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Flows of Nonlinear Fluids Through Porous Media

The conventional "cylindrical tube" model of porous media is shown to be inapplicable to flows of nonlinear fluids. A more precise geometric description of the converging-diverging pore geometry appears to enable accurate pressure drop-flow rate descriptions but requires further development.

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SCOPE

The understanding of flows through porous media represents an interesting challenge in chemical reactor design and in some polymer engineering and separational processes. Further, it may play a primary role in formulating optimum strategies for management of underground reservoirs, as in petroleum production and in the exploitation and control of aquifers. Consequently, an extensive literature has been developed on this subject. The two major features of this literature may be described as follows.

Analyses of non-Newtonian fluid flows through porous media under laminar flow conditions have almost universally been developed by coupling a specific model of the pore structure of the medium with a specific model of the rheological properties of the fluid being employed in the flow process. Most commonly the pore structure is modeled by means of the cylindrical equivalent capillary, a cylindrical duct of length and diameter such that it exhibits the same resistance to flow as the actual interstices in the real porous medium. Thoughtful and clear descriptions of this modeling are presented in the paper by Ergun (1952), the book by Bird, Stewart, and Lightfoot (1960), in the research papers of Christopher and Middleman (1965) and of Gaitonde and Middleman (1967), and in the comprehensive article of Savins (1969). This latter paper shows how a particular rheological model of the viscosity function of the fluid may be replaced with

a generalized analysis similar to that employed in laminar and turbulent flows of non-Newtonian fluids through tubes.

Separately, there has been a growing awareness of the fact that the actual pores in a granular bed are, of course, not isolated circular cylinders of constant cross-sectional area; they are interconnected and noncircular, and the changes in cross-sectional area to which a fluid element is exposed as it moves through the bed may occur rapidly and be of large magnitude. An early attempt to apply some of these considerations to the flow of viscoelastic fluids was published by Marshall and Metzner (1967); the subject of the flow of Newtonian fluids through pores of complex geometry has recently been extended substantially, and its current status is presented in a series of papers by Dullien (1975), Payatakes, Tien, and Turien (1973a, b, c), Dullien and Azzam (1973a, b), and Batra, Fulford, and Dullien (1970). At the present time, therefore, a designer may choose between analyses which consider either nonlinear fluid properties or some of the complexities of real pore geometries, but generally not both.

It is shown in the present work that, conceptually, the cylindrical capillary model of the pore structure must be a grossly inadequate representation for use when flows of nonlinear fluids are to be described and that this inadequacy may lead to order-of-magnitude errors in pressure drop-flow rate predictions. As a step toward resolution of this problem, a more realistic model of the pore structure is introduced and applied to flows of nonlinear but purely viscous fluids through porous media.

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CONCLUSIONS AND SIGNIFICANCE

A sinusoidal model of pore structure is developed and applied to the interpretation of both literature data and new experimental results obtained in the present study. An appreciable improvement in predictive ability is obtained. In the case of data for highly nonlinear emulsions, the average percentage deviation between predicted and experimental friction factors is 2.7% as compared to 29.1% when the capillary model is used; these deviations correspond to average errors of 12 and 300%, respectively, in the prediction of flow rate at a given pressure drop.

When applied to flows through a particular porous medium, under either inertial or noninertial flow conditions,

no greater a number of empirical parameters than involved in the simple and conventional capillary model of pore structure are required for use of the present analysis. However, these parameters would be expected to depend upon the porosity of the medium, and this dependence has not yet been evaluated. A surprising result of the present work is an indication that inertial effects may arise at extremely low Reynolds numbers in the case of highly nonlinear fluids.

Overall, the present study is believed to contribute to a clearer conceptual description of flows of nonlinear fluids through porous media, but it requires further development.

DEFINITION OF THE PROBLEM, BACKGROUND

A very great variety of phenomena associated with flows through porous media have been discovered, and it appears prudent to begin by defining the question to be addressed as precisely as possible. Specifically, we shall consider continuum effects in circular pores and omit any consideration of three special problems:

1. Surface effects at the fluid-solid interface include the phenomena of adsorption (thereby constricting the pore when its diameter is comparable to the thickness of the adsorbed layer), surface slippage, plugging of the finer pores, and partial separation of multicomponent solutions or mixtures. The state of knowledge in this area is summarized by Burcik (1965, 1967, 1968), McKinley et al. (1966), Kozicki et al. (1967), Savins (1969), Maerker (1973), and McAuliffe (1973). These problems may be of substantial importance in media having very fine pores, as encountered in petroleum reservoirs and in flows through aquifers. However, they are clearly distinct from continuum effects and should be studied separately, preferably after an adequate understanding of the continuum problem has been obtained, since it is not clear that any other mechanism for uncoupling the two is available.

2. Viscoelastic effects in flows through porous media may lead to exploitable mobility-control techniques in which bypassing of the denser portions of a porous medium (and short-circuiting through the largest pores) may be minimized (Marshall and Metzner, 1967). Again, however, we probably need to understand the flows of inelastic fluid continua before much light can be shed on this question. Some results are, however, available to the reader in the above study and in those of Dauben and Menzie (1967), Savins (1969), Wissler (1971), Gregory and Grisley (1967), and Park (1973).

3. For flows of emulsions through porous media, a variety of special problems arises when the droplet size of the emulsified phase approaches or exceeds the diameter of the constricted portion of the pore (Melrose and Brandner, 1974). It is believed that the maximum dimensions of the droplets comprising the dispersed phase in the present work are small as compared to the pore sizes, usually several orders of magnitude smaller (Falco et al., 1974), and that these complications are therefore absent.

The above restrictions reduce the present study to one of flows of fluids which may be described by the rheological constitutive equations

$$S = -pg + P \quad (1)$$

$$P = 2\bar{\mu} d \quad (2)$$

in which the viscosity function $\bar{\mu}$ is determined by the scalar invariants of the deformation rate tensor d . We will follow convention in assuming that the dependence of $\bar{\mu}$ on deformation rate is given by a function of the second invariant II_d only and that the simple power law may be used to describe this dependency.

The equivalent capillary model of the pore structure through which the fluid flow occurs yields, when combined with the power law model of the fluid properties

$$f = \frac{\Delta p D_p \epsilon^3 \rho}{L G^2 (1 - \epsilon)} \quad (3)$$

$$Re = \frac{D_p G^{2-n} \rho^{n-1}}{150 H (1 - \epsilon)} \quad (4)$$

as shown by Christopher and Middleman (1965). The shear rate at the wall of the pores may be determined from

$$\left(\frac{-\partial v_x}{\partial r} \right)_R = \frac{3(3n+1)(1-\epsilon)V_o}{n D_p \epsilon^2} \quad (5)$$

For flows free from surface, inertial and viscoelastic effects the above definitions lead to

$$f Re = 1.0 \quad (6)$$

Although a number of authors appear to have used these equations successfully in the correlation of data, several points should be noted. First, even if every fluid element had to traverse each successive particle completely from one pole to the other, the ratio of actual length of the streamlines to the length of the bed would be only $\pi/2$; thus, a tortuosity as great as 25/12, as commonly used, must be compensating for some limitation or inadequacy of the model as well as describing the ratio of the length of the streamlines to the bed length. Secondly, when an investigator has been faced with the inadequacy of Equations (1) to (4), he has almost invariably chosen to relate this difficulty to limitations in the power law model of the fluid properties, and it should be useful to refute this fallacy once and for all, as follows.

The flow rate Q_P through a single pore of radius R and length L may be obtained by means of two integrations as

$$v_x = \int_0^{v_x} dv_x = - \int_R^r \left(\frac{P_{rx}}{K} \right)^{1/n} dr$$

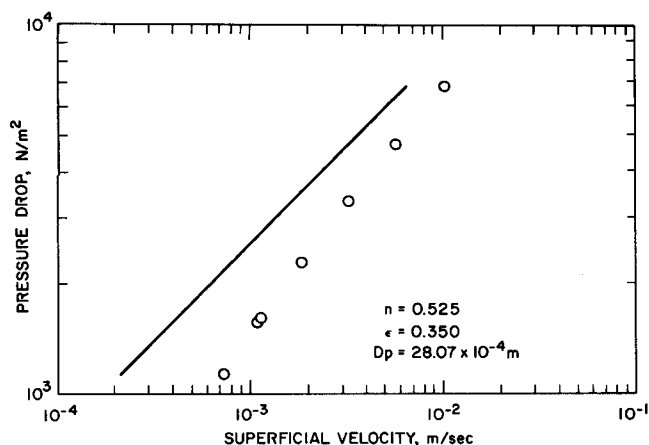


Fig. 1. Comparison of predictions based upon the capillary model (solid line) with experimental results. Data are for a 0.5% solution of Natrosol 250H from the work of Sadowski (1963).

$$= - \int_R^r \left[\frac{1}{2K} \left(\frac{-\partial p}{\partial x} \right) \right]^{1/n} (r)^{1/n} dr \quad (7)$$

and

$$\frac{Q_p}{2\pi} = \int_0^R (v_x r) dr \quad (8)$$

The parameters n and K must, of course, be determined at the deformation rate levels of interest in a particular problem; from Equations (7) and (8) it is clear that the range extends from $-\partial v_x / \partial r$ (or P_{rx}) equal to zero (at $r = 0$) to a maximum value at $r = R$. However, one also sees that the integrand of Equation (8) approaches zero as r approaches zero, and the flow rate Q_p is therefore insensitive to errors in the integrand for small values of r . In fact, it is easy to show that the range $0 < r < R/4$ only contributes between 12% (at $n = 1.0$) and 6% (at $n = 0$) of the total volumetric flow rate. Inspection of all of the $P_{rx} - d_{rx}$ data available on fluids whose flow through porous media has been studied shows that if the power law parameters have been properly determined over a range of deformation rates downward from the level specified by Equation (5), then only rarely has the breakdown of this constitutive equation been great enough to be measurable, much less significant. This does not prove that the power law will always suffice; it does show that substantially none of the assertions to the contrary are factual.

Turning to the question of just how good the available correlations based on a capillary model of the porous medium and a power law model of the fluid behavior are, we should consider carefully the following independent results and analyses.

1. An especially good set of experimental data for polymer solution flows through packed beds are those of Sadowski (1963, 1965). Inspection of his published rheological data (Figure 1 of his paper) reveals an excellent approximation of the fluid properties to power law behavior, and an analysis of all his measurements, based on Equations (7) and (8), confirms that any errors due to deviation from a power law description must be too small to be measurable. These data were correlated by using a standard friction factor-Reynolds number diagram [based on Equations (3) and (4)] by Christopher and Middleman (1965) with an average deviation of only 12%. By contrast, Savins (1969) has depicted some of the same results on a plot of the predicted pressure drop vs. that experimentally observed, and, on these coordinates, many of the data appear to fall outside the $\pm 20\%$ error lines indicated. In Savins' figure the experimental pressure drop is usually smaller than that predicted by Equations (3) and

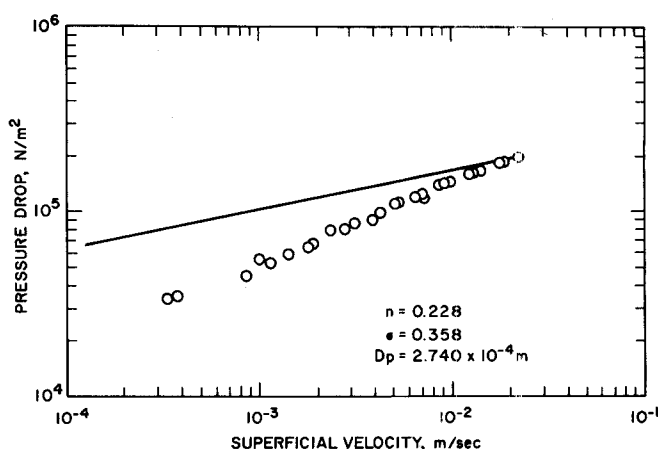


Fig. 2. Comparison of capillary model with experimental results (cont.). Data are for an emulsion from the work of Sheffield (1975).

(4), and the error appears to increase as flow rate decreases. Figure 1 compares the actual pressure drop-superficial velocity results for one set of Sadowski's data with predictions based on the model embodied in Equations (3) and (4); it is clear here that the predicted behavior deviates greatly from that observed experimentally. We should note that as the measured flow rates are higher than those predicted neither inertial or viscoelastic effects are likely to be responsible for the deviation from theory.

2. Data obtained by using emulsions are especially revealing because the accuracy of the experimental results is frequently so high as to reveal even subtle trends in the data. Uzoigwe and Marsden (1970), using a format derived from the same principles employed in obtaining Equations (3) and (4), presented their results in terms of predicted superficial velocities as compared to experimental velocities at a given pressure drop. The comment is made that, in most cases, the model predicts velocities which are lower than those measured experimentally, just as in Figure 1. The average error in the superficial velocities appears to increase as the degree of non-Newtonian behavior of the emulsions increases and ranges from 5% for a Newtonian emulsion to 14.5% for the most highly non-Newtonian emulsion ($n = 0.84$). Gogarty (1967a, b) studied the flow of surfactant-stabilized dispersions of water in hydrocarbons, similar in structure to the emulsions used by Uzoigwe and Marsden and to the fluid used in our own studies. Gogarty found that his fluids exhibited shear thinning behavior both in viscometric and porous medium experiments, but that the apparent value of the power law index was enormously different in the two experiments. Gogarty was thus unable to correlate his data by combining the capillary model, on an *a priori* basis, with a suitable rheological model, and he developed an empirical set of correction factors. The fluids employed by Gogarty were highly non-Newtonian ($n = 0.35$ to 0.54), and, thus, the failure of the capillary model for these fluids is consistent with the observation of Uzoigwe and Marsden which indicated increasing deviation from the capillary model with decreasing values of the power law index. The generality of the experimental results reported by Gogarty were confirmed in this study by employing both emulsions and porous media having appreciably different characteristics from those of his study; one set of these results is depicted in Figure 2. We note that the trends are qualitatively similar to those observed in Figure 1 and that a prediction of the flow rate obtainable at a given pressure drop (more usual pragmatically than the inverse calculation of pressure drop at a given flow rate) may be in error by as much as 1 to 2 orders of magnitude. Extrapolation of the results to flow rates of interest in petroleum production

(in the range of 10 ft/day or 3.5×10^{-5} m/s) would incur yet greater discrepancies between theory and experiment.

Clearly, the predictive abilities of Equations (3) and (4) appear to be severely limited. Especially disturbing is the very small scatter of the data on friction factor-Reynolds number diagrams and the much larger level of error evident in Figure 1. The trends noted in the data for emulsions (increases in level of the error as the nonlinearity of the fluid increases) provide the key to the resolution of this enigma. We must observe that Equations (3) and (4), although of the standard format commonly employed in portrayal of results for Newtonian fluids, may be extremely misleading in the case of highly shear thinning non-Newtonian systems. To illustrate this assertion, let us suppose a set of data yields superficial velocities which are larger than predicted by a factor of 10. The friction factor computed from Equation (3) will be lower than expected by a factor of 100 because of the increased value of G . If the fluid happens to be Newtonian ($n = 1$), the Reynolds number [Equation (4)] will be increased by a factor of 10^{2-1} , leaving the data point tenfold removed from the correlating line [Equation (6)]. However, as the fluid becomes progressively more shear thinning ($n \rightarrow 0$), the shift in Reynolds number increases; indeed, as the rheological parameter n approaches zero, the shift in Reynolds number will be such as to place this erroneous point precisely back onto the line of perfect correlation! For the fluid of Figure 2 ($n = 0.23$), a tenfold actual error will show up only as a deviation of about 40% from the Reynolds number friction-factor correlation. Lest there be the slightest doubt on this point, let us emphasize that there has been no falsification of any results and that the friction factor-Reynolds number coordinates remain as the most convenient means for correlating a wide variety of data. We must, however, be aware of the diminished sensitivity of these coordinates as the power law index decreases toward zero.

ANALYSIS

As a fluid element moves forward through the interstices of a porous medium it will be exposed to a channel whose dimensions (size and shape of the cross section) change progressively, and which, indeed, is not portrayed at all closely by the usual idealization of a cylindrical capillary tube of constant diameter. As a first approximation to the actual stochastic pore geometry let us assume that the major effects may be portrayed by a tube which is of cylindrical cross section but whose diameter varies sinusoidally with axial position:

$$D = D_{\min} \left(\frac{1 + b \sin x}{1 - b} \right) \quad (9)$$

The parameter b is defined as

$$b = \frac{D_{\max} - D_{\min}}{D_{\max} + D_{\min}} \quad (10)$$

In this analysis the maximum pore diameter is assumed to equal the particle diameter; thus

$$b = \frac{D_p - D_{\min}}{D_p + D_{\min}} \quad (11)$$

It is instructive to compare this model of the pore structure with the cylindrical (parallel walled) capillary model which led to Equations (3) to (6). In the capillary model the actual geometry is replaced by an average diameter so chosen that the regions of small diameter, in which the pressure gradient is highest, are exactly counterbalanced by the regions of largest diameter and lowest pressure gradient. For laminar, well-developed flow of purely vis-

cous fluids through a capillary tube

$$-\frac{\partial p}{\partial x} \propto \frac{Q_p^n}{D^{3n+1}} \quad (12)$$

or

$$\Delta p = c_1 Q_p^n \int_0^L \frac{dx}{D^{3n+1}} \quad (13)$$

Equation (13) shows that in the Newtonian case ($n = 1.0$) the average pore diameter employed in the capillary model of flows through porous media is one in which the correct average pressure drop is obtained by averaging the term $D^{1/4}$ along the length of the pore. If we consider nonlinear fluids, the corresponding term to be averaged is $1/D^{3n+1}$, clearly a greatly different quantity. Equations (12) and (13), based upon the assumption of a well-developed laminar flow, may be too simple to represent all real flows, but even this greatly simplified model reveals that departure of the data from the curve predicted by Equations (3) to (6), as in Figures 1 and 2, is to be expected. Further, Equation (13) suggests that the explicit diameter-distance relationship chosen will be quite important in view of the numerical value of the exponent of the diameter term. As a first approximation, the sinusoidal geometry depicted by Equations (9) and (11) has been chosen for the present analysis, but this may be in need of subsequent refinement. For example, the geometry chosen by Payatakes et al. (1973), though more complex, may be more realistic.

Newtonian Fluid Flow in Sinusoidal Pores

We begin, as in Equation (12), by assuming that under conditions of slow laminar motion the local pressure gradient in a converging or diverging pore is the same as that for well-developed flow in a cylindrical pore. This approximation has also been employed in the porous medium flow studies of Dullien (1975). An algorithm which enables the avoidance of this approximation has been published by Payatakes et al. (1973a, b), but its extension to non-Newtonian fluids is not trivial. Use of Poiseuille's law to describe the local pressure gradient—diameter relationship

$$-\frac{\partial p}{\partial x} = \frac{128\mu Q_p T}{\pi D^4} \quad (14)$$

when combined with Equations (9) and (11) gives after considerable algebraic manipulation

$$Q_p = \frac{\pi \Delta p D_{\min}^4 (1 - b^2)^{7/2}}{64\mu TL (1 - b)^4 (2 + 3b^2)} \quad (15)$$

The tortuosity term T has been inserted in Equations (14) and (15) to account for the effective pore length TL .

The superficial velocity through the porous medium may be determined from

$$V_o = Q_p n_p \quad (16)$$

in which n_p denotes the number of pores per unit cross-sectional area of the porous medium. It may be shown (Sheffield, 1975) that this quantity is related to the other porous medium parameters by

$$n_p = \frac{8\epsilon(1 - b)^2}{\pi D_{\min}^2 (2 + b^2)} \quad (17)$$

Combining Equations (15), (16), and (17), one obtains

$$V_o = \frac{\epsilon}{4} \left[\frac{D_{\min}}{2} \right]^2 \left[\frac{\Delta p}{2\mu TL} \right] \left[\frac{4(1 - b^2)^{7/2}}{(1 - b)^2 (2 + 3b^2) (2 + b^2)} \right] \quad (18)$$

Equation (18) is similar in form to the traditional equation derived from the capillary model (Bird et al., 1960)

$$V_o = \frac{D_p^2 \Delta p \epsilon^3}{72\mu CL (1 - \epsilon)^2} \quad (19)$$

the only differences being a different definition of diameter in the second term and the presence of a factor which is solely dependent on the parameter b to account for the converging-diverging nature of the new model. Combination of Equations (18) and (19) enables one to relate the parameters b and D_{\min} to the other properties of the porous medium:

$$D_{\min} = 2 \left[\frac{2T D_p^2 \epsilon^2 (1 - b)^2 (2 + 3b^2) (2 + b^2)}{150 (1 - \epsilon)^2 (1 - b^2)^{7/2}} \right]^{1/2} \quad (20)$$

To obtain Equation (20) the constant C in Equation (19) was taken as 25/12.

Behavior of Power Law Fluids in Sinusoidal Pores

The power law analogue of Equation (14) may be written as

$$Q_p = \frac{\pi n}{3n + 1} \left(\frac{D}{2} \right)^{\frac{3n+1}{n}} \left[\frac{1}{2KT} \left(\frac{-\partial p}{\partial x} \right) \right]^{1/n} \quad (21)$$

Let us consider the movement of an element of the fluid as it emerges from the throat of a pore and proceeds forward into the diverging region. As this occurs, the element must contract axially and stretch radially in order to continue to conform to the pore-wall geometry. Considering the resistance to deformation to be given as an apparent viscosity, one may note that a fluid element having a flow behavior index of less than unity becomes progressively more resistant to deformation as one moves radially from the pore wall toward the center line. Consequently, this process of conforming to the pore-wall geometry at a given flow rate becomes progressively more difficult as the power law index decreases toward zero. In other words, power law fluids will be expected to exhibit a greater tendency to separate in a diverging flow and to channel through the central region of a pore than do Newtonian fluids at the same flow rate or Reynolds number; the Reynolds number at which the fluid channels through the central region of the pore, with a diameter everywhere not differing greatly from the pore throat diameter, will tend to zero as the flow behavior index approaches zero.

Under conditions of fluid channeling, the effective diameter of a pore, at all points except the throat, will be lower than the local value of the actual geometric diameter D . We may define the pore geometry in terms of such an effective diameter by analogy to Equation (9) as

$$D_{\text{eff}} = D_{\min} \left[\frac{1 + b' \sin x}{1 - b'} \right] \quad (22)$$

in which b' denotes an empirical factor which one would expect to depend on the rate of flow through the porous medium (Reynolds number) and the degree to which the fluid being tested deviates from Newtonian behavior. The factor b' , which must always be less than or equal to the fixed parameter b , may therefore be introduced to evaluate the effective pore shape to which the non-Newtonian fluid is assumed to conform and may be determined from experimental data.

In applying Equation (21) under conditions of separation of the fluid from the wall of the pore, it is assumed that the pressure gradient will be inversely proportional to the $(3n + 1)$ power of the instantaneous value of the ef-

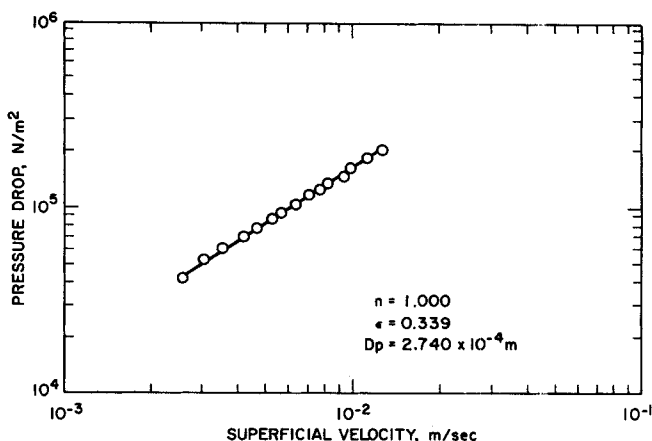


Fig. 3. Comparison of predictions based upon a capillary model of the porous medium with experimental measurements. Data are for glycerin from the study of Sheffield (1975).

fective pore diameter D_{eff} :

$$-\frac{\partial p}{\partial x} = \frac{A_{PL}}{(D_{\text{eff}})^{3n+1}} \quad (23)$$

The pressure drop over a bed of length L becomes

$$\Delta p = \frac{L A_{PL}}{2\pi} \left[\frac{1 - b'}{D_{\min}} \right]^{3n+1} \int_0^{2\pi} \frac{dx}{(1 + b' \sin x)^{3n+1}} \quad (24)$$

Combining Equations (16), (17), and (21) to (23) one obtains, as a generalization of Equation (18)

$$V_o = \frac{8n\epsilon}{3n + 1} \left(\frac{1}{2} \right)^{\frac{3n+1}{n}} \left[\frac{A_{PL}}{2KT} \right]^{\frac{1}{n}} \left[\frac{(1 - b)^2}{D_{\min}^2 (2 + b^2)} \right] \quad (25)$$

in which D_{\min} is defined by Equation (20), as before, and the term A_{PL} may be obtained from Equation (24).

An estimate of the wall shear rate in the porous medium may be obtained from Equations (24) and (25) as

$$\left(\frac{-\partial v_x}{\partial r} \right)_R = \frac{V_o (3n + 1) (1 - b')^3 (2 + b^2)}{n \epsilon D_{\min} (1 - b)^2} \quad (26)$$

and the corresponding stress becomes

$$(P_{rx})_R = \frac{\pi \Delta p}{2TL \int_0^{2\pi} \frac{dx}{(1 + b' \sin x)^{3n+1}}} \left[\frac{D_{\min}}{1 - b'} \right] \quad (27)$$

One possible definition of a friction factor for flows through porous media is

$$f = \frac{(P_{rx})_R}{\rho V_o^2} \quad (28)$$

The corresponding Reynolds number [Equation (6)] is

$$Re = \frac{D_{\min}^n V_o^{2-n} \rho}{K} \left[\frac{1}{2(1 - b')} \right]^{3n} \left[\frac{8n\epsilon(1 - b)^2}{(3n + 1)(2 + b^2)} \right]^n \quad (29)$$

For Newtonian fluids in noninertial flows, Equation (29) reduces to

$$Re = \frac{D_p G}{\mu(1 - \epsilon)} \left[\frac{\epsilon^2}{2(2 + b^2)} \right]$$

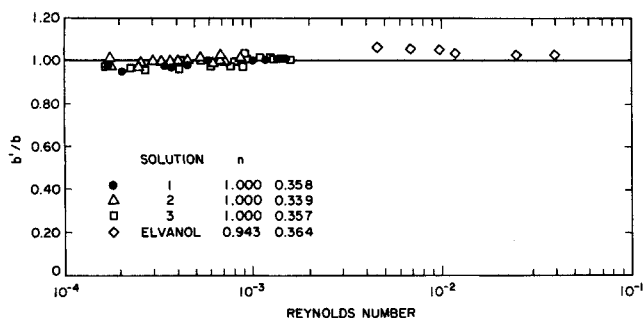


Fig. 4. Comparison of predictions based upon the converging-diverging pore model with noninertial experimental data for Newtonian and nearly Newtonian fluids. Data from Sadowski ($n = 0.943$) and Sheffield ($n = 1.00$).

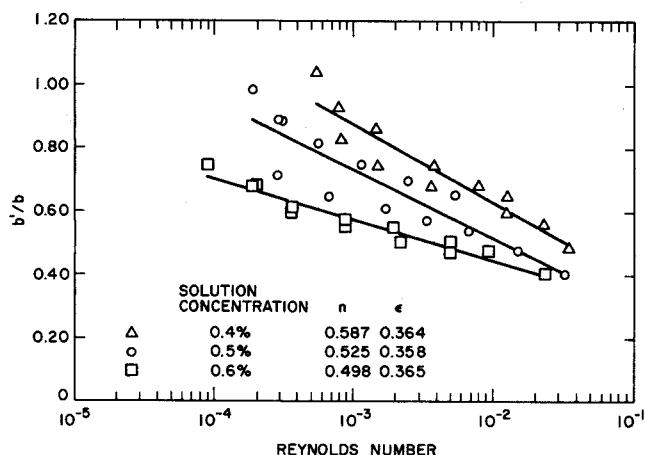


Fig. 5. Application of the converging-diverging pore model to non-linear polymer solutions. Data from Sadowski (1963) for various concentrations of Natrosol 250H.

$$\left[\frac{2T(2 + 3b^2)(2 + b^2)}{150(1 - b^2)^{7/2}} \right]^{1/2} \quad (30)$$

This Reynolds number may be compared to that of Christopher and Middleman as

$$Re = \frac{D_p G}{\mu(1 - \epsilon)} \left[\frac{1}{150} \right] \quad (31)$$

Comparison of Equations (30) and (31) indicates that, for any given porous medium, the two Newtonian fluid Reynolds numbers will differ by only a constant which is a function of porosity, tortuosity, and the geometric parameter b .

EXPERIMENTAL EVALUATION OF ANALYSIS: CHOICE OF FLUID SYSTEMS

An extensive evaluation of all experimental results available in the literature for flows of polymer solutions, molten polymers and emulsions was carried out to select those systems which could be used to obtain the most definitive assessment possible of the converging-diverging pore model. Additionally, two highly nonlinear emulsions having a water:oil ratio of 1.4 were prepared in the present study. According to Falco et al. (1974), these are expected to have a lamellar structure and were found, with the exception of a small fraction of the data of Park (1973), to be more highly nonlinear than any other laminar flow system studied to date in porous media ($n = 0.23$ and 0.26). The porous media used had a particle diameter of 2.74×10^{-4} m, a length of 6.35×10^{-3} m, a void fraction ranging from 0.339 to 0.359, and permeabilities ranging from 44.53 to 56.06×10^{-12} m². A 90% by weight glycerin

solution was also used to evaluate the modeling of the flow through these media.

Overall, the data chosen to evaluate the analysis covered the following ranges in system properties:

Non-Newtonian behavior	: $0.23 < n < 1.0$
Diameter of particles comprising the porous media	: $0.55 \times 10^{-4} < D_p < 28.1 \times 10^{-4}$ m
Void fraction of porous media	: $0.17 < \epsilon < 0.37$

RESULTS

Figure 3 compares the behavior predicted by using the cylindrical capillary model of the porous medium, as shown by the solid line, with experimental data for flow of a Newtonian fluid through one of the porous media used in the present study. The ability of this simple model to portray the results satisfactorily is in stark contrast to its breakdown in the case of nonlinear fluids (Figures 1 and 2).

All of the Newtonian fluid results obtained in the present study, as well as data from Sadowski (1963) for a nearly Newtonian fluid, are compared with the converging-diverging pore analysis in Figure 4. Under the noninertial conditions used [a Newtonian fluid should exhibit no inertial effects until the Reynolds number defined by Equation (30) exceeds 0.1], the ratio b'/b must be unity if the analysis is valid; the average deviation of the Newtonian data from the requirement that b'/b be equal to unity is only 1.5%; the average deviation of the Elvanol data is 4.4%, and the maximum deviation of any point from the curve is 6.1%. These calculations presupposed a tortuosity of unity [Equation (25)], and the values for the parameter b ranged between 0.552 and 0.576; this corresponds to pores whose ratio of maximum-to-minimum diameter falls between 3.47 and 3.72. Thus, the converging-diverging pore model appears to correlate noninertial flows of Newtonian fluids at least as well as the capillary model, has the same number of adjustable parameters (the tortuosity T which, perhaps, may not always be set equal to unity), but involves a more reasonable numerical value for this parameter.

Turning to nonlinear fluid systems, Figure 5 shows results for three polymer solutions of progressively increasing nonlinearity. As noted in the analysis, the Reynolds number corresponding to the onset of inertial effects, as revealed by values of the ratio $(b'/b) < 1.0$, should decline from its value for Newtonian fluids (0.1) toward zero as the fluid behavior index decreases toward zero. This is seen to be the case, although a limitation of these, and other highly nonlinear data, is that none appear to extend well into the noninertial region in which the ratio b'/b would be equal to unity over a range of Reynolds numbers. Figure 6 shows similar results for the emulsions used in this study. The results indicate that at Reynolds numbers below 10^{-6} , the fluid effective geometry is the actual pore geometry, and, as the Reynolds number increases, the fluid begins to channel through the pores. At a Reynolds number slightly greater than 10^{-3} , the fluid is effectively passing through a capillary of diameter D_{min} . In spite of substantial efforts to do so, no data could be obtained at Reynolds numbers appreciably below 10^{-6} or greater than 3×10^{-3} . For all of the data available from Sadowski (seven fluids) and the present work (two fluids), the average percentage deviation of the data from a semi-logarithmic correlating line as shown in Figures 5 and 6 was slightly below 6%.

A typical set of data from the study of Gogarty is given in Figure 7. These indicate that not only does the ratio

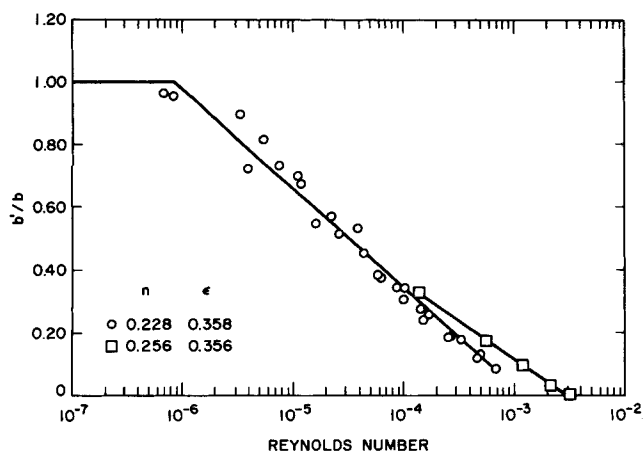


Fig. 6. Results for microemulsion flows through porous media. Data of Sheffield (1975).

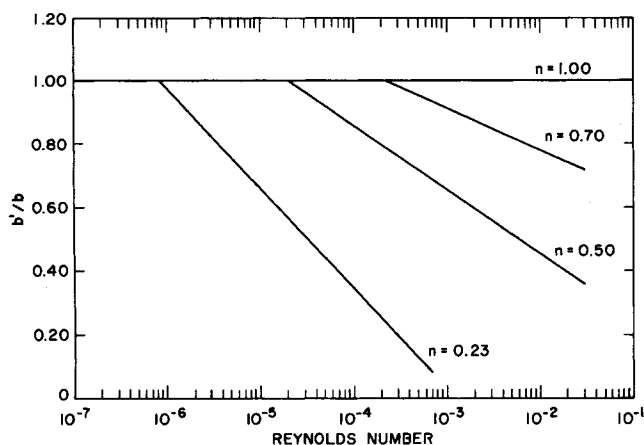


Fig. 8. Design chart for nonlinear fluids flow through porous media for which $0.34 < \epsilon < 0.38$.

b'/b depend upon the Reynolds number and the power law parameter n , but also upon either particle diameter D_p or the porosity ϵ . Since the data of Sadowski indicated no effect of D_p over a wider range of particle diameters, the effect is presumably one of the void fraction ϵ . The large difference in porosities between the data in Figures 5 and 6 on the one hand, and Figure 7 on the other, also leads to major changes in the value of b'/b at a given Reynolds number. It is additionally reasonable that the much denser packing of the particles in the porous media used by Gogarty would lead to the presence of much smaller throat diameters for the pores in the medium, and therefore to much greater values for the parameter b . This is indeed the case; values for b are in the range 0.69 to 0.74, as compared to 0.55 to 0.58 for the less densely packed beds considered earlier.

Figure 8, based on the data of Sadowski and of the present work, summarizes the results in design chart form for the more porous beds studied ($0.34 < \epsilon < 0.38$). At this stage of understanding of the problem, a different chart of this kind, or its numerical equivalent, is needed for each different level of bed porosity.

APPLICATION TO DESIGN

The analysis presented could be used in the following manner to make *a priori* predictions of the pressure drop-flow rate relationships for nonlinear fluids in porous media:

1. The physical properties of the fluid (K , n , ρ) and of the porous medium (D_p , ϵ) must be determined by using standard techniques.
2. The coupled parameters b and D_{\min} are computed

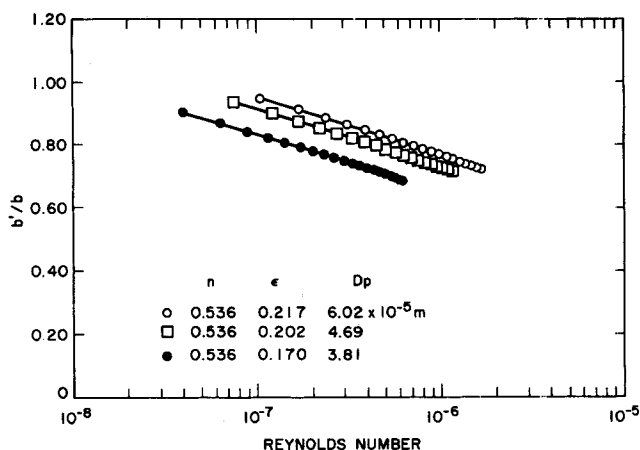


Fig. 7. Results for microemulsions (cont.). Data for fluid B of Gogarty (1967a) as taken from smoothed curves.

from Equations (11) and (20), taking T as equal to unity in Equation (20).

3. The second pair of coupled parameters b' and Re [Equation (29)] are determined in conjunction with a design chart such as Figure 8 for a particular chosen velocity V_o .

4. The integral in Equation (24) is evaluated (numerically) to provide a relationship between the parameter A_{PL} and the pressure drop Δp . This is used with Equation (25) to obtain the desired pressure drop at the velocity chosen in step 3. Again, in Equation (25), the tortuosity T is taken as unity.

5. As a final step always involving calculations for non-Newtonian fluids the shear rate is computed [Equation (26) to ensure that the rheological parameters K and n were actually evaluated at the levels of shear rate encountered in the problem.

The above procedure simplifies appreciably if the Reynolds number is sufficiently low for the ratio b'/b to be taken as unity. It appears likely that this would normally be the case, for example, in calculations dealing with problems encountered in either aquifer flows or in petroleum production operations. We wish to emphasize again that under such noninertial conditions use of this converging-diverging pore model requires no information other than that also required for calculations employing the capillary model of the porous medium.

EVALUATION OF THE CONVERGING-DIVERGING PORE MODEL

Newtonian Fluids

The glycerin solutions studied in this work were all confined to the noninertial region ($Re < 0.1$). The average errors in friction factor or pressure drop predicted for these three fluids were 1.91, 1.63, and 2.33%; when the capillary model is used instead, the corresponding errors were 2.01, 1.53, and 2.32%, respectively. In other words, both models are of equivalent value when applied to these simple systems.

Nonlinear Fluids

The converging-diverging pore model predictions for the emulsion flow studies conducted during this work predict the experimental friction factors with an average deviation of 2.67%, while the average deviation in the capillary model friction factor for the same data was 29.1%. The deviations in the capillary model friction factor ranged from -10.6 to 59.2%, while the range of deviations in the converging-diverging pore model predictions was from -5.84 to 7.76%. The deviations in the capillary model friction factor steadily decreased with increasing Reynolds

number; the errors in the predictions of the converging-diverging model were of a random nature. Thus, it is clear that the capillary model does not represent the flow of non-Newtonian fluids in a porous medium as accurately as the converging-diverging pore model does.

The results of the friction factor-Reynolds number calculations based on the converging-diverging model of the porous medium for Sadowski's fluids gave an average deviation of 6.8%, while the maximum error was never above 20%. Sadowski's correlation, based on a capillary model but employing one more parameter than used in the present study (two more if $T = 1.00$), correlated the data with an average deviation of 18%. The recorrelation by Christopher and Middleman gave average deviations of 12% but was believed to deal with noninertial flows and therefore employed one less parameter than required in the present work. Finally, we must note that although friction factor-Reynolds number correlations provide for a convenient and general representation of a variety of results, they do tend to mask the actual errors involved, and the overall results are not as precise as the above figures suggest. The 2.67% average deviation in the friction factors calculated for the emulsion data corresponds to an error of 12% in the velocity predicted at a given pressure drop; likewise, the 0.6% Natrosol data of Figure 5 were correlated with an average deviation of 9.70% (less precisely than all of the Sadowski data taken together), and this corresponds to a 23% average error in prediction of velocity at a given pressure drop level. The 29.1% average error in friction factor, with the capillary model used, corresponds to a 300% average error in prediction of the flow rate at a given pressure drop level.

Overall, it is seen that the converging-diverging pore model appears to correlate data appreciably more precisely than the conventional capillary model of the porous medium, but a significant number of unresolved problems remain. Paramount among them is the extraordinarily low value of the Reynolds number at which inertial effects appear to arise (Figures 5 to 8); are these real or merely a spurious masking of some other deficiency of the model? Secondly, the simple sinusoidal form chosen for the pore geometry [Equation (9)] cannot, surely, be applicable to all porous media. Does this underly, for example, the sensitivity of the model to porosity as noted in the discussion of Figure 7, or are the details of the pore geometry chosen not of critical importance overall? The noncircularity of the pores, as well as the distribution of cross-sectional area changes, could be of significance if the flow rates are highly sensitive to geometric details. Further, several excellent sets of data for nonlinear fluids were omitted from consideration because of the apparent occurrence of viscoelastic and major inertial effects. These phenomena deserve analysis. Finally, if data such as those shown in Figures 5 to 7 could be obtained at Reynolds numbers which are lower by one or more orders of magnitude than presently available an *a priori* evaluation of the model, with no adjustable parameters, would be possible.

In summary, the present work conclusively demonstrates the inapplicability of the conventional capillary model to flows of nonlinear fluids through porous media. A plausible alternative has been suggested and shown to be applicable at least over a restricted range of conditions. The study is, however, of an introductory nature, and a substantial number of problems remain unresolved.

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NOTATION

- A_{PL} = pressure gradient proportionality constant, power law fluids
 b = fixed geometric parameter, Equation (11)
 b' = effective geometric parameter, Equation (22)
 C = tortuosity term in capillary model of porous medium, Equation (19)
 D = actual local pore diameter
 D_{eff} = effective pore diameter
 D_{max} = maximum pore diameter
 D_{min} = minimum pore diameter
 D_p = particle diameter
 d = deformation rate tensor; shearing deformation rate in cylindrical coordinates denoted as d_{rz}
 f = friction factor
 G = superficial mass velocity, ρV_o
 g = metric tensor
 H = Darcy's law viscosity function for power law fluids (see Christopher and Middleman, 1965)
 K = rheological power law parameter (consistency index)
 L = length of porous medium
 n = rheological power law parameter (flow behavior index)
 n_p = number of pores per unit cross-sectional area of medium
 P = deviatoric stress tensor; shearing stress component (in cylindrical coordinates) given as P_{rz}
 p = isotropic pressure
 Δp = pressure difference
 Q_p = volumetric flow rate in one pore or tube
 R = radius of pore
 r = radial position in a pore
 Re = Reynolds number [Equation (29) employed in all figures]
 S = total stress tensor
 T = tortuosity
 V_o = superficial velocity
 v_z = local axial velocity in a pore
 x = dimensional axial position parameter
 ϵ = porosity or void fraction
 μ = viscosity coefficient, Newtonian fluid
 $\bar{\mu}$ = apparent viscosity, nonlinear fluid
 ρ = fluid density
 II_d = second invariant of deformation rate tensor

$$II_d = \frac{1}{2}[tr d^2 - (tr d)^2]$$

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An Examination of the Adaptive Random Search Technique

Random search procedures have recently been successfully applied to the optimization of a variety of chemical engineering problems, including optimization of chemical processes by flow sheet simulation. These procedures represent the independent variables as random variables described by probability distributions. The adaptive random search procedure centers the distribution for each variable about the best search point found and examines this region for a better point. Thus, this technique has the ability of continuously moving the search region toward the optimum, which is particularly advantageous in following constraints.

This study examines the efficiency of the adaptive random search technique as applied to six different problems, which have been previously solved by various other techniques.

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SCOPE

Recently, random search techniques have been applied to the optimization of chemical processes (Luus and Jaakola, 1973a; Gaines and Gaddy, 1976). A random search procedure is characterized by random determination of the independent variables in a given search region. Unlike many techniques, the random search does not require approximations, auxiliary functions, or an extensive knowl-

edge of the problem under study in order to obtain an optimum solution. The success of a random procedure is not jeopardized by a nonlinear objective function or nonlinear implicit constraints.

The adaptive or pseudo random search selects new search points by sampling a probability distribution centered about the best known value of the objective func-