η = dimensionless radial position = $y^+/Re^* = y/R$
θ = angle in cylindrical coordinates
σ = constant

LITERATURE CITED

Bennett, C. O., and J. E. Meyers, *Momentum, Heat, and Mass Transfer*, McGraw Hill (1967).
Bogue, D. C., and A. B. Metzner, "Velocity Profiles in Turbulent Pipe Flow," *Ind. Eng. Chem. Fundam.*, **2**, 143 (1963).
Hinze, J. O., *Turbulence*, McGraw-Hill, New York, (1975).
Laufer, J., "The Structure of Turbulence in Fully Developed Pipe Flow," NACA TN 2954 (June, 1953).
Nikuradse, J., "Gesetymassigkeiten der turbulenten Stromung in glotten Rohren," Forschungslaft No. 356, V.D.I. Verlag, Berline, (1932).
Ross, D., "A New Analysis of Nikuradse's Experiments on Turbulent Flow in Smooth Pipes," p. 651, Proc. Third Midwestern Conference on Fluid Mechanics, Univ. of Minnesota (1953).
Schlichting, H., *Boundary-Layer Theory*, McGraw Hill, New York (1968).

Manuscript received August 2, 1979; revision received and accepted August 17, 1979.

Non-Newtonian Viscous Properties of Methacoal Suspensions

RON DARBY
and
BETH ANN ROGERS

Chemical Engineering Department
Texas A&M University
College Station, Texas 77843

Rapidly expanding interest in transportation of coal by water slurry pipeline has brought into focus technical problems relating to the flow and stability properties of suspensions, liquid-solid separation, slurry preparation, etc. There are economical, political and environmental problems associated with acquiring and disposing of large volumes of water as well. One alternative to water slurries was proposed by L. J. Keller (1977). In a patented process called Methacoal, methanol is used to produce a relatively stable, fluid, highly shear thinning suspension of coal or lignite solids. Potential advantages of Methacoal over water slurries include superior flow and stability, easier liquid-solid separation, and the absence of a water requirement if the methanol is assumed to be recycled. Several economic analyses of the Methacoal process have been made (Blaustein 1976, Gambill 1975, Jimeson 1976, Jimeson 1977, Bank 1977, and Vanston et al. 1978), and all but one were generally favorable.

Since there are no data on the properties of Methacoal, we undertook an evaluation of the rheological properties of such suspensions. It is well known that the viscosity of concentrated suspensions depends upon solids fraction, particle size and size distribution, and shear rate, and the influence of these variables upon the viscosity of Methacoal suspensions was studied. In addition, we examined the effect of initial solids moisture content.

Approximately 60 Methacoal samples were made, using Texas lignite and reagent grade methanol, in a process modeled that described in Keller's patent. The details are given by Darby (1978). Conditions were varied to produce a range of solids concentration and particle size distribution, from solids dried initially to nominally 0, 5, 10 and 15% moisture. The resulting suspensions were relatively stable, homogeneous, and fluid.

Viscosities of the suspensions were measured as a function of shear rate in a specially constructed concentric cylinder (Couette) attachment on a Weissenberg Rheogoniometer, over a range of about two decades in shear rate, at room temperature (23 ± 1°C). Data were analyzed by standard methods (e.g. Darby 1976), and an example of the data is shown in Figure 1. The data points were obtained in a repetitive cycle, initially increasing the shear rate (round symbols), then decreasing it, (triangles) and finally increasing it again (squares). Any spread of data between cycles generally indicated some degree of settling during the run, in which case the first series of points (higher viscosities) were taken to represent that sample. Particle sizes were nominally less than about 300-400μm, with the majority less than 100μm. Particle size distributions were measured by a sedimentation method. More extensive data are given by Darby (1978), and a complete tabulation of all data is given by Rogers (in prep.).

The apparent viscosity of the suspension, relative to that of the suspending fluid, is assumed to be a function of the solids volume fraction (C_v), particle size distribution (PSD), shear ($\dot{\gamma}$), and lignite moisture content (M)

$$\eta_r = \eta/\eta_0 = \eta_r(C_v, \dot{\gamma}, PSD, M) \tag{1}$$

Although particle-shape would be an additional parameter, magnified photomicrographs of the suspension particles showed them to be roughly spherical, with no unique features.

The non-Newtonian character of the suspensions may be represented by any of several empirical models which are frequently used to describe the viscosity of suspensions. Four such models are:

Power Law:
$$\eta_r = m\dot{\gamma}^{n-1} \tag{2}$$

Bingham Plastic:
$$\eta_r = (\tau_b/\dot{\gamma}) + \mu_b \tag{3}$$

Casson:
$$\eta_r = [\sqrt{\tau_c/\dot{\gamma}} + \sqrt{\mu_c}]^2 \tag{4}$$

Herschel-Bulkley:
$$\eta_r = (\tau_1/\dot{\gamma}) + m_1\,\dot{\gamma}^{n_1-1} \tag{5}$$

The last three exhibit a yield stress, which is consistent with qualitative observations of the suspensions. Furthermore, the Bingham and Casson models exhibit a high shear limiting viscosity, which is physically realistic, whereas the other two do not. Thus, on physical grounds, the Bingham and Casson models should be preferred, if the data must be extrapolated.

The apparent viscosity data were all fit by each of these four models by least squares regression methods. The Power Law model gave the best fit most often, although the other models also fit the data quite well, and in some cases better than the Power Law. The solid line in Figure 1 represents the Power Law fit, and dashed line is the Bingham best fit. The other two models fell between these two in every case. The influence of solids fraction and particle size distribution would be reflected by a dependence of the model parameters upon these variables.

A series of runs were made in which only the solids volume fraction was varied, over range of 0.32 to 0.52. A wide variety of expressions have been proposed in the literature for relating suspension viscosity to solids volume fraction (e.g. Jinescu 1974, Rutgers 1962), although these generally assume Newtonian behavior. Attempts to fit a number of these expressions to the data met with very little success. We found, however, that the relative suspension viscosity at a given shear rate and particle size distribution could be represented quite well by a simple exponential function of C_v, with an exponential constant of 18.3.

0001—1541—80—3244—0310—$00.75.

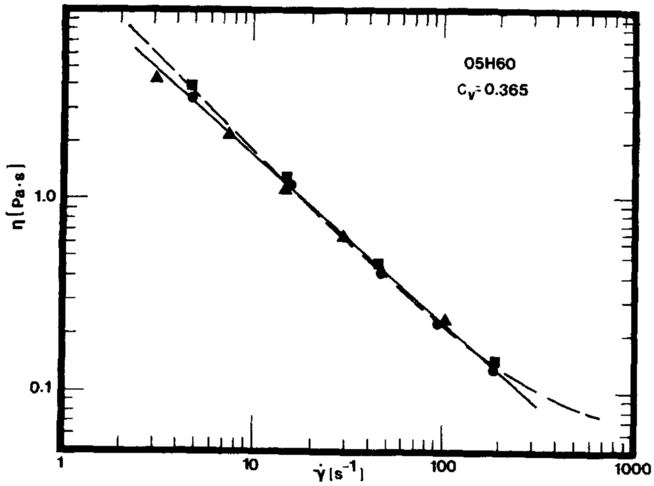

Figure 1. Example of apparent viscosity versus shear rate data.

TABLE 1. MODEL CORRELATION PARAMETERS

		5%M	15%M
Power Law			
$m = M \exp(18.3\, C_v + p_m\, \phi_{30})$	M	0.0515	0.184
$n = N \exp(p_n\, \phi_{30})$	p_m	15.2	12.9
Bingham	N	2.56	0.96
$\tau_b = Y_0 \exp(18.3\, C_v + b_0\, \phi_{30})$	Y_b	0.214	0.473
$\mu_b = V_b \exp(18.3\, C_r + b_1\, \phi_{30})$	b_0	12.4	11.9
	V_b	0.226	0.627
Casson	b_1	−2.44	3.18
$\tau_c = Y_c \exp(18.3\, C_v + c_0\, \phi_{30})$	Y_c	0.0603	0.181
$\mu_c = V_c \exp(18.3\, C_r + c_1\, \phi_{30})$	c_0	15.2	13.5
	V_c	0.0800	0.0703
	c_1	−8.28	−0.015

Thus, the concentration dependence of each of the model parameters, P_k, (except the Power Law flow index, which was independent of C_r) could be expressed by the function

$$P_k = P_{ko} \exp(18.3\, C_r) \tag{6}$$

This is equivalent to factoring the relative apparent viscosity function into a function of solids fraction and a shear rate dependent function

$$\eta_r(C_r, \dot{\gamma}) = \eta_r(\dot{\gamma})\exp(18.3\, C_r) \tag{7}$$

where the parameters of the function $\eta_r(\dot{\gamma})$ (e.g. the P_{ko}) remain dependent upon particle size distribution, however.

Attempts at correlating the model parameters with mean particle size or standard deviation of the distribution were generally unsuccessful. In general, solids less than 10-50μm in diameter usually form stable homogeneous (non-settling) suspensions, whereas larger particles tend to form unstable, inhomogeneous (settling) suspensions (e.g. Stepanoff 1969, Wasp et. al. 1977). Further, when a distribution of particle size is present, a minimum viscosity and minimum pressure drop in pipe flow occurs when there is a significant fraction of fines (less than 10-15μm) (e.g. Eveson 1959, Elliot and Gliddon 1970). Also, the maximum solids loading occurs for a bimodal distribution when the size ratio is about 10:1. Hence it should be possible to characterize the effect of size distribution simply in terms of the fraction of fines in suspension. Here, this was taken to be the fraction less than 30μm in diameter (ϕ_{30}), since this represents approximately one tenth the size of the largest particles, and could be accurately determined from the particle size distribution measurements.

A correlation of the Power Law parameters with ϕ_{30} is shown in Figure 2. The Bingham and Casson yield parameter correla-

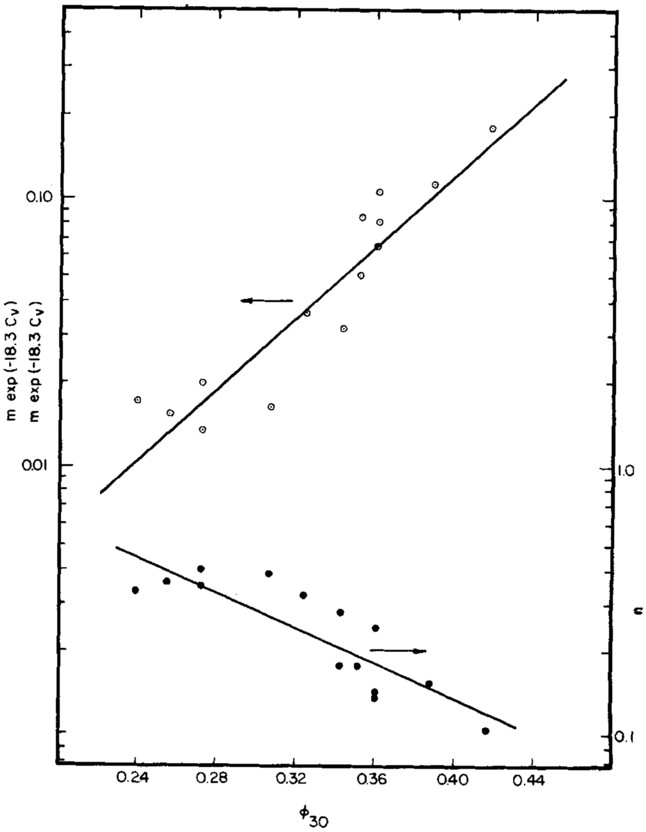

Figure 2. Reduced Power Law model parameters as a function of the fraction of solids less than 30μm, 5% lignite moisture.

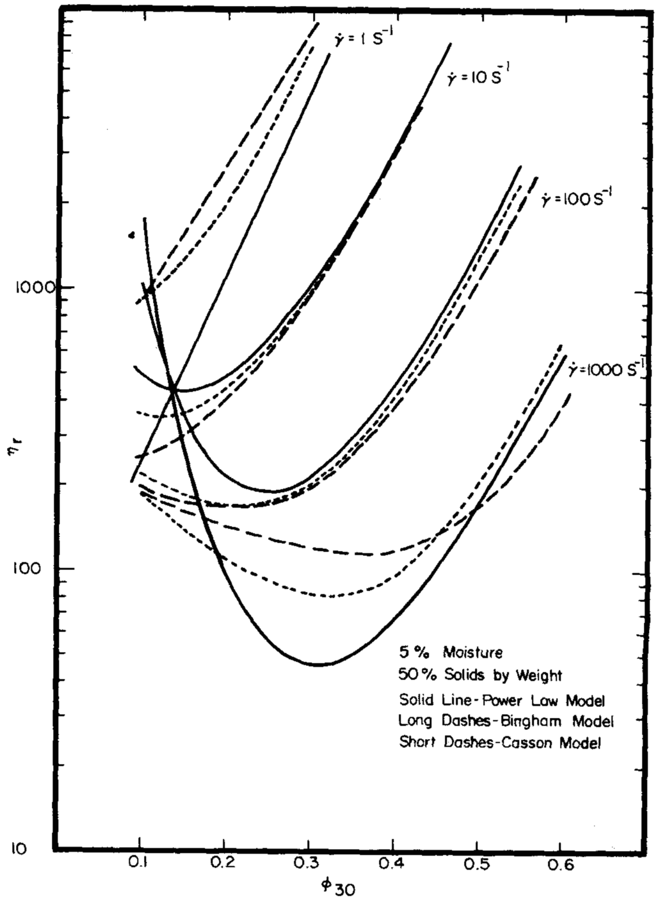

Figure 3. Suspension relative viscosity for 50% by weight solids, 5% initial lignite moisture.

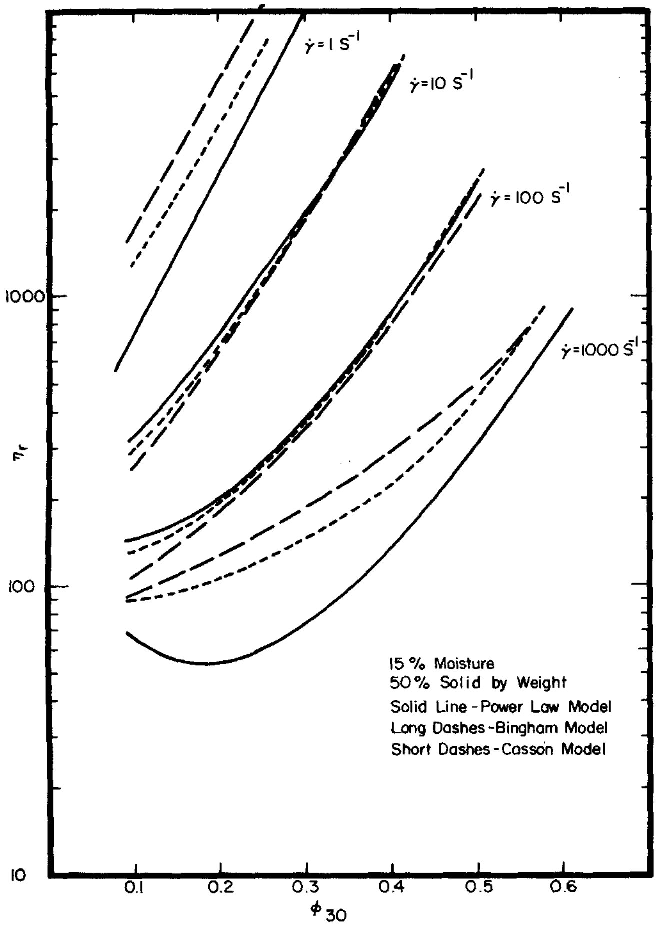

Figure 4. Suspension relative viscosity for 50% by weight solids, 15% initial lignite moisture.

tions were similar to that for m and the limiting viscosity parameter correlations similar to that for n. The Herschel-Bulkley parameters did not correlate nearly as well as these, however. A least squares regression analysis indicated that the most significant fit of these correlations is again a simple exponential function of ϕ_{30}. The final combined correlation equations, along with values of the correlating parameters for the 5% and 15% moisture lignite, are shown in Table 1. The values in this table show that the influence of the amount of fines (ϕ_{30}) upon the viscosity parameters is significantly greater for the suspensions made with the drier lignite.

The final correlations were used to calculate the relative suspension viscosities at a fixed solids fraction (corresponding to a weight fraction of 50%) over an extended range of ϕ_{30} and shear rate values, for the Power Law, Bingham, and Casson models. The results are shown in Figure 3 for the 5% moisture lignite, and in Figure 4 for 15% moisture. The values predicted from the correlation equations and the measurements are in good agreement over the range of measurements ($2 < \dot{\gamma} < 200 s^{-1}$, $0.24 < \phi_{30} < 0.42$) but tend to diverge outside of this range due to the differences in the limiting behavior of the models. Figure 3 indicates a minimum viscosity at a particular value of ϕ_{30}, which depends upon shear rate. Figure 4 for the 15% moisture sample shows a tendency toward this type of behavior as well, although it is much less pronounced.

All of the suspensions were "stable," in that they did not hard pack on standing for extensive periods. However, for the coarser suspensions, varying degrees of settling were observed in visometer. The majority of the suspensions remained quite fluid after extended periods, however, even with some settling. This enhanced stability is attributed to the high degree of adsorption of methanol on the lignite (Lowry 1963), which is inversely related to the moisture content of the lignite. This was evidenced by a significant heat evolution when dried lignite was mixed with methanol, the amount of which varied inversely with the lignite moisture content. Adsorption of methanol on the large surface area of the fine particles would tend to reduce the effective density of these particles as well as modify the degree of solid-solid contact. Therefore, the stability and fluidity of the suspensions is increased. However, too many fines would tie up a significant fraction of the liquid by adsorption, resulting in a thick, pasty suspension. These conclusions are consistent with observations and the resulting correlations.

ACKNOWLEDGMENT

Financial support for this study was provided by the Texas A&M Engineering Experiment Station, the Texas A&M Center for Energy and Mineral Resources, and the U.S. Department of Energy.

LITERATURE CITED

Banks, W. F. and J. F. Horton, "Efficiency Improvements in Pipeline Transportation Systems," Technical Report, Contract EY-76-C-03-1171, U.S. DOE, (September, 1977).

Blaustein, B., "Comparative Analysis of Transporting Coal: Coal-Water Slurry vs Coal-Methanol Fuel Suspension," Pittsburgh Energy Center, U.S. DOE, (January, 1976).

Darby, R., *Viscoelastic Fluids*, p. 302, Marcel Dekker, Incorporated, New York, (1976).

Darby, R., "Viscous Rheological Properties of Methacoal Suspensions from Texas Lignite," Final Report to DOE for ERDA Grant No. EF-77-G-01-2725, (September, 1978).

Elliot, D. E. and B. J. Gliddon, "Hydraulic Transport of Coal at High Concentrations," paper G.2 in Hydrotransport I, First International Conference on the Hydraulic Transport of Solids in Pipes, BHRA Bedford, England, (September, 1970).

Eveson, G. F., in C. C. Mill (ed), *Rheology of Disperse Systems*, 61, Pergammon Press, London, (1959).

Gambill, W. R., "The Methacoal Concept-A-Preliminary Assessment," HNL, Oak Ridge National Laboratories, (March, 1975).

Jimeson, R. M., "Methacoal Enhances the Transportation and Use of Coal Resources, Paper presented at IIASA Conference on Coal Resources, Meshduretchensk, U.S.S.R., (Nov. 8-Dec. 2, 1977).

Jimeson, R. M., "Methacoal: Synthetic Energy Symbiotic with Water Deficient Coal Resource Regions," paper presented at AIChE Third National Conference on Water Reuse, Cincinnati, (June 27-30, 1976).

Jinescu, V. V., "The Rheology of Suspensions," *Int. Chem. Eng.*, **14**, 397 (1974).

Keller, L. J, "Fuel Composition and Method of Manufacture," U. S. Patent No. 4 045 092, (August 30, 1977).

Lowry, H. H. (ed.), *Chemistry of Coal Utilization*, Suppl. vol., John Wiley & Sons, New York (1963).

Rogers, B. A., "Viscous Rheological Properties of Methacoal Suspensions," M. S. Thesis, Texas A&M University (in preparation).

Rutgers, I. R., "Relative Viscosity and Concentration," *Rheol. Acta*, **2**, 305 (1962).

Rutgers, I. R., "Relative Viscosity of Suspensions of Rigid Spheres in Newtonian Fluids," *Rheol. Acta*, **2**, 202 (1962).

Stepanoff, A. J., *Gravity Flow of Bulk Solids and Transport of Solids in Suspension*, John Wiley & Sons, (1969).

Vanston, J. H. Jr., D. G. Hannan, J. W. Barnes, I. Bilsky, H. B. H. Cooper, Jr., M. E. Leesley, T. D. Lies, D. M. White, C. D. Zinn, "Systems Study and Assessment of Methacoal as a Coal Transportation Alternative," Research Report No. 11. Center for Energy Studies, University of Texas at Austin (March, 1978).

Wasp, E. J., J. P. Kenny, and R. L. Gandhi, *Solid-Liquid Flow—Slurry Pipeline Transportation*, Trans. Tech. Publications, Switzerland, (1977).

Manuscript received May 18, 1979; revision received August 9, and accepted August 20, 1979.