LITERATURE CITED

Aris, R., "On Shape Factors for Irregular Particles," *Chem. Eng. Sci.*, **6**, 262 (1957).

Aris, R., "The Mathematical theory of diffusion and reaction in permeable catalysts," vol. 1, p. 247, Oxford University Press (1975).

Georgakis, C. and Aris, R., "Diffusion reaction and the P.S.S.H.," *Math. Biosciences*, **25**, 237 (1975).

Knudsen, C. W., Roberts, G. W. and Satterfield, C. N., "Effect of Geometry on Catalyst Effectiveness Factor," *Ind. Eng. Chem. Fundamental*, **5**, 325 (1966).

Petersen, E. E., "A General Criterion for Diffusion Influenced Chemical Reactions in Porous Solids," *Chem. Eng. Sci.*, **20**, 587 (1965).

Roberts, G. W. and Satterfield, C. N., "Effectiveness Factor for Porous Catalyst," *Ind. Eng. Chem. Fundamental*, **4**, 288 (1965).

Roberts, G. W. and Satterfield, C. N., "Effectiveness Factor for Porous Catalyst," *Ibid.*, **5**, 317 (1966).

Villadsen, J. V. and Stewart, W. E., "Solution of Boundary Value Problems by Orthogonal Collocation," *Chem. Eng. Sci.*, **22**, 1483 (1967).

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Flow Through Permeable Beds Consisting of Layers of Different Size Spheres

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TABLE 1. HEIGHTS OF BED LAYERS, (MM)

Bed	3 mm spheres	1 mm spheres	Total height
A	27.9	111.8	139.7
B	55.9	83.8	139.7
C	83.8	55.9	139.7
D	111.8	27.9	139.7

This paper is concerned with the flow—pressure drop characteristics for non-Darcy flow in permeable beds consisting of two horizontal layers of different size spheres. To identify the extent of the interaction between the layers, the measured results are compared with those from a computational model which assumes that the constituent layers behave as if they are unrelated parallel flow paths. In the absence of alternative approaches, such a model would, in all likelihood, be employed to predict the characteristics of multilayer beds by making use of available information for single layer (that is, homogeneous) beds. Therefore, an experimental test of the computational model is of some practical relevance.

EXPERIMENTS

The layered beds were contained in a test section having a 14×14 cm^2 cross section and a length of 35.6 cm. The lower and upper layers, respectively, consisted of 1 mm* and 3 mm spherical glass beads. Four layered beds were investigated, and the heights of the constituent layers are listed in Table 1. Experiments were also carried out for beds consisting exclusively of either 1 or 3 mm beads. These homogeneous bed experiments encompassed bed heights of 27.9, . . . and 139.7 mm.

The test section, made of Plexiglas, was instrumented with thirty pressure taps, fifteen in the top wall and fifteen in the bottom wall. The taps were spaced at 2.5 cm intervals along the spanwise center line of the respective wall. Packing of the spheres was accomplished by removal of the top wall. The packing process (see Bahrami, 1975) involved a succession of pourings and smoothings and, as a final stage, a tamping procedure.

The working fluid was water. It was supplied to the test section by means of a constant head tank. Adjustment of the water level in the tank permitted control of the rate of mass flow through the porous bed. Measurement of the mass flow was accomplished by the direct weighing method. The water temperature was maintained constant in the range 20°C ± 0.3° during the entire course of the experiments. Care was taken to remove air bubbles from the bed prior to the initiation of data collection. The pressure distributions were read from a manometer bank by a cathetometer capable of discriminating heights to within 0.05 mm.

RESULTS AND DISCUSSION

The homogeneous bed experiments were performed first, and the measured flow—pressure drop data were examined with the aid of the Forchheimer relation, which is a nonlinear generalization of Darcy's law. For data analysis, a particularly convenient form of the Forchheimer law is

$$\frac{1}{\mu V}\left[-\frac{dp}{dx}\right] = \frac{1}{k} + \frac{c}{\sqrt{k}}\left(\frac{V}{\nu}\right) \qquad (1)$$

where dp/dx is the streamwise pressure gradient. If data plotted on a graph of $(-dp/dx)/\mu V$ vs. V/ν yield a straight line relationship, then the Forchheimer relation is obeyed. Furthermore, the values of c and k can be deduced from the slope and the intercept of the straight line.

All of the homogeneous bed data were found to be well correlated by the Forchheimer relation (Bahrami, 1975). In round numbers, the respective permeabilities of the 1 and 3 mm beds were 0.7×10^{-5} and 7×10^{-5} cm^2. The c values were in the range from 0.3 to 0.5.

For the layered beds, it may first be noted that for all the layer configurations and flow rates that were investigated, the pressure gradients measured along the top and bottom walls of the test section were within a fraction of a percent of each other. Furthermore, the p vs. x distributions were linear. Thus, the flows in the two layers were driven by a common and constant pressure gradient.

* Nominal value; the beads were sifted between 16 and 20 mesh sieves having openings of 1 and 0.841 mm, respectively.

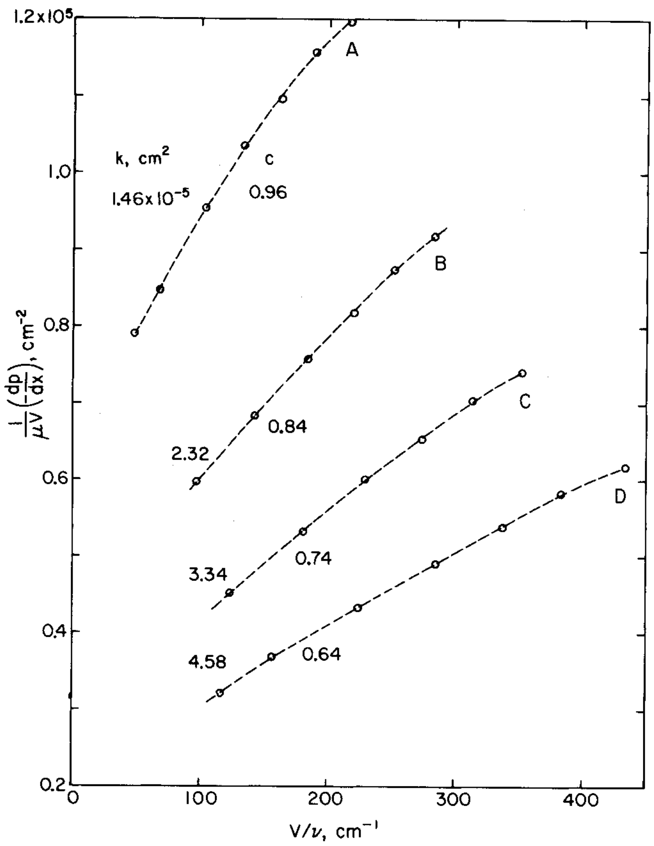

Fig. 1. Examination of the applicability of the Forchheimer relation to layered beds.

A first approach to organizing and correlating the stratified bed results is to consider the use of the Forchheimer relation. In this connection, one point of uncertainty is the proper method of evaluating the mean filter velocity V, especially since the respective mean velocities for the two layers are significantly different (by a factor of 4 or 5 in the present experiments). From the standpoint of applications, a mean velocity based on the overall mass rate of flow through the bed (that is, through both layers) and on the total cross-sectional area is the most practical choice. Mean filter velocities evaluated in this way were employed in conjunction with the measured pressure gradients to compute $(-dp/dx)/\mu V$ and V/ν for all the stratified bed data runs.

This information is presented in Figure 1, where dashed lines have been passed through the data points to provide continuity. The respective beds A, B, C, and D are identified in Table 1. Inspection of the figure shows that for all of the beds, the variation of $(-dp/dx)/\mu V$ with V/ν is not linear; rather, there is a small but unmistakable curvature. As can be seen from Equation (1), the presence of the curvature is an indication that k or c, or both, are not constant; rather, they are velocity dependent. Thus, the constant coefficient Forchheimer relation does not appear to hold for a layered bed when the velocity V is evaluated as the average for the cross section as a whole.

If the curvature were to be ignored and least-squares straight lines were passed through the data, then effective values of k and c could be found. These are listed in Figure 1. The effective k values for the layered beds are bounded between the k values for the constituent single-layer beds, 0.7×10^{-5} and 7×10^{-5} cm², respectively. On the other hand, the effective c values are substantially higher than those of the constituent beds (0.3 to 0.5) and than those reported elsewhere for single-layer beds.

An alternate approach to interpreting the layered-bed results is to examine them in light of a computational model which assumes that the constituent layers behave as if they are unrelated parallel flow paths. One way of using such a model is to write the Forchheimer relation (1) for each of the layers. With these equations, it can be shown, in agreement with the experimental results of Figure 1, that the flow pressure drop characteristics of a layered bed are not representable by a constant coefficient Forchheimer relation in which the velocity is the average for the entire bed cross section.

Such a model can also be used for predicting the mass flow through layered beds by employing flow—pressure drop information for the constituent beds. Suppose that for a specific fluid, flow—pressure drop information is available for single-layer beds consisting of particles of size i. This information can be plotted in the form $(\dot{m}/A)_i$ vs. $(dp/dx)_i$. Suppose also that similar information is available for flow of the same fluid through single-layer beds that are made up of particles of size j. To maximize the usefulness of the single-layer information, the fluid density ρ and viscosity μ for the j bed data should be the same as for the i bed data.

Next, consider a two-layer bed in which one layer consists of particles of size i and the other layer consists of particles of size j. The respective layers have cross sections A_i and A_j. Furthermore, suppose that the pressure gradient dp/dx, common to both layers, is known. Then, for a first estimate, the mass flow rate $\dot{m}$ through the bed may be evaluated as

$$\dot{m} = (\dot{m}/A)_i A_i + (\dot{m}/A)_j A_j \qquad (2)$$

where $(\dot{m}/A)_i$ and $(\dot{m}/A)_j$ are for the single-layer beds and correspond to the aforementioned common value of dp/dx. The respective single-layer inputs to Equation (2) are for cross sections which closely approximate the A_i and A_j of the two-layer bed. Equation (2) superposes the mass flows from the two constituent layers as if each layer were a separate entity, replete with its bounding

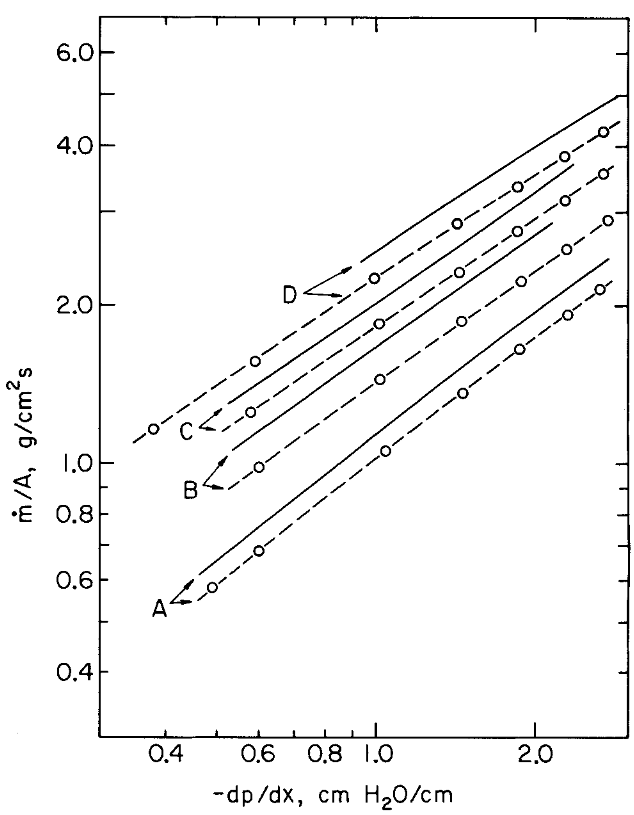

Fig. 2. Comparison of predicted and measured mass flow rates for layered beds.

walls. In effect, the actual interface between the constituent layers is replaced by a rigid wall that separates the layers.

Values of $\dot{m}$ were evaluated from Equation (2) by using the data from the homogeneous bed experiments for the 1 and 3 mm spheres. The resulting mass flow predictions* for each of the layered beds are shown as solid lines in Figure 2. The experimental data for the layered beds are plotted as open circles and are connected by dashed lines for continuity. For each layered bed, the information represented by the solid and dashed lines thus involves the results of three separate sets of experiments.

Inspection of Figure 2 indicates that, in general, the measured mass flow for the two-layer bed is smaller than that predicted by adding the flows that would pass through the constituent layers if they were separate entities. The deviations are of the order of 10%. The finding that the measured mass flow is smaller can be made plausible by comparing the local permeability in the zone adjacent to the interface between the layers with that adjacent to a solid wall. In the interface zone, owing to the intermeshing of the larger and smaller spheres, the local permeability will be diminished and the mass flow correspondingly reduced. On the other hand, for a bed of spheres, the packing is less dense adjacent to a solid wall than in the interior of the bed. As a consequence, the permeability for streamwise flow is relatively high adjacent to a solid wall. Inasmuch as the predictive model, in effect, replaces the actual interface with a solid wall, the foregoing arguments are suggestive of an overprediction of the mass flow, although it is not likely that such arguments can fully account for the magnitudes of the deviations evidenced in Figure 2. Any intrusions of smaller spheres into the bed of larger spheres would lower the local permeability and reduce the mass flow.

* The area A in the ordinate variable is the total cross section of the layered bed.

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NOTATION

A = cross-sectional area
c = Forchheimer inertia coefficient
k = permeability
$\dot{m}$ = mass flow rate
p = pressure
V = mean filter velocity (superficial velocity)
x = axial coordinate
μ = viscosity
ν = kinematic viscosity
ρ = density

LITERATURE CITED

Bahrami, P. A., "Flow of Viscous Fluid Through Layered Beds of Spheres," M.S. thesis, Univ. Minn., Minneapolis (1975).

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Chemical Absorption Kinetics Over a Wide Range of Contact Time: Absorption of Carbon Dioxide into Aqueous Solutions of Monoethanolamine

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When a gas, after being absorbed in liquid, may react not only with a reactant dissolved in the liquid but also with a product of this first reaction, then the second (gas with product) reaction will only influence the rate of gas absorption after an exposure time long enough for the product of the first reaction to form. This situation would be expected in many processes involving chemical absorption. However, previous experimental investigations on chemical absorption have been restricted to those with short contact times. In the present work, experiments for the carbon dioxide—monoethanolamine system were performed by using a laminar liquid jet, a wetted wall column, and a quiescent liquid absorber in order to examine the gas absorption mechanism and the reaction kinetics over a wide range of contact times. It is suggested from measured values of the enhancement factor that the present absorption process should be discussed by gas absorption with an irreversible consecutive reaction of the second order and that the reaction rate constant ratio can be estimated.

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CHEMICAL ABSORPTION MECHANISM

The overall reaction occurring in the liquid phase is expressed by (Astarita, 1967)

$$CO_2 + 2RNH_2 \rightarrow RNHCOORNH_3 \quad (1)$$

$$CO_2 + RNHCOORNH_3 + 2H_2O \rightarrow 2HCO_3RNH_3 \quad (2)$$

where R refers to $(HCOH_2CH_2)$.

The rate constant of the reaction (1) has been obtained by several investigators (Jensen et al., 1954; Emmert and Pigford, 1962; Clarke, 1964). However, the reported value of the reaction rate constant at 25°C ranges from 5 400 to 8 500 l/(g-mole) (s). In the present work, the reaction rate constant was measured with a laminar liquid jet absorber and applied to analyses of chemical absorption data.