

POROSITY AND HYDRAULIC RESISTANCE OF A BIDISPERSE MIXTURE

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UDC 539.6,539.217

The known nonmonotonic dependence of a bidisperse mixture porosity on the portion of the small fraction at arbitrary diameter ratios between small and large particles has been modeled. The expressions for the mixture porosity obtained in the course of modeling have been compared to the experimental data. With the use of the derived expressions for the mixture porosity, the hydraulic resistance of a layer from a bidisperse material has been calculated. It has been shown that in accordance with the experiment at a small ratio between the sizes of small and large particles the dependence of the hydraulic resistance of a bidisperse layer on the portion of the small fraction has a maximum which is higher than the resistance in the absence of large particles. The agreement between theory and experiment is quite satisfactory.

The characteristics of porous structures play an important part in natural phenomena (sediments) and in many fields of technology and nature (composite materials, heterogeneous catalysis, filtration of suspensions, transport and storage of granular materials, sediments). As a rule, the question is of irregular, static packings that naturally arise in the form of random ensembles of particles in the course of mixing, precipitation, crushing, and other natural and technological processes. Usually, to characterize them, one restricts oneself to two parameters: the porosity and the specific surface, which are used to estimate the other quantities of practical interest (e.g., pore size). The experiments show that for fairly large identical particles of a certain shape (e.g., balls) the volume porosity in the general case is, on average, a constant independent of the particle size [1–3].

The density of a random packing from particles with radii of two sizes r and R depends not only on the radii ratio r/R , but also on the ratio between the volumes of particles of these two components V_r/V_R . The sensitivity of the packing density to the composition shows up, of course, also for multicomponent mixtures, which is observed in experiments but is hardly amenable to theoretical forecast. It is natural to use statistical methods for studying such packings. Here an important role can be played by computer experiment [4–11].

As a starting point for constructing a model of a random multicomponent packing [12, 13], the expressions for the bidisperse mixture porosity are often used. The equations for the bidisperse system porosity ϵ_m have been derived by Jeshar [1, 14] for the case where $r/R \rightarrow 0$ (independently, the derivation was repeated in [15] with some development for the case of different porosities for the small and large fractions). For finite values of r/R , empirical corrections were applied to the Jeshar formulas [14, 15]. Below, we give the derivation for the porosity of a bidisperse mixture at an arbitrary value of r/R and V_r/V_R based on certain physical notions about the interaction of particles in the layer. Calculations confirm the known experimental facts of the nonmonotonic dependence of the hydraulic resistance of the layer of bidisperse particles on the portion of the small fraction.

Theory. A mixture of two kinds of particles with radii r (small fraction, volume V_r) and R (large fraction, volume V_R) forms a layer of volume V , with the total volume of particles in the layer being

$$V_p = V_r + V_R. \quad (1)$$

The porosity of such a mixture is

$$\epsilon_m = \frac{V - V_p}{V}. \quad (2)$$

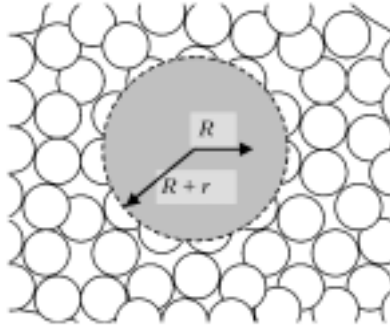


Fig. 1. Schematic representation of the cell in a bidisperse mixture with a predominant portion of small particles. The shaded circle schematizes the volume V_d with porosity ϵ_d .

It is assumed that both fractions, taken separately, have the same porosity ϵ_0 . Proceeding from the dimensional theory, we may expect the existence of the following function:

$$\frac{\epsilon_m}{\epsilon_0} = F\left(\epsilon_0, \frac{r}{R}, \frac{V_r}{V_p}\right). \quad (3)$$

Jeshar [1] calculated $\epsilon_m/\epsilon_0 = F(\epsilon_0, 0, V_r/V_p)$, i.e., without the variable r/R . Below, ϵ_m/ϵ_0 will be determined with account for this variable.

Following Brauer [1], it is convenient to consider separately the two limiting cases: $V_r/V_R \rightarrow 0$ and $V_r/V_R \rightarrow 1$.

The layer consists mainly of small particles. The number of large particles is so small that there are no contacts between them (Fig. 1). Each of the large particles together with a certain number of small particles around it forms a spatial region with a porosity other than ϵ_0 . As a result of this, the total volume V is divided into regions V_d and V_o with porosity values ϵ_d and ϵ_0 , respectively. From the definitions $V_d + V_o = V$, $V_d(1 - \epsilon_d) + V_o(1 - \epsilon_0) = V_p$, and $V_p = V(1 - \epsilon_m)$, follow the expression for the layer-average porosity

$$\epsilon_m = \epsilon_d \left(1 - \frac{V_o}{V}\right) + \epsilon_0 \frac{V_o}{V}. \quad (4)$$

To calculate the porosity ϵ_d , let us consider the vicinity near a large particle. Suppose that the region in which the influence of the large particle is strong is a sphere of radius $R + a_1 r$. Here a_1 is a constant of the order of unity, which will be estimated below. Taking this into account, the considered volume around the large particle is

$$V_d = V_R \left(1 + a_1 \frac{r}{R}\right)^3 \quad (5)$$

and, accordingly

$$V_o = V - V_d = V - V_R \left(1 + a_1 \frac{r}{R}\right)^3. \quad (6)$$

Hence

$$\frac{V_o}{V} = 1 - \frac{V_p}{V} (1 - \alpha) \left(1 + a_1 \frac{r}{R}\right)^3, \quad \alpha = V_r/V_p. \quad (7)$$

Substitution of (7) into Eq. (4) gives the following expression for the layer porosity:

$$\frac{\varepsilon_m}{\varepsilon_0} = \frac{1 - (1 - \alpha) \Psi}{1 - \varepsilon_0 (1 - \alpha) \Psi}, \quad (8)$$

where

$$\Psi = (1 - \varepsilon_d / \varepsilon_0) (1 + a_1 r / R)^3. \quad (9)$$

In calculating Ψ , the value of ε_d must be known. By definition,

$$1 - \varepsilon_d = \frac{R^3 + nr^3}{(R + a_1 r)^3}, \quad (10)$$

and n is the number of small particles in the disturbance region having, under the influence of the large particle, a packing density other than ε_0 . It can be estimated from the balance of the number of small particles neighboring the large one:

$$a_3 (R + a_2 r)^2 \propto nr^2 \quad (11)$$

with the additional constants $a_2 \approx a_1$ and a_3 . From Eq. (11) it follows that

$$n = a_3 \left(1 + \frac{R}{a_2 r}\right)^2. \quad (12)$$

On the other hand, for a monodisperse material n can be interpreted as a coordination number (number of contacts of an arbitrary particle with neighbors [1–3]) equal, on average, to 8. This leads to the estimate

$$\frac{a_3}{a_2} (1 + a_2)^2 \approx 8, \quad (13)$$

and the equation for ε_d takes on the form

$$\varepsilon_d = 1 - \frac{1 + \frac{a_3}{a_2} (1 + a_2 x)^2 x}{(1 + a_1 x)^3}, \quad x = \frac{r}{R}. \quad (14)$$

The requirement that for the monodisperse case ($x = 1$) $\varepsilon_d = \varepsilon_0$ be fulfilled permits estimating the constant

$$a_1 = \sqrt[3]{\frac{1 + \frac{a_3}{a_2} (1 + a_2)^2}{1 - \varepsilon_0}} - 1. \quad (15)$$

The porosity $\varepsilon_{d,R}$ in the layer adjoining a large particle can be calculated as a side result:

$$\varepsilon_{d,R} = 1 - \frac{\frac{a_3}{a_2} (1 + a_2 x)^2 x}{(1 + a_1 x)^3 - 1}. \quad (16)$$

Figure 2 presents the porosities ε_d and $\varepsilon_{d,R}$ as functions of the ratio between the radii of particles of the small and large fractions. The increase in the porosity $\varepsilon_{d,R}$ at $r/R \rightarrow 0$ can be interpreted as the known fact [1] that the fill-

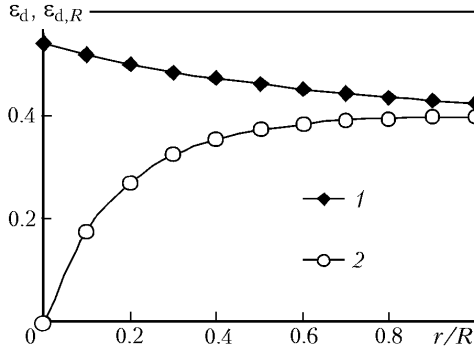


Fig. 2. Porosity values ε_d (1) and $\varepsilon_{d,R}$ (2) as a function of the diameter ratio in the bidisperse mixture.

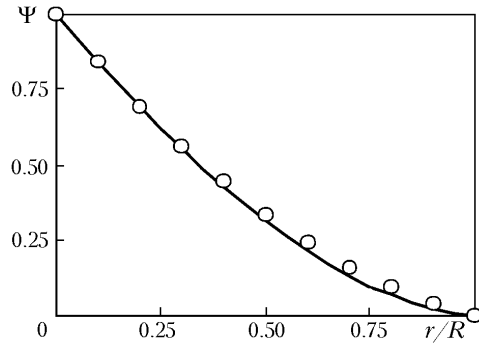


Fig. 3. Function $\Psi(x)$ (curve) and its parabolic approximation (dots).

ing porosity near a flat wall increases. In this case, expression (16) will become $\varepsilon_{d,R} = 1 - a_3/3a_1a_2^2$. The linear approximation in (14) as $x \rightarrow 0$ gives $\varepsilon_d = (3a_1 - a_3/a_2^2)x$, which as $r/R \rightarrow 0$ leads to

$$\Psi \approx 1 - \frac{3a_1(1 - \varepsilon_0) - a_3/a_2^2}{\varepsilon_0} x = 1 - ax. \quad (17)$$

The calculation of the function $\Psi(x)$ with the use of Eq. (9) is given in Fig. 3. It is expedient to propose a simple approximation of this function with account for the expansion of $\Psi(x)$ into a series. With sufficient accuracy (Fig. 3), we can write

$$\Psi = 1 - ax + (a - 1)x^2. \quad (18)$$

At $r/R = 0$ and $\Psi = 1$ Eq. (18) becomes the equation $\varepsilon_m/\varepsilon_0 = \alpha/(1 - \varepsilon_0(1 - \alpha))$ derived by Jeshar [14]. In the other limiting case where $r = R$, $\varepsilon_m = \varepsilon_0$ holds independent of the impurity content.

Small content of the small fraction. In this case, one should proceed from the fact that the change in the porosity may be due to two reasons:

- (1) particles of the small fraction fill the space between large particles;
- (2) in the course of mixing, small particles occupy the space between large ones and change the packing structure of the large particles.

In the case of very small sizes of added particles, item (1) is more preferable. But if the added particles are not so small compared to the particles of the main large fraction, then the moving of large particles apart by small ones (item (2)) will be the more significant, the larger the portion of small particles in the mixture.

Let the volume V be initially filled only with particles of the large fraction. In the absence of particles of the small fraction the volume will be equal to

$$V = V_{R,0}/(1 - \varepsilon_0), \quad (19)$$

where $V_{R,0}$ is the volume of large particles in the initial situation.

Let us consider two opposite situations that can be realized upon the addition of the small fraction:

1. $r/R \rightarrow 0$. In this case, the volume of large particles in the filled volume remains the same: $V_R = V_{R,0}$, since the small particles only fill the interparticle voids without changing the packing of the large particles.

2. $r \approx R$. The added particles are comparable to those that are already present in the volume V . This means that the introduction of new particles (impurity) requires removal of the particles that were in the volume before the mixing. Therefore, $V_R \approx V_{R,0} - V_r$ and the existence of a function of the following form is supposed:

$$V_R = F(V_{R,0}, V_r, r/R), \quad (20)$$

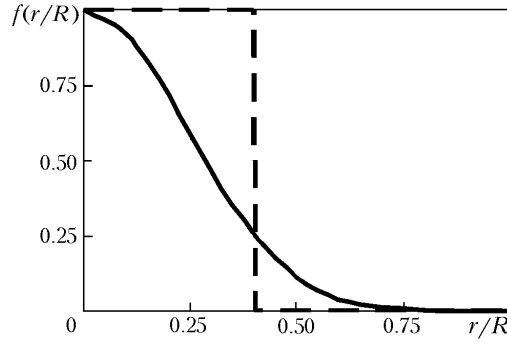


Fig. 4. Form of the function $f(r/R)$ in the ideal (dashed line) and real (solid line) representations.

This function describes the change in V_R upon the introduction of a small fraction of volume V_r . Such a function cannot be found analytically and will be introduced semiempirically.

First of all, we seek the probability function $f(r/R)$ for a small particle to penetrate into the packing without changing it. For the ideal packing of large particles, it would be a step function, as is shown by the dashed line in Fig. 4. For a random structure, the passage of the penetration probability equal to 1 to 0 will occur in some interval of the argument r/R (solid curve). Particles with r/R that is already small can be kept by the matrix of large particles. The probability of this should be proportional to the ratio between the surface of the midsection of the small particle and the area of the pore formed by large particles:

$$f \approx 1 - \left(\frac{r}{a_4 R} \right)^2. \quad (21)$$

Nonsharpness of the passage of $f(r/R)$ from 1 to 0 is achieved by reformulating (21) to the form

$$f \approx \exp \left(- \left(\frac{r}{a_4 R} \right)^2 \right). \quad (22)$$

With increasing r/R the new (small) particles can be built in the matrix only upon removal of large particles from it. Such a replacement of some volume of large particles by small ones should be the function $(r/R)^3$. Accordingly, at $r/R \rightarrow 1$

$$f \approx 1 - \left(\frac{r}{R} \right)^3. \quad (23)$$

The combination of formulas (22) and (23) yields

$$f \approx \left(1 - \left(\frac{r}{R} \right)^3 \right) \exp \left(- \left(\frac{r}{a_4 R} \right)^2 \right), \quad (24)$$

which reflects both of the above-mentioned limits.

An approximate value for $a_4 = 0.4$ can be determined if we take for estimations a regular cubic packing of large particles, for which the radius of small particles that get stuck in it is equal to $0.4R$. With the use of the function $f(r/R)$, the reduction of the volume V_R , containing both cases can be given in the form

$$V_R = V_{R,0} - V_r \left(1 - f \left(\frac{r}{R} \right) \right). \quad (25)$$

Combining of Eqs. (1) and (25) leads to the equalities

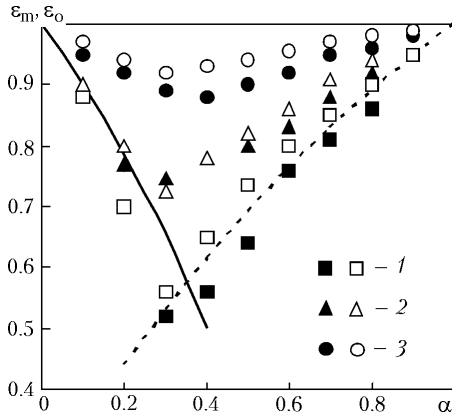


Fig. 5. Comparison of the calculated and measured porosity values for bidisperse mixtures of particles with different diameter ratios (full circles show Jeshar's data [14] and open ones — Furnas's data [17], the curves represent the calculated data): 1) $r/R = 0.1$; 2) 0.3; 3) 0.5.

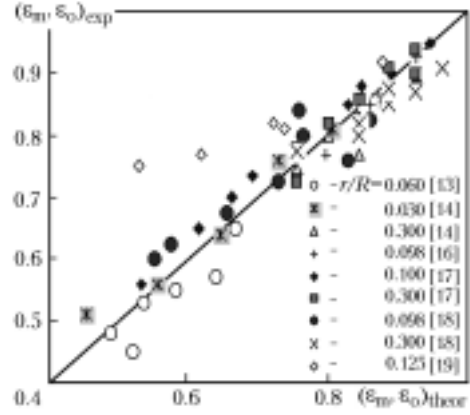


Fig. 6. Comparison of the theoretical porosity values of the bidisperse mixture to those obtained by different authors.

$$V_p = V(1 - \varepsilon_0) + V_p f(r/R) \quad \text{or} \quad 1 - \varepsilon_m = 1 - \varepsilon_0 + (1 - \varepsilon_m) \alpha f(r/R),$$

which yields the formula

$$\frac{\varepsilon_m}{\varepsilon_0} = 1 - \frac{\alpha f}{1 - \alpha f} \left(\frac{1 - \varepsilon_0}{\varepsilon_0} \right). \quad (26)$$

Jeshar's result [1] corresponds to the case where $r/R \rightarrow 0$, or otherwise, $f = 1$. If the added particles coincide with the large ones, i.e., $r = R$, then $f = 0$, and, consequently, $\varepsilon_m = \varepsilon_0$ independently of α .

Comparison to the Experiment. The summary of the formulas needed for mixture porosity calculations can be given in the form $\frac{\varepsilon_m}{\varepsilon_0} = \begin{cases} F_0, & F_0 > F_1 \\ F_1, & F_1 > F_0 \end{cases}$, and F_0 therewith is given (for $\alpha \rightarrow 0$) by formula (8) and F_1 (for $\alpha \rightarrow 1$) by formula (26). With the values of the three constants redetermined in the course of comparison to the experimental data, whose approximate estimates are given above, the functions used in F_0 and F_1 have the form

$$f \approx \left(1 - \left(\frac{r}{R} \right)^3 \right) \exp \left(- \left(\frac{r}{0.36R} \right)^2 \right), \quad \Psi = 1 - 1.55x + 0.55x^2.$$

With the use of the formulas derived, the dependence $\varepsilon_m(\alpha)/\varepsilon_0$ has been calculated. The corresponding curves for various values of r/R are given in Fig. 5. The $\varepsilon_m(\alpha)/\varepsilon_0$ curves that ascend with increasing α originate from formula (8), and the descending ones originate from formula (26). At some $\alpha = \alpha_s$ the curves intersect. At this point, the range of applicability of both formulas changes. It is seen that α_s weakly depends on r/R . At $r/R = 0$, one can calculate $\alpha_s = \varepsilon_0/(1 + \varepsilon_0)$ and extend this to arbitrary values of r/R . The same figure shows Jeshar's [14] and Furnas's [17] measurement data. The agreement between the theoretical and experimental data is good.

Figure 6 compares the theoretical values and those measured by different authors [12–18] for the porosity of bidisperse mixtures. The statistical straggling of the results presented in different literature sources does not reveal any systematic tendency. The only exceptions are the measurements [19], where in the experiments the authors used not balls, but particles of complex form, which, having one and the same size, gave a porosity equal to 0.32.

Hydraulic Resistance. The dependence of the porosity on the small fraction strongly influences the change in the hydraulic resistance to the filtration flow. It is known [1, 3] that the hydraulic resistance is proportional to some

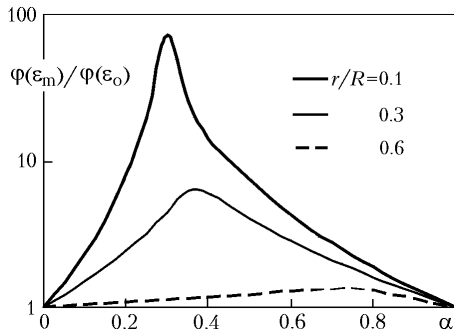


Fig. 7. Changes in the porosity function depending on the portion of small particles at various diameter ratios between particles in the bidisperse mixture.

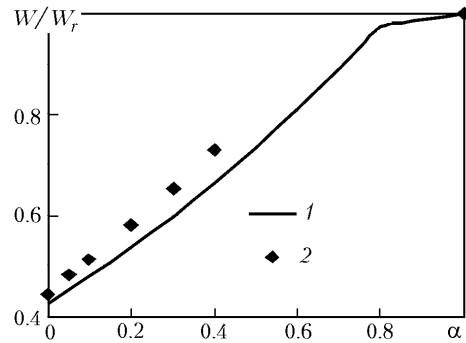


Fig. 8. Comparison of the calculated (1) and experimental (2) values [20] of the hydraulic resistance of the bidisperse mixture ($r/R = 0.65$) as a function of the portion of the small fraction.

monotonically decreasing function of the porosity and inversely proportional to the squared Sauter-mean diameter of particles in the mixture:

$$W \propto \frac{\varphi(\epsilon)}{d_S^2}. \quad (27)$$

The Sauter diameter is expressed in terms of the total volume of particles V_p and the total surface area of particles A_p as $d_S = 6V_p/A_p$. For an arbitrary disperse mixture, the expression for d_S can easily be obtained in the form

$$\frac{1}{d_S^2} = \frac{1}{4R^2} \left[1 + \alpha \left(\frac{R}{r} - 1 \right) \right]^2. \quad (28)$$

The function $d_S(\alpha)$ changes from $d_S(0) = 2R$ to $d_S(1) = 2r$. For $\varphi(\epsilon)$, numerous formulas constructed, as a rule, on an empirical basis are known. Of these, the best known function is the Kármán–Kozeny formula [1–3] $\varphi(\epsilon) = (1 - \epsilon)^2/\epsilon^3$, which was derived on the basis of the concept of a hydraulic radius with minimal use of empiricism. The applicability of this formula lies in the range $0.3 \leq \epsilon \leq 0.7$. In this range, with a correlation higher than 99%, the Kármán–Kozeny function can be approximated by the monomial

$$\varphi(\epsilon) = 0.056\epsilon^{-5}. \quad (29)$$

For a mixture, as a value for the porosity, one should take ϵ_m . The porosity function will depend on α , as shown in Fig. 7.

The general formula for the specific hydraulic resistance to the filtrate can be written in view of (27)–(29) in the form

$$\frac{W}{W_R} = \left(\frac{\epsilon_o}{\epsilon_m} \right)^5 \left[1 + \alpha \left(\frac{R}{r} - 1 \right) \right]^2, \quad (30)$$

where W_R means the resistance in the absence of the small fraction.

Figure 8 shows the calculations by formula (30) compared to Rudolf's measurements [20]. The agreement is fairly good, although in the experiments of [20] not balls of specific size, but a mixture of two closely classed powders was used. For the first of them, 80% of all particles had radii between 7 and 11 μm , and for the second one — 10–15 μm . The average particle sizes of these powders were 8 and 12 μm . And they were used in the calculations.

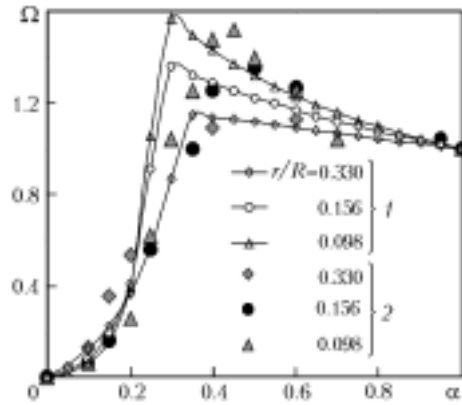


Fig. 9. Comparison of the calculated (1) and experimental (2) values [16] of the hydraulic resistance of the bidisperse mixture as a function of the portion of the small fraction at various values of r/R .

The ratio between these sizes is fairly high ($r/R \approx 0.65$), and the increase in the resistance with increasing portion of the small fraction is monotonic.

A different picture is observed for small values of r/R . Figure 9 shows the theoretical and experimental [16] dependences of the relative resistance $\Omega = (W - W_R)/(W_r - W_R)$ on the portion of the small fraction in bidisperse mixtures for various values of the parameter r/R . A distinguishing feature of the curves is their nonmonotonic character at low r/R , which is natural to associate with the dependence of the maximum of the function φ on the portion of the small fraction at various r/R . It is necessary to note the systematic deviation of the calculated maximum of the dependence $\Omega(\alpha)$ compared to the experiment. Let us indicate two possible reasons for this: first, in the experiments, not strictly bidisperse systems, but mixtures of powders were used (only the ratio between the average sizes is given); second, the nonmonotonic character of the resistance shows up at relatively low values of r/R , at which a porosity so small that the applicability of the Kármán-Kozeny formula is doubtful is achieved.

Thus, for quantitative modeling of the known nonmonotonic dependence of the bidisperse mixture porosity on the ratio between the portions of the small and large fractions the following mechanisms of interaction of particles have been used: 1) filling of the pores in the matrix from large particles with particles of the small fraction; 2) porosity inhomogeneity of the layers from small particles near an arbitrary large particle; 3) replacement of large particles in the packing by small particles, leading to a change in the packing structure.

The expressions for the mixture porosity depending on the ratio between the portions of small and large particles and on the diameter ratio between small and large particles obtained in the course of modeling have been compared to the experimental data. The agreement between the theory and the experiment is quite satisfactory. With the use of the derived expressions for the mixture porosity ε_m , the hydraulic resistance of the layer from a bidisperse material has been calculated. It has been shown that, in accordance with the experiment, at a small size ratio between small and large particles the dependence of the hydraulic resistance of the bidisperse layer on the portion of the small fraction has a maximum that is higher than the resistance values in the absence of large particles.

NOTATION

a, a_1, a_2, a_3, a_4 , constants; A_p , total surface area of particles, m^2 ; d_S , Sauter diameter, m ; f, F, F_0, F_1, Ψ , functions; r, R , particle radii of the two fractions, m ; x , ratio between particle radii; V , volume, m^3 ; W , specific hydraulic resistance of the layer, m^{-2} ; α , portion of small particles in the mixture; ε , porosity; Ω , relative specific hydraulic resistance of the layer; $\varphi(\varepsilon)$, porosity function. Subscripts: d, disturbance; m, mixture; o, original; p, particles; r, small particles; R, large particles; s, intersection; exp, experimental; theor, theoretical.

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