8]) that takes into account only the high fluxes between close particles and ignores the other (weaker) fluxes.

With a finite-dimensional model one can write a high-speed (a few minutes per program run) computer program for simulating a composite filled with randomly distributed particles and calculating its effective conductance and, after a large number (hundreds) of runs collect a body of data sufficient for statistical processing.

Assume the following:

1. High-conductivity inclusions are modeled by disks D_i that are uniformly distributed without overlap and are close-packed. The latter means that the characteristic distance between disks is much smaller than their characteristic size. This approach is a frequently used method for modeling a random distribution of particles known as the successive addition procedure [9].

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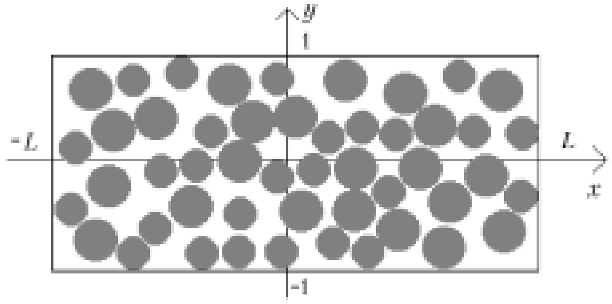


Fig. 1. Composite model.

2. The potential φ on the disk D_i is constant ($\varphi = t_i$ on D_i); the values of t_i are unknown and are determined from the problem solution. This condition corresponds to the "stiff" expansion in the averaging theory [10].

3. Only the fluxes between adjacent (neighboring) disks are taken into account; all the other fluxes are neglected. The possibility of taking into account the fluxes between adjacent disks was substantiated in [7].

Problem Formulation. We consider a model of a composite with randomly distributed, perfectly conducting inclusions (Fig. 1). Let the composite occupy the region $\Pi = [-L, L] \times [-1, 1]$. Denote the disks that model inclusions as $\{D_i, i = 1, ..., N\}$, where N is the total number of disks. Then $Q = \Pi \setminus \bigcup_{i=1}^{N} D_i$ is the region occupied by the matrix. Let us introduce the space of functions V taking (unknown) constant values on inclusions:

$$V_p = \left\{ \varphi \in H^1(Q) : \varphi(\mathbf{x}) = \text{const} \quad \text{on} \quad D_i, \varphi(x, \pm 1) = \pm 1 \right\}. \tag{1}$$

The condition $\varphi(x, \pm 1) = \pm 1$ means the application of potentials ± 1 to the bounds $y = \pm 1$ respectively.

Let us consider the energy integral

$$I(\varphi) = \frac{1}{2} \int_Q |\nabla\varphi|^2 d\mathbf{x} \to \min, \quad \varphi \in V_p. \tag{2}$$

The problem of minimization of (1), (2) can be written in the form of the boundary-value problem:

$$\Delta\varphi = 0 \quad \text{in} \quad Q, \tag{3}$$

$$\varphi(\mathbf{x}) = t_i \quad \text{on} \quad \partial D_i, \tag{4}$$

$$\int_{\partial D_i} \partial\varphi/\partial\mathbf{n} d\mathbf{x} = 0, \tag{5}$$

$$\varphi(x, \pm 1) = \pm 1, \tag{6}$$

$$\partial\varphi/\partial\mathbf{n}(\pm L, y) = 0. \tag{7}$$

Hereinafter, $x = \pm$L denotes the vertical (left and right) bounds of the region Π, and $y = \pm 1$ denotes the horizontal bounds (see Fig. 1).

Formulas for calculating the effective constant. We define the effective conductance having the meaning of the total flux through the bound $y = 1$ on a per length unit basis as

$$a = \frac{1}{2} \int\limits_{y=1} \frac{\partial \varphi}{\partial \mathbf{n}} d\mathbf{x} .$$

The quantity a can be related to the energy integral $I(\varphi)$ (2) usually used in introducing effective characteristics in averaging theory. Multiplying (3) by φ and integrating the result by parts, we obtain

$$0 = -\int\limits_{Q} |\nabla\varphi|^2 d\mathbf{x} + \int\limits_{y=\pm 1} \frac{\partial \varphi}{\partial \mathbf{n}} \varphi d\mathbf{x} + \sum_{i=1}^{N} \int\limits_{\partial D_i} \frac{\partial \varphi}{\partial \mathbf{n}} \varphi d\mathbf{x} . \quad (8)$$

For the boundary integrals, using (4)–(6), we have

$$\int\limits_{\partial D_i} \frac{\partial \varphi}{\partial \mathbf{n}} \varphi d\mathbf{x} = t_i \int\limits_{\partial D_i} \frac{\partial \varphi}{\partial \mathbf{n}} d\mathbf{x} , \quad (9)$$

$$\int\limits_{y=\pm 1} \frac{\partial \varphi}{\partial \mathbf{n}} \varphi d\mathbf{x} = \int\limits_{y=1} \frac{\partial \varphi}{\partial \mathbf{n}} d\mathbf{x} - \int\limits_{y=-1} \frac{\partial \varphi}{\partial \mathbf{n}} d\mathbf{x} . \quad (10)$$

Integration of (3) by parts taking account of (5) and (10) yields

$$\int\limits_{y=\pm 1} \frac{\partial \varphi}{\partial \mathbf{n}} \varphi d\mathbf{x} = 2 \int\limits_{y=1} \frac{\partial \varphi}{\partial \mathbf{n}} d\mathbf{x} . \quad (11)$$

From (8), (9), and (11) we get

$$\int\limits_{y=1} \frac{\partial \varphi}{\partial \mathbf{n}} d\mathbf{x} = \frac{1}{2} \int\limits_{Q} |\nabla\varphi|^2 d\mathbf{x} , \quad (12)$$

where φ is the solution of problem (3)–(7) (or, which is the same, (1), (2)).

For convenience, we use the quantity $A = 2aL$ having the meaning of the total flux through the upper bound of the rectangle Π. By virtue of (12), the value of A can be calculated by one of the following formulas:

$$A = \frac{1}{2} \int\limits_{Q} |\nabla\varphi|^2 d\mathbf{x} , \quad (13)$$

$$A = \int\limits_{y=1} \frac{\partial \varphi}{\partial \mathbf{n}} d\mathbf{x} , \quad (14)$$

from where, in particular,

$$A = \frac{1}{2} \min \int\limits_{Q} |\nabla\varphi|^2 d\mathbf{x} , \quad \varphi \in V_p . \quad (15)$$

Note that in the case under consideration the matrix conductivity is equal to one.

Discrete problem. We express A in terms of fluxes $p = \nabla\varphi$, which permits using condition 3 for calculating A. According to this condition, only the fluxes in the channels connecting adjacent disks are taken into account; the other fluxes are neglected. In so doing, one should know how to calculate the flux between two disks on which con-

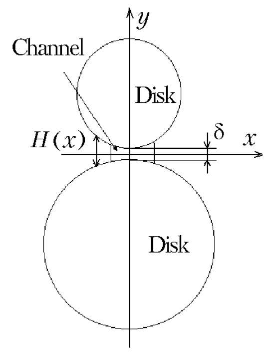


Fig. 2. Layout of two adjoining disks.

stant potentials are given. A simple method for calculating the flux in this case was proposed in [4] for identical disks (this method was justified in [7]). In considering disks of different sizes, the method of [4] can be used to calculate the flux between disks (the ith and jth) of different radii R_i and R_j (Fig. 2). Following [4], we approximate the disks by parabolas $y = \delta/2 + \rho_i x^2/2$ and $y = \delta/2 + \rho_j x^2/2$, where $\rho_i = 1/R_i$ and $\rho_j = 1/R_j$. The distance between the disks is

$$H(x) = \delta_{ij} + \frac{(\rho_i + \rho_j)\, x^2}{2}. \tag{16}$$

Assume that the local flux (potential gradient) between the disks is defined as

$$\mathbf{p} = \left(0, \frac{t_i - t_j}{H(x)}\right), \tag{17}$$

i.e., the first component of the vector **p** is assumed to be equal to zero and the second component is assumed to be proportional to the potential difference on the disks and inversely proportional to the distance between the disks along the y-axis. Then the total flux is

$$J_{ij} = (t_i - t_j)\int_{-S}^{S} \frac{dx}{\delta + (\rho_i + \rho_j)\, x^2/2} = \frac{1}{((\rho_i + \rho_j)/2)^{1/2}\, \delta^{1/2}} \arctan\left(\frac{(\rho_i + \rho_j)/2}{\delta^{1/2}}\right)\Bigg|_{-S}^{S}. \tag{18}$$

The sum of curvatures $\rho_i + \rho_j$ is large enough, but the distance δ between disks is small, by virtue of which the last factor on the right side of (18) approaches π. As a result, we obtain (taking into account that $\rho_i = 1/R_i$ and $\rho_j = 1/R_j$)

$$J_{ij} = (t_i - t_j)\, G_{ij},$$

where

$$G_{ij} = \pi \frac{\sqrt{2R_i R_j/(R_i + R_j)}}{\sqrt{\delta}} \tag{19}$$

is the flux per unit potential difference of the disks.

Formula (19) can be obtained as the principal term of asymptotics as $\delta \to 0$ from the formula for the pair capacitance of two disks separated by a distance δ [11].

Construction of the Network Problem. Let us introduce a discrete network corresponding to the input continuous problem. In accordance with the hypothesis on the particle distribution, we will generate some random distribution of disks of a given radius (or several given radii) in the rectangle $\Pi = [-L, L] \times [-1, 1]$. The center **x** of each disk is generated as a random point uniformly distributed in Π. If a generated disk (with a center **x** and a radius R) intersects the previously generated disks, then this attempt "does not count"; if there is no intersection, then the generated disk is added to the list of disks

$$L = \{(\mathbf{x}_i, R_i)\ ,\quad i = 1, ..., N\}\ . \tag{20}$$

The generation procedure will terminate when the specific volume of the disks becomes equal to the given value of V:

$$V = \sum_{i=1}^{N} \pi R_i^2 / |\Pi|\ .$$

We next calculate the distance δ_{ij} between the generated disks and introduce fluxes, using the following rule:

$$g_{ij} = \begin{cases} G_{ij}\ , & \delta \le \delta^*\ ; \\ 0\ , & \delta > \delta^*\ . \end{cases} \tag{21}$$

The quantities g_{ij} in (21) describe the flux between the ith and jth disks. In choosing δ^*, one should take into account the "neighbor–neighbor" contacts and ignore the other contacts between the disks. The numerical simulation performed by us shows that a disk usually has 5–6 close neighbors the distance to which can be any, however small. The distance to the other disks can be much larger (of the order of a few radii of the disks). Numerical simulation of random distributions of disks has shown that the choice of δ^* in (21) in the (0.3–0.5)R interval steadily distinguishes the "neighbor–neighbor" relation even for disks of different sizes if the radii differ by a factor of no more than 5. In numerical counting, the "neighbor–neighbor" relation was additionally checked visually (a few test runs of the program with a display of disk distribution images were realized).

As a result, we obtain the discrete model (weighted graph)

$$G = \{\mathbf{x}_i, g_{ij};\quad i, j = 1, ..., N\}\ , \tag{22}$$

consisting of nodes $\mathbf{x}_i$ (disk images) and edges ("neighbor–neighbor" links) with their corresponding specific fluxes g_{ij}. The discrete model (22) does not contain the geometric characteristics of the disks and the distances between them in explicit form, and this information is taken into account in the values of g_{ij}.

Some of the disks are situated near the boundary, and, therefore, one should take into account the flux in the "disk–y = ±1 boundary" system. Let us include this case in the previous one, considering the boundary as a disk of infinite radius. We will call the near-boundary disk projection onto the +1 boundary the "quasi-disk." Likewise, we determine the "quasi-disks" at the $y = -1$ boundary. The flux in the "disk–quasi-disk" system is calculated by formula (19), in which we assume $R = \infty$ for the "quasi-disk."

To close the model, we need equations for determining the potentials t_1 in the network nodes $\mathbf{x}_i$ (22). To this end, we make use of (2). According to condition (3), only the fluxes between adjoining disks contribute to integral (2). From (21) it follows that the discrete analog of integral (2) for the network is written as $\frac{1}{4}\sum_{i,j=1}^{N} g_{ij}(t_i - t_j)^2$. The factor 1/4 instead of 1/2 is due to the fact that summing over i, j = 1, ..., N, we twice pass through the channel between the disks. As a result, we obtain the problem

$$\frac{1}{4}\sum_{i,j=1}^{N} g_{ij}\,(t_i - t_j)^2 \to \min\ , \tag{23}$$

corresponding to (2). The boundary conditions for (23) have the form

$$t_i = \pm 1 \ \text{ for } \ i \in S^{\pm}\ . \tag{24}$$

Here $S^{\pm}$ denotes the boundary disks, i.e., the disks touching the $y = \pm 1$ boundaries and the "quasi-disks g_{ij}" lying at the $y = \pm 1$ boundaries. Designate the remaining (internal) disks as $I = \{\mathbf{x}_i, i = 1, ..., N\} \backslash (S^+ \cup S^-)$.

Problem (23) is equivalent to the following system of linear equations (Kirchhoff equations):

$$\sum_{j=1}^{N} g_{ij}(t_i - t_j) = 0 \quad \text{for} \quad i \in I, \quad t_i = \pm 1, \quad i \in S^{\pm}. \tag{25}$$

Formula (14) for calculating the effective conductance takes on the form

$$A = \sum_{i \in S^+} \sum_{j \in I} g_{ij}(1 - t_j) = 0, \quad i \in I, \tag{26}$$

since $t_i = 1$ for $i \in S^+$.

Note that problem (25), (26) can be obtained by introducing finite elements of a special kind (equal to zero outside the channels between the disks and given by formula (17) in the channels).

Numerical Simulation. The author has developed a computer program that performs the following operations:

1) generation of a system of random distribution of disks;
2) calculation of coefficients g_{ij} for the generated system of disks;
3) solution of the linear system (24), (25);
4) computation of the effective conductance A by formula (26).

The program parameters are: diameter of disks (in the case of using disks of several parameters) and their volume content V.

As a result of one run of the program, we obtain the effective conductance $A(\omega)$ corresponding to the random distribution ω of disks generated at the given start of the program. To collect statistics, we repeatedly ran the program (100–300 times) and collected data $\{A(\omega), \omega \in \Omega\}$. Using the data collected upon execution of the given number of program runs, we computed: the mathematical expectation (effective conductance) $A = MA(\omega)$, the mean deviation $DA = M|A(\omega) - A|$, and the conductance maximum $m = \max_{\omega \in \Omega} A(\omega)$. All of the above quantities were calculated at given R and V, in which connection they were their functions: A, DA, $m = A$, DA, $m(R, V)$.

Results of the numerical simulation of the monodispersion medium. In this problem, we considered disks of one radius, which could be different in different sets of numerical experiments. The volume content V increased from zero with some step δV. For each value of V the effective conductance $A(\omega)$ was calculated. The data on the region Π are presented below.

Calculations were performed for disks of different sizes in order to control the numerical count. Theoretically, the effective conductance should not depend on the particle radius, which was just checked. From the results of the calculation of the value of A (Figs. 3–5) it is seen that it is really independent of the disk radii (the graphs in Fig. 3b and c practically coincide).

In the graphs presented in Figs. 3–5, two regions are distinguished: $V \le 0.25$, where the flux through the composite is small, and $V \ge 0.35$, where the flux becomes high. In the [0.25; 0.35] interval, there is a slow increase in the effective conductance of the composite, i.e., the $A = A(V)$ curve demonstrates the behavior of the flux through the composite that is typical of percolation theory [12]. The percolation limit in all calculations turned out to be equal to $V_0 \approx 0.3$. Figures 3–5 also present the graphs of the maximum (in realizations) values of the conductance $m(\omega)$ and the quantities $A \pm DA$. It is seen that they correlate with the averaged conductance.

Determination of the functional dependence A(V). Let us use the power law $A(V) = [a(V - V_0)]^b$ and the exponential dependence $A(V) = a \exp [b(V - V_0)]$ to describe the graph presented in Fig. 3b. We determine the coefficients a and b by the least-square technique. The power law gives a better agreement with the numerically obtained effective conductances than does the exponential dependence. The values of $A(V)$ are given in Table 1. For the power law

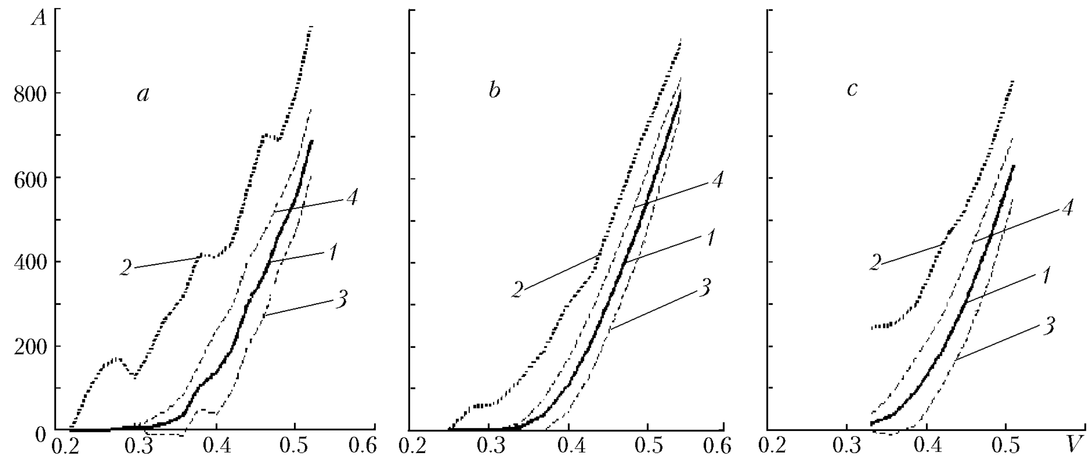


Fig. 3. Dependences of the effective conductance $A(V)$ (1), the maximum conductance m = max $A(V)$, $A(V) - DA$ (3), and $A(V) + DA$ (4) on the volume content of disks V at R = 25/200 (a), 15/200 (b), and 20/200 (b).

TABLE 1. Effective Conductance Value Depending on the Volume Content of Disks ($A(V)$ denotes the numerical experiment, A_1 and A_2 have been calculated by formulas (27) and (28))

V	$A(V)$	A_1	A_2
0.300	0	0	—
0.325	10	5	—
0.350	20	25	—
0.375	42	63	—
0.400	105	117	107
0.425	190	190	177
0.450	275	282	270
0.475	400	392	382
0.500	520	525	517

$$A_1\,(V) = [a\,(V - 0.3)]^b \tag{27}$$

a = 85 and b = 2.17. Taking into account that the model was obtained under the condition of close packing of particles, we give the calculations of the same quantities under the condition of data neglect in the vicinity of the percolation threshold, using only the data at $V \geq 0.4$ (this is a rather high filling, its limiting value is about 0.55). In this case, a = 75 and b = 2.25. It is seen that the values of the coefficients a and b have changed insignificantly. Taking into account that the model is based on the assumption of a close packing of particles, we can recommend using the formula

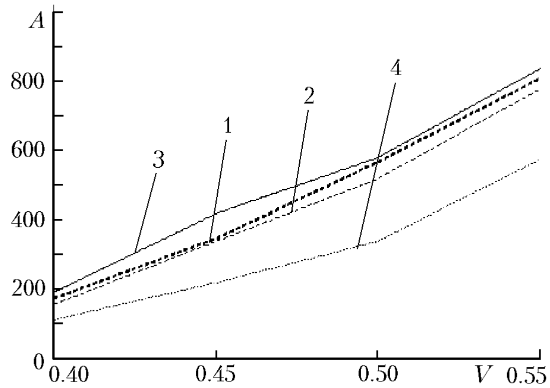
$$A_2\,(V) = [75\,(V - 0.3)]^{2.27} \tag{28}$$

at a volume content of particles $V > 0.4$. The values of the effective conductances calculated by formulas (27) and (28) are also given in Table 1.

Polydispersion composites. A composite filled with inclusions of different sizes is called dispersive. In the model considered, polydispersion is modeled by the scatter in the region Π of disks of different radii. The distribution of radii can be continuous and discrete. We considered the discrete distribution. Mixtures of two kinds of disks were used (the characteristics of the mixtures are given in Table 2). The volume content of disks was varied over the 0.4–0.55 range. The latter value is close to the largest one possible. The graph of the effective conductance A as a function of the volume content V is given in Fig. 4. Plotted on the x-axis is the specific content of disks with R_1 =

TABLE 2. Characteristics of Polydispersive Mixtures

Kind of mixture	R_1	V_1, %	R_2	V_2, %
Mixture 1	25	33	15	67
Mixture 2	25	67	15	33
Mixture 3	35	33	15	67
Mixture 4	35	67	15	33



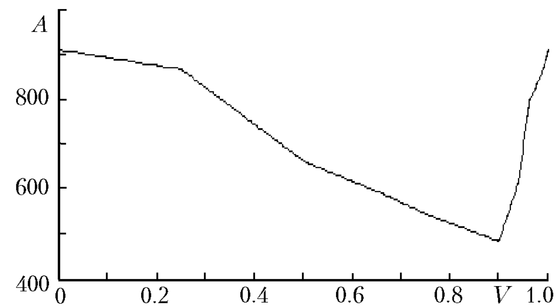


Fig. 4. Effective conductance A versus the volume content of disks V. To the curve numbers correspond the mixture numbers in Table 2.

Fig. 5. Effective conductance of the polydispersive composite (mixture of disks of radii 25/200 and 10/200) depending on its dispersive content.

25/200. The total volume content of disks is 0.55. The $A(V)$ curves for mixtures 1–3 are closely spaced. For them the polydispersion does not influence the effective coefficient of the medium. The curve for mixture 4 is distant from the others, i.e., here this influence is appreciable.

We investigated mixtures of disks at their total volume content $V = 0.55$. The disk radii were reduced to $R_1 = 25$ and $R_2 = 10$ in order to increase their number and, owing to this, decrease the statistical straggling of the calculation results. The relative contents V_1 and V_2 ($V_1 + V_2 = 1$) of disks of the first and second kinds were varied from 0 to 1, and the respective effective constants were calculated. Figure 5 presents the graphs of the effective conductance versus the relative volume content of disks. It shows equal values of A at $V_1 = 0$ and $V_1 = 1$ (the size effect is absent, which is consistent with the theory) and the decrease in the effective conductance at $V_1 = 0.9$ by about one half compared to the monodisperse composites.

Description of the Numerical Calculation. In the numerical calculations, $\Pi = 550 \times 400$ (accordingly, $L = 550/400 = 1.375$). The choice of such region dimensions is connected with the graphical information display. Disk centers were generated by a procedure of the kind of random(n) + random(1), where random(n) generates pseudorandom integral numbers from the set $\{0, 1, ..., n\}$, and random(1) generates real numbers from the [0, 1] interval.

We used disks of radii 35, 25, 20, 15, and 10 (which in dimensional coordinates was 35/200, 25/200, 20/200, 15/200, and 10/200, since [−1, 1] in the definition of the region Π corresponded to [0, 400] in modeling). In the calculations, the total number of disks varied from 50 to 150.

CONCLUSIONS

1. Numerical calculations of the effective conductance of a flat medium with randomly distributed absolutely conducting disks have been performed.

2. The use of the network model of a high-contrast composite has made it possible to calculate the effective conductance of the composite for a large number of random configurations and obtain fairly exact values of the effective conductance depending on the volume content and dispersive composition of the disks.

3. It has been found that the dependence of the effective conductance on the volume content has the form characteristic of percolation theory. Its parameters have been determined.

4. The dependence of the effective conductance on the dispersive composition of the disks has been found.

NOTATION

a, specific flux; $A = 2aL$, total flux through the upper bound of rectangle Π; D, mean deviation; g_{ij}, specific flux between two disks; G, discrete model (weighted graph); H^1, V_p, space of functions; I, internal disks; $I(\varphi)$, functional; L, list of disks; m, maximum conductance; M, mathematical expectation; N, total number of disks; $\mathbf{p} = \nabla\varphi$, local flux (vector); $Q = \Pi \setminus \bigcup_{i=1}^{N} D_i$, region occupied by the matrix; $S^{\pm}$, boundary disks; t_i, disk D_i potential; φ, potential; $\Pi = [-L, L] \times [-1, 1]$, area occupied by the composite; $|\Pi|$, area of Π.

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