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THE RHEOLOGICAL BEHAVIOR OF THORIUM OXIDE SLURRIES IN LAMINAR FLOW

ABSTRACT

The flow behavior of aqueous thorium oxide slurries is an integral part of the information needed in designing the blanket system of a thorium oxide slurry breeder reactor. As a result of an investigation designed to determine these rheological properties, it was found that thorium oxide slurries are plastic non-Newtonian fluids at ambient temperatures. It was found that the viscosity can be expressed in terms of shear stress, τ , and concentration, S , by the equation

$$\mu_S = \mu_L \left[\frac{1.2 + S(c\tau^{-0.2} + 1)^3}{1.2 - 2S(c\tau^{-0.2} + 1)^3} \right]$$

The laminar flow of thorium oxide slurries in circular conduits is described by the equation

$$\frac{8V}{g_c D} = \frac{4}{\mu_L \tau_D^3} \int_{\tau_y}^{\tau_D} \tau^3 \left[\frac{1.2 - 2S(c\tau^{-0.2} + 1)^3}{1.2 + S(c\tau^{-0.2} + 1)^3} \right] d\tau,$$

and the yield stress is related to concentration by the equation

$$\tau_y = \left[\frac{c}{(1.2/2S)^{1/3} - 1} \right]^5$$

The constant c in the above expressions is unique for a given slurry (independent of concentration) and may be determined from any one viscosity, flow rate, or yield stress of that slurry.

INTRODUCTION

Fluids are classified rheologically as Newtonian fluids or non-Newtonian fluids. Newtonian fluids in laminar motion are characterized by the velocity gradient, dv/dr , being directly proportional to the shear stress acting on the fluid, τ , or

$$-\frac{1}{g_c} \left(\frac{dv}{dr} \right) = \frac{\tau}{\mu} \quad (1)$$

Equation (1) is known as Newton's law and μ is termed the coefficient of viscosity. A plot of the velocity gradient versus shear stress is known as a shear diagram. Fig. 1a illustrates a shear diagram of a Newtonian fluid; the reciprocal of the slope of this curve represents the viscosity.

Non-Newtonian fluids are classified and compared by means of shear diagrams. Shear diagrams of several types of non-Newtonians are illustrated in Figs. 1b-1e. Fig. 1b illustrates a general shear diagram for the plastic class of non-Newtonians. Using the definition of viscosity expressed in Equation (1), it may be seen that the viscosity of plastic fluids remains infinitely large until a definite value of shear stress is reached. Further increase in shear stress is then accompanied by a decrease in the viscosity. The shear stress at the point where the viscosity becomes finite is designated as the yield stress, τ_y . Bingham plastics, illustrated in Fig. 1c, may be considered special cases of the plastic class in which the shear diagram is a straight line above the yield stress. The reciprocal of the slope of this line is termed the coefficient of rigidity, η , and has the units of viscosity. The fluid illustrated in Fig. 1d represents the pseudoplastic class of non-Newtonians in which the viscosity is a finite value, μ_0 , at rest and decreases

Fig. 1a Newtonian Fluid

Fig. 1b Plastic Fluid

Fig. 1c Bingham Plastic Fluid

Fig. 1d Psuedo Plastic Fluid

Fig. 1e Dilatant Fluid

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as the shear stress is increased. Fig. 1e represents the dilatant class which differs from the pseudoplastic class only in that the viscosity increases as the shear stress increases. For a more extensive discussion of non-Newtonian classes and shear diagrams see ORNL CF 55-4-182.⁶

A particular application of the viscosity concept is the prediction of pressure drop caused by the laminar flow of fluids in lengths of circular conduit. Mooney⁹ has derived a general expression, equation (2), relating the coordinates of shear diagrams with the average velocity in a pipe, V_a , the diameter of the pipe, D , and the pressure drop, ΔP , across a length L of the pipe.

$$-\frac{1}{g_c} \left(\frac{dv}{dr} \right) = \frac{3}{4} \left(\frac{8V_a}{g_c D} \right) + \frac{\tau_D}{4} \left(\frac{d(8V_a/g_c D)}{d\tau_D} \right) \quad (2)$$

where τ_D equals $D \Delta P / 4L$. An integral form of equation (2) is

$$\frac{8V_a}{g_c D} = \frac{4}{g_c \tau_D^3} \int_{\tau_y}^{\tau_w} - \left(\frac{dv}{dr} \right) \tau^2 d\tau \quad (3)$$

When equation (1) is introduced into equation (3), the integration yields

$$\frac{8V_a}{g_c D} = \frac{\tau_D}{\mu} \quad (4)$$

A plot of $8V_a/g_c D$ versus τ_D is termed a pseudo shear diagram.

Pseudo shear diagrams for all types of non-Newtonian fluids may be obtained from shear diagrams by graphical integration of equation (3). In some cases, however, the relationship between shear stress and velocity gradient may be expressed mathematically and equation (3) integrated directly.

For Bingham plastics the true shear diagram is described by equation (5) and the integration of equation (3) yields equation (6).

$$-\frac{1}{g_c} \frac{dV}{dr} = \gamma (\gamma - \gamma_y) \quad (\text{when } \gamma \geq \gamma_y) \quad (5)$$

$$-\frac{1}{g_c} \frac{dV}{dr} = 0 \quad (\text{when } \gamma < \gamma_y)$$

$$\frac{8V}{g_c D} = \frac{1}{\gamma} \left(\gamma_D - \frac{4}{3} \gamma_y + \frac{1}{3} \frac{\gamma_y^4}{\gamma_D^3} \right) \quad (6)$$

Farrow and Lowe⁷ have suggested that some pseudoplastic fluids are described by the relationship of equation (7) and hence the pseudo shear diagrams of these fluids have the form of equation (8), where n is a constant greater than unity and typical of a particular suspension.

$$-\frac{1}{g_c} \left(\frac{dV}{dr} \right) = \frac{\gamma^n}{\mu_0} \quad (7)$$

$$\frac{8V}{g_c D} = \frac{\gamma_D^n}{\mu_0} \left(\frac{4}{3+n} \right) \gamma_D^n \quad (8)$$

Other workers have proposed analytical expressions describing pseudoplastic materials. Powell and Eyring¹¹ have suggested the form of equation (9)

$$-\frac{1}{g_c} \left[\left(\frac{dV}{dr} \right) + \frac{1}{B} \sinh^{-1} \left(\frac{1}{A} \frac{dV}{dr} \right) \right] = \frac{\gamma}{C} \quad (9)$$

where A , B and C are constants of a particular fluid. Christiansen et al⁴ have combined equations (9) and (1) and integrated to obtain the pseudo shear diagrams in terms of the three constants. Their results are presented in graphical form.

Reiner and Rivlin¹² have derived an expression for the form of psuedo shear diagrams for psuedoplastics based on a model suggested by Hatchek.⁸ Their expression is

$$\frac{8V}{g_c D} = \gamma_D / \left[(\mu_0 - \mu_\infty) \left(1 - \frac{\gamma_D^F}{4\mu_0} \right) + \mu_\infty \right], \quad (10)$$

where μ_0 is the rest viscosity, μ_∞ is the viscosity at infinite shear, and F is a constant. Hatchek's model assumed that the particles in a suspension carry with them fluid envelopes, sometimes referred to as lyospheres, which have a maximum volume when the fluid is resting and which decrease toward a minimum in volume with increasing velocity gradient. The further assumptions used in the derivation of equation (10) are that the effective volume fraction of solids decreases with shear exponentially, and that the Einstein's equation relating the viscosity of a Newtonian suspension to the volume fraction of solids applies.

Asbeck et al¹ have recently found that the viscosity of paints at infinite shear could be correlated with concentration by the equation

$$\mu_\infty = \mu_L \log^{-1} \frac{ukS}{1-S} \quad (11)$$

where μ_L is the viscosity of the suspending fluid, S is the volume fraction of pure solids, and u and k are constants for a particular system. The equation was extended to express the viscosity of paint suspensions as a function of concentration and "shearing velocity", E , for "shearing velocities" greater than 300 sec^{-1} . Their final expression is

$$\log \mu = \log \mu_L + \frac{k u S}{u - S} + \frac{\alpha S(u - S)}{\mu_L k S u (\sigma - S) E^{1/2}} \quad (12)$$

where α and σ represent two additional constants of the system.

Efforts have also been made to correlate the yield stresses of non-Newtonians with concentration. Bingham³ reports that the yield stress of a certain clay increased in proportion to the increase in concentration after a minimum value of concentration was reached. Babbitt and Caldwell² found a similar phenomenon in clay, although the increase in yield stress was not strictly linear. Norton, Johnson and Lawrence¹⁰ report that the yield stresses of certain kaolinite slurries increase with the third power of concentration.

Model and Analysis:

The model used in this paper for the analysis of non-Newtonian flow is similar to the one proposed by Hatchek. That is, it is postulated that the dispersed particles in a non-Newtonian suspension are solvated and the amount of liquid carried in the solvated envelope of bound water varies with shear stress. In order to simplify the analysis, it will be further postulated that the solid particles of the suspension are spherical in shape and of uniform radius, r , and that at any given shear stress, the envelope is of uniform thickness, t . If it is postulated that the viscosity of the suspension is a function only of the volume fraction of solids present, as in the case of Newtonian suspensions, only two functions remain unknown in order to complete the analysis; namely, the expression for the viscosity of a suspension in terms of the volume fraction of effective solids and the relationship between shear stress and envelope thickness, t .

A relationship between suspension viscosity and volume fraction of solids has been proposed by Robinson¹³ as

$$\frac{\mu_S - \mu_L}{\mu_L} = \frac{xS}{1 - yS} \quad (13)$$

where x is termed a frictional coefficient and y is the relative sediment volume or volume ratio of sediment to solids. Selecting Einstein's value for x and the porosity of the settled bed as 0.40, as in the case of randomly packed spheres, x becomes 2.5 and y is 1.667, and equation (13) may then be written as

$$\frac{\mu_S - \mu_L}{\mu_L} = \frac{2.5S}{1 - 1.667S} \quad (14)$$

Rearranging equation (14) and substituting the volume fraction of effective solids S_T for S , the viscosity of a suspension of uniform solvated spheres is

$$\mu_S = \mu_L \left[\frac{1.2 + S_T}{1.2 - 2S_T} \right] \quad (15)$$

With the postulate that the dispersed particles are spherical and the envelope thickness uniform, the envelope thickness may be related to particle radius by the volume ratio of liquid in the envelopes to solid particles, ν , as

$$t = r \left[(\nu + 1)^{1/3} - 1 \right] \quad (16)$$

The total volume fraction of effective solids, S_T , is the sum of the volume fractions of pure solid, S , and fraction of liquid immobilized in the envelope or

$$S_T = S(1 + \nu) \quad (17)$$

Combining equations (16) and (17), the volume fraction of effective solids may be written in terms of volume fraction of pure solids and the ratio t/r as

$$S_T = S(t/r + 1)^3 \quad (18)$$

Combining equations (15) and (18) yields an expression for the viscosity of a non-Newtonian fluid in terms of viscosity of the suspending liquid, volume fraction of pure solids, and the shear stress dependent ratio t/r or

$$\mu_S = \mu_L \left[\frac{1.2 + S(t/r + 1)^3}{1.2 - 2S(t/r + 1)^3} \right]. \quad (19)$$

Determination of the Function t/r :

The relationship between shear stress and envelope thickness is not known quantitatively and therefore must be determined experimentally. A suspension of thorium oxide particles in water containing approximately 1500 ppm of sulfate added as thorium sulfate was conditioned by circulation for over 100 hours at 300°C in a pressurized pump-loop system. Shear diagrams for various concentrations of thorium oxide were prepared from data obtained using a pipe line-type viscometer in which a tube 67.5 inches long with 0.125 inches inside diameter was employed as a test section. The ends of the tube were flat and corrections for the kinetic effects were made using the relationship

$$\frac{D \Delta P}{4L} = \left(\frac{D \Delta P}{4L} \right)_{\text{measured}} - \frac{m \rho D}{L} \frac{v_a^2}{4g_c} \quad (20)$$

where m was taken as 1.12 as recommended by Bingham³ for Newtonian fluids.

The data are shown as psuedo-shear diagrams in Fig. 2 where $8V_a/g_c D$, the average rate of shearing strain, is plotted versus τ_D , the shear stress at the wall. True shear diagrams, shown in Fig. 3, were constructed from the psuedo shear diagrams using the Mooney equation (2).

Lines of constant viscosity (a through m) were constructed on the true shear diagrams covering the complete range of viscosity. Values of shear stress

Fig. 2. Psuedo Shear Diagram as a Function of Solidity, S, for Thorium Oxide-Water Slurry at 250°C.

Fig.3. Shear Diagrams as a Function of Solidity, S , for Thorium Oxide-Water Slurry Including Lines of Constant Viscosity "a" Through "m".

were evaluated as function of concentration and viscosity (the intercepts of the constant viscosity lines and shear diagrams). Shown in Table I are values of the volume fraction of pure solids, S , for each slurry and values of volume fraction of total effective solids, S_T , obtained from equation (15) for each viscosity. From equation (17), the ratio of envelope volume to particle volume, ν , was evaluated at each shear stress. These values are tabulated for each concentration and shear stress in Table I. Plots of ν versus γ are shown in Fig. 4. An illustrative calculation of the preceding quantities is given in Appendix A.

To determine the functional relationship between envelope thickness and shear stress, the ratio t/r was plotted versus γ on logarithmic grids (Fig. 5). It may be seen that the values plot as a straight line and the slope has a value of approximately -0.2. Similar plots for other thorium oxide slurries have been made and it was found that the lines were essentially parallel.

With this in mind, it is possible to express the relationship between envelope thickness and shear stress quantitatively as

$$\frac{t}{r} = c\gamma^{-0.2} \quad (21)$$

Combining equations (19) and (21) the final expression for the viscosity of a non-Newtonian slurry is

$$\mu_S = \mu_L \left[\frac{1.2 + S(c\gamma^{-0.2} + 1)^3}{1.2 - 2S(c\gamma^{-0.2} + 1)^3} \right] \quad (22)$$

Equation (22) may be combined with Newton's law and the form of the shear diagrams of these materials becomes

$$-\frac{1}{g_c} \frac{dV}{dr} = \frac{\gamma}{\mu_L} \left[\frac{1.2 - 2S(c\gamma^{-0.2} + 1)^3}{1.2 + S(c\gamma^{-0.2} + 1)^3} \right] \quad (23)$$

TABLE I
VALUES OF S , μ_S AND γ FROM FIG. 3 AND
COMPUTED VALUES OF S_T AND ν

Curve	μ_S lb _M /ft-sec	S_T	$S = .0350$		$S = .0552$		$S = .0739$		$S = .0903$		$S = .0991$		$S = .1138$	
			ν	γ lb _F /ft ²	ν	γ lb _F /ft ²	ν	γ lb _F /ft ²	ν	γ lb _F /ft ²	ν	γ lb _F /ft ²	ν	γ lb _F /ft ²
a	.00145	.287	7.20	.116										
b	.00185	.346	8.88	.067										
c	.00238	.393	10.24	.045	6.13	.230								
d	.00290	.427	11.21	.039	6.73	.155								
e	.00350	.454	11.96	.033	7.23	.125	5.13	.400						
f	.00425	.479			7.68	.105	5.48	.325	4.31	.700				
g	.00490	.494			7.95	.095	5.68	.286	4.47	.636	3.98	.830		
h	.00605	.513			8.30	.086	5.94	.242	4.68	.563	4.18	.755		
i	.00728	.527			8.55	.078	6.13	.212	4.83	.506	4.32	.695	3.63	1.196
j	.00947	.543					6.34	.190	5.01	.446	4.48	.616	3.77	1.073
k	.0126	.557					6.53	.173	5.16	.393	4.62	.540	3.88	.950
l	.01975	.572					6.74	.154	5.33	.340	4.77	.455	4.02	.805
m	.0516	.589					6.97	.140	5.53	.290	4.94	.375	4.17	.665
n	∞	.600	16.15	.020	9.87	.060	7.12	.130	5.64	.270	5.05	.340	4.27	.590

Fig. 4. Bound Water as a Function of Shear Stress for Thorium Oxide-Water Slurry.

Fig. 5. Relationship of t/r Ratio to Shear Stress for Thorium Oxide - Water Slurry (Log Plot)

Then from Mooney's equation, in the integral form, the expression for the psuedo shear diagram may be written

$$\frac{8V_a}{g_c D} = \frac{4}{\mu_L \tau_w^3} \int_{\tau_y}^{\tau_w} \tau^3 \left[\frac{1.2 - 2S(c\tau^{-0.2} + 1)^3}{1.2 + S(c\tau^{-0.2} + 1)^3} \right] d\tau. \quad (24)$$

The lower limit of the integral of equation (24), τ_y may be evaluated from equation (23) when dV/dr is zero or

$$\tau_y = \left[\frac{c}{(1.2/2S)^{1/3} - 1} \right]^5. \quad (25)$$

Unfortunately, equation (24) would be excessively laborious to evaluate numerically due to the form of the function of τ in the integral. Therefore the integral of equation (24) is being evaluated using the ORACLE. The results will be plotted as the function $(8V_a/D)(\mu_L/4g_c)$ versus τ_w in families of S for various values of c . These plots, together with illustrative calculation will appear in a future report.

Conclusions:

The quantitative relationship between suspension viscosity, liquid viscosity, solidity and shear stress, expressed as equation (22), makes it possible to calculate the viscosity of a non-Newtonian of the type exhibited by thorium oxide slurries as a function of shear stress and concentration knowing only one experimentally determinable constant, c . The constant c may be determined from any one measured viscosity or yield stress at a known concentration from equation (22). From a more practical standpoint, the analysis makes possible

the calculation of the pressure drop in any pipe for any concentration of slurry, from the measurement of one pressure drop in one pipe of one concentration using equation (24). A valuable corollary of this conclusion is that from the yield stress at one concentration, the yield stress at any concentration may be predicted, equation (25). This application has been proposed in a previous memo.⁵

A conclusion of secondary importance is that non-Newtonian fluids satisfying the relationship of equation (22), such as thorium oxide slurries, are neither pseudoplastic or strictly Bingham plastic in behavior. In order to satisfy equation (22), a fluid must necessarily have a yield stress at all concentrations of solid above infinite dilution. From equation (25) it may be seen that τ_y equals zero only when S equals zero and hence the fluid cannot be a pseudoplastic. That the fluid cannot satisfy equation (22) and be Bingham plastic may be seen from a comparison of equation (5) for a Bingham plastic with equation (23). It should be noted, however, that a material can comply with equation (22) and still appear to be a Bingham plastic as demonstrated by the shear diagrams for low solidity in Fig. 3.

Several limitations of the proposed analysis are evident. First, the data thus far obtained have been limited to low temperatures. Therefore the applicability of the analysis to high temperatures, i.e., 100-300°C where the rheological properties of thorium oxide slurries are most pertinent, has not been proven. Second, the data verifying the analysis have been obtained only with systems of thorium oxide and water thereby limiting its generality. A third limitation of the analysis results from the relatively small range of the variables of concentration and shear stress explored. It is possible that

the relationship between t/r and γ deviates from that shown in Fig. 5 at values of shear stress less than 0.02 lb/ft^2 and greater than 1.1 lb/ft^2 . However, a similar plot for a different thorium oxide system indicated the validity for shear-stress up to 4.5 lb/ft^2 and solidities up to 0.155.

Finally, the actual existence or importance of the lyosphere envelope, which is an integral part of the analytical model, has been seriously questioned by many authorities. Regardless of its actual existence, however, the utility of the lyosphere concept is undeniable, and it is believed that the results disclosed in this paper demonstrate that utility further.

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NOMENCLATURE

A,B,C	Constants in Powell-Eyring equation	-
D	Diameter of circular conduit	ft
E	"Shearing velocity" in Asbeck equation	sec ⁻¹
F	Constant in Reiner-Riwlin equation	-
L	Length of circular conduit	ft
ΔP	Pressure drop across length L of circular conduit	lb _F /ft ²
S	Volume fraction pure solids or solidity	ft ³ /ft ³
S _L	Volume fraction effective solids or effective solidity	ft ³ /ft ³
dV/dr	Rate of shearing strain or velocity gradient	sec ⁻¹
V _a	Average velocity in circular conduit	ft/sec
c	Constant	-
g _c	Conversion factor	$\frac{32.2 \text{ ft-lb}_M}{\text{sec}^2 - \text{lb}_F}$
k,u,α,σ	Constants in Asbeck equations	-
m	Coefficient	-
n	Exponent in Farrow-Lowe equation	-
r	Radius of particles	ft
t	Thickness of liquid envelope	ft
x	Frictional coefficient in Robinson equation	-
y	Relative sediment volume	ft ³ /ft ³
γ	Coefficient of rigidity	lb _M /ft-sec
μ	Viscosity of Newtonian fluid	lb _M /ft-sec
μ _L	Viscosity of suspending liquid	lb _M /ft-sec

μ_o	Viscosity of psuedoplastic at no shear stress	$lb_M/ft\text{-}sec$
μ_S	Viscosity of suspension	$lb_M/ft\text{-}sec$
μ_∞	Viscosity of psuedoplastic at infinite shear stress	$lb_M/ft\text{-}sec$
ν	Volume ratio of envelope to pure solids	ft^3/ft^3
ρ	Density of fluid	lb_M/ft^3
τ	Shear stress	lb_F/ft^2
τ_D	Shear stress at diameter D in circular conduit	lb_F/ft^2
τ_y	Yield stress	lb_F/ft^2

APPENDIX

Sample Calculation of the Quantity t/r

1. The viscosity, μ , of any point on line a of Fig. 3 may be computed from equation (1) as

$$-\frac{1}{g_c} \left(\frac{dV}{dr} \right) = \frac{\gamma}{\mu} \text{ or } \mu = \frac{\gamma}{-\frac{1}{g_c} \left(\frac{dV}{dr} \right)}$$

$$\text{or } \mu = \frac{0.290}{200} = 0.00145 \frac{\text{lb}_M}{\text{ft-sec}}$$

2. The volume fraction of effective solids represented by line a of Fig. 3 may be calculated from equation (13) as

$$\mu_S = \mu_L \left[\frac{1.2 + S_T}{1.2 - 2S_T} \right] \text{ or } S_T = \frac{1.2 \left(\frac{\mu_S}{\mu_L} - 1 \right)}{\frac{2\mu_S}{\mu_L} + 1}$$

or using $\mu_L = 0.000597 \text{ lb}_M/\text{ft-sec}$ at 25°C .

$$S_T = \frac{1.2 \left(\frac{0.00145}{0.000597} - 1 \right)}{2 \left(\frac{0.00145}{0.000597} \right) + 1} = .287$$

3. The shear diagram for the suspension having a concentration, S , of 0.0350 intersects line a at $\gamma = 0.116 \text{ lb}_F/\text{ft}^2$, Fig. 3.
4. The ratio of envelope volume to solids volume for the suspension having a concentration, S , of 0.0350 at the shear stress γ of $0.116 \text{ lb}_F/\text{ft}^2$ may be computed from equation (17) as

$$S_T = S(1 + \nu) \text{ or } \nu = \frac{S_T}{S} - 1$$

$$\text{or } \nu = \frac{0.287}{0.035} - 1 = 7.20$$

5. The ratio t/r for the suspension having a concentration, S , of 0.0350 at the shear stress, γ , of $0.116 \text{ lb}_f/\text{ft}^2$ may be computed from equation (16) as

$$\frac{t}{r} = [(\nu + 1)^{1/3} - 1]$$

$$\frac{t}{r} = [(7.20 + 1)^{1/3} - 1] = 1.016$$