R & D NOTES

Particle Settling in Viscous Non-Newtonian Hydroxyethyl Cellulose Polymer Solutions

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Oil production along the Gulf Coast and elsewhere from formations that are essentially unconsolidated sand often requires some means of "sand control." That is, oil production must be free of sand, not merely to minimize solids handling problems, but to insure long and productive well performance. Gravel packing is often the preferred means of sand control. With this method, gravel, properly sized to prevent the production of formation sand, is injected into the well bore where it builds a filter between the formation sand and in most cases a central metal screen. The gravel is usually pumped into the well as a concentrated slurry using a viscous aqueous polymer solution. For example, Natrosol 250HHR (Hercules, Inc.) a hydroxyethyl cellulose (HEC) polymer, is used by Shell Oil in its "AQUAPAC" (Lybarger et al., 1974). The success of the gravel pack depends on the "down-hole" properties of the polymer and suspension.

In part to provide background for a study of the settling behavior of slurries, measurements were made of the settling velocity of single particles in the viscous non-Newtonian polymer solutions of Natrosol 250HHR. However, particle settling data also allows calculation of apparent viscosities at low shear rates. And, because of the wide variation in settling velocity with polymer concentration, provides a convenient on-site test of polymer quality during the placement operation. That is, the effects of viscosity reducers and the particular time-temperature history for a given polymer solution may be readily estimated. Since particle suspensions in viscous polymer solutions are also used for fracturing and in other *in situ* energy applications, there are a range of uses for the kinds of results summarized here.

EXPERIMENTAL RESULTS

Figure 1 shows the influence of shear rate, $\dot{\gamma}$, on apparent viscosity η_a, for concentrations of 0.25%w, 0.50%w, 0.75%w and 1.0%w of Natrosol 250HHR. Note in particular the wide range of apparent viscosities (shear stress divided by shear rate) for the more concentrated solutions. At high shear rates, the data fall on straight lines and the solutions follow the "power-law" model. As shear rates decrease the curves level out to an asymptote, the "zero-shear" viscosity, which in the case of a 0.75%w solution is about 3.5 Pa·s, while for 0.50%w, it is about 0.45 Pa·s. Note that the zero-shear viscosity asymptote moves to higher shear rates as polymer concentration decreases. For example, while the asymptote hasn't been achieved even at shear rates, $\dot{\gamma}$, as low as 0.1 s^{-1} for 1%w solutions, apparent viscosity is nearly constant for 0.25%w solutions between $\dot{\gamma}$ = 1 to 10 s^{-1}. A typical AQUAPAC fluid at 80 lb (36 kg) HEC/1,000 gal (3.8 m^3) is somewhat more viscous than that of 0.75%w. At formation temperature it could have an apparent viscosity similar to that of a 0.5%w solution at room temperature. Values at the higher shear rates were measured with Fann and Brookfield viscometers. At the lowest shear rates, apparent viscosities were calculated from Stokes' Law settling velocities.

Measured settling velocities, V_T, for particles of different sizes and densities are presented in Figure 2. All of the particles are spheres except for the gravel. Sizes for both the gravel and UCAR props are taken as the average value for the two screens used as indicated on the figure. (These particles are often used in gravel packing.) Settling velocities for gravel and UCAR props are seen to be in close agreement. Results for Teflon form a smooth set of curves, while there is more scatter in the results for nylon spheres which are only 14% more dense than water, so that very low settling velocities could be achieved. However, as settling velocities decrease they become subject to much more error, from inhomogeneities or minute air bubbles in solution and the viscosity calculation is also made more uncertain by the difficulty of accurately specifying the actual density and diameter of the progressively smaller particles used.

The zero-shear viscosity, η_o, can be estimated by measuring V_T for spherical particles settling in the polymer solution at low shear rates within the range of validity of Stokes' Law (Williams, 1965; Sutterby, 1966; Uhlherr et al., 1976). Then,

$$\eta_0 = \eta_s = \frac{D_p^2(\rho_s - \rho)g}{18V_T} \qquad \text{for } Re_p = \frac{D_p\rho V_T}{} \leq 0.1$$

With measurements for several different particle sizes, the calculated values of η_s can be extrapolated back to zero-shear. V_T values were initially measured for teflon spheres of 1.5, 3.2 and 4.7 mm over length increments of 0.13 m or more in 0.05 m diameter tubes. The Reynolds number limitation is met for the 1.0 and 0.75%w solutions and η_s increases slightly with decreasing shear rate. For the 0.5%w solution, shear rates were high (at 7 to 23 s^{-1}) and the Reynolds number was 0.4 for the largest sphere. To improve zero shear estimates experiments were carried out with spherical nylon (1.6 to 4.8 mm) for all solutions and glass (UCAR props −0.77 to 2.2 mm) particles in 0.25 and 0.75% solutions. The results show that η_s tends to increase slightly with particle size and shear rate even when Re_p is very small (Torrest, 1982). However, from Figure 1 it is evident that apparent viscosities determined with the Brookfield viscometer and settling particles intergrade smoothly as the zero shear asymptotes are approached.

DISCUSSION

The influence of particle size on terminal settling velocities for several polymer concentrations is shown in Figure 3. The data is from Figure 2 but replotted to show the relation between V_T and D_p. Since from Stokes' Law $V_T \sim D_p^2$, on Figure 3 lines of slope 2 have been drawn through the data points for the smallest particles at each concentration with the data taken from smoothed curves of Figure 2. Data points clearly fall on or near the lines of slope 2 for all but the lowest polymer concentration, with a tendency to fall below the lines for the larger particles, that is, as the shear rate increases. Since the maximum shear stress at the sphere surface for a Newtonian fluid is $D_p(\rho_s - \rho)g/6$, (Williams, 1965), then with Stokes' Law the maximum shear rate is $(3V_T/D_p)$ and this value was used for Figure 1. Numerical values for this surface shear rate for 1mm gravel particles vary from about 105 s^{-1} for the 0.25%w solution (for which $Re_p > 0.1$) to 12 for 0.5%w and 1.2 s^{-1} for 0.75%w. For the 0.25%w solution, 105 s^{-1} is much closer to the zero shear asymptote. For the nylon spheres, $Re_p > 0.1$ only for the 0.25%w solution and varies from 0.19 at 9.1 s^{-1} to 1.4 at 15.6 s^{-1} for the 3.97 mm particle (Torrest, 1982). The approach of the data to the lines of slope 2 are really just indications that for small particles falling in viscous polymer solutions, the shear rates are low enough to keep the viscosities closer to the zero shear asymptotes than the power law region which holds roughly above $\dot{\gamma} \simeq 50\ s^{-1}$ (but in fact depends on polymer concentration).

In a recent paper, Novotny (1977) has combined the definition

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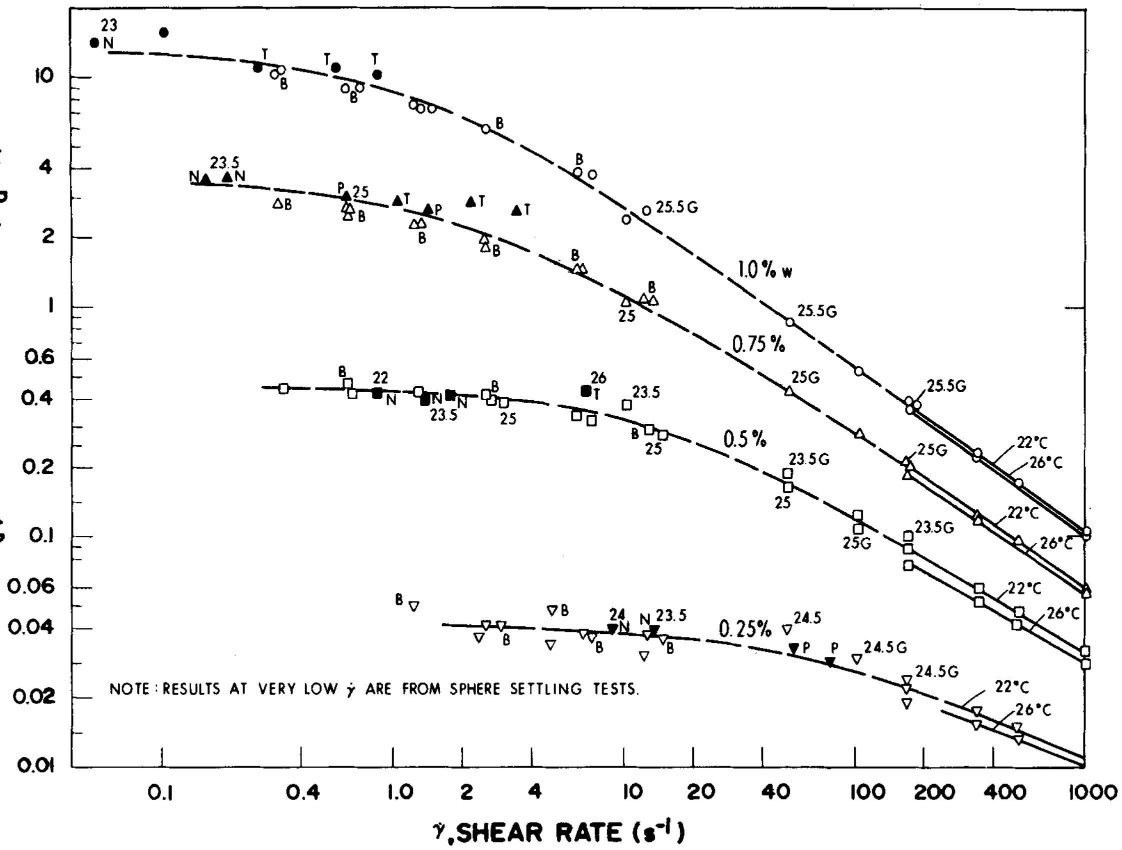

Figure 1. Apparent viscosity of Natrosol 250HHR solutions: *N* and *T* at data points denote nylon and teflon spheres; *P* = UCAR props; *B* = Brookfield viscometer; *G* = Fann viscometer (with gear changer).

tion of apparent viscosity for a power law fluid, $\eta_a = m(\dot{\gamma})^{n-1}$, with the effective shear rate on a settling particle so that $V_T \sim (D_p)^{n+1/n}$. Since Stokes' Law can be used at sufficiently low shear rates to estimate zero-shear viscosity, while the power law representation is valid only above some minimum shear rate, Novotny's procedure is, in general, not valid. In our case, for Natrosol 250 HHR solutions HHR solutions at 22°C n = 0.43 for 0.50%w and 0.34 at 0.75%w. These lead to velocities proportional to particle diameter raised to a power greater than three—which is clearly inconsistent with the data. Gravel pack gravel does not fall fast enough in the viscous HEC solutions to exhibit shear rates in the power law range. However, the lines drawn on Figure 3 allow us to extrapolate back to gravel diameters typical of 20–40 mesh gravel (at about 0.6mm) and this has been done with the dashed lines.

Figure 4 shows the influence of polymer concentration on settling velocity. This is a composite of Figures 2 and 3, and some additional data for 20–40 mesh gravel and calculations. Note in particular that V_T on a log scale is essentially linear in polymer

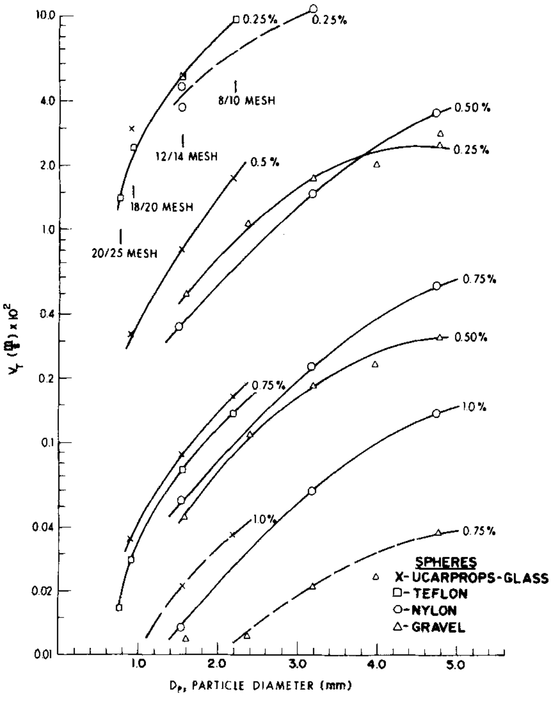

Figure 2. Settling velocities for several kinds of particles in Natrosol 250HHR solutions.

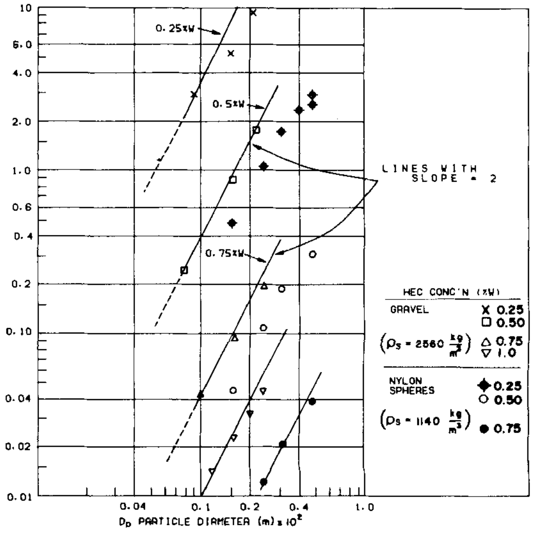

Figure 3. Particle settling velocities in Natrosol 250HHR solutions and the applicability of Stokes' Law.

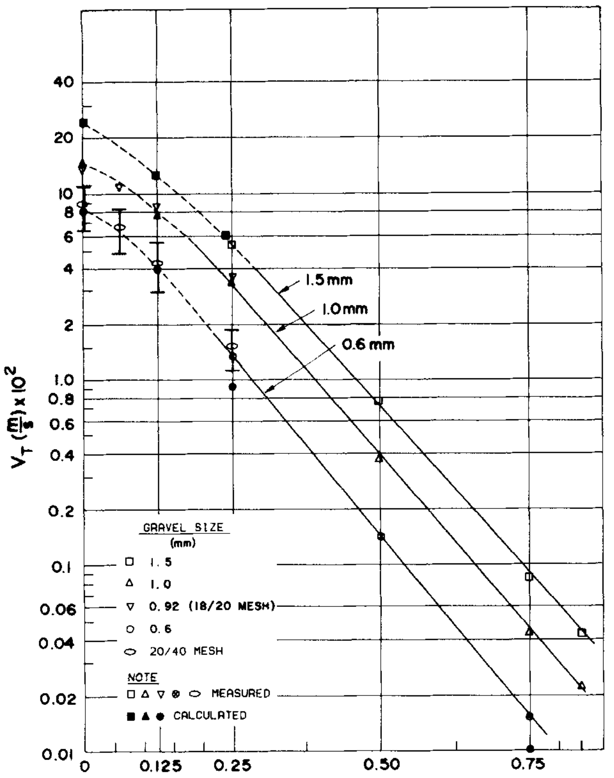

Figure 4. Influence of polymer concentration on settling velocity for several particle sizes.

concentration, at least above 0.25%w. At lower concentrations, the apparent viscosity is low so that Stokes' Law with η_a chosen for the actual shear rate doesn't provide a good estimate. Instead an estimate is made using an empirical relationship for the drag coefficient (Bird et al., 1963) valid for $2 < Re_p < 500$. Then, with V_T calculated using an estimate of η_a, the shear rate is checked to see if it is high enough to require a reduction of η_a. An iteration or two gives satisfactory agreement with experimental values with the straight lines at higher concentrations curving down as the solutions become more dilute until they are water without polymer. Note that the average velocity of 20–40 gravel is essentially the same as that for 0.6 mm gravel (as extrapolated from measurements). Vertical lines through the data points show approximate extremes of the velocities.

The large variation in settling velocity for gravel particles with variation in polymer concentration could be used as a convenient test of fluid quality in field applications. For example, V_T varies by about 2 orders of magnitude between 0.5%w HEC and water. Hence, measurement of the settling velocity of particles of known size would give a reasonable estimate of polymer concentration and viscosity. Using a pinch of 20–40 mesh gravel the average settling velocity is increased by about 100-fold from that for 0.75%w to 0.25%w for which V_T = 15 mm/s although the apparent viscosity is still about 0.025 to 0.04 Pa·s at 0.25%w.

NOTATION

D_p = particle diameter
n = power law exponent
V_T = particle settling velocity
Re_p = particle Reynolds number
η_a = apparent viscosity, Pa·s
η_s = Stokes viscosity, Pa·s
ρ = liquid density
ρ_s = solid density
$\dot{\gamma}$ = shear rate, s^{-1}

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Effects of Association on Enthalpy of Coal-Derived Liquids: Cryoscopic Molecular Weight As a Function of Concentration

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INTRODUCTION

A major difficulty in estimating physical and thermophysical properties of petroleum fractions and coal-derived liquids is the fact that such mixtures represent an essentially infinite continua of components and as such they cannot be defined by component mole or mass fraction. In the petroleum industry, this problem has been overcome by the use of easy to measure characterization variables such as density (or API gravity) and the Watson Characterization Factor, K, defined as

$$K = \frac{\sqrt[3]{T_b(^0R)}}{d}$$

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