DEVELOPMENT OF DEVICES FOR IN-LINE VISCOSITY MEASUREMENTS OF WASTES

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ABSTRACT

In applications involving the transport and evaluation of Hanford wastes, performance of the pipeline and pumping systems depends upon the viscosity of the waste stream. These streams are typically heterogeneous and involve a multiphase mixture of solids, liquids and, often, gases. These slurries are opaque and it is usually difficult to take a representative sample that can be tested off line.

The goal of this research has been to develop on line sensors for the measurement of slurry viscosity. The principle of the technique rests with the conservation of linear momentum for steady pressure driven flow in a tube. Here, the pressure drop in the tube determines the local shear stress. A simultaneous measurement of the velocity and a robust technique for differentiating it provides the local shear rate distribution. Evaluating the shear rate and the shear stress at points along the tube yields the shear stress shear rate relation, which is equivalent to the shear viscosity shear rate relation. The superiority of experimental techniques based upon this principle is that a single measurement of the velocity profile and the pressure drop provides the viscosity over a wide range of shear rates – up to two decades in practice. This permits the accurate characterization of complex liquids including slurries.

Implementing this technique for slurries requires measurement methods that provide for the accurate velocity determination over the entire tube radius and that the system can work in the presence of particles and with opaque systems. We have developed two techniques: one based on magnetic resonance imaging (MRI) and the other based on ultrasonic Doppler velocimetry (UDV).

For the MRI, we use phase encode imaging to directly measure the velocity profile. Measurements have been on a variety of non-Newtonian fluids, including those showing shear thinning at high shear rates and a Newtonian plateau at low shear rates. We have also examined a suspension that has a well-defined yield stress. The MRI technique provides excellent agreement with standard rheological measurements. We are able to identify design parameters that can be used to build an instrument and make a priori viscosity measurements without reference to a particular model of fluid behavior.

For the UDV, we have developed an instrument that is capable of making measurements of velocity in tubes. We have shown that these measurements can reproduce the parabolic velocity profile found for Newtonian fluids. We have also been able to make accurate measurements of

the velocity of fluids which exhibit power law behavior with the concomitant blunted velocity profiles.

INTRODUCTION

Process monitoring rheometers are generally divided into on-line and in-line devices. The most popular method of these measures the pressure drop during the flow of the fluid through a die or capillary. One viscosity data point corresponding to the capillary/tube wall shear rate is determined for one tube size and fluid flow rate. At the tube wall, the shear rate is determined using the Weissenberg-Rabinowitsch equation while the shear stress is calculated using a pressure drop per unit tube length measurement and the conservation of linear momentum. Using tubes of different diameters or changing the fluid flow rate permits the material to be characterized at different shear rates. In the case of complex liquids, shear viscosity data over a wide range of shear rates is needed. Our approach to obtain such data is to recognize that a fluid undergoing tube flow experiences shear rates ranging from zero at the tube center to a maximum at the wall and that, independent of the constitutive relation, the local shear stress distribution can be calculated from the pressure drop. To determine the shear rate we measure the velocity profile and take its radial gradient. We use two techniques to measure the velocity. Nuclear magnetic resonance imaging (NMRI) is non-invasive, chemically sensitive and does not need to contact the pipe. These characteristics make NMRI an attractive tool for material characterization. It can provide spatially resolved, rapid measurements for a wide range of materials including food products, polymer melts, filled systems and slurries that are either optically opaque or transparent. Ultrasonic Doppler Velocimetry is an alternative technique for determining velocity profiles in tubes. This technique offers the prospect of being less expensive than NMRI and, perhaps, more easily implemented. At the same time, it is not as well tested as NMRI.

THEORY

For steady, fully developed laminar flow in a tube the conservation of linear momentum gives the shear stress ,τ, as a function of the radial position in the tube, r,

$$\tau(r) = - \Delta P \, (r/2L) \qquad \text{Eq. (1)}$$

where ΔP is the downstream pressure subtracted by the upstream pressure over the pipe length L. The shear rate, $\dot{\gamma}$, at every point is obtained by differentiating the velocity,v, with respect to r,

$$\dot{\gamma}\,(r) = dv/dr \qquad \text{Eq. (2)}$$

Comparing equations. (1) and (2) and evaluating both expressions at each radial position gives the shear stress dependence upon the shear rate. Or, if the shear stress is divided by the shear rate to give the shear viscosity, η, shear viscosity data is provided over shear rates that theoretically range from zero at the tube center to a maximum at the tube wall. In practice, the range of shear rate data obtained from one velocity profile measurement is mediated by the fluid flow rate, fluid properties, tube diameter and timing diagram parameters. Accurate determination of the smallest velocity gradients, those near the tube center, is dominated by the

velocity resolution. Physical sources of velocity dispersion (e.g. diffusion and unsteady flow) and imperfections in the experiment, such as those brought upon by NMR hardware (e.g. eddy currents), cause an apparent spreading in the measured velocity at a given radius and affect the accuracy of the velocity gradient calculations.

ULTRASONIC DOPPLER VELOCIMETRY (UDV)

Velocity profiles for pipe flow obtained from ultrasonic Doppler velocimetry (UDV) may be used, together with measured pressure drops, to construct a rheogram for the fluid. The basic approach used was the same as above, with the exception that velocity profiles were obtained by UDV instead of by NMR. Velocity profiles were measured in fully developed, laminar pipe flow. Each such profile was then inverted to obtain the shear rate as a function of distance from the centerline of the pipe. The shear rate curve, together with the measured pressure drop was then used in constructing the rheogram. As with NMR, the measurement was non-invasive and real-time, requiring no sampling or laboratory measurements.

As described in (1), ultrasonic Doppler velocimetry (UDV) relies on measurement of the Doppler frequency shift of moving tracer particles, or scatterers, within a pipe flow (Figure 1). Using a short ultrasonic pulse system, the cross-sectional velocity profile can be obtained from the Doppler shift at each point in range. The Doppler frequency shift is given by

$$f_D = \frac{2v\cos\theta}{c} f \qquad \text{Eq. (3)}$$

where v is the particle velocity, c is the speed of sound in the fluid, f is the ultrasonic frequency, and θ is the angle of the transducer with respect to the pipe centerline. For example, the Doppler shift for a particle moving with velocity of 1 m/sec in water with speed of sound equal to 1500 m/sec, a transducer angle of 45°, and an ultrasonic frequency of 5 MHz is 4.72 kHz.

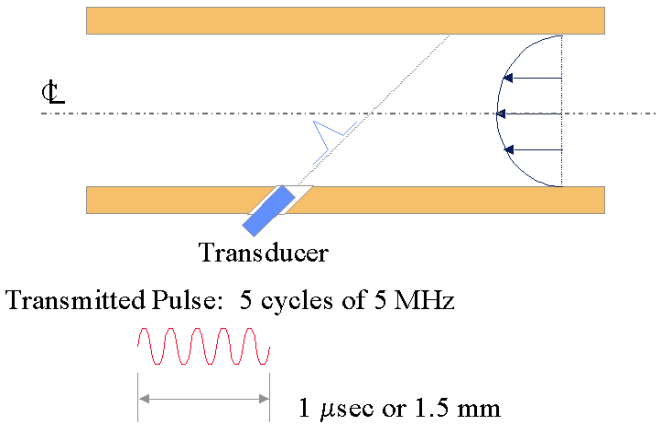

Figure 1. Schematic of UDV experiment.

In order to obtain good range resolution it is essential to transmit a short ultrasonic pulse. The range resolution is approximately equal to one-half of the spatial width of the pulse. For *N* sine-wave cycles of wavelength λ, the range resolution is approximately

$$\Delta_R = N\frac{\lambda}{2} \qquad \text{Eq. (3)}$$

For example, 5 cycles of 5 MHz (0.3 mm wavelength) yields a range resolution of approximately 0.75 mm. Obtaining such high range resolution creates a problem for measurement of the Doppler shift which is on the order of kHz. A gated sine-wave of only a few cycles has a relatively wide frequency bandwidth *B* of approximately,

$$B = \frac{f}{N} \qquad \text{Eq. (4)}$$

which is 1 MHz for 5 cycles of a 5 MHz wave. The Doppler shift would need to be on the order of or larger than 1 MHz to be distinguishably measurable from the frequency spectrum of the echo returned from the particles. Doppler shifts this large would only be expected if the fluid velocity were on the same order as the speed of sound in the fluid. Velocities of interest in pipe flow problems are much lower than the speed of sound. While reducing the bandwidth would allow for good Doppler velocity resolution it would yield a poor range-resolution. To resolve this conflict, we have opted to transmit and receive multiple pulses, i.e., to observe the fluid over a much longer time interval and obtain a higher number of observations from each point in the flow field.

In this approach, the pulse rate has prescribed limits. The maximum pulse rate for an unambiguous signal is set by the pulse transit time. The minimum pulse rate is that which will resolve the Doppler frequency for the highest particle velocity (two points per cycle to prevent aliasing, or $2f_D$). Combined, these requirements set the maximum flow velocity that can be measured. In a 2-inch diameter pipe with transducer geometry and sonic velocity given in the earlier example, this velocity limit is just over 1m/s.

The shear rate as a function of distance from the centerline of the pipe was obtained from the velocity profile by a curve-fitting procedure. The velocity profiles from UDV were fitted using a different method than was used with the NMR data. The shear rate $\dot{\gamma}$ versus radial position was approximated as a cubic spline. This form of curve is very general (constrained only so that the function and its first and second derivatives are continuous) and any kind of rheology may be represented by it. Once a set of knots for the spline is chosen, it is possible to represent the spline curve as a linear combination of independent, simple functions

$$\dot{\gamma}(r) = \sum_{j=0}^{N} \alpha_j \varphi_j(r) \qquad \text{Eq. (5)}$$

where *r* is the reduced distance from the centerline r/R, R being the radius of the pipe, the φ_j were cubic b-spline basis functions and the α_j were coefficients estimated from the velocity data

by multi-linear regression. The basis functions were easily integrated to obtain a general representation of the velocity profile containing the unknown coefficients:

$$\frac{v(r)}{R} = \alpha_{N+1} - \sum_{j=1}^{N} \left\{ \alpha_j \int_{r_0}^{r} \varphi_j(x)dx \right\} \qquad \text{Eq. (6)}$$

It is the above expression that was fit to the UDV-determined velocity profiles. The shear rate curve $\dot{\gamma}(r)$ was then obtained directly from the original linear combination without differentiating.

After the shear rate curve was determined, the inverse rheogram was recovered using the relation

$$\dot{\gamma}(r) = \dot{\gamma}\left(\frac{\tau}{\tau_w}\right), \qquad \text{Eq. (7)}$$

as discussed above, where reduced distance r=r/R and τ_w was the wall shear stress determined from the measured pressure drop -ΔP

$$\tau_w = -\Delta P\left(\frac{R}{2L}\right), \qquad \text{Eq. (8)}$$

L being the distance between the pressure measurement points.

EXPERIMENTAL AND SAMPLE RESULTS

NMRI

The NMRI velocity profiles were obtained from the proton signal using a General Electric CSI-II/TecMag Libra spectrometer connected to a 0.6 Tesla Oxford superconducting magnet (corresponding to 25.96 MHz 1H resonance frequency) with a PowerMac/MacNMR user interface (TecMag, Houston, USA). The horizontal magnet has a clear inner bore diameter of 330 mm. 0.2 G/mm. The PGSE method was used for all fluids except the CMC solution studies where a pulsed gradient stimulated-echo (PGSTE) was used. The stimulated-echo is able to reduce displacement measurement inaccuracies resulting from eddy currents that generally arise when using unshielded magnetic field gradients (2, 3).

The closed flow loop system consisted of both nonmagnetic stainless steel (VNE Corporation, Janesville, WI) and acrylic (Laird Plastics, Santa Clara, CA) tubing of the same inside diameter, 22 mm. The acrylic section (length 0.75 m) was used within the bore of the magnet. Six meters of straight tubing preceded the imaging section of the magnet (length/diameter 270) to ensure fully developed flow. A positive displacement pump (SPS-20 Sine Pump, Orange, CA) was used the shaft rotational speed being controlled by a variable frequency drive and gear motor (Statco Engineering & Fabricators, Inc., Huntington Beach, CA). Approximately 15 m of 25.4 mm ID Tygon tubing was attached between the outlet of the pump and the first stainless steel

section of the flow loop and submerged in water to dampen the pressure pulsations. Differential pressure measurements were measured at two hole pressure taps on both sides of the bore using a Taylor differential pressure transmitter (Model 3400T Series Transmitter, Taylor Instrument Company, Rochester, NY). The length between the taps was changed to ensure the pressure drop exceeded seven inches of water, the minimum pressure drop the transducer could measure. The differential pressure transmitter was connected to the hole pressure taps using high pressure tubing approximately 4 m in length. The 4-20 mA signal from the transmitter was connected to a resistor and a voltmeter to measure the voltage across the resistor. The transducers were calibrated by applying known liquid level columns of water and measuring the resulting voltages.

Typical results for the NMRI experiments are shown in Figure 2 (4). For these experiments a polymer solution, 1% aqueous polyethylene oxide (PEO) was used. Ten measurements were obtained at a constant flow rate of 26 ml/s. The velocity resolution was maintained at 2.2 mm/s while the radial resolution was 0.2 mm, 0.3 mm, 0.4 mm, 0.5 mm and 1 mm, yielding velocity profiles with 110, 73, 56, 44 and 22 equally spaced velocity data points, respectively. Two velocity profiles were measured at each radial resolution setting. One velocity data point corresponding to r=0 was removed for each measurement. Figure 2 compares five experiments to shear viscosity data obtained with a Haake using the cone and plate geometry. Although it is difficult to distinguish the individual data points from each NMRI experiment as well as the Haake RS100 data, the overall coincidence indicates that these data sets were nearly identical.

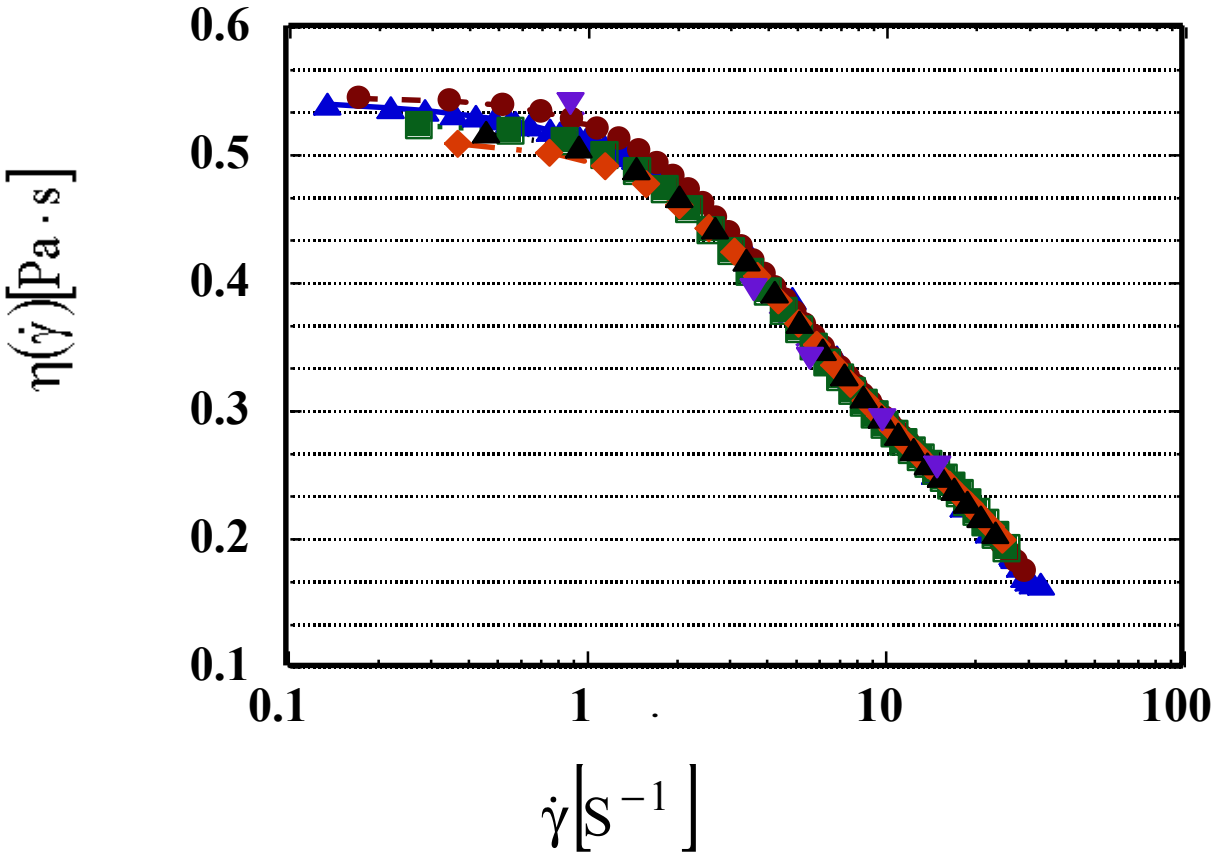

Figure 2. Shear viscosity data for polyethylene oxide solution. The ordinate is shear viscosity in Pa s.

UDV

A flow loop system was set up in our laboratory for testing the UDV device. The fluid used in the experiments was a 0.1-wt. percent solution of Carbopol EZ-1 (from B.F. Goodrich) in deionized water neutralized with sodium hydroxide to form a gel. The fluid was seeded with glass beads ("Ballotini Impact Beads, AH-Spec" from PQ Corporation, with diameters between 45 and 90 microns) to a concentration of 0.032-vol. percent. The beads served to scatter the ultrasound pulses back to the transducer. The fluid was circulated in a loop constructed of 2.093" I.D. tubing. A plastic block containing the ultrasound transducer was mounted in a long straight section of pipe, approximately 110 pipe diameters downstream of the nearest disturbance. A pair of single-leg water manometers measured the pressure drop per unit length in the pipe.

The rheograms of the pseudoplastic fluid were constructed from UDV velocity profiles. Measured and fitted velocities for one case, in cm/sec are plotted versus reduced radial distance r/R on the left side of Figure 3.

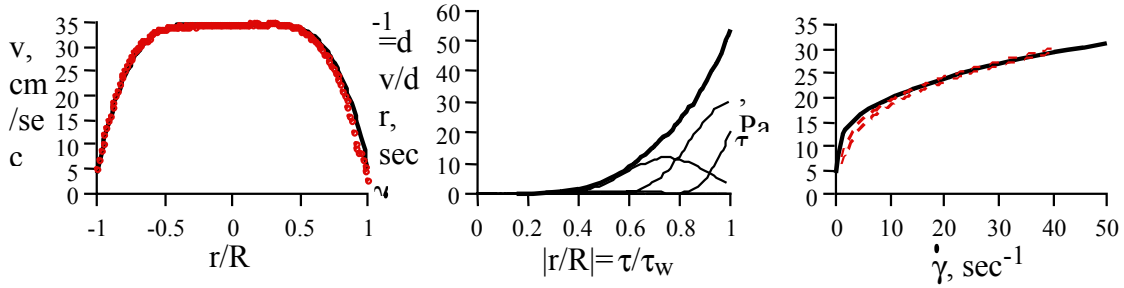

Figure 3. Left: UDV velocity versus distance from pipe centerline. Center: Shear rate versus distance. Right: Shear stress versus shear rate.

The points are experimental velocities from UDV, the solid line is the curve fit profile. The corresponding volumetric flow, pressure drop and wall shear stress were:

Flow rate, LPM	Pressure drop, inches H_20/inch	Wall Shear Stress, Pa
33.1	0.2424	31.59

Plotted in the center of Figure 3 is the corresponding shear rate versus radial position curve, this curve being that of the derivative of the left-hand plot. The fine lines in the central plot are the contributions of the individual basis functions to the total. On the right of Figure 3 is the resulting rheogram of shear stress versus shear rate. The solid line is the UDV-derived result, the dashed lines are measurements made on two grab samples using a cup and bob viscometer. The agreement of the UDV result with viscometer was good except for shear rates less than about 10 sec^{-1}. The low shear rates correspond to flow in the central plug region, where the slope in velocity was small and relatively uncertain.

CONCLUSION

In this paper, we have shown that both NMRI and UDV can be used to accurately determine the shear viscosity of complex liquids. Future work remains on looking at effects of signal dispersion on the ability to obtain accurate measurements as well as extending the range of data that can be obtained from a single measurement. It is important to emphasize that for either an NMRI or UDV experiment, data are obtained over a wide range of shear rates from a single measurement.

ACKNOWLEDGEMENT

This work was supported by a grant from the EMSP program at the Department of Energy.

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