FINAL REPORT

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DEPARTMENT OF CHEMICAL ENGINEERING & MATERIALS SCIENCE

UNIVERSTIY OF CALIFORNIA DAVIS

NON-INVASIVE DIAGNOSTICS FOR MEASURING PHYSICAL PROPERTIES AND PROCESSES IN HIGH LEVEL WASTES

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EXECUTIVE SUMMARY

This research demonstrated the usefulness of tomographic techniques for determining the physical properties of slurry suspensions. Of particular interest was the measurement of the viscosity of suspensions in complex liquids and modeling these. We undertook a long range program that used two techniques, magnetic resonance imaging and ultrasonic pulsed Doppler velocimetry. Our laboratory originally developed both of these for the measurement of viscosity of complex liquids and suspensions. We have shown that the relationship between shear viscosity and shear rate can be determined over a wide range of shear rates from a single measurement. We have also demonstrated these techniques for many non-Newtonian fluids which demonstrate highly shear thinning behavior. This technique was extended to determine the yield stress with systems of interacting particles. To model complex slurries that may be found in wastes applications, we have also used complex slurries that are found in industrial applications.

Magnetic resonance imaging (MRI) benefits from being non-invasive and also able to probe deep into opaque liquids that have high concentrations of solid particles. The sole disadvantage of this technique is that it has been difficult to implement in an industrial setting as a result of the cost and size of equipment. Recently, new developments with permanent magnets should make it possible to develop systems that do not rely on superconducting magnets that operate at liquid helium temperatures. The techniques developed during the course of these studies will be directly applicable to this.

Ultrasonic pulsed Doppler velocimetry (UPDV) enjoys some of the benefits of MRI, works with opaque systems and is non-invasive, while also being low cost and easily installed. The principal limitation of this technique is that for highly concentrated slurries, the ultrasonic pulse cannot penetrate deeply into the sample, which limits somewhat the range of shear rates that can be accessed.

We have also developed a technique to model systems comprised of particles of various sizes. Our most recent work in this area is aimed at developing continuum models that can be applied to complex flows.

SUMMARY OF ACCOMPLISHEMENTS

This work accomplished beyond question that MRI and UPDV can be used to measure the viscosity of complex slurries. By viscosity, we mean the relationship between the viscosity and the shear rate over a range of shear rates that allows the determination of the low shear rate Newtonian regime, the shear thinning (power law) regime and the high shear rate Newtonian plateau. We have also demonstrated that the yield stress can be determined. These were the objectives set out in this work.

PROJECT ACTIVITIES

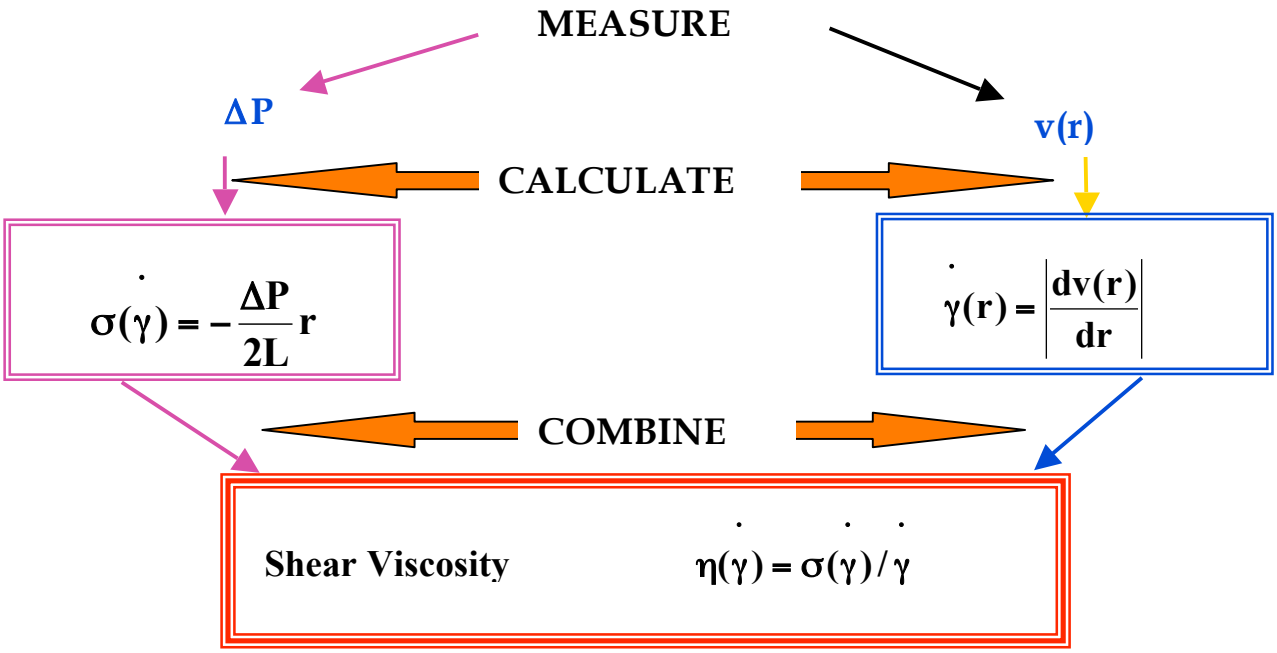

Figure 1. Schematic for measuring viscosity using either MRI or UPDV.

The technique used for measuring the viscosity using both magnetic resonance imaging (MRI) and ultrasonic pulsed Doppler velocimetry (UPDV) is shown schematically in Figure 1. The liquid or slurry is made to flow through a circular pipe at steady state. It is assumed that the flow is fully developed, that is there are no transients and that there are no effects due to the length of the pipe. Under such conditions, two measurements are made, the velocity profile, v, as a function of radial position, r, and the pressure drop, ΔP, over a length L. The velocity profile is the most critical measurement and the one that was developed for this application here. There are several methods that can be used with MRI to determine the velocity, including time-of-flight and phase encoding. These yield accurate values of v(r). For UPDV, the technique itself is designed for velocity profile determinations. The pressure drop is determined using differential pressure transducers that can be easily purchased. With these two measurements in hand three calculations are made. First, the shear stress in the pipe, σ, as a function of the radial position, r, can be ascertained regardless of the constitutive nature of the slurry / liquid.

Secondly, the shear rate, $\dot{\gamma}$, as a function of the radius can be calculated by differentiating the velocity with respect to the radius. Lastly, from the local value of the shear stress and the shear rate, the local value of the viscosity can be determined by dividing the former by the latter. This means that, for example, for a shear thinning power law fluid, which many slurries and complex liquids are, the viscosity variation across the pipe can be measured. This variation can be quite large, as shown by the theoretical calculation in Figure 2.

Figure 2 shows that the viscosity of a shear thinning material can vary by orders of magnitude across the radius of a pipe. At the wall, the viscosity is lowest where the shear rate is highest. Near the pipe center, where the shear rate approaches zero, the viscosity increases. Capturing this behavior is at the heart of this technique.

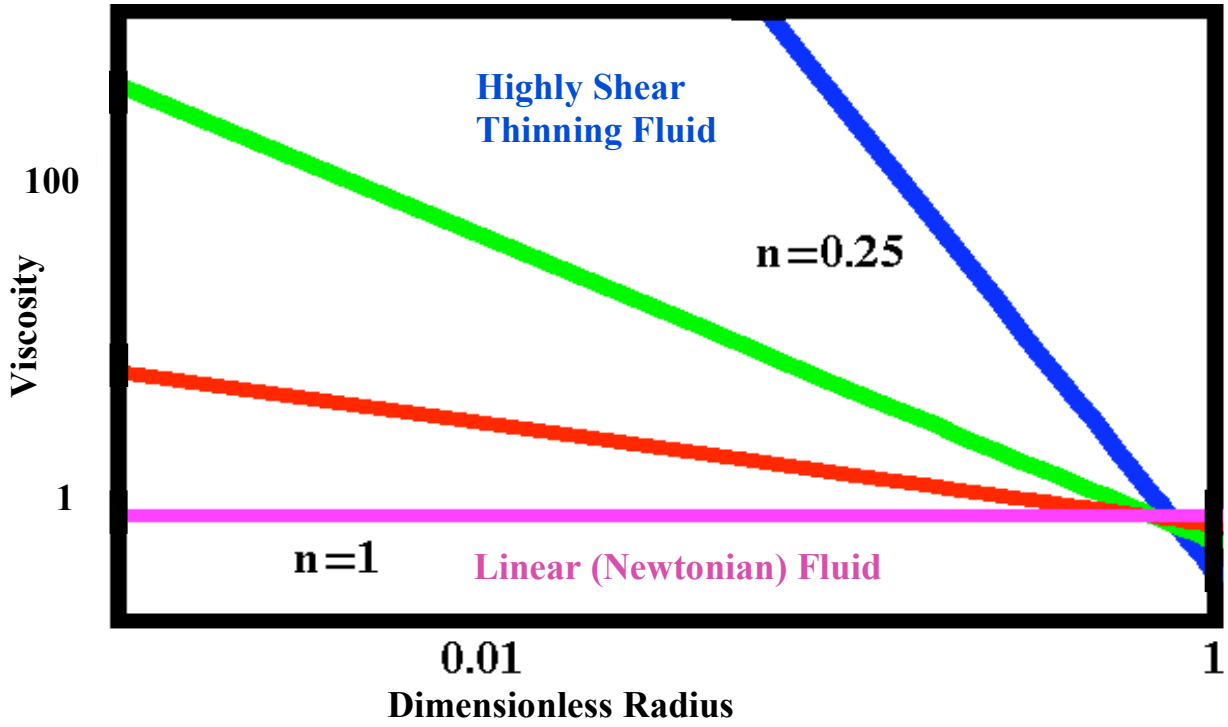

Figure 2. Theoretical viscosity variation across a pipe for a power law fluid. Values of the power law exponent used for these calculations were 0.25, 0.5, 0.75 and 1.0.

Typical data from MRI experiments are given in Figure 3. Here we show the shear stress and the shear rate as functions of radius. These data are for a highly complex liquid with properties that mimic slurries. To determine the viscosity at each radial position the shear stress is divided by the shear rate and the value of the viscosity at that radial position is calculated. Since the shear rate is known at each value of r, it is a simple matter to convert the viscosity as a function of radius to viscosity as a function of shear rate, as shown in Figure 4. Here, we also show the effect of one experimental parameter, the radial resolution, on the measurements.

Figure 4 shows that a wide range of properties of highly complex fluids can be obtained from MRI measurements. All of these data were measured at a single volume flow rate, 26 ml/s. At low shear rates, the material behaves as a Newtonian fluid with a constant viscosity. The value of this viscosity is independent of the radial resolution used, indicating that the measurement is highly reproducible. Further, we compare the MRI results with those obtained using a conventional rotational rheometer, the HAAKE RS100. In all cases, the agreement is excellent. This agreement among the MRI data and the rotational data persists at higher shear rates where the viscosity decreases with shear rate. Although we show data on a semi-logarithmic plot, in fact at the higher shear rates, the material is behaving as a power law fluid

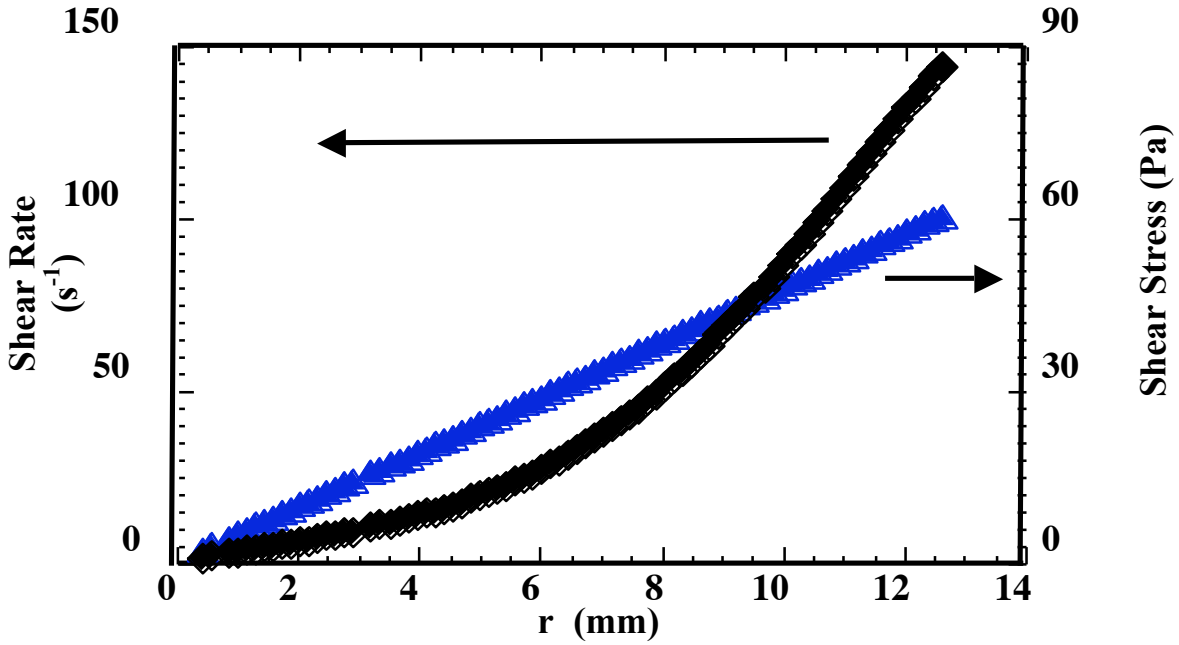

Figure 3. Shear stress and shear rate versus radius for a 1 % polyethylene oxide solution by MRI.

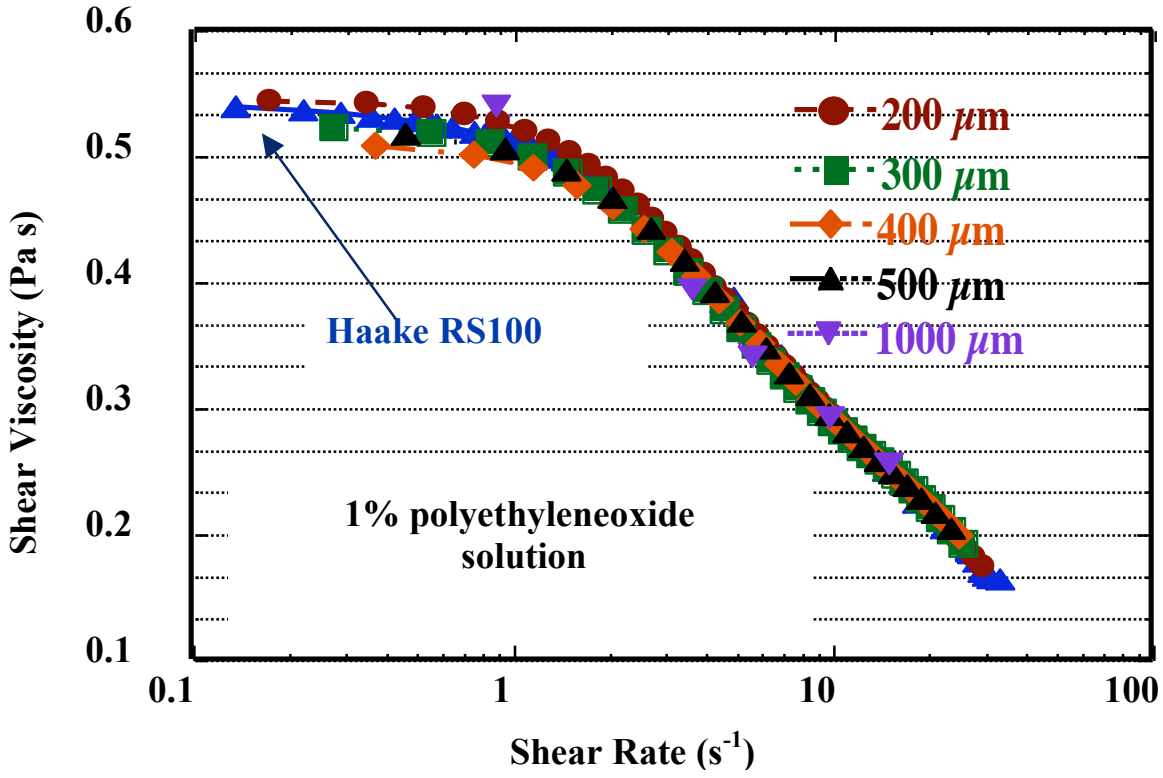

Figure 4. Shear viscosity versus shear rate for a 1% polyethylene oxide solution obtained by MRI using different radial resolutions.

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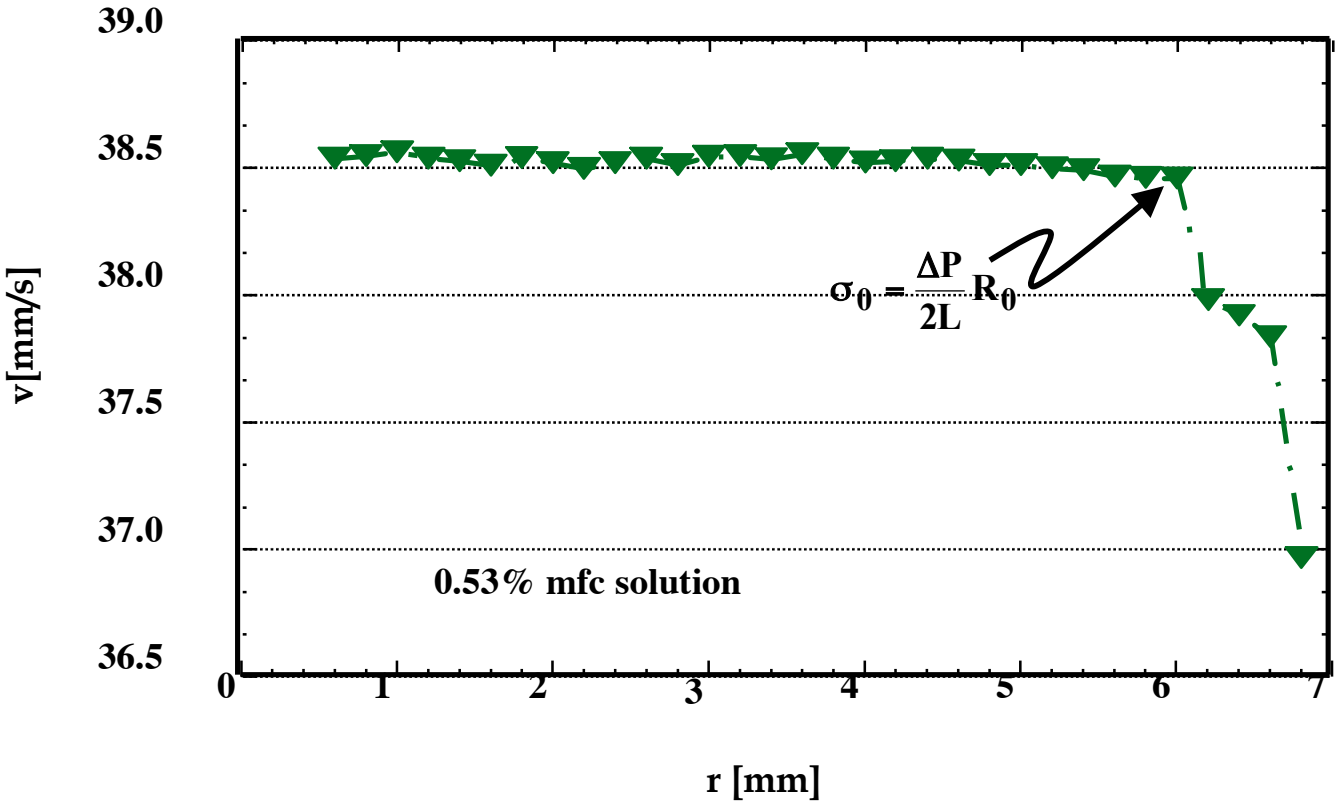

Figure 5. MRI velocity profile for a model fluid that shows yield stress behavior.

Figure 5 shows the versatility of the MRI (and, as we shall see, the UPDV method) for measuring not just the shear viscosity. In this figure, the velocity profile of a model fluid that exhibits a yield stress is shown. Near the center of the pipe, the velocity profile is flat, to within the measurement error. Near the pipe wall, all of the shearing occurs as the velocity falls from the maximum near the pipe center to zero at the wall. If the point in the pipe at which the velocity decrease begins to occur is identified, then the corresponding shear stress can be found, as shown in the figure. This value is the yield stress. Comparing the value found from the figure, 3.42 Pa ±0.34 with that obtained using the HAAKE RS100, 3.15 Pa±0.08, we see that there is excellent agreement.

For the MRI, similar data for the viscosity and the yield stress can be found for a wide variety of systems. It is then important to show that this technique can be applied to highly loaded slurries. This is amply demonstrated in Figure 6, which shows the velocity profile obtained by MRI for a 36% by volume particle suspension. Clearly, MRI is able to measure the velocity accurately over the pipe radius.

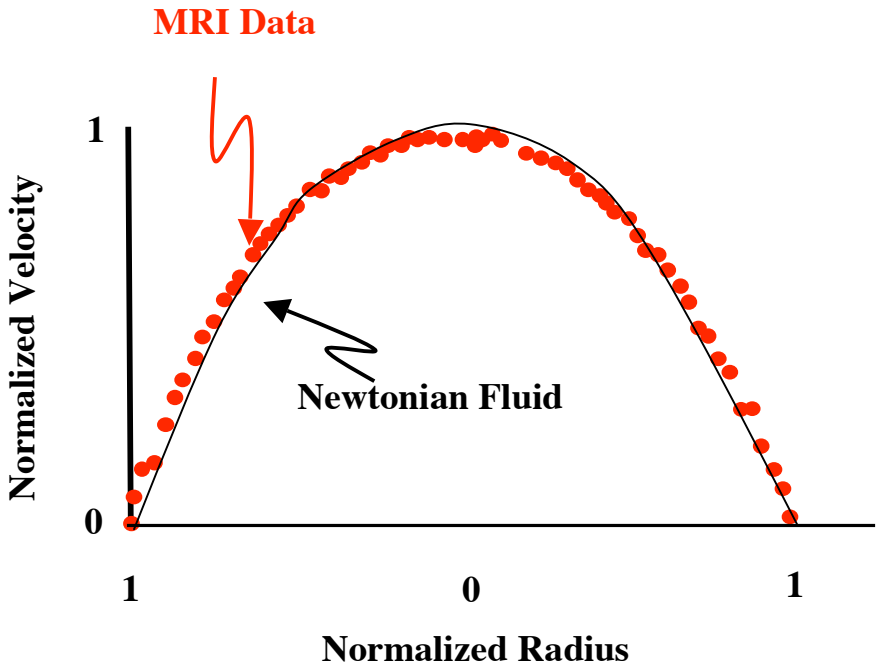

Figure 6. Velocity profile for a suspension of solid particles at 36% by volume loading in a Newtonian oil.

The foundation of the UPDV measurement is the same. Here, we will only demonstrate two features of this. First that excellent agreement can be obtained between the UPDV technique for measuring the viscosity and a standard method. Secondly, that the UPDV technique can be applied to studies of the yield stress. The experimental configuration is essentially the same: steady pipe flow. The UPDV technique is used to measure velocity profiles and the pressure drop is recorded. Figure 7 shows typical results for a complex suspension. Three sets of data based upon UPDV are given for different flow rates. In all cases, the data overlap showing excellent reproducibility. Accuracy is found by comparing the UPDV results with those obtain from a conventional capillary rheometer, which are indicated by the four triangles. Excellent agreement is found at all values of the shear rate. Such data were obtained for a wide variety of suspensions and complex liquids clearly confirming the findings for the MRI.

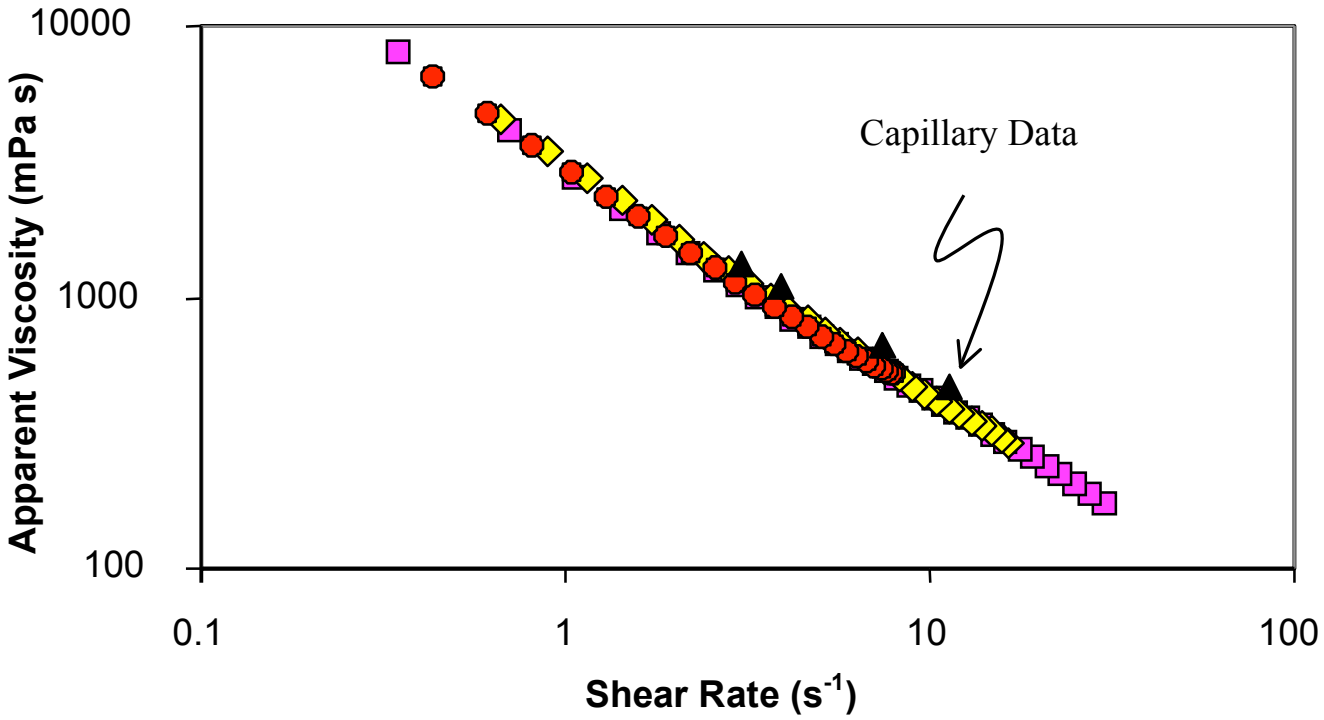

Figure 8. Comparison of UPDV data with data obtained from a conventional capillary viscometer.

Similar to the MRI-based technique, it is possible to determine the yield stress for complex slurries. Table 1 shows such data for a slurry that was designed to exhibit a measurable yield stress. This shows that the yield stress is independent of the volume flow rate (maximum velocity, vmax) and is therefore a material property. It also indicates that it increases with solids concentration, as expected.

Table 1. Yield stress data for suspensions at various solids concentration.

Percent Solids	v_{max} (cm/s)	Yield Stress (Pa)
17	4	5.0
13	4	3.0
13	7	3.3
13	13	3.4
9	7	1.9
9	17	2.2

The data in Table 1 and the yield stress data from MRI measurements show that this material property can be obtained from velocity profile – pressure drop measurements in pipe flow. As with the velocity measurements, these clearly showed the applicability of the techniques to a wide range of systems such as those that would be found in many slurry applications.

The last set of data to show are the design data for an instrument based on either the MRI or the UPDV techniques. With both techniques, the ultimate non-equipment (that is, pump, transducers, etc.) limitation results from the data near the center of the pipe. Here, the velocity profile is nearly flat. Calculating the shear rate, that is the velocity gradient, is difficult and the resolution of the technique is important. That resolution is represented by δv, the velocity resolution. The minimum shear rate at which viscosity data can be obtained can be nondimensionalized by the average velocity,

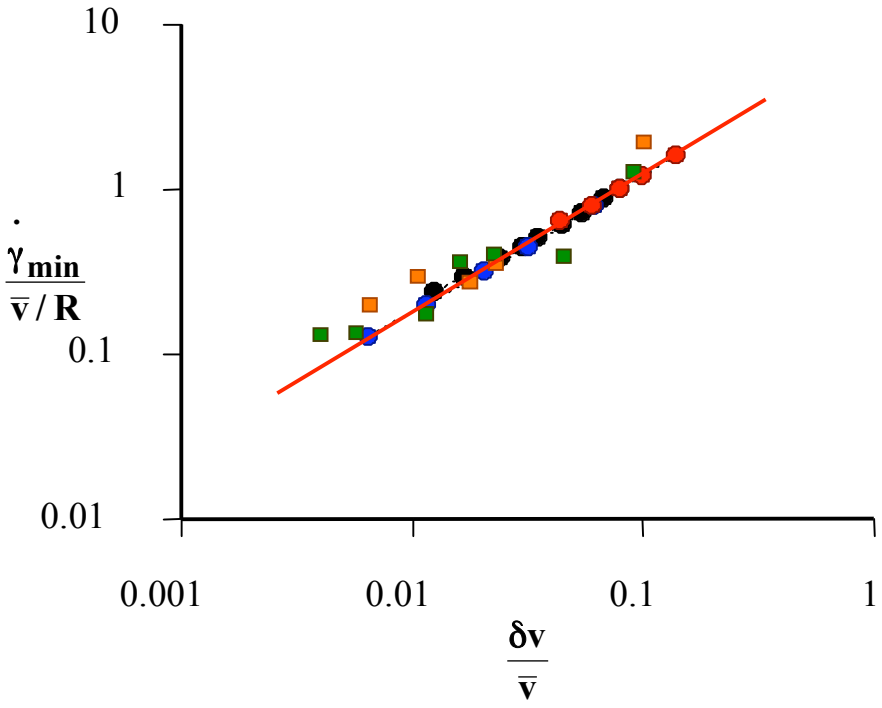

Figure 9. Dimensionless minimum shear rate at which accurate viscosity data can be obtained as a function of the velocity resolution.

represented by v with an overbar, and the pipe radius, R. The velocity resolution is also nondimensionalized by the average velocity. The correlation shown in Figure 9 is for all of the data obtained using MRI and UPDV. This represents a correlation for obtaining data that fall within 5% of benchmark data obtained by conventional rheological techniques. With the correlation in Figure 9, it is possible to design a system for determining the viscosity using either UPDV or MRI *a priori*.

These techniques comprise a set of tools that can be applied to remotely and non-invasively determine the viscosity of complex materials that include particle suspensions that are opaque. The sole limitations to these techniques are: the ability to pump the materials, appropriate choice of δv and choosing the proper pressure transducers. This work has definitively shown that this technique can be applied to predict phenomena such as plugging and can also potentially be designed to study chemical characteristics (MRI) and detect large particles that may cause processing problems (MRI & UPDV).

In addition to this experimental work, which comprised most of the studies, we developed models for multiphase systems. This work has shown the effect of sizes and size distribution on the rheology of spherical particle suspensions. We have also provide a review of the rheology of concentrated slurries and are currently pursuing a new approach to modeling such systems that promises to allow us to make calculations in complex and time dependent flows.

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6. Pointwise Observations for Rheological Characterization, 70th Annual Meeting of the Society of Rheology, October, 1998, Monterey, CA, with D. F. Arola, G. A. Barrall, A. Shekarriz, M. J. McCarthy.

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