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PROCESSES OF HEAT AND MASS TRANSFER IN POROUS MEDIA

MODELING OF THE STRUCTURE OF A BULK LAYER

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The characteristics of porous layers of a dispersed material have been investigated by the method of computer modeling. A linear growth in the porosity with increase in the adhesive force has been revealed. The statistical distribution functions of the size of pores and their shape have been computed and analyzed for different values of the adhesion forces. The distinctive features of the orientation of pores for different adhesive forces have been noted.

In technical practice, the layers of a dispersed material are formed almost without exception in the course of the processes of charging, settling, or filtration of a suspension that are statistical in nature. Thus, the layer structure is randomness-dependent and must be described by means of probability theory. One can only make statements on certain characteristics of the average state of the layer, for example, the layer porosity.

The experiments show that, for fairly large particles of a definite shape (for example, spheres), the volume porosity ε_v is, on the average, a constant in the case of satisfaction of certain conditions for the dimensions of the layer: the height of the layer is not too large so that the gravity force does not cause its compression by forcing particles into the layer matrix; compaction in formation of the layer is minimum; the dimensions of the layer are large as compared to the particle size. The fact that the volume porosity of the layers formed from particles is in the interval 0.36–0.44 even for the most elementary geometric particles demonstrates that these conditions are difficult to control [1–5].

Porosity, though undeniably the most important characteristic of the layer structure, which finally determines the hydraulic resistance to the liquid flow through the layer, is not the only quantity of interest. The pore size and shape are also important characteristics, particularly for problems of catalysis and chemical engineering [4–6]. All these characteristics are difficult to theoretically predict. Significant results have recently been obtained in the works of A. P. Mozhaev [7, 8].

Additional difficulties are presented by the fact that physical effects and primarily the force of cohesion between particles, which grows in importance as their size decreases, play a large role in organization of the structure apart from the geometric factors (particle size and shape) [4, 5]. Thus, for glass spheres, the measured values of volume porosity increase from 0.4 to 0.8 as the particle size changes from 160 to 10 μm. Other examples of the influence of the forces of interaction of particles are the dependence of the sediment density on the layer height, the particle size, and the chemical composition of the liquid phase and the dependence of the filtered layer of a solid material on the pressure difference in a filter flow [9–12].

To illustrate the process of formation of the structure of a bulk layer from spheres it is more convenient to consider a two-dimensional analog — coverage of a plane surface with circles. Such a formulation makes it possible to determine the surface porosity ε_s. We note that the circles are considered here as projections of spheres lying on the plane. The centers of the spheres are equidistant from the projection plane (within the distance of their radius). Such a consideration differs from the regular formulation [6, 7] in which one analyzes the porosity of a charge of an

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arbitrary section on the plane. In this case, circles of different diameter will be found on the cut plane by virtue of the random arrangement of the spheres.

A relationship between ε_s and ε_v can be established in the following manner. Let a cube with a side equal to unity be filled with spheres of radius $R \ll 1$. The packing will be considered to be random. If the number of spheres in the cube is N_s, the volume porosity will be considered as

$$\varepsilon_v = 1 - \frac{4}{3}\pi R^3 N_s . \tag{1}$$

We mentally arrange these spheres uniformly at a constant distance between the centers of neighboring spheres $2L$, so that

$$\frac{4}{3}\pi L^3 N_s = 1 . \tag{2}$$

Let us pass a section parallel to one plane of the cube. On the surface of such a section, there are N_s spheres. The distance between the centers of two neighboring spheres is equal to $2L$; we have

$$\pi L^2 N_s = 1 . \tag{3}$$

The surface porosity will be found as

$$\varepsilon_s = 1 - \pi R^2 N_s . \tag{4}$$

Expressions (2) and (3) yield

$$N_s = \frac{4}{3} L N_v . \tag{5}$$

Then (4) will be written in the form

$$\varepsilon_s = 1 - \frac{4}{3}\pi R^2 L N_v . \tag{6}$$

From (1) and (6), we obtain

$$\frac{1-\varepsilon_v}{1-\varepsilon_s} = \frac{R}{L} . \tag{7}$$

Considering now one sphere of radius R in the spherical volume of radius L, we have

$$1 - \varepsilon_v = \left(\frac{R}{L}\right)^3 . \tag{8}$$

From (7) and (8), we obtain the final expression

$$\varepsilon_v = 1 - (1-\varepsilon_s)^{3/2} . \tag{9}$$

Formula (9) has been obtained not in an exactly rigorous manner. In particular, it is easy to note that spherical volumes of radius L partially fill the cube and the circles of radius L do not cover the entire section selected. This circumstance is equally disregarded in the normalization equalities (2) and (3), which leads to a compensation in equality (5). According to formula (9), the limiting impracticable cases of absolutely close ($\varepsilon_v \to 0$ simultaneously with $\varepsilon_s \to 0$) and absolutely loose ($\varepsilon_v \to 1$ and $\varepsilon_s \to 1$) packings are fulfilled. When the construction is different, we obtain dependences analogous to (9) in structure but with a certain factor of $(1-\varepsilon_s)^{3/2}$. For example, when cubes are used

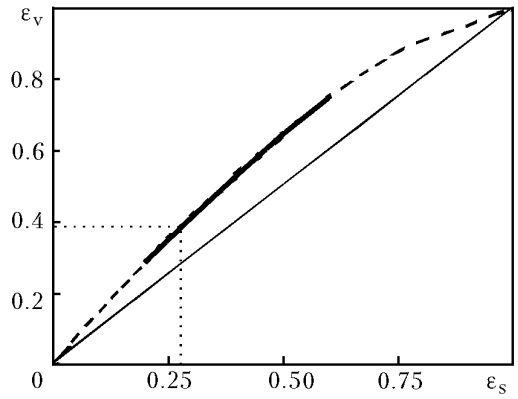


Fig. 1. Relationship between the surface and volume porosities.

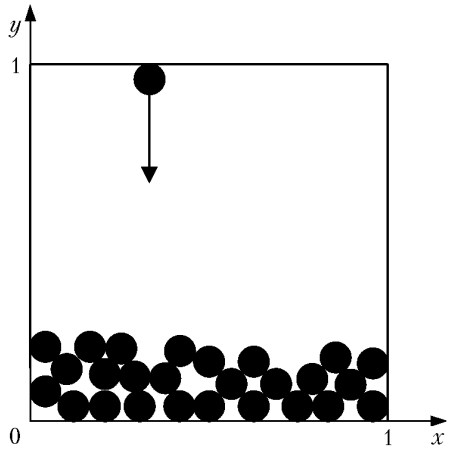


Fig. 2. Modeled situation.

instead of the spherical volumes in (2) and (3), this coefficient is equal to 0.75. The limiting transition $\varepsilon_v \to 0$ for $\varepsilon_s \to 0$ is absent. The curve of ε_v as a function of ε_s in accordance with (9) is shown in Fig. 1, where the portion of practical interest is shown by the semiheavy line. The straight line $\varepsilon_v = \varepsilon_s$ demonstrates the equality of the volume and surface porosities [3, 4, 6] in the arbitrary section of the charge.

The derivation of (9) assumes the uniformity and isotropy of the packing in the statistical meaning, i.e., the constancy of the characteristics of the packing on scales much larger than the sphere size. This property, as will be shown, may not be filled if the adhesive force acts between the particles. The monodispersity of spheres adopted in derivation is another important property. The subsequent consideration is restricted to this case.

The two-dimensional picture reflects, if not completely, the layer structure in a three-dimensional representation, much as the representations of the layer structure follows from an analysis of the picture of the layer sections.

In what follows, we will describe the algorithm of the program and the results of computer modeling.

Algorithm. The imitation system consists of a square with a side equal to unity and a set of equally large circles of prescribed diameter that are characterized by their position in the cube. The circles fall singly in the square until they hit the lower side of the cube (at the beginning of filling of the square) or the surface of the layer already formed. The circles once stopped cannot be set in motion. The coordinate of the origin of a falling circle is (x, 1), where x is a random uniformly distributed quantity between 0 and 1 (Fig. 2). The coordinate of a freely falling circle is $y = v_y t$, where the rate of fall v_y (it is implied that the fall occurs by the action of the mass force, just as in sedimentation) is selected together with the time step Δt such that the inequality $v_y \Delta t << 1$ is fulfilled and a certain calculation accuracy is ensured.

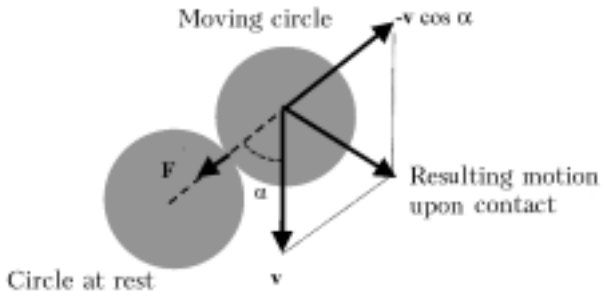


Fig. 3. Motion of a circle upon contact with the circle already settled.

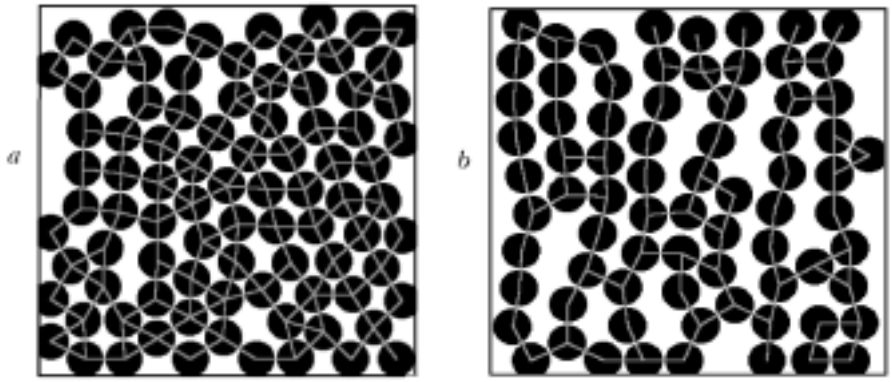


Fig. 4. Layer structures made of circles and formed for different values of the dimensionless adhesion coefficient: a) k_{ad} = 0 and b) 0.5.

If the circle comes against an obstacle in its motion, its trajectory is corrected in a certain manner (Fig. 3). Let the moving circle have a momentum $m\mathbf{v}$ before its contact with an immobile circle. Upon collision, it acquires the momentum $-m|\mathbf{v}|\cos\alpha$ along the line of centers of the two circles in the direction from the center of the circle at rest and continues its motion in the direction of the vector that is a result of the summation of the vectors **v** and **v** cos α, as is shown in Fig. 3. The forces of cohesion between the circles **F** are assumed to begin to act at the instant of contact of the two circles in question. For the moving circle to separate from the circle at rest the momentum acquired by the moving circle must overcome this force, i.e., the condition

$$k_{ad} = \frac{|\mathbf{F}|\,\Delta t}{m\,|\mathbf{v}|} \tag{10}$$

must be satisfied.

For a prescribed cohesion force and parameters of motion of a particle this relation determines the limiting value of the slip angle required for further motion of the circle. We call $k_{ad} = \cos\alpha$ the adhesion parameter of the dimensionless coefficient of cohesion. In the course of computer experiments, we can vary the value of cos α itself directly, thus changing the relation between the cohesion force and the momentum of the particle.

If it crosses the region that is already occupied by the circles settled earlier after the described correction of the circle's trajectory, this means that the motion of the circle ceases and the circle in question is involved in the community of the settled circles. Thereafter the next circle is set in motion on the upper side of the square. The process continues until the entire square is filled.

The program realizing the algorithm described has been composed in the C Builder 5.0 (C++) language.

Quantities Computed. ***1. Porosity***. The porosity has been calculated by subtraction of the area of all the circles filling the square from the area of the square itself.

2. Computation of the Laws of Statistical Distribution of the Parameters of Pores. In establishing the laws of statistical distribution of the parameters of pores, we must primarily identify each pore. For this purpose we con-

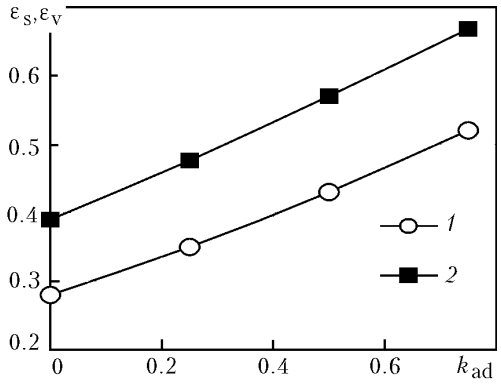


Fig. 5. Values of the surface (1) and volume (2) porosity vs. adhesion parameter.

struct a plane graph connecting the centers of the contacting circles. Then we carry out the following procedure: we take one node of the graph (center of one circle) and, beginning with the last node on the right, trace counterclockwise from one node to another continuously until we meet the starting node. This means that the boundary is traced and the pore is determined. The nodes making up the boundary checked are eliminated from the number of selected nodes as the starting ones for the next tracings. For control we repeat the same procedure with change in the direction of tracing, then we take the next possible node, etc. We include in the statistics only the internal pores, i.e., those not having any side of a rectangle as one of its boundaries.

(1) The perimeter of the pores is computed by summation of the lengths of all the straight lines that connect the centers of particles forming a pore. This procedure yields a result somewhat differing from the true value of the perimeter but considerably simplifies the computation. For a symmetric pore from six circles of radius R, the program will compute a value of $12R$ (instead of the true value equal to $4\pi R$). Such an accuracy is considered to be satisfactory.

(2) The pore area is computed by subdivision of the area of a polygon formed by the straight lines of the graph into triangles whose area is computed based on the lengths of two sides and the angle between them. If any interior angle of the graph is larger than π, for this angle a triangle from the sides is eliminated from consideration of the pore area.

(3) The parameter of the pore shape is computed based on the following determination: $Sh = 2\sqrt{\pi S}/P$. For a circle we have $Sh = 1$.

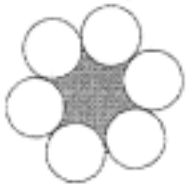
Computation Results. The statistics of distribution of the characteristics of pores has been established in a twofold manner. First, we processed all the pores resulting from the realization of filling of the square; second, we carried out a fairly large number of realizations. The number of the necessary realizations was established from the requirement that the statistical straggling (confidence interval with a probability of 95%) be no higher than 2 to 3%. Computer experiments show that the number of necessary realizations depends on the coefficient of adhesion of particles: the sufficient number of replications of the experiment is no larger than 50 for $k_{ad} = 0$ and increases to 100 for $k_{ad} = 0.5$ or higher.

Another important parameter is the particle size. It must be fairly small as compared to the square length, equal to unity. Experience shows that, for particles with a radius smaller than 0.025, further decrease in the particle size does not cause the statistical characteristics of the layer to change. It is precisely with these adjusting parameters that the calculations have been performed.

Figure 4 gives examples of a random arrangement of circles in the square for two values of the adhesion coefficient. The packing of circles is closer for $k_{ad} = 0$ than that for $k_{ad} = 0.5$. Porosity increases with the force of cohesion between particles. The dependence of the porosity on k_{ad} is shown in Fig. 5.

The quantity k_{ad} influences not only the porosity but also other characteristics of the layer structure. Figure 6 compares the distribution histograms of the number of joints n (it is equal to the number of circles forming the pore), the shape factor of the pore Sh, and the reduced area of the pores. These results demonstrate that the distributions of different parameters are much more diffuse in the layer with mutually cohesive particles than in the case of adhesion-neutral particles.

TABLE 1. Characteristics of Certain Ideal Pores

Form of a pore	S	P	Sh
	$R^2\left[\sqrt{3}-\frac{\pi}{2}\right]=0.16R^2$	πR	0.45
	$R^2[4-\pi]=0.86R^2$	$2\pi R$	0.52
	$R^2\left[6\sqrt{3}-\pi\right]=7.25R^2$	$4\pi R$	0.57

Discussion of the Computation Results. Certain characteristics of the simplest pores are given in Table 1 for analysis of the results of computer modeling. The graphs of the given pores are regular polygons. Consequently, we can classify the pores as regular or ideal ones. Pores of such kind can be found in a sufficient number in the layer structure for $k_{ad} = 0$. Pores formed by three circles correspond to the packing with $\varepsilon_s = 0.1$. The area of an individual pore in relation to the area of a circle (normalized area) is only 0.05. A square (or cubic in three-dimensional representation) packing has $\varepsilon_s = 0.21$ and a normalized area of 0.27. The probability of a hexagonal packing (case $k_{ad} = 0$) must be very low, since the pore area in this case exceeds the circle area more than twice. Accordingly we have $\varepsilon_s = 0.7$ in such a packing. The shape factor grows in such ideal pores (it is equal to unity in the limit) with the number of particles.

The modeling shows the presence of a small number of pores formed by six circles (approximately 6% of their total number for the case $k_{ad} = 0$ and 12% for $k_{ad} = 0.5$) or even by seven circles (of the order of 2% of the total number for $k_{ad} = 0$ and of 8% for $k_{ad} = 0.5$). Consideration of the pictures of different realizations (for example, in Fig. 4) shows that these pores are far from regular ones in shape. They are often curved or extended along the direction of fall of the circles (y axis). A rectangle with sides a and b with the shape index

$$Sh = \sqrt{\pi}\,\frac{\sqrt{b/a}}{1+b/a}$$

would be an appropriate geometric shape for such pores.

We have $Sh \to 0$ in the case of strongly extended pores when $b/a \to 0$ or $b/a \to \infty$. Therefore, it is not surprising that the shape factor for $k_{ad} = 0.5$ is virtually equally frequent in the range between 0 and 1.1. For the case $k_{ad} = 0$ the shape factor is mainly concentrated in the region between 0.7 and 1.1.

Mutually cohesive circles are characterized by the possibility of forming pores from a large number of circles, up to 12. The maximum number of pores is formed from five particles, whatever the value of the coefficient k_{ad} is. The value of the porosity is virtually in proportion to k_{ad} (see Fig. 5). An increase in the porosity with adhesive force is a well-known fact embodied in the theory and practice of flocculation. This effect has been investigated theoretically and experimentally from the viewpoint of the structure of filter beds and cakes [7–10].

For adhesion-neutral particles, we have $\varepsilon_s = 0.28$. Based on formula (9), this corresponds to a value of $\varepsilon_v = 0.39$ lying between the bounds of the measured values of volume porosity. The porosity values for other values of the coefficients k_{ad} have been computed from formula (9) and are given in Fig. 5.

The quantity k_{ad} also has an effect on the distribution of the pore areas (Fig. 6). The maximum number of the pores (32%) has an area equal to $0.25\pi R^2$ if $k_{ad} = 0$, and one-third of all the pores (also the maximum number)

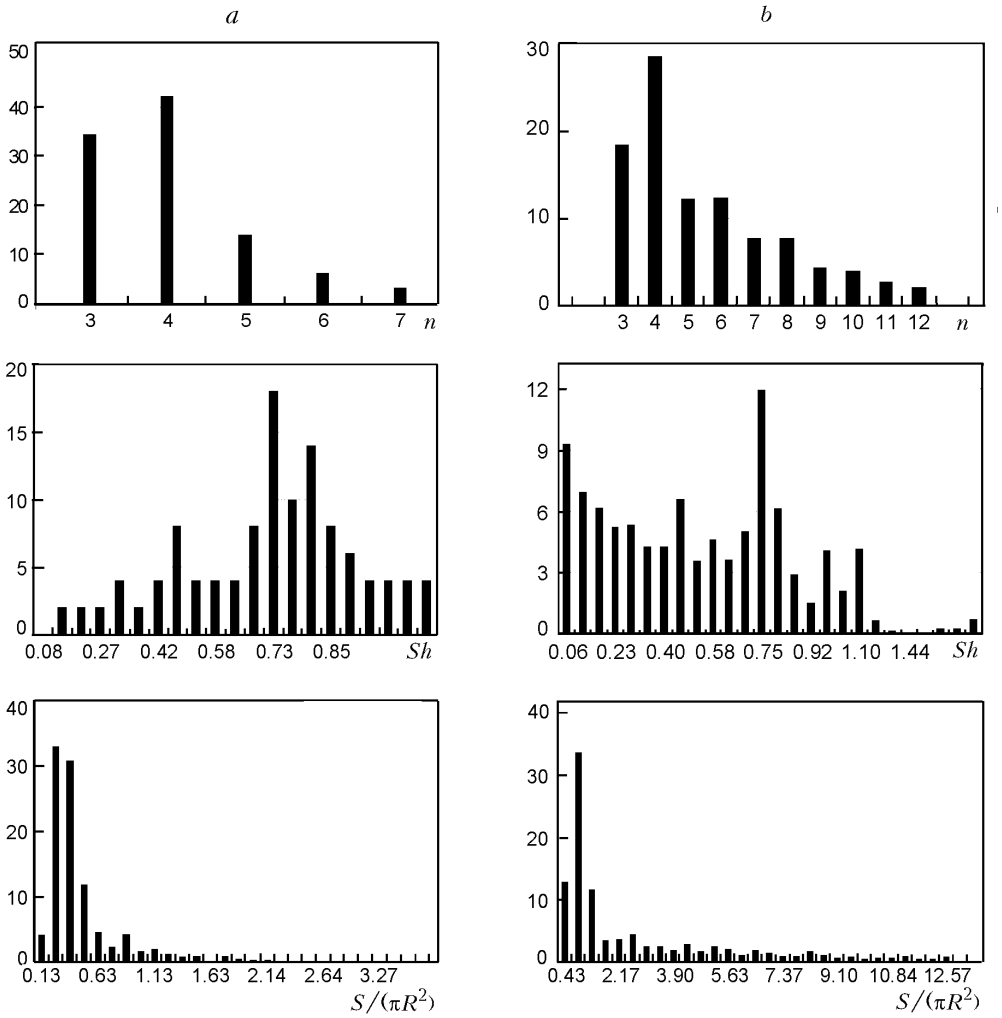


Fig. 6. Distribution histograms of different parameters: a) k_{ad} = 0 and b) 0.5. The fraction of pores of a given form is plotted on the vertical axis in percent.

has an area equal to $0.86\pi R^2$ for k_{ad} = 0.5. The distribution of the pore areas becomes wider with increase in k_{ad}. Figure 7 shows the distribution functions of the normalized pore areas D_s and the parameters of shape D_{sh} that have been composed based on the histograms of Fig. 6. In considering these distributions, we are interested in the following details:

1. The lower bound of the normalized pore areas is finite, i.e., the domain of definition of the distribution function is bounded on the left not by zero. The larger k_{ad}, the higher the lower value of a possible pore area. An analogous remark is also true for the maximum possible area. We observed no pores of area larger than $3.64\pi R^2$ for k_{ad} = 0 and larger than $12.57\pi R^2$ for k_{ad} = 0.5.

2. The value D_s = 50% for the case k_{ad} = 0 is attained when $S = 0.31\pi R^2$ and for k_{ad} = 0.5 when $S = \pi R^2$. Thus, the higher the adhesive force, the larger the average pore size. The distribution width can be determined by the relation of the values of the argument for D_s = 75 and 25%. It is approximately equal to 2.5 for k_{ad} = 0 and nearly to 6.4 for k_{ad} = 0.5.

3. The shape factors have nearly equal values (Sh = 0.1 for k_{ad} = 0 and Sh = 0.06 for k_{ad} = 0.5) as their lower bound, but the probability that such low values will be attained by mutually cohesive circles is higher than that for adhesion-neutral ones. The shape factor Sh for the case k_{ad} = 0 does not exceed a value of 1.2, and values up to Sh = 1.6 are observed for k_{ad} = 0.5.

4. For k_{ad} = 0, the values of the factor Sh are concentrated in the vicinity of ≅0.75 (here the value for which D_{sh} = 50% is attained), whereas for k_{ad} = 0.5 the distribution function is almost straight. This means that the shape

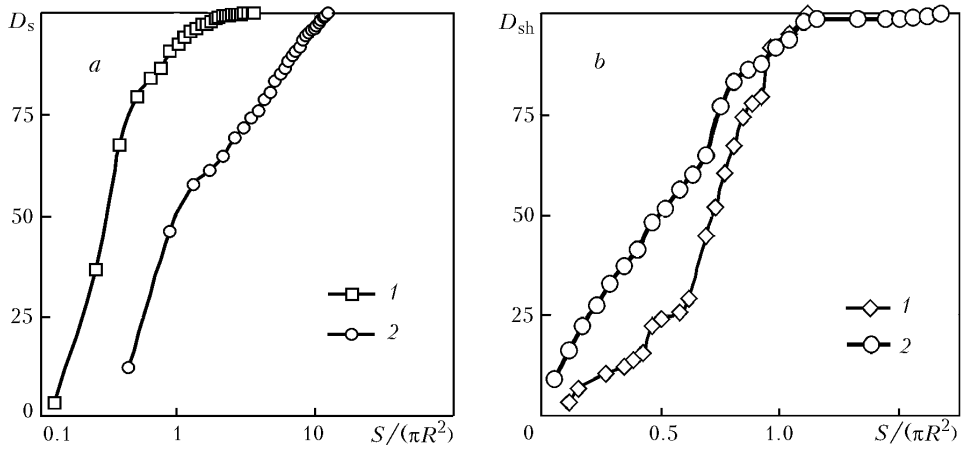


Fig. 7. Distribution functions of the relative pore areas (a) and the parameter of the pore shape (b): 1) k_{ad} = 0 and 2) 0.5.

factors in the interval between 0.06 and 0.75 are nearly equally probable in this case. The value D_{sh} = 50% is attained when Sh = 0.5. Just as above, the distribution width for D_{sh} can be determined by the relation of the argument values for D_{sh} = 75 and 25%. It will be equal to 1.33 for k_{ad} = 0 and will increase to 3.75 for k_{ad} = 0.5. These indices reflect the apparent fact that pores are farther from a circle in shape in the case of mutually cohesive circles than those in the case of noninteracting circles.

Many of the observations made here for a plane layer of circles are undeniably true for three-dimensional random packings, e.g., the volume porosity as a function of the adhesive force is a well-established fact correlating with the computer experiment presented, which needs to be extended to the three-dimensional case. Another line of investigation would be inclusion of the external influences on the layer — shaking, gravity, and others — into consideration.

CONCLUSIONS

1. We have developed and realized the method of computer modeling of random packings with allowance for the forces of cohesion between particles.

2. We have found the relationship between the surface and volume porosities.

3. It has been established that the porosity grows nearly in proportion to the adhesive force.

4. It has been shown that, when the adhesive force is large, pores acquire the shape of polygons extended in the direction of motion of particles.

5. We have found the statistical distribution functions of different characteristics of the pores.

6. It has been found that the distribution grows with adhesion force.

NOTATION

a and b, sides of the rectangle, m; D_s, distribution functions of the normalized pore areas, %; D_{sh}, distribution functions of the shape factors, %; **F**, vector of the adhesive force, N; k_{ad}, adhesion coefficient; L, half-distance between the centers of two neighboring spheres, m; m, mass of a circle, kg; n, number of pore nodes; N_s, number of spheres in the section; N_v, number of spheres in the cube; P, perimeter of a pore, m; R, radius of spheres or circles, m; S, pore area, m^2; Sh, parameter of the pore shape; t, time, sec; Δt, time step, sec; **v**, velocity vector, $m \cdot sec^{-1}$; v_y, component of the velocity vector along the y axis, $m \cdot sec^{-1}$; x, y, coordinates in the square, m; α, slip angle, rad; ε_v, volume porosity; ε_s, surface porosity. Subscripts: ad, adhesive; s, surface; sh, shape; v, volume.

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