

A Generalized Blake-Kozeny Equation for Multisized Spherical Particles

Michael J. MacDonald, Chao-Feng Chu, Pierre P. Guilloit, and Ka M. Ng
Dept. of Chemical Engineering, University of Massachusetts, Amherst, MA 01003

Darcy's law is widely used for laminar, single-phase flow in porous media. It relates the volumetric flow rate of the fluid Q and the pressure gradient ∇p in a linear fashion.

$$\frac{Q}{A} = -\frac{k}{\mu} |\nabla p| \quad (1)$$

Here, A is the cross-sectional area for fluid flow, μ is the viscosity of the fluid, and k is the permeability of the porous medium. For media consisting of individual particles, the permeability can be related to the porosity, ϵ , and particle diameter, D_p . In this case, a special form of Darcy's law, the Blake-Kozeny equation, provides a more explicit relationship.

$$\frac{Q}{A} = -\frac{D_p^2}{150 \mu} \frac{\epsilon^3}{(1-\epsilon)^2} |\nabla p| \quad (2)$$

While Eq. 2 provides reasonably good predictions for flow in a limited class of porous media, it is not applicable to those made up of multisized particles. This is a serious omission. Most porous media in nature, such as soil, have a wide particle size distribution. In a chemical plant, crystallization is often followed by filtration to recover the product crystals. Thus, the filter cake is made up of particles with an exponential particle size distribution. Despite this obvious limitation, surprisingly little has been done on the prediction of the permeability of a porous medium with multisized particles.

The only existing set of data is due to Standish and Collins (1983), who measured the permeability of a ternary mixture of glass beads under laminar flow conditions. The same group also performed experimental work for turbulent flows (Standish and Mellor, 1980; Standish and Leyshon, 1981). There have been a couple of suggestions for extending the Blake-Kozeny equation to multisized particles. Standish and Collins (1983) proposed that for a mixture the particle diameter in the Blake-Kozeny equation should be replaced by the harmonic mean. Leitzement et al. (1985) further suggested that in calculating the harmonic mean each particle size should be weighted with the fractional volume of those particles in the mixture.

The objectives of this study were twofold. First, in view of the paucity of data in the literature, additional experiments were conducted for laminar flow through a porous medium made up of multisized particles. Second, a theoretical analysis was performed to provide a physical basis for correlating the permeability data. We focused primarily on spheres; thus, this study is not applicable for particles, such as fibers with a large aspect ratio, whose shape deviates significantly from that of a sphere. The development parallels that presented in Bird et al. (1960, p. 196).

Theory

We begin by considering a packed bed of spherical particles with a size distribution, $n(D_p)$. Thus, $n(D_p)dD_p$ represents the number of particles per unit volume of bed whose diameters range between D_p and $D_p + dD_p$. The i th moment of the particle size distribution is:

$$M_i = \int_0^\infty D_p^i n(D_p) dD_p \quad (3)$$

If a horizontal cut is made across the bed, one sees circular discs with different diameters, x , on the sectional plane. The size distribution of these discs $f(x)$ is given by:

$$f(x) = \int_0^\infty P(x/D_p) P(D_p) dD_p \quad (4)$$

Here, $P(D_p)$ is the probability density function such that $P(D_p)dD_p$ is the probability that the diameter of a given particle in the bed ranges between D_p and $D_p + dD_p$. We have:

$$P(D_p) = \frac{n(D_p)}{\int_0^\infty n(D_p) dD_p} = \frac{n(D_p)}{M_0} \quad (5)$$

where M_0 is the total number of particles per unit volume of bed. $P(x/D_p)$ is the conditional probability density function such that $P(x/D_p)dx$ is the probability that, given a sphere

Correspondence concerning this note should be addressed to K. M. Ng.

with diameter D_p , the diameter of a given disc ranges between x and $x+dx$. Note that discs of the same size can originate from spheres of different sizes because the disc size depends on the vertical position at which a sphere is cut. Of course, the size of a disc from a given sphere is always less than or equal to the diameter of that sphere. Thus, we have:

$$P(x/D_p) = \frac{x}{D_p \sqrt{D_p^2 - x^2}} [1 - H(x - D_p)] \quad (6)$$

The quotient on the right side of Eq. 6 [see Ng (1986) for derivation] represents the size distribution of the discs if the particles were of a uniform size, D_p . H is the Heaviside function and the term within the squared brackets ensures that, for a given sphere diameter, discs of diameters larger than D_p are not counted. Substituting Eqs. 5 and 6 into Eq. 4, we get:

$$f(x) = \int_0^\infty \frac{x}{D_p \sqrt{D_p^2 - x^2}} \frac{n(D_p)}{M_0} [1 - H(x - D_p)] dD_p \quad (7)$$

If one assumes an isotropic porous medium, the areal porosity in a sectional plane is equal to the overall bed porosity. The relationship between the number of discs per unit cross-section area N_c and the bed porosity is:

$$1 - \epsilon = N_c \frac{\pi}{4} \int_0^\infty x^2 f(x) dx \quad (8)$$

Substituting Eq. 7 into Eq. 8, we obtain after integration:

$$N_c = \frac{6}{\pi} (1 - \epsilon) \left(\frac{M_0}{M_2} \right) \quad (9)$$

where M_2 is the second moment of the particle size distribution. Then, the wetted perimeter per unit area of bed S can be determined in a similar manner.

$$S = N_c \pi \int_0^\infty x f(x) dx = N_c \frac{\pi^2}{4} \left(\frac{M_1}{M_0} \right) \quad (10)$$

where M_1 is the first moment of $n(D_p)$.

We are now ready for the flow analysis. The hydraulic radius R_h is defined as the following:

$$R_h = \frac{\text{cross-sectional area for flow}}{\text{wetted perimeter}} \quad (11)$$

The cross-sectional area for flow is equal to ϵ , while the wetted perimeter per unit area of the bed is S . Substituting ϵ and S from Eq. 10 into Eq. 11, we get:

$$R_h = \frac{2}{3\pi} \frac{\epsilon}{1 - \epsilon} \frac{M_2}{M_1} \quad (12)$$

If a pressure drop Δp is applied to the system with length L , the velocity profile $u(r)$ is given by the Hagen-Poiseuille equation:

$$u(r) = \frac{r^2 - R_h^2}{4\mu} \frac{\Delta p}{L} \quad (13)$$

where r is the radial position. The average velocity over the area available for flow is:

$$\bar{u} = \frac{\int_0^{2\pi} \int_0^{R_h} u(r) r dr d\theta}{\pi R_h^2} = \frac{1}{32\mu} (4R_h)^2 \frac{\Delta p}{L} \quad (14)$$

The fluid superficial velocity, $v (= Q/A)$ can be evaluated as $v = \epsilon \bar{u}$, or

$$v = \frac{2}{9\pi^2 \mu} \frac{\epsilon^3}{(1 - \epsilon)^2} \left(\frac{M_2}{M_1} \right)^2 \frac{\Delta p}{L} \quad (15)$$

Equation 15 represents a generalized form of the Blake-Kozeny equation. Comparison of Eqs. 2 and 15 shows that D_p in Eq. 2 is replaced by the ratio of the second moment to the first moment (M_2/M_1) in Eq. 15. Note that as a particle size distribution gets narrower and narrower, the ratio M_2/M_1 approaches a single value D_p .

The other difference between the two expressions is the proportionality constant: 1/150 for Blake-Kozeny and $2/9\pi^2 (= 1/44.4)$ for the generalized expression. The discrepancy might seem large but 1/150 was actually determined from experimental data. Arguing that tortuosity was not accounted for in this simplified analysis, Bird et al. (1960) adjusted the proportionality constant in their derivation from 1/72 to 1/150. In addition, other sources of error are present. For sphere packs, the converging-diverging nature of the channels can have a significant impact on flow (Payatakes et al., 1973). The fact that the porous medium is actually made up of interconnected channels is another neglected factor (Ng and Payatakes, 1985). Also, there are limitations to the hydraulic radius approach for flow in noncircular channels (Kays, 1966). For laminar flows, although the form of the Hagen-Poiseuille equation is retained, a shape factor is needed to account for the channel shape. For all these reasons, we will adjust our constant to match experimental data. Based on additional experimental results, Carman (1937) suggested a value of 1/180 for the proportionality constant. After reviewing a large number of experimental data, MacDonald et al. (1979) concluded that using a constant of 1/180 provides better predictions and this value is chosen for the current work.

Experimental Studies

Apparatus

The experimental setup used to determine the flow rate through a packed tube is shown in Figure 1. The apparatus is similar to that used in Chu and Ng (1989). It consisted of a cylindrical glass tube, 42-cm-long and 1.56-cm-dia., packed with spheres to a height of L , two beakers each with a side-opening and a reservoir. The beakers were connected to the column by plastic tubings. Valves were available for flow control. The fluid used in our experiments was water, which flowed continuously from the reservoir to beaker 1 to maintain the water level there. The overflow from beaker 2 was collected to determine the water flow rate through the bed. Thus, the

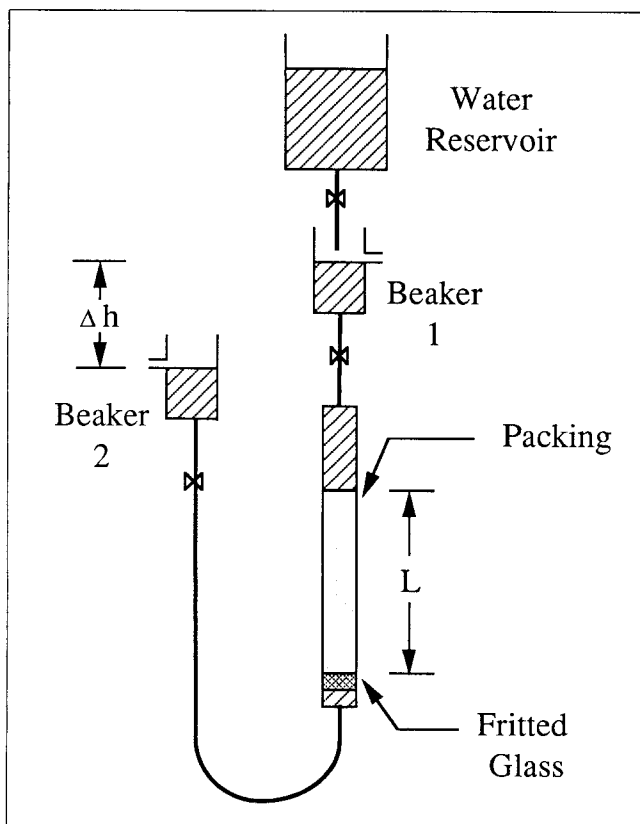


Figure 1. Experimental setup.

water levels in the two beakers were separated by a height of Δh apart, providing a constant hydrostatic head across the bed. A fritted glass disc of pore size 70–100 μm was used as a support for the spheres at the bottom of the tube.

The packings were made up of solid glass spheres (Potters Industries, Inc.). The spheres as received were sieved before use so that particles with a relatively narrow size range could be obtained. The size ranges were 840–710, 710–595, 595–500, 355–300 and 180–125 μm , and their corresponding arithmetic mean sizes were 775, 652.5, 547.5, 327.5 and 152.5 μm . Three ternary mixtures of particles were obtained by mixing these five sets of spheres: mixture no. 1 (775, 547.5 and 327.5 μm); mixture no. 2 (775, 652.5 and 327.5 μm); and mixture no. 3 (775, 327.5 and 152.5 μm).

Procedure

The composition of one of the three ternary mixtures was first selected. About 10 g of glass beads was needed for each packing. Air tended to get trapped in the sphere pack if water was added to a dry packing. For this reason, the empty tube was filled with water before adding particles to it. The disadvantage was the possibility of particle segregation as they fell through water. The following steps were followed to minimize the formation of heterogeneous regions in the packing. The spheres were added in five batches, each formed by mixing the appropriate amount of the three constituent particles and weighing approximately 2 g. Particles for each batch were added in small spoonfuls. After settling in the bed, the tube was lightly tapped on the side to promote a close packing. The flow rate was then measured after all 10 g was added. The

height of the hydrostatic head and the height of the packing were also recorded. Note that the magnitude of the hydrostatic head was chosen to ensure laminar flow. The Reynolds number ($= \rho v D_p / \mu$), based on the arithmetic mean size of the largest particles in the ternary mixture, in all the experiments was 0.5 or less. The entire procedure was repeated twice, by adding another 10 g of the ternary mixture each time to the existing packing. Therefore, the final sphere pack weighed approximately 30 g.

Data Analysis

From the data, the porosity and permeability of the bed were calculated. The density of the glass beads was 2.50 g/cm^3 ; since the height and diameter of the packing were known, the porosity could be determined readily. The permeability was calculated using Darcy's Law, Eq. 1, as follows. The pressure drop was provided by the hydrostatic head; thus, we have:

$$\Delta p = \rho g \Delta h \quad (16)$$

where ρ is the fluid density, g the acceleration due to gravity, and Δh the difference between the two fluid levels. In addition to the packing, the valves, tubings, and the fritted glass disc also presented resistance to flow. Let α be the combined resistances of these miscellaneous items. If we assume that α is dependent linearly on the volumetric flow rate of the fluid under laminar flow, we can rewrite Eq. 1 as:

$$Q = \frac{1}{\frac{L}{Ak} + \alpha} \frac{\rho g \Delta h}{\mu} \quad (17)$$

Because data were obtained for three different packing lengths, the resistance α can be eliminated from the flow rate data of any two different packing lengths, L_1 and L_2 . The permeability is given by:

$$k = \frac{L_2 - L_1}{A} \frac{\mu}{\rho g \Delta h} \frac{Q_1 Q_2}{Q_1 - Q_2} \quad (18)$$

The third packing length and flow rate provided additional data for checking consistency and reproducibility.

Results

The porosity and permeability results are presented in ternary diagrams, where the composition of a given mixture, in weight percent, can be easily looked up from the scale along each edge. Figure 2a shows the porosity values for mixture no. 1. At the vertices of the triangle, the porosity is approximately 0.40, a well-known value for a random packing of uniform spheres. Any point along an edge, except for a vertex, represents a binary mixture. There seems to be a minimum porosity between the coarse and fine particles (around 0.331) and between the medium and fine particles (around 0.359). The presence of a minimum porosity between the coarse and medium particles is less obvious although the minimum value seems to be around 0.378. Any point within the triangle represents a ternary mixture. The following overall picture can be obtained with all the data points. The porosity is around 0.4 at all three

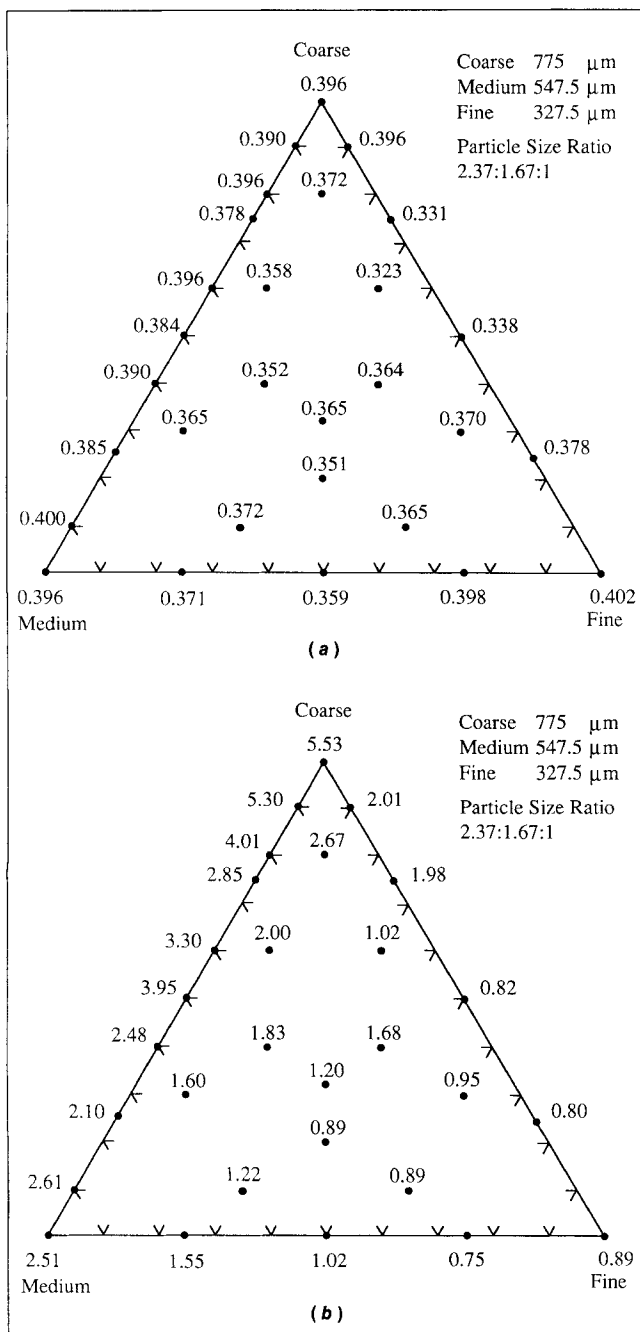


Figure 2. Ternary diagrams for mixture no. 1: (a) porosity; (b) permeability.

corners. The minimum porosity occurs by mixing the coarse and fine particles. Moving away from the coarse particles-fine particles edge tends to increase the porosity. These data in general agree with the results of Standish and Borger (1979).

The corresponding permeability values (in 10^{-6} cm^2) for mixture no. 1 are shown in Figure 2b. The trends of these data are in general agreement with the results of Standish and Collins (1983). Let us again begin at the vertices. As expected, the permeability of the fine particles ($0.89 \times 10^{-6} \text{ cm}^2$) is smaller than the medium ones ($2.51 \times 10^{-6} \text{ cm}^2$) and the coarse ones ($5.53 \times 10^{-6} \text{ cm}^2$). There seems to be a minimum permeability

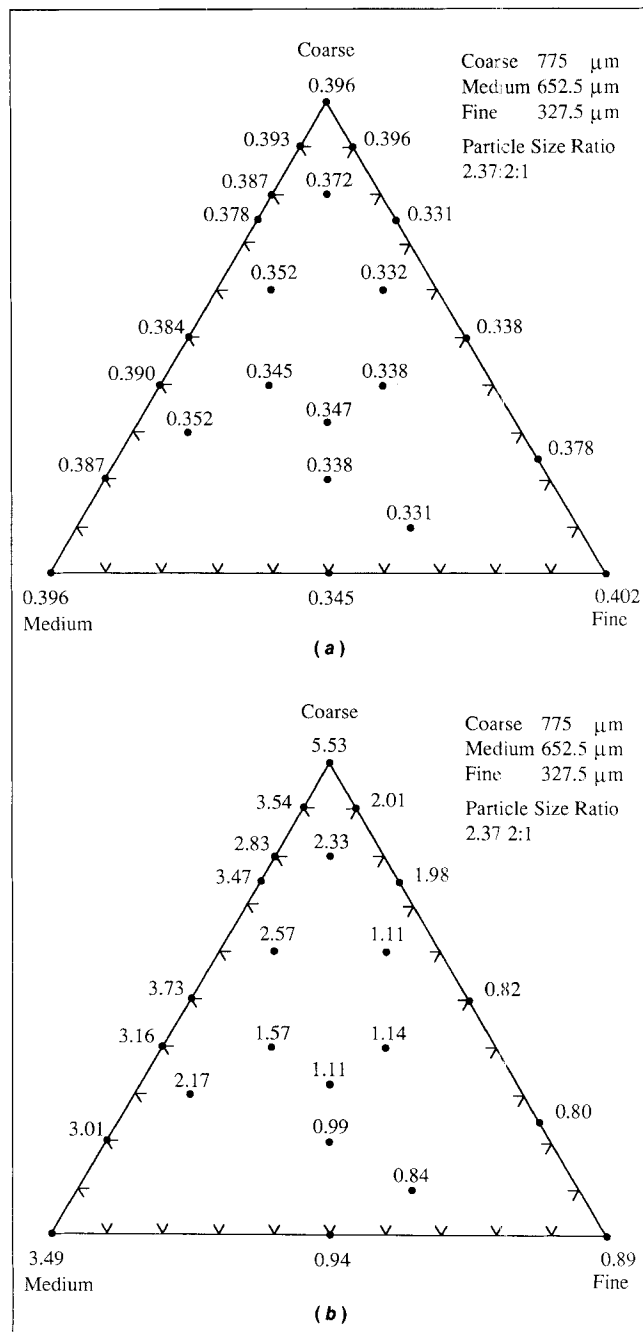


Figure 3. Ternary diagrams for mixture no. 2: (a) porosity; (b) permeability.

between the coarse and fine particles (around $0.80 \times 10^{-6} \text{ cm}^2$) and between the medium and fine particles (around $0.75 \times 10^{-6} \text{ cm}^2$). The presence of a minimum permeability between the coarse and medium particles is less obvious although the minimum value seems to be around $2.10 \times 10^{-6} \text{ cm}^2$. Note that for a binary mixture the minimum permeability point does not correspond to the composition with the minimum porosity. For example, a 25% fine-75% coarse mixture gives the minimum porosity (Figure 2a), while a 75% fine-25% coarse mixture gives the minimum permeability (Figure 2b). Obviously, this is because permeability depends on both the bed porosity

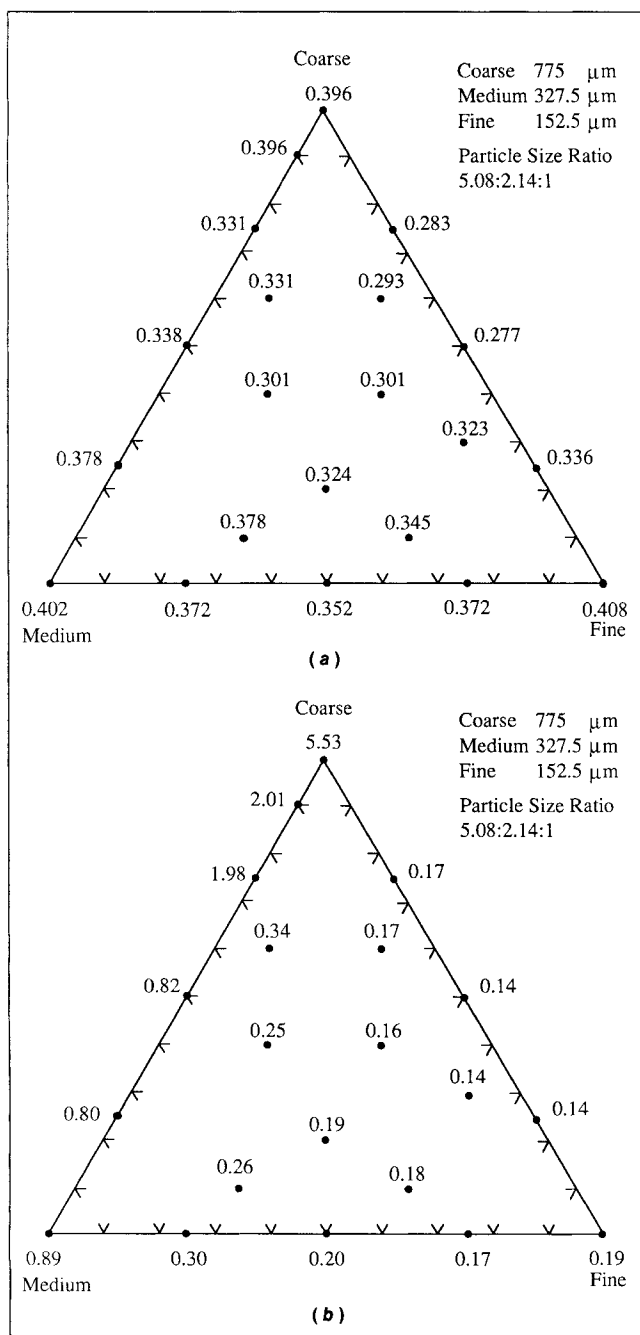


Figure 4. Ternary diagrams for mixture no. 3: (a) porosity; (b) permeability.

and particle size. Examination of all the data points indicates that the permeability generally increases with particle size, but can be modified by changes in the overall porosity.

The porosity and permeability also depend on the ratio of the diameters of the constituent particles. The data for mixtures no. 2 and no. 3 are shown in Figures 3 and 4, respectively. The results are qualitatively similar to those of mixture no. 1. Most interesting is mixture no. 3 which has a much higher particle size ratio (5.08:2.14:1) than the other two mixtures. The minimum measured porosity of 0.277 is significantly smaller than 0.323 and 0.331, the minimum values in mixture

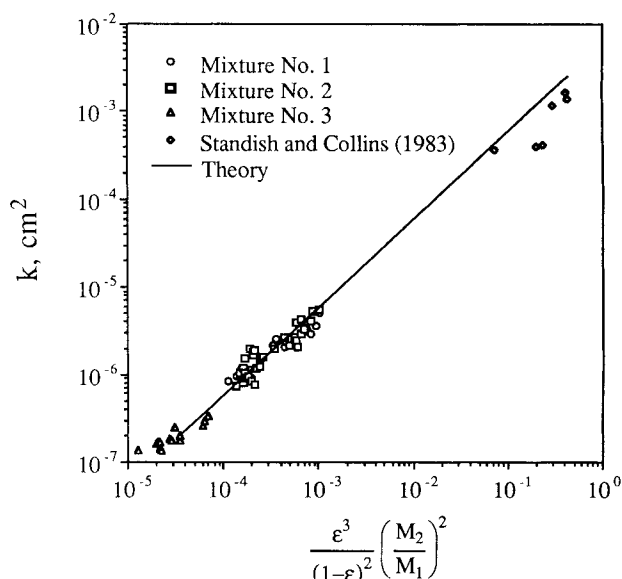


Figure 5. Measured permeability of ternary mixtures of solids.

nos. 1 and 2, respectively. Clearly, a large difference in particle size leads to a denser packing.

To confirm the validity of our theory, the permeability data are plotted against $\epsilon^3/(1-\epsilon)^2 (M_2/M_1)^2$ in a log-log graph in Figure 5. Also shown are representative data points obtained by Standish and Collins (1983). The solid line corresponds to Eq. 15 except that the proportionality constant used is 1/180, rather than the value (1/44.4) derived in our theory. All of the data, although scattered, cluster around the straight line representing the theory. As mentioned previously, the ratio of M_2 and M_1 is equal to a single value D_p if the particle size distribution actually corresponds to particles of a single size. Therefore, the straight line also represents the best available equation for uniform-sized particles. In view of the amount of scatter experienced even for packings with uniform-sized particles (MacDonald et al, 1979), the agreement is reasonably good. The fact that the five sets of spheres used in our experiments were not exactly of a uniform size could also contribute to some scattering.

Conclusion

The theory presented here provides a simple and natural extension of the Blake-Kozeny equation for estimating the permeability of a mixture of multisized particles. A porosity value needed in our theory was determined experimentally to avoid a potential source of uncertainty in evaluating the theory. The need for experimental testing can be inconvenient in the conceptual design of solids processes (Rajagopal et al., 1988). Fortunately, a number of theories for estimating the porosity of a multisized mixture of spheres (Ouchiya and Tanaka, 1981, 1984; Yu and Standish, 1988) are available, which can be used to close our theory.

Acknowledgment

We express our appreciation to the National Science Foundation, Grant No. CTS-8821793, for partial support of this research.

Notation

A = area
 D_p = particle diameter
 $f(x)$ = size distribution function of discs
 g = acceleration due to gravity
 k = permeability
 L = length of packing
 M_i = i th moment of particle size distribution
 n = particle size distribution
 N_c = number of discs per cross-sectional area of bed
 p = pressure
 P = probability
 Q = volumetric flow rate
 R_h = hydraulic radius
 r = radial coordinate
 S = wetted perimeter per unit area of the bed, Eq. 10
 u = interstitial velocity
 v = superficial velocity
 x = diameter of a disc on a horizontal cut of a packed bed

Greek letters

α = combined flow resistances
 Δh = hydrostatic head
 ϵ = porosity
 μ = liquid viscosity
 ρ = density of fluid

Literature Cited

Bird, R. B., W. E. Stewart, and E. N. Lightfoot, *Transport Phenomena*, Wiley, New York (1960).
Carman, P. C., "Fluid Flow Through Granular Beds," *Trans. Inst. Chem. Eng.*, **15**, 150 (1937).
Chu, C. F., and K. M. Ng, "Flow in Packed Tubes with a Small Tube to Particle Diameter Ratio," *AIChE J.*, **35**, 148 (1989).

Kays, W. M., *Convective Heat and Mass Transfer*, McGraw-Hill, New York (1966).
Leitzelment, M., C. S. Lo, and J. Dodds, "Porosity and Permeability of Ternary Mixtures of Particles," *Powder Technol.*, **41**, 159 (1985).
MacDonald, I. F., M. S. El-Sayed, K. Mow, and F. A. L. Dullien, "Flow through Porous Media—the Ergun Equation Revisited," *Ind. Eng. Chem. Fundam.*, **18**, 199 (1979).
Ng, K. M., "A Model for Flow Regime Transitions in Cocurrent Downflow Trickle-Bed Reactors," *AIChE J.*, **32**, 115 (1986).
Ng, K. M., and A. C. Payatakes, "Critical Evaluation of the Flow Rate-Pressure Drop Relation Assumed in Permeability Models," *AIChE J.*, **31**, 1569 (1985).
Ouchiya, N., and T. Tanaka, "Porosity of a Mass of Solid Particles having a Range of Sizes," *Ind. Eng. Chem. Fundam.*, **20**, 66 (1981).
Ouchiya, N., and T. Tanaka, "Porosity Estimation for Random Packings of Spherical Particles," *Ind. Eng. Chem. Fundam.*, **23**, 490 (1984).
Payatakes, A. C., C. Tien, and R. M. Turian, "Numerical Solution of Steady State Incompressible Newtonian Flow Through Periodically Constricted Tubes," *AIChE J.*, **19**, 67 (1973).
Rajagopal, S., K. M. Ng, and J. M. Douglas, "Design of Solids Processes—Potash Production," *Ind. Eng. Chem. Res.*, **27**, 2071 (1988).
Standish, N., and Borger, D. E., "The Porosity of Particulate Mixtures," *Powder Technol.*, **22**, 121 (1979).
Standish, N., and D. N. Collins, "The Permeability of Ternary Particulate Mixtures for Laminar Flow," *Powder Technol.*, **36**, 55 (1983).
Standish, N., and D. G. Mellor, "The Permeability of Ternary Coke Mixtures," *Powder Technol.*, **27**, 61 (1980).
Standish, N., and P. J. Leyshon, "The Permeability of Quaternary Particulate Mixtures," *Powder Technol.*, **30**, 118 (1981).
Yu, A. B., and N. Standish, "An Analytical-Parametric Theory of the Random Packing of Particles," *Powder Technol.*, **55**, 171 (1988).

Manuscript received June 3, 1991, and revision received Aug. 19, 1991.