"the flow anomaly is explained by utilizing the Carreau viscosity equation . . . in conjunction with the modified Rabinowitsch equation."

Flow of Viscoelastic Fluids through Porous Media

by
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ABSTRACT

In the laminar flow of purely viscous non-Newtonian fluids through porous media, the effects of shear thinning on pressure drop are well understood and can be readily accounted for with appropriate modification of the Blake-Kozeny equation. For viscoelastic fluids at large deformation rates, a gross increase in pressure drop over and above the prediction from the purely viscous theory has been observed. This deviation was observed for two sets of viscoelastic fluids: aqueous polymer solutions (shear thinning elastic) and corn syrup-Separan mixtures (elastic but constant viscosity).

A modified Rabinowitsch equation employing two geometric parameters, originally proposed by Kozicki et al. (1967), is used in conjunction with the Carreau viscosity equation to describe the anomalous flow resistance observed for the flow of aqueous polymer solutions through a packed bed. For fluids which are highly shear thinning and elastic, a prediction based solely on the zero shear viscosity represents the upper bound of the viscous contributions. The difference in pressure drop between the experiment and the upper bound prediction can be attributed to the elastic effect.

The Deborah number, an elastic parameter based on Maxwellian relaxation time, does not uniquely correlate the observed excess flow resistance for the highly elastic but nearly constant viscosity fluids (Boger fluids). An alternative correlation is proposed using a modified Deborah number based on the zero shear viscosity.

INTRODUCTION

Much of the interest in the study of flow of non-Newtonian fluids through porous media is generated by the increasing use of high molecular weight polymers to improve water flooding efficiency in enhanced oil recovery processes.

The reduction of water fingering and thus enhancement of oil recovery can be achieved by improving mobility ratio which is defined as the permeability of the porous medium divided by the viscosity of the fluid. A mobility ratio less than or equal to one ensures a piston-like displacement whereas at values greater than one, the more mobile water fingers through the oil zone, leaving some oil unrecovered. In theory, the mobility ratio can be controlled by reducing the oil viscosity or increasing water viscosity or through permeability changes. Virtually, all efforts have been aimed at increasing the viscosity of the displacid phase.

In water polymer flooding, the function of the polymer is to reduce the mobility of the injected water by raising the viscosity of water and lowering the permeability to water. Water soluble polymers have been proven to be so far the most efficient of any materials considered. The large size of the macromolecules suggests significant energy dissipation under the flow conditions and thus a substantial viscosity increase at low polymer concentrations. Currently, polysaccharide and partly hydrolysed polyacrlyamide are most often used.

Some enhanced oil recovery projects involving injection of non-Newtonian fluids have been moderately successful but most of these have performed below expectation. Hence, there is a need for a thorough study of the stability of non-Newtonian fluids at reservoir conditions and also a new look at the mechanism of the flow of non-Newtonian fluids in porous media.

In the laminar flow of purely viscous non-Newtonian fluids through porous media, the effects of shear thinning or pseudoplastic behavior on pressure drop are well understood and can be accounted for with appropriate modification of Darcy's Law or of the Blake-Kozeny equation. For viscoelastic fluids at high deformation rates, a gross increase in pressure drop over and above the prediction from the purely viscous theory has been observed. Various mechanisms, including elastic effects, polymer adsorption, gel formation on surfaces and shear thickening effects have been proposed to explain the sudden increase in the flow resistance. So far, none of these mechanisms have been shown conclusively to be the major factor in controlling the flow of viscoelastic fluids through porous media.

To further complicate the situation, most viscoelastic fluids not only exhibit elastic characteristics but are also highly shear thinning. It is not clear whether the flow anomaly observed in the flow through porous media is due solely to elastic effects, the combination of elastic and shear thinning effects or simply the inadequacy of the purely viscous theory in the highly shear thinning region.

In this study, the flow anomaly is explained by utilizing the Carreau viscosity equation, which possesses a zero shear viscosity and a characteristic time parameter, in conjunction with the modified Rabinowitsch equation originally proposed by Kozicki et al. (1967). A modified elastic number will be used for correlation.

THEORY

Kozicki et al. (1967) extended the Rabinowitsch equation for laminar pipe flow and porous media utilizing the concept of geometric parameters. In the absence of slip effect, the equation relating flow rate and pressure drop in a packed bed for any time-independent purely viscous fluid can be written as

$$2V/r_H = 2(1 + \xi)/k_i \; \tau_w^{\xi} \int_0^{\tau_w} (\tau^{\xi}/\eta)\, d\tau \qquad (1)$$

The aspect factor = 3, same as for circular pipe and the impermeability factor k_i = 4.6 to 5.2 for beds randomly packed with spherical particles. For a bed with small particle to column diameter ratio, the hydraulic radius can be shown to be (Hanna, et al., 1977).

$$r_H = \in d / 6(1 - \in) + 4(d/D) \qquad (2)$$

For an inelastic power law fluid, it can be shown from Eq. (1) that the apparent shear rates for capillary tubes and packed beds, determined at the same wall shear stress, are simply related by a shift factor $k_i/2$ that is

$$(2V/r_H)\text{ capillary tube} = (k_i/2)\,(2V/r_H)\text{ packed bed} \qquad (3)$$

Eq. (3) allows for the prediction of flow rate or pressure drop for a power law fluid flowing through a packed bed from viscometric data and vice versa.

Unfortunately, not all non-Newtonian fluids behave as power law fluids. Many polymer solutions exhibit a constant viscosity at low shear rates and a shear thinning characteristic at moderately high shear rates. In a complex flow situations such as in a packed bed, the apparent shear rates encountered in the tortuous paths could vary from zero to as high as several orders of magnitude. Hence, a realistic fluid model even for inelastic fluids, should possess a zero shear viscosity and a characteristic time constant.

Furthermore, most shear thinning polymer solutions are invariably viscoelastic. For this kind of fluid, the curve obtained in a packed bed is no longer parallel to the viscometric flow curve as predicted from Eq. (3). A sudden increase in pressure drop over and above the inelastic prediction beyond a certain point in the flow curve has been observed (Marshall and Metzner, 1967). The deviation from inelastic behavior has been correlated in terms of the elastic parameter, Deborah number (Marshall and Metzner, 1967) or Ellis (Sadowski and Bird, 1965; Hong, 1981). However, there has been little agreement with regard to the increase in pressure drop as a function of the elastic parameter and also the critical value which marks the onset of the flow anomaly (Gaitonde and Middleman, 1967, Siskovic et al., 1971). Marshall and Metzner (1967) found that for De <0.05, the power law model (purely viscous) describes the flow of a non-Newtonian fluid which may be viscoelastic, in a porous medium. However, if De >0.1, the power law model shows significant deviations. Siskovic et al. (1971) and Kemblowski and Martl (1974) suggested a critical Deborah number in the vicinity of 0.2.

Duda and Vrentas (1972, 1973) have shown that any rheological model which does not contain a characteristic time parameter will not predict an excess pressure drop with increase in flow rate in converging-diverging flow channel. Consequently, the Carreau viscosity equation has been chosen here to represent the viscous behavior of aqueous polymer solutions which exhibit a highly shear thinning viscosity. The Carreau viscosity equation is written as (Carreau, 1972)

$$(\eta-\eta_\infty)/(\eta_o-\eta_\infty) = [1+(\lambda\dot{\gamma})^2]^{(n-1)/2} \quad (4)$$

The equation describes all the viscous features which most of the shear thinning polymer solutions are known to exhibit. It contains the two limiting viscosities, η_o and η_∞ the power law index n, characterising the shear thinning region and a characteristic time parameter λ which is considered to be essential to empirically account for elastic effects (Abdel-Khalik et al., 1974). Chhabra et al. (1980; 1981) have successfully used the fluid model to study the creeping motion of a spherical particle in a viscoelastic fluid medium.

In integrating Eq. (1) using the Carreau viscosity equation, Eq. (4), it is preferable to express the integration variable in terms of shear rate using the relation $\tau = \eta\,\dot{\gamma}$. Also, the infinite shear viscosity η_∞ can be neglected as it is much smaller than the zero shear viscosity η_o for most aqueous polymer solutions. Analytical integration can be obtained for certain values of power law index n. The resulting expression for four values of n are presented in Table 1. Relationships for other values of n can be determined either by graphical interpolation or by numerical integration. Thus, knowing the fluid parameters η_o, λ and n, the flow rate-pressure drop relation in a packed bed for a fluid that can be described by the Carreau viscosity equation can be readily predicted.

Table 1. Flow rate-pressure drop relationships in packed beds using Carreau viscosity model

n	$2V/r_H$
1.0	$2\dot{\gamma}_w/k_i$
0.667	$A[1-B/3\{2+(1+y_w)^{0.5}(y_w-2)\}]$
0.333	$A[1-B/2\{y_w-\ln(1+y_w)\}]$
0	$A[1-B\{(2+y_w)(1+y_w)^{-0.5}-2\}]$
Where y_w =	$(\lambda\dot{\gamma}_w)^2$; $A=8\dot{\gamma}_w/3k_i$; $B=\eta_o^3/\lambda^4\ \tau_w{}^3\dot{\gamma}_w$

EXPERIMENT

The packed bed used in this study consisted of a 59.5 cm. long glass tube of 5.08 cm. internal diameter. Glass spheres of diameters 1.09 mm. and 5.03 mm. were used as packing materials. The bed particles were contained between wire screens placed at both ends of the cylinder cross section. The bed porosity was calculated from the weight and density of spheres and bed dimensions. The porosities for the 1.09 mm. and 5.03 mm. sphere beds are 0.39 and 4.0, respectively. The pressure tap connections were situated about 13 cm. from each end of the packed column, in order to provide a fully developed pressure drop measurement without the need for an end effect correction. The test fluid was prepared in a conical mixing tank fitted with a three-blade impeller to ensure continuous mixing of the solutions. The test fluid was fed into a head tank located directly above the packed column. The operating pressure was maintained by regulating, using a regulator connected to a standard test gauge, the compressed air admitted to the liquid surface in the head tank. The pressure drops were separated by a distance of 32.3 cm. Pressure drops were measured either with a differential pressure gauge or manometer. The manometer fluid has a specific gravity of 1.58. The volumetric flow rate through the bed was determined by collection and weighing.

An inelastic Newtonian corn syrup, MCY 41N (Fielders Starches, Australia) and a purely viscous inelastic fluid, 0.5 percent aqueous Methocel HG 90 (Dow Chemical Pty. Ltd) were used to calibrate the packed bed, to establish the accuracy of the experimental techniques and to indicate the general trend in flow behavior observed with these types of rheological fluids.

Two types of viscoelastic fluids were employed in the study. One was aqueous Separan AP 30 solution (Dow Chemicals) which exhibited a significant shear thinning as well as elastic characteristics. The other was Separan AP 30 dissolved in a small quantity of water and mixed with a large amount of glucose syrup (Boger fluid) in order to achieve elastic but constant viscosity fluid.

The test fluids were both characterized immediately before and after being used in the packed bed experiment using a cone and plate Weissenberg Rheogoniometer (WRG, Sangamo Controls, Ltd. England). No significant degradation was observed during the course of the experiment. Shear stress and first normal stress difference were measured as a function of shear rate. Characterization was carried out in a temperature controlled room at 22°C. The temperature in the cone and plate was adjusted to the temperature of each run in the packed bed using a cooling chamber and a cooling system provided with the equipment. The temperature was measured using a chromel alumel thermocouple.

RESULTS AND DISCUSSION

Table 2 shows the composition of all test fluids used in the study. The viscous properties for all test fluids are presented in Table 3 in the form of the power law parameters n, K, Carreau time constant λ and the zero shear viscosity η_0. The first normal stress difference N_1 is shown as a function of shear rate in Figure 1. All first normal stress difference-shear rate data show a bigger slope compared to the corresponding shear stress-shear rate data. The slope n'' approaches a value of 2 in the very low shear rate region, indicating ideal second order fluid behavior.

The Carreau viscosity equation is not a good rheological model for a nearly constant viscosity because the power N, defined as $(1 - n)/2$, is very close to zero when n is unity. Hence, the shear rate dependence in the Carreau viscosity equation becomes rather insensitive.

The Carreau viscosity equation was used to represent the viscous behavior of the 0.85 percent and 1.62 percent aqueous Separan AP 30 solutions (R8 and R9, respectively) which exhibited a high shear thinning characteristic. The zero shear viscosity η_0 was found by extrapolation of the viscosity data to $\dot{\gamma} = 0$. The power law index n was determined from the slope of the shear thinning region. The characteristic time constant λ was calculated from regression analysis. The viscosity versus shear rate plot for 0.85 percent aqueous Separan AP 30 solution is shown in Figure 2. The viscosity data indicate that the lowest data point is still in the shear thinning region. An extrapola-

Table 2. Compositions of Test Fluids

Code	Test Fluid Composition
R1	Corn Syrup (MCY41N)
R2	0.644% Carboxymethyl Cellulose and 10.15% Water in Corn Syrup MCY41N
R3	0.31%Separan AP-30 and 7.48% Water in Corn Syrup MCY41N
R4	0.037% Separan AP-30 and 7.12% Water in Corn Syrup MCY41N
R5	0.028% Separan AP-30 and 3.57% Water in Sweetol 41N
R6	0.036% Separan AP-30 and 4.32% Water in Sweetol 41N
R7	0.052% Separan AP-30 and 6.59% Water in Sweetol 41N
R8	0.85% Separan AP-30 and 99.15% Water
R9	1.62% Separan AP-30 and 98.38% Water
R10	0.75% Separan AP-30,0.75% CMC and 98.50% Water
R11	0.5% Methocel HG-90 and 99.5% Water

Table 3. Rheological Parameters of Test Fluids

Code	n	K [$Ns^n\ m^{-2}$]	n_o [$Ns\ m^{-2}$]	λ [s]	Shear Rate range [s^{-1}]
R1	1.00	11.0	11.49	–	0.18 – 71.1
R2	0.74	17.0	20.6	–	2.15 – 34.1
R3	0.77	13.0	18.0	3.37	1.08 – 17.0
R4	0.98	4.8	4.4	1.00	0.43 – 43.0
R5	0.96	3.8	3.85	0.69	0.34 – 54.0
R6	0.95	3.2	3.0	0.16	0.86 – 86.0
R7	0.91	2.5	2.5	0.91	0.34 – 27.1
R8	0.41	1.9	7.60 (8.20)*	10.1 (11.0)*	1.4 – 68.2
R9	0.42	6.0	26.5	14.50	0.043 – 271
R10	0.61	1.11	4.0	7.52	0.034 – 54.1
R11	0.73	0.008	0.28	–	1.71 – 43.0

* Refitting of Carreau model with one less data point and using λ= 11.0 s as obtained by Chhabra (1980)

tion to zero shear rate to yield a value of η_o is not valid for this case. Chhabra (1980) has used the same fluid composition in his falling sphere experiments and has quoted a value of η_o of 8.2 Pa-s for this fluid. This value of η_o is used in the Carreau model fitting in Figure 2. The fitting in Figure 2 is not as good as that obtained for 1.62 percent aqueous Separan AP 30 solution and solution of 0.75 percent Separan AP 30, 0.75 percent CMC in water (R10). It could be observed that an improvement can be obtained by adjusting the value of η_o. A value of η_o of 7.6 Pa-s was also used for the same set of data.

The viscometric and packed bed data obtained for the Newtonian corn syrup are presented in Figure 3. It is evident that the flow curve obtained is parallel to that for the cone and plate viscometer separated by a geometric shift factor $k_i/2$, in accordance with Eq. (3). The shift factor is found experimentally to be 2.6. This value of $k_i/2$ is within the range of values reported in the literature. Similar results are obtained for the inelastic power law fluid R2.

In the case of viscoelastic fluids, the flow curve obtained in a packed bed is found to be no longer parallel to that obtained from the viscometric flow curve as predicted by Eq. (3). An increase in pressure drop over and above the inelastic prediction was observed beyond certain critical point in the flow curve (Tiu et al., 1984; Marshal and Metzner, 1967). Since the shift factor is a geometric parameter, shown to be independent of fluid properties, it should also be used to describe the flow of viscoelastic fluids through the same packed bed. Figure 4 shows a similar plot for the 0.85 percent aqueous Separan AP 30 solution. The viscometric flow curve, which has been converted to τ_w versus 8V/D, appears on the right hand side of the plot. If the fluid were represented by a power law model using the rheogoniometer data in the high shear region, the expected flow curve for the packed bed would be parallel to the viscometric flow curve in the high shear region in accordance with Eq. (3). Since the power law model is invalid in the low shear rate region, the Carreau viscosity model is a better rheological equation to represent the viscous behavior of the 0.85 percent Separan AP 30 solution over the range of shear rates encountered in the packed bed experiments. The integration of Eq. (1) using the Carreau equation would yield different flow curves for the packed bed depending upon the values of the material constant η_o, λ and n in the model. A slight adjustment of the slope in the power law region would easily alter the values of η_o and λ. Two sets of material constant were obtained for the 0.85 percent Separan AP 30 solution (set A: η_o = 7.6 Pa-s, λ = 10.1 s, and n = 0.41; set B: η_o = 8.2 Pa-s, λ = 11.0 s, and n = 0.49). Both sets of results were determined by regression analysis with slightly different weighing assigned to the low shear rate data. It can be seen from Figure 4 that the two sets of material constants yield two flow curves of different slopes. The result obtained with set A is almost identical to the power law prediction using the high shear rate data. The most sensitive parameter appears to be η_o. This is not unreasonable since the local shear rate varies significantly inside the bed as the fluid traverses the tortuous paths. The use of an average apparent shear rate may be an oversimplification in describing the kinematics of the flow inside the bed. Set B, which has a higher value of η_o, predicts a flow curve with a larger slope than either that predicted by set A or by the power law parameters alone. Results obtained for 1.62 percent aqueous Separan AP 30 solution show similar agreement. The trend is in quantitative agreement with the observed excess pressure drop. It is expected that a slight increase in the value of η_o will shift the predicted flow curve towards the experimental packed bed as in Figure 4. In the limit of zero shear rate, most polymer solutions exhibit a constant viscosity η_o. Using this parameter alone would predict a packed bed flow curve identical to the Newtonian fluid with a slope of unity. This represents the upper bound of the viscous behavior manifested by a viscoelastic fluid in a packed bed. The difference between the experimental observation and the viscous upper bound could be attributed to the elastic effect, which may be correlated with an elastic parameter such as the Deborah number.

Marshall and Metzner (1967) first presented the effect of elasticity in the form of the product of friction factor and Reynolds number against Deborah number. There are many forms of friction factor and Reynolds number for packed beds defined in the literature. The expressions

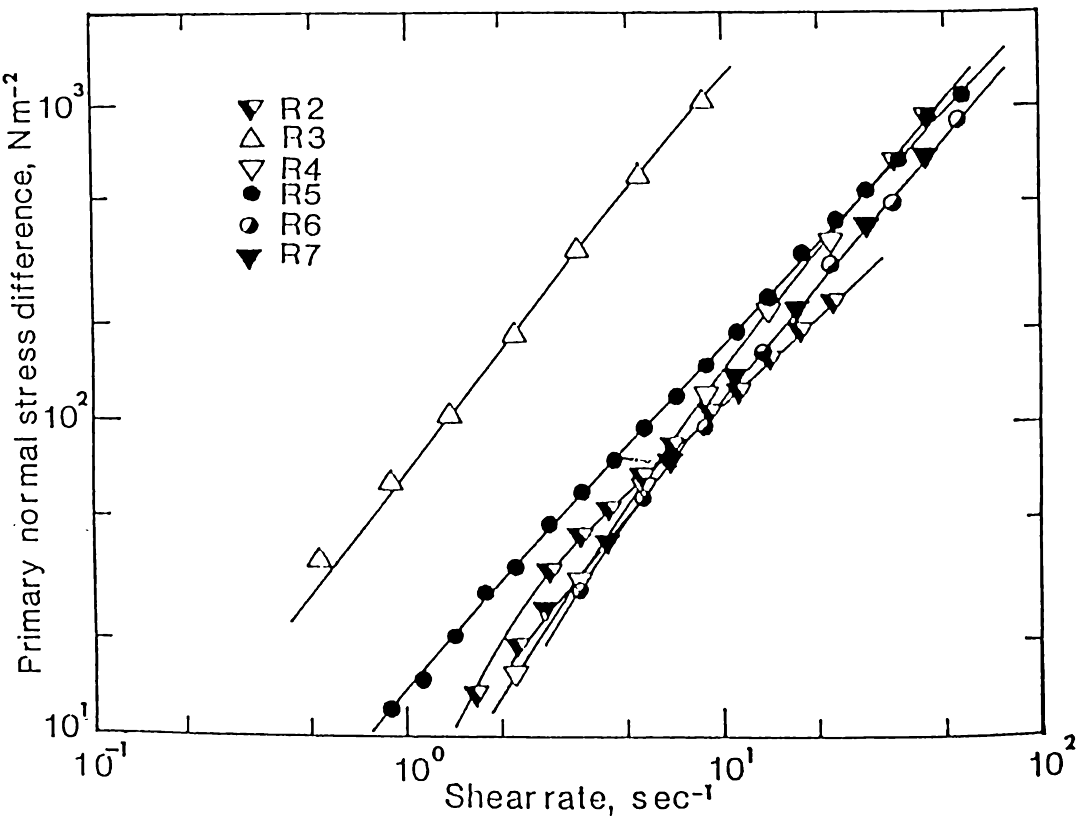

Figure 1. Primary normal stress difference – shear rate data for viscoelastic fluids R2 – R7 at 22°C

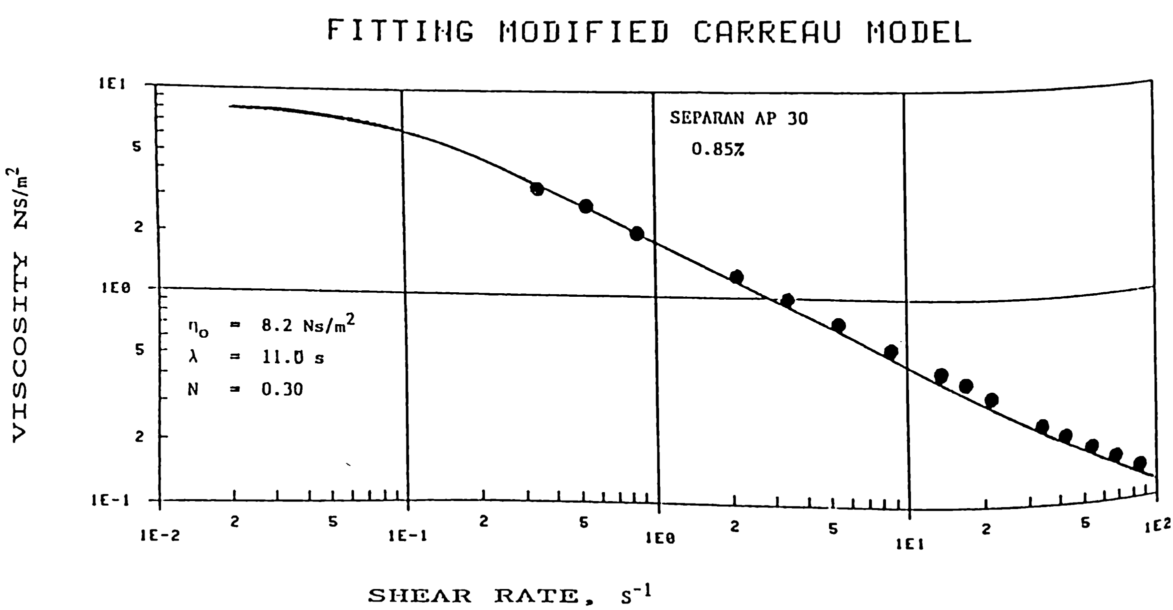

Figure 2. The viscosity data as fitted with the modified Carreau model ($\eta_\infty = 0$) for the 0.85% Separan AP 30 (R8)

for f and Re used here are analogous to laminar pipe flow for power law fluids (Kozicki et al., 1967)

$$f = 16/Re \tag{5}$$

where

$$f = 2\,\tau_w/\rho V^2 \tag{6}$$

and

$$Re = 2^{3-n}\, r_H{}^n\, V^{2-n}\, \rho/(k_i/2)^n \left(\frac{3n+1}{4n}\right)^n K \tag{7}$$

The definition of Re differs only from the pipe flow by a factor $(k_i/2)^n$. Eqs. (5-7) reduce to the Blake-Kozeny for Newtonian flow through packed bed when n = 1.0.

Hence, the experimental results are first presented in a friction factor-Reynolds number plot as shown in Figure 5. Notice that except for R1 which is a Newtonian fluid and R2 which is a very weak viscoelastic fluid, all other fluids show a significantly higher friction factor compared to the inelastic prediction, according to Eq. (5). Although a wide range of Reynolds numbers was covered in the experiment, all data are well within the laminar flow regime.

Marshall and Metzner (1967) defined their Deborah number as the ratio of the duration of fluid memory (relaxation time) θ_f to duration of deformation (processing time) θ_p which maybe used to describe the transition from the viscous to viscoelastic flow. For a porous medium, the Deborah number is

$$De = \theta_f/\theta_p = \theta_f V/d \tag{8}$$

The fluid relaxation time was calculated from the Maxwell fluid model as

$$\theta_f = N_1/2T\dot{\gamma} = N_1/N\dot{\gamma}^2 \tag{9}$$

The fluid processing time was assumed to be inversely proportional to the apparent shear rate in the packed bed, V/d. In the present study, the apparent shear rate is defined in terms of the hydraulic radius as $2V/r_H$. Hence, a modified Deborah number defined as

$$De = V\theta_f/4r_H \tag{10}$$

is used in the correlation of the viscoelastic data. Figure 6 shows the plots of the difference between the measured f-Re and the inelastic prediction which is 16 as a function of De_m for the five viscoelastic fluids(R3-R7). Contrary to the results by Marshall and Metzner, there appears to be no unique relationship existing between f-Re and De or De_m. Hong (1981) obtained similar results using Deborah and Ellis numbers as the elastic parameters in the correlation.

The Deborah number defined either by Eq. (8) or Eq. (10) suffers two disadvantages. Firstly, very few viscoelastic fluids can be described by the idealized Maxwell fluid model. The relaxation time calculated based solely on the steady shear and normal stress data may not represent the true material time constant. Other time parameters based on molecular theory and the various inelastic models such as the Ellis fluid model have been used in literature. Secondly, Eq. (9) predicts a decrease in the relaxation time when the shear rate is increased, whereas the Maxwell model has a constant relaxation time. Since none of the test fluids exhibit a truly constant viscosity, it is not surprising that neither definitions of Deborah number as given by Eq. (8) and Eq. (10) would give a meaningful correlation. As Wang et al. (1979) have indicated, the failure of the Deborah number to correlate the excess flow resistance in porous media may be the result of the oversimplification of the Maxwell model and the vast difference in the nature of the flows between porous media and the steady relaxation time experiment.

An alternative definition of Deborah number is proposed here. Instead of using the apparent viscosity in the calculation of θ_f as indicated by Eq. (9), the zero viscosity is used. Thus,

$$f = N_1/2\eta_o\,\dot{\gamma}^2 \tag{11}$$

and

$$De_o = De_m\,[\eta/\eta_o] \tag{12}$$

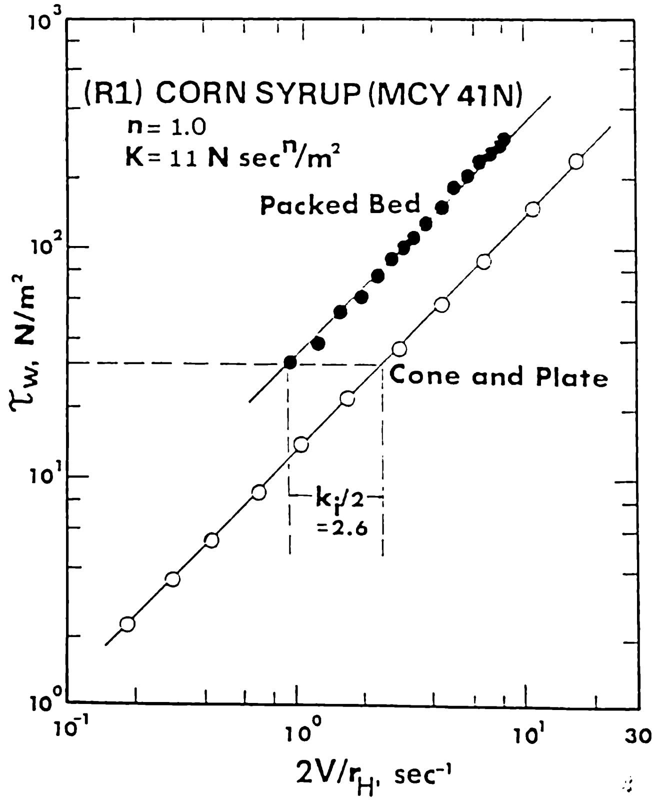

Figure 3. τ_w versus $2V/r_H$ for corn syrup MCY 41 N

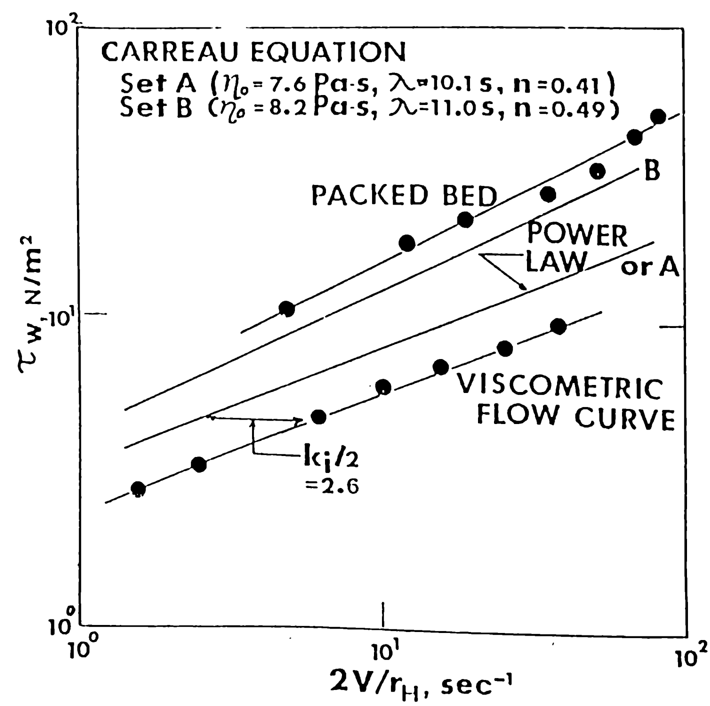

Figure 4. τ_w versus $2V/r_H$ for 0.85% aqueous Separan AP 30 as predicted using the Carreau viscosity equation

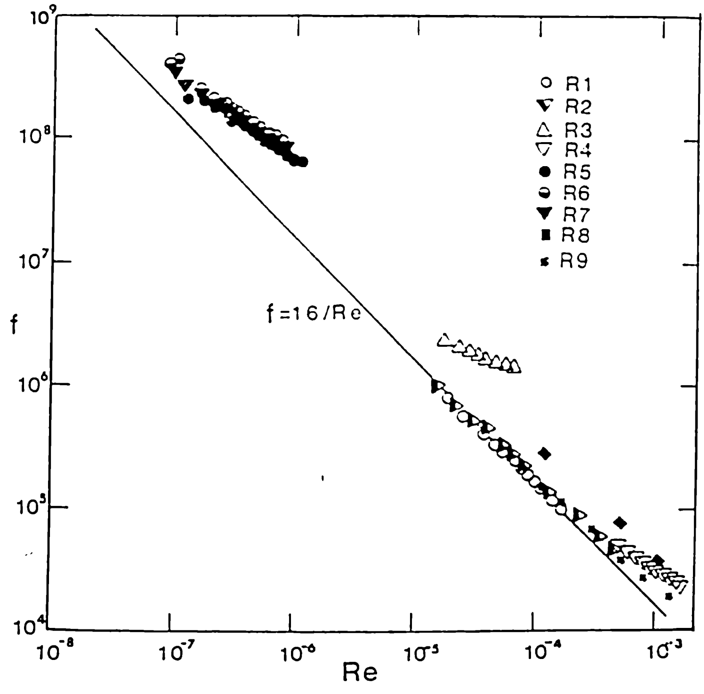

Figure 5. Friction factor versus Reynolds number for all test fluids

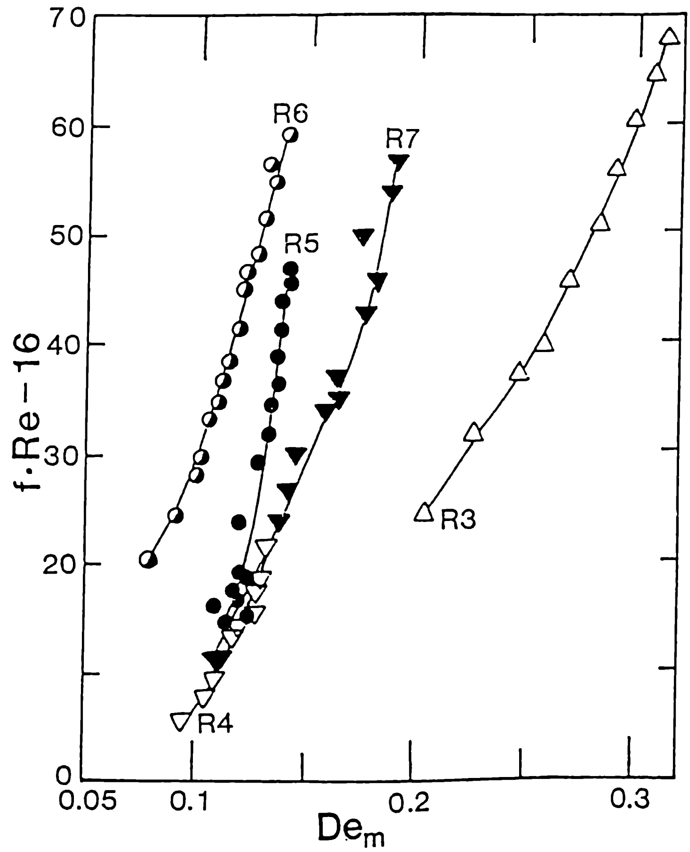

Figure 6. (f. Re −16) versus De_m for five viscoelastic fluids

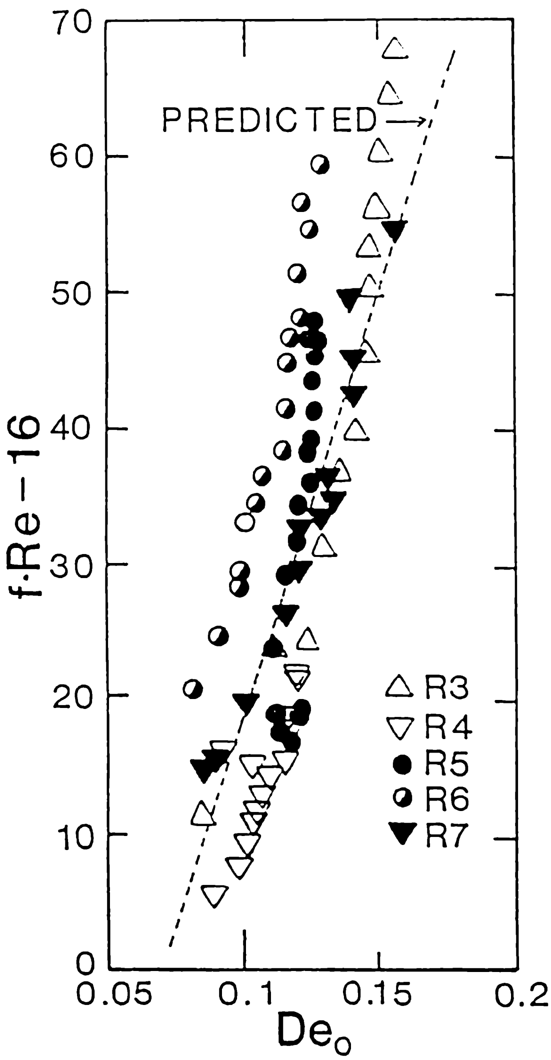

Figure 7. (f. Re − 16) versus De_0 for five viscoelastic fluids

The definition of De_0 eliminates the variation of fluid viscosity due to shear thinning effects. It is expected to bring the data for fluids with lower values of n closer to those exhibiting nearly constant viscosity. Figure 7 shows the replotting of data from Figure 6 with the abcissa changed to De_0. A significant improvement in the correlation is realized. The broken line in the figure is obtained by least square fitting of all data. However, in view of the data scattering normally associated with packed bed experiments, the accuracy of the correlation as shown in Figure 7 should not be overemphasized. Nonetheless, Eq. (12) suggests a possible definition of Deborah number for viscoelastic fluids which, though empirical in nature, could be used to study the viscoelastic effects encountered in a non-viscometric flow field.

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