

# ELECTRICAL PROPERTIES OF DIELECTRIC MATRIX WITH SPHERICAL INCLUSIONS OF METAL

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UDC 621.315.612:537.226.1

The dielectric permittivity and electrical conductivity of a dielectric matrix with spherical inclusions of metal are calculated. The theoretical formula is verified experimentally and compared with other known formulas.

Structurally inhomogeneous materials consisting of several phases are used in various branches of engineering. Such materials include inhomogeneous and porous dielectrics and also dielectrics with inclusions of conducting particles. The wide application of many of these materials is impeded by a lack of information on their physical properties, and, in particular, their electrical properties. The electrophysical properties of multiphase materials have been investigated experimentally [1-5], and attempts at theoretical calculation have been made [1-3, 6-13]. In [9-13], in particular, theoretical formulas are presented for the averaged parameters of a two-phase system consisting of a dielectric matrix with conducting inclusions. The results obtained depend very greatly on the model of the system adopted: For example, the formulas for the averaged parameters of the medium derived in [9-11] assume cubic conducting inclusions (Fig. 1a), while those in [10, 12, 13] assume spherical inclusions.

In the present paper, the dielectric permittivity and electrical conductivity of a system consisting of a dielectric matrix with spherical inclusions of metal are calculated. The results obtained are verified experimentally, and previously derived formulas are examined.

The model of the two-phase system used for the calculations is as follows. The sample whose averaged parameters are to be established is a cube of side 1 cm. Hence its electrical resistance (when a voltage is applied across opposite faces) is equal to the resistivity. The spherical inclusions in the dielectric are all of the same diameter  $d$ , and are equally spaced; their centers lie at the points of the simplest three-dimensional cubic lattice (Fig. 1b).

In deriving formulas for the dielectric constants of the system, it is assumed that there is no through conduction of the dielectric, and that the electrical resistance of the metal is practically zero.

When a voltage is applied across opposite faces of the cube, the electrical capacitance of the sample is

$$C = k\epsilon_{av}, \quad (1)$$

where  $\epsilon_{av}$  is the averaged dielectric permittivity of the system, which has to be found.

If we now imagine the sample to be divided into cylindrical columns perpendicular to the electrodes, each containing a row of metallic inclusions, we obtain a number of parallel-connected capacitors, all but one of which contain inclusions. Assuming that over the length of the cube there are  $n$  inclusions, we have  $n^2$  columns containing inclusions (Fig. 1b). Therefore,

$$C = C_d + n^2 C_{col}, \quad (2)$$

where  $C_{col}$  is the capacitance of a column of material containing  $n$  spherical metallic inclusions;  $C_d$  is the capacitance of the body obtained when all the columns containing the inclusions are removed from the sample.

Assuming initially that the electric field between the inclusions does not extend beyond the limits of the column, i.e., that the force tube is a cylinder of diameter  $d$ , the capacitance of the part of the sample not containing inclusions is

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Agrophysical Scientific-Research Institute, Leningrad. Translated from *Inzhenerno-Fizicheskii Zhurnal*, Vol. 31, No. 6, pp. 1000-1007, December, 1976. Original article submitted December 8, 1975.

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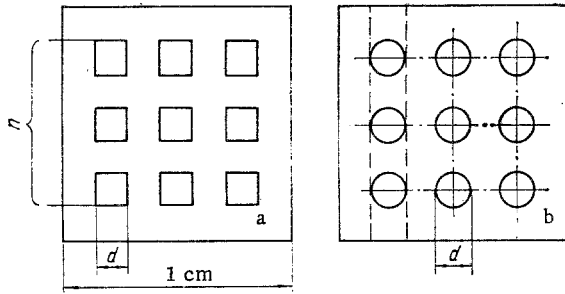


Fig. 1. Model of dielectric matrix with inclusions:  
a) cubic; b) spherical.

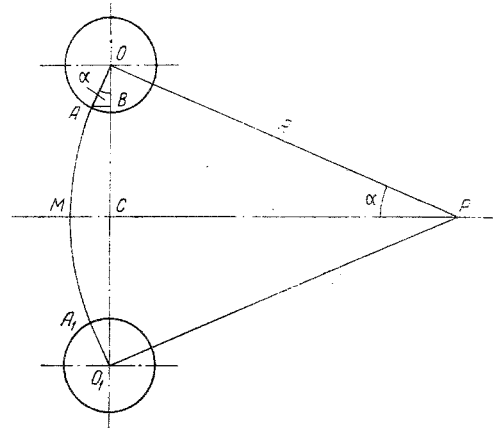


Fig. 2. Diagram for calculating the capacitance of an elementary capacitor.

$$C_d = k\epsilon_0 S_0 = k\epsilon_0 \left( 1 - n^2 d^2 \frac{\pi}{4} \right) = k\epsilon_0 (1 - 0.785 c_{sp}^2). \quad (3)$$

Here  $\epsilon_0$  is the dielectric permittivity of the matrix phase;  $S_0$  is the area of the electrode, excluding the ends of the columns;  $\pi d^2/4 \approx 0.785$ ;  $d^2$  is the cross-sectional area of each column;  $c_{sp} = nd$ .

Note that the calculation of  $S_0$  in this way is accurate only for cubic inclusions (neglecting edge effects); this is also true of the formulas derived in [11].

The capacitance of each column  $C_{col} = C'/(n+1)$ , where  $n+1$  is the number of dielectric intervals between the metallic inclusions in the columns;  $C'$  is the capacitance of each such interval. (We assume that each column consists of  $n+1$  series-connected elementary capacitors in which the electrodes are metallic inclusions.)

It is possible to calculate the capacitance of the capacitor formed by two charged insulated spheres using a fairly complicated method [16]. In our case, however, the spheres are not perfectly insulated, and therefore we derive the capacitance between them using the scheme shown in Fig. 2. For a sufficiently large distance between the inclusions, the lines of force of the field normal to the spherical surface form arcs of circles [16]. The radius of each of these (if there is an angle  $\alpha$  between the column axis and the tangent to the lines of force at the point of contact with the surface of the sphere) is

$$R = OD = OC/\sin \alpha = \frac{1}{\sin \alpha} \left( \frac{l}{2} + \frac{d}{2} \right) = \frac{l+d}{2 \sin \alpha},$$

where  $l$  is the distance between inclusions.

The length of the lines of force is

$$l_x = L - d = 2R\alpha - d = \frac{2\alpha(l+d)}{2 \sin \alpha} - d = \frac{\alpha(l+d) - d \sin \alpha}{\sin \alpha},$$

where  $l$  is the length of the arc  $OAMA_1O_1$ .

A capacitor with annular electrodes of infinitesimal width has capacitance

$$dC = k\epsilon_0 dS/l_x = \frac{k\epsilon_0 \sin^2 \alpha \cos \alpha d\alpha \pi d^2}{2[(l+d)\alpha - d \sin \alpha]} = \frac{k\epsilon_0 \pi d^2 \sin^2 \alpha \cos \alpha d\alpha}{2\{[(l+d)/d]\alpha - \sin \alpha\}},$$

since

$$dS = 2\pi ABd \quad (AB) = (\pi d^2 \sin \alpha \cos \alpha d\alpha)/2.$$

Then

$$C' = \frac{k\epsilon_0 \pi d}{2} \int_0^{\alpha_1} \frac{\sin^2 \alpha \cos \alpha d\alpha}{[(l+d)/d]\alpha - \sin \alpha}. \quad (4)$$

TABLE 1. Dependence of Some Auxiliary Calculational Values on the Concentration of Metallic Inclusions

$\theta$	0,1	0,2	0,3	0,4	0,5	0,524
$c_{sp}$	0,576	0,726	0,832	0,915	0,985	1
$T$	1,74	1,38	1,20	1,093	1,016	1
$J$	0,9187	1,625	3,0	5,909	20,845	—

TABLE 2. Values of the Function  $f(\theta) = \epsilon_{av}/\epsilon_0$  Calculated from Eqs. (8), (9), (11), (12), and (14)

$\theta$ , vol. %	10	30	40	50	70	90
Eq. (8)	1,576	3,37	8,919	32,54	—	—
Eq. (9)	1,333	2,285		4,0	8,0	28
Eq. (11)	1,184	1,935		3,368	7,38	26,76
Eq. (12)	1,376	3,25	7,0		—	—
Eq. (14)	1,372	3,43		8,0	37	1000

To simplify the problem we assume that only lines of force with  $\alpha_1$  not more than  $45^\circ$  contribute to the capacitance, i. e., the other lines of force are associated with leakage, which we neglect. Then

$$C' = \frac{k\epsilon_0\pi d}{2} \int_0^{\pi/4} \frac{\sin^2 \alpha \cos \alpha d\alpha}{[(l+d)/d] \alpha - \sin \alpha} = \frac{k\epsilon_0\pi d}{2} \int_0^{\pi/4} \frac{\sin^2 \alpha \cos \alpha d\alpha}{T\alpha - \sin \alpha},$$

where

$$T = \frac{l+d}{d} = \frac{(1-nd)/(n+1) + d}{d} = \frac{1-c_{sp}}{c_{sp}} + 1. \quad (5)$$

Let  $\int_0^{\pi/4} \frac{\sin^2 \alpha \cos \alpha d\alpha}{T\alpha - \sin \alpha} = J$ ; then

$$C' = \frac{k\epsilon_0\pi d J}{2}. \quad (6)$$

We now find  $c_{sp}$

$$\theta = \pi n^2 d^3 / 6; \quad c_{sp} = \sqrt[3]{1.91\theta}. \quad (7)$$

Results calculated for  $c_{sp}$  and  $T$  for various volume concentrations of inclusions are shown in Table 1, together with values of the integral  $J$  found by a numerical method. Then

$$C_{col} = k\epsilon_0\pi d J / 2; \quad C_M = n^2 C_{cal} = \frac{k\epsilon_0\pi d n^2 J}{2(n+1)} \approx \frac{k\epsilon_0\pi c_{sp} J}{2};$$

$$C = C_M + C_d = k\epsilon_0 (1 - 0.785c_{sp}^2 + \pi J c_{sp} / 2).$$

Comparing this with Eqs. (1) and (2), we have finally

$$\epsilon_{av} = \epsilon_0 (1 - 0.785c_{sp}^2 + \pi J c_{sp} / 2) = \epsilon_0 f(c_{sp}). \quad (8)$$

Thus, by means of Eqs. (7) and (8) the average dielectric permittivity of a two-phase composite system can be calculated as a function of the parameters of the matrix phase and the concentration of metallic inclusions.

Note that it is impossible to calculate  $\epsilon_{av}$  for concentrations  $\theta$  above 0.524. In fact, for inclusions arranged in a simple cubic lattice, the maximum concentration of the inclusions (when the spheres are touching) is

$$\theta_{max} = V_{sp} / V_{cub} = \pi d^3 / 6d^3 = 0.524.$$

In other words, using Eq. (8) it is possible to calculate the average dielectric permittivity of the system up to the maximum concentration of inclusions which is permissible in the model assumed. For closest packing

of the metallic spheres, the limiting concentration rises to 0.74 [17], but this is to change the model of the system and hence also the results obtained.

In [10], a formula for  $\epsilon_{av}$  was derived for spherical inclusions of a second phase; for metallic spheres it takes the form

$$\epsilon_{av} = \epsilon_0 (1 + 2\theta)/(1 - \theta). \quad (9)$$

It applies for concentrations in the range from 0 to 1 but, as is evident from what has been said, this is a shortcoming. The formula of [9] is said to apply to both cubic and spherical inclusions:

$$\epsilon_{av} = \epsilon_0 \left[ 1 + \frac{\theta}{(1 - \theta)(3 + \epsilon_0)/(\epsilon_1 - \epsilon_0)} \right]. \quad (10)$$

It can be rearranged into a form identical to Eq. (9), which means that, like the formula in [11],

$$\epsilon_{av} = \epsilon_0 \left[ 1 - c^2 + \frac{c^2}{1 - c} \right], \quad c = \sqrt[3]{\theta}, \quad (11)$$

it can be used for inclusion concentrations between 0 and 1 only in the case of cubic inclusions with parallel faces (since, as the concentration of inclusions increases toward 1, the dielectric layer between the cubes may be as thin as required, right up to contact between the metal particles).

Other formulas were proposed in [12-14] for the calculation of the dielectric constants of emulsions and suspensions of conducting spherical particles (drops of aqueous salt solution dispersed in oil [12], mercury and soot conglomerates in oils and paraffins [13-14]); in [12]

$$\epsilon_{av} = \epsilon_0 \frac{1 + \theta}{1 - 2\theta}; \quad f(\theta) = \frac{\epsilon_{av}}{\epsilon_0} = \frac{1 + \theta}{1 - 2\theta}, \quad (12)$$

in [13] (after reducing it to a form convenient for calculations)

$$\epsilon_{av} = \epsilon_0 + \frac{\theta(2 + \epsilon_0)}{1 - \theta}, \quad (13)$$

also in [15] the "mixing rule"

$$f(\theta) = 1/(1 - \theta)^2, \quad (14)$$

and in [18] the conductivity equation

$$\sigma/\sigma_0 = (-\pi/2) \lg(\pi/6 - \theta) + \dots \quad (15)$$

The virtue of the formulas in [12, 14, 15] is that they may be reduced to an explicit relation between  $f(\theta)$  and  $\theta$ , i.e., they are expressed in the form  $\epsilon_{av}/\epsilon_0 = f(\theta)$ . The main shortcoming of Eq. (13) is that it does not reduce to such a simple and convenient form. Like Eq. (8) derived in the present work, Eqs. (12) and (15) are valid up to concentrations of 0.5, which corresponds to a system with spherical conducting inclusions (the case of simple cubic packing of the particles). But the concentration limit of Eq. (12) is somewhat lower than that of Eq. (8), while Eq. (15) neglects the conductivity of a portion of the dielectric matrix, which reduces its accuracy.

Table 2 presents values of the function  $f(\theta) = \epsilon_{av}/\epsilon_0$  calculated from Eqs. (8), (9), (11), (12), (13), and (14). In these equations  $\epsilon$  should be taken to denote both the dielectric permittivity and the electrical conductivity. It is assumed that in the case of electrical conductivity (the current problem), the metal always has infinitely large conductivity. Unless the difference between the conductivity of the dielectric and the inclusions is very large (more than two orders of magnitude), it is necessary to take the finite values of the metal and dielectric permittivity explicitly into account.

If the ratio between the resistivities of the metal and the dielectric is more than 100, the complication involved is not justified, and Eq. (8) provides sufficient accuracy.

Analysis of the data of Table 2 shows that Eq. (12) gives the results closest to those of Eq. (8).

As an experimental verification of Eq. (8), the dielectric permittivity of glass-metal samples (CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> glass and Ni) was determined. The samples were obtained by wet vibrogrinding of the mixture,

dry pressing with binder (paraffin, polyvinylbutyral), and sintering in a hydrogen tubular furnace at 900-1000°C. The content of metal was 6.2 and 9.1 vol.%. The measurements were carried out on an E9-4 Q-meter at a frequency of 1 MHz. The values of  $\epsilon_{av}$  obtained were 9.75 and 10.3, while for samples of the glass alone sintered in the same conditions the value of  $\epsilon$  obtained was 10.6 (this high value of  $\epsilon$  for a nonalkali glass is due to the incomplete removal of the binder in the H<sub>2</sub> medium and the appearance of free carbon). The calculated values of  $\epsilon_{av}$  for these samples were 7.78 and 8.25, which are 15-20% lower than the experimental values. The discrepancy may be the result of the following factors: 1) The model assumed for the system is simplified: in reality, the metallic particles may be far from spherical in form and in addition their positions are not strictly ordered but chaotic; 2) in Eq. (8) the first part, corresponding to the capacitance and conductivity of the volume of the sample that does not contain inclusions [see Eq. (2)], is known to be too high, and the second part is also not sufficiently accurate: the choice of  $\alpha_1 = \pi/4$  as the upper limit of integration is to some extent arbitrary.

It is also necessary to bear in mind two factors that have opposite effects on the experimental value of  $\epsilon$ : The presence of a small amount of carbon in the sample after sintering in hydrogen increases the value of  $\epsilon$ , while the presence of porosity reduces it. As the same time, a muffle furnace with an oxidizing gas medium cannot be used, because of the risk of oxidizing the metal particles.

What has been said above leads us to conclude that in the case of high concentrations of spherical inclusions, Eq. (8) is preferable to the other formulas, if only because of its consistency. For inclusions of complex shape, it is necessary to introduce a correction. It is very desirable to verify the proposed model by other experimental methods, for example, using an electrolytic bath, but this lies outside the scope of the present work.

Note that, for simplicity, phenomena complicating the flow of a current through the dielectric (accumulation of space charge and distortion of the electric field, intersurface polarization, etc.) were neglected in deriving the formulas in the present work. These factors must be taken into account, in particular, in calculating the dielectric permittivity and electrical conductivity of a dielectric with inclusions in the case of a variable electric field at low and medium frequencies.

#### NOTATION

$d$ , diameter of the spherical particles of metal;  $n$ , number of inclusions over the length of the cubic sample;  $c_{sp}$ , volume constant of the two-phase system;  $C$ , electrical capacitance of the sample or its individual parts;  $\epsilon$ , dielectric permittivity;  $\alpha$ , angle between the tangent to the lines of force and the axis joining the centers of two adjacent inclusions;  $R$ , radius of a line of force;  $l$ , distance between inclusions;  $\theta$ , volume content of metallic phase.

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## RESISTANCE TO MOTION OF BODIES IN A FLUIDIZED BED

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UDC 621.785:66.096.5

The force resisting the motion of bodies of revolution in a horizontal direction in a fluidized bed is measured. A generalization of the experimental data is used to obtain a relationship estimating the resistive forces arising in a fluidized bed for flow around obstacles.

In industrial apparatuses, fluidized beds are used to wash around a variety of bodies (heat-exchanger surfaces, pipes for distributing reagents, immobile sediments, sensors, and so on). In some kiln constructions components being heat treated are moved through a fluidized bed. In all cases, in order to calculate the strength of the mountings, supports, and other structural elements, one requires information on the forces arising from the flow around bodies of a fluidized bed. The data published in the literature on this topic are very limited and relate to the average vertical forces acting on fixed model bodies immersed in the fluidized bed [1-3]. At the same time, as correctly noted in [1, 3], the instantaneous forces arising from the flow around a body of a dispersed material that is transported upward in the wake of a bubble are much greater than the average forces, in some cases by more than an order of magnitude. In order to be able to calculate the instantaneous forces in a fluidized bed, we need to know, besides the velocity of ascent of the bubbles, the dependence of the force on the velocity of motion the surrounding medium, on the characteristics of the medium, and on the size and shape of the body around which the medium is flowing. The present paper reports an experimental study of this question.

The experiments were performed in a column of cross section  $275 \times 70$  mm which had a gas-distributing grid made from a sheet of felt of thickness 6 mm. The dispersed material was three fractions of quartz glass ( $d = 0.15, 0.23, \text{ and } 0.63$  mm;  $u_0 = 4, 6, \text{ and } 32$  cm/sec) and one fraction of silica gel ( $d = 0.19$  mm;  $u_0 = 2$  cm/sec). Fluidization was by means of air at room temperature for  $N$  from 1 to 5. The initial height to which the column was filled was 29 cm. A dynamometer sensor was moved backward and forward along the horizontal (more accurately, along an arc of radius 45 cm) by means of a special crank/connecting rod

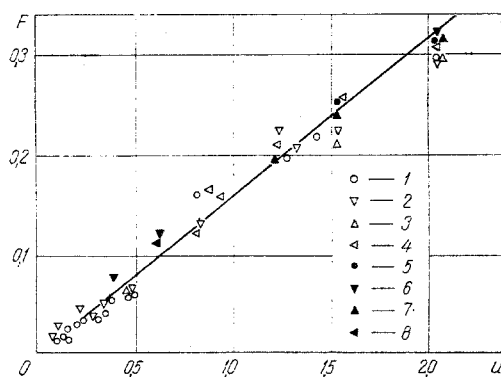


Fig. 1. Dependence of force of resistance  $F$  (N) for a sphere on its velocity of motion  $u$  (m/sec) in a fluidized bed of quartz sand  $d = 0.23$  mm: points 1, 2, 3, and 4 correspond, respectively, to fluidization numbers 1.2, 2, 3, and 5 for a height above the grid of 125 mm. Points 5, 6, 7, and 8 correspond to fluidization numbers 1.2, 2, 3, and 5 for a height above the grid of 225 mm.

Translated from *Inzhenerno-Fizicheskii Zhurnal*, Vol. 31, No. 6, pp. 1008-1012, December, 1976.  
 Original article submitted April 21, 1976.

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