ANL/ET/CP-94482
CONF-970962--

AN ULTRASONIC INSTRUMENT FOR MEASURING DENSITY AND VISCOSITY OF TANK WASTE

S. H. Sheen, H. T. Chien, and A. C. Raptis

MASTER

Energy Technology Division
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

INTRODUCTION

An estimated 381,000 m^3/1.1 x 10^9 Ci of radioactive waste are stored in high-level waste tanks at the Hanford, Savannah River, Idaho National Engineering and Environmental Laboratory, and West Valley facilities. This nuclear waste has created one of the most complex waste management and cleanup problems that face the United States. Release of radioactive materials into the environment from underground waste tanks requires immediate cleanup and waste retrieval. Hydraulic mobilization with mixer pumps will be used to retrieve waste slurries and salt cakes from storage tanks. To ensure that transport lines in the hydraulic system will not become plugged, the physical properties of the slurries must be monitored. Characterization of a slurry flow requires reliable measurement of slurry density, mass flow, viscosity, and volume percent of solids. Such measurements are preferably made with on-line nonintrusive sensors that can provide continuous real-time monitoring. With the support of the U.S. Department of Energy (DOE) Office of Environmental Management (EM-50), Argonne National Laboratory (ANL) is developing an ultrasonic instrument for in-line monitoring of physical properties of radioactive tank waste.

The instrument is based on a patented ANL ultrasonic viscometer [1] that measures both density and viscosity. But while the ANL viscometer has been demonstrated in the high-viscosity range (>100 cP), it lacks sensitivity in the low-viscosity range. Because the typical viscosity of tank waste is <30 cP, the focus of the developmental effort describe here is on improving the ability of the ANL viscometer to measure low-viscosity. Sensitivity of the ANL viscometer depends on the shear impedance of the transducer wedge; thus, the initial effort is to evaluate various wedge materials. In this paper, we summarize this evaluation and propose an optimal wedge design. The baseline technology of the viscometer and its limitations are described, and results of laboratory calibration tests are presented.

THE ANL ULTRASONIC VISCOMETER

ANL's ultrasonic viscometer, a nonintrusive in-line device that measures both fluid density and viscosity, is based on longitudinal- and shear- impedance measurements. The technique of measuring liquid shear impedance was first applied by Moore and McSkimin [2]. Figure 1 shows the basic design of the ultrasonic viscometer and its signal processing scheme. Two transducer wedges are mounted on a pipe, opposite one another and flush with the inner surface of the pipe. Each wedge shown in Fig. 2 uses an offset surface to provide

DISCLAIMER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

coefficient, from which the product of density and viscosity is deduced. The operating frequencies are 5 MHz for shear and 1 MHz for longitudinal operation. Oblique incidence was commonly used because of better sensitivity, but mode-converted waves often occur in wedges that do not exhibit perfect crystal structure and well-polished surfaces. Therefore, for practical applications, we use normal-incidence.

Longitudinal Waves and Acoustic Impedance of Fluid

Acoustic impedance Z_l of a fluid is the product of fluid density ρ and phase velocity V of sound in the fluid; it can be determined by measuring the reflection coefficient R at the boundary of the fluid and the transducer wedge. If we select the normal incidence configuration, R is given as

$$R = \frac{Z_l - Z_w}{Z_l + Z_w} \quad , \tag{1}$$

where Z_W is the acoustic impedance of the wedge in which longitudinal waves propagate from the transducer to the fluid. If the phase velocity in the fluid can be determined accurately from other measurements (such as time-of-flight of longitudinal waves traveling in the fluid), the fluid density can be derived from

$$\rho = \frac{Z_w(1-|R|)}{V(1+|R|)} \quad , \tag{2}$$

where the absolute value of the reflection coefficient is used because, in principle, R is a complex number. However, in practice, if we assume that wave attenuation in the wedge and fluid can be neglected, only the real parts of R and Z_W are used in the density calculation.

Shear Waves and Shear Impedance of Fluid

Use of the ultrasonic shear reflectance method to obtain the shear mechanical properties of fluids has been the subject of many studies of Newtonian [3] and non-Newtonian [4] fluids. Consider the case where gated SH plane waves propagate in a wedge at an incident angle that is normal to the polished surface in contact with the fluid and are reflected back. The shear reflection coefficient can be expressed by Eq. 1, with shear impedances replacing acoustic impedances. The shear impedances of the wedge, Z_{WS}, and fluid, Z_{lS}, are given by the equations

$$Z_{ws} = \sqrt{\rho_w C_{44}} \tag{3}$$

and

$$Z_{ls} = \sqrt{j\omega\rho\eta} \, , \tag{4}$$

where ρ_W is the density of the wedge material, C_{44} is the stiffness constant of the wedge, ω is the radial frequency of the shear wave, and η is the fluid viscosity. By using Eq. 4, we assume that the fluid behaves as a Newtonian fluid; more complex expressions are expected for non-Newtonian fluids [5]. The shear impedance of a fluid is a complex value that consists of amplitude and phase. Because the phase change is very small for a single reflection, we consider only the amplitude variation. The shear reflection coefficient, R_S, which is a measurable quantity, can be used to calculate the product of the density and viscosity as follows:

$$\sqrt{\rho\eta} = \sqrt{\frac{\rho_w C_{44}}{2\omega}} \frac{1-\sqrt{1-2K^2}}{K} \quad , \tag{5}$$

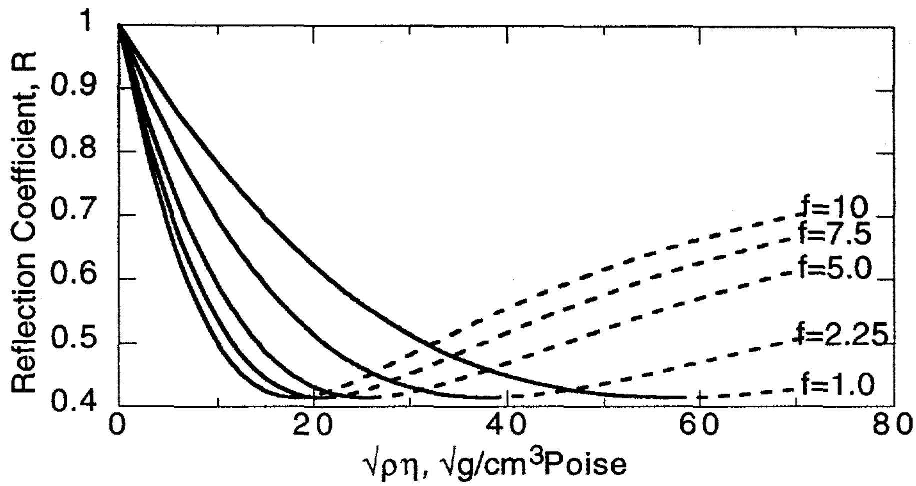

Figure 3. Reflection coefficient as a function of fluid density-viscosity product for various operating shear wave frequencies.

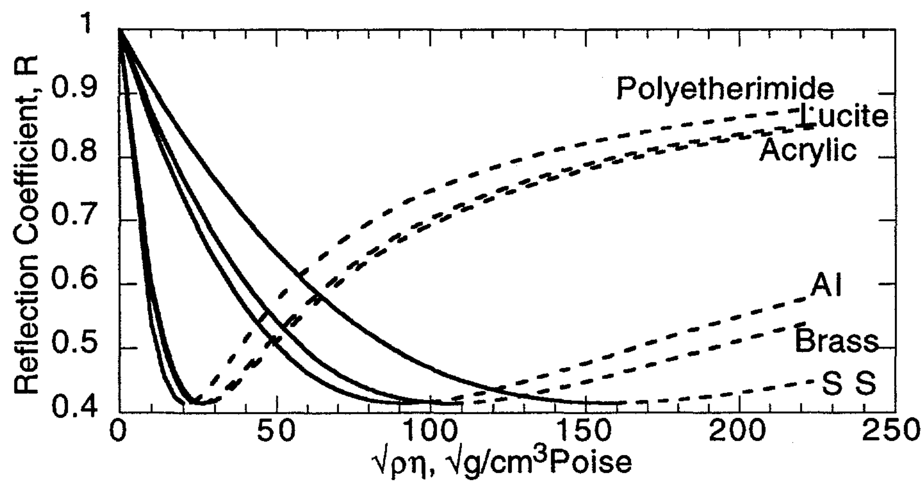

Figure 4. Reflection coefficient vs. square root of fluid density-viscosity product for various wedge materials (SS = stainless steel).

where $K = \dfrac{1-|R_s|^2}{1+|R_s|^2}$.

Equation 5 predicts the measurement sensitivity and range of the shear reflectance method. Figures 3 and 4 show the dependence of the reflection coefficient on the product of density and viscosity for various operating shear frequencies and wedge materials, respectively. In principle, lower shear-impedance materials and higher operating shear frequencies provide better sensitivity but a smaller measurement range. However, for tank-waste applications, the choice of Lucite and 10 MHz is not sufficient to achieve the desired sensitivity.

LABORATORY TESTS AND RESULTS

The feasibility of this technology has been demonstrated and reported [6]. The primary effort of this program is to evaluate wedge materials and determine the sensitivity and accuracy of the wedges in measuring density and viscosity. Table 1 lists the tested wedge materials and their acoustic properties. For the tests, transducers (longitudinal and shear)

Table 2. Liquids used for density calibration tests.

Liquid[1]	Chemical Constituents	Density (g/cm^3)	Longitudinal-Wave Phase velocity (cm/μsec)
R-827	Kerosene Chloronaphthlene Napthol	0.818	0.12766
G-1000	2-Butoxy Ethanol 51.9% Ethylene Glycol 47.2% BASACID Green <1%	1.002	0.15906
Y-120	Chloronaphthlene 99% Kerosene <1% Mono Azo Dye <1%	1.194	0.14272
B-175	Diazene-42 99% Diazene-200 <1% Solvent Blue 36 <1%	1.730	0.11452

[1]Supplied by ALTA Robbins, Anaheim, CA.

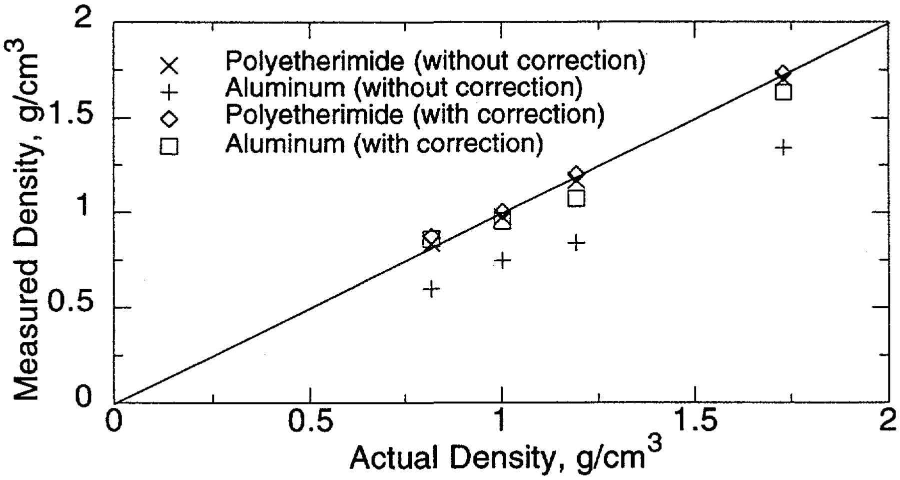

Figure 6. Density calibration results for polyetherimide and aluminum. A wedge correction factor of 4% was applied.

change; thus, phase velocity alone cannot be used to predict liquid density. However, by combining phase velocity and acoustic impedance measurements, we can obtain an accurate measurement of liquid density. Figure 6 shows the density calibration results for polyetherimide and aluminum wedges. The polyetherimide wedge gives an accuracy better than 0.5% for the test liquids, but results from the aluminum wedge are significantly lower than the actual values. The discrepancy in the results obtained for the aluminum wedge may be due to wetting problems and wedge geometry, which consistently give a reflection coefficient measurement that is 4% higher than the actual value. If we apply the 4% correction to both wedges, which are of the same design, the discrepancy is significantly reduced, as shown in Fig. 6.

Viscosity Measurement

When compared with the density wedge, the wedge designed to measure viscosity is scaled down in a ratio of longitudinal-to-shear velocities. Earlier measurements [6] show that an aluminum wedge gives a change of ≈1% in reflection amplitude for a 250-cP viscosity change. This sensitivity is very poor, especially for low-viscosity measurements, but it can be improved if a low-impedance wedge and high operating frequency are used. Figure 7

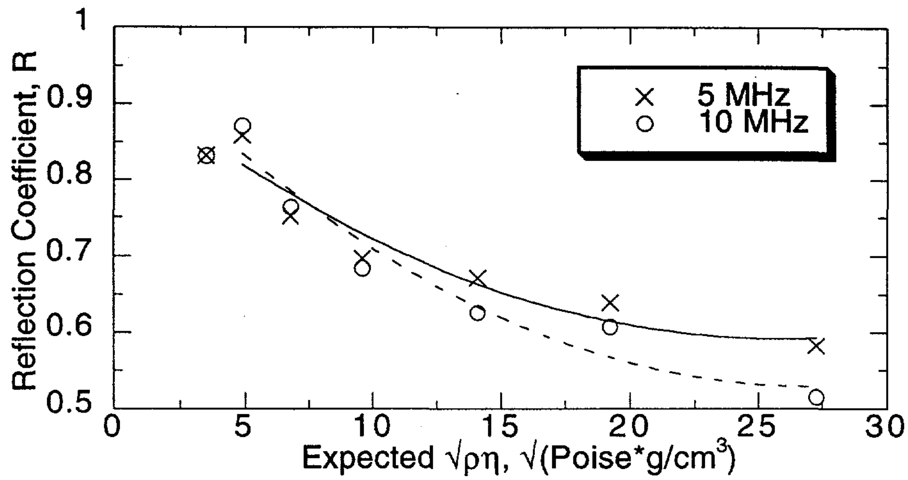

Figure 7. Measured reflection coefficients with polyetherimide wedge at two operating frequencies.

Table 3. Liquids used for viscosity calibration tests.

Liquid[1]	Chemical Constituent	Density (g/cm³)	Viscosity (cP)
N600	Mineral oil	0.8876	1381
N1000	PAO oil	0.8479	2823
N2000	Poly(1-butene)	0.8753	5248
N4000	Poly(1-butene)	0.8812	10450
N8000	Poly(1-butene)	0.8873	22390
N15000	Poly(1-butene)	0.8919	41360
N30000	Poly(1-butene)	0.8954	83040

[1]Supplied by Cannon Instrument Company, State College, PA.

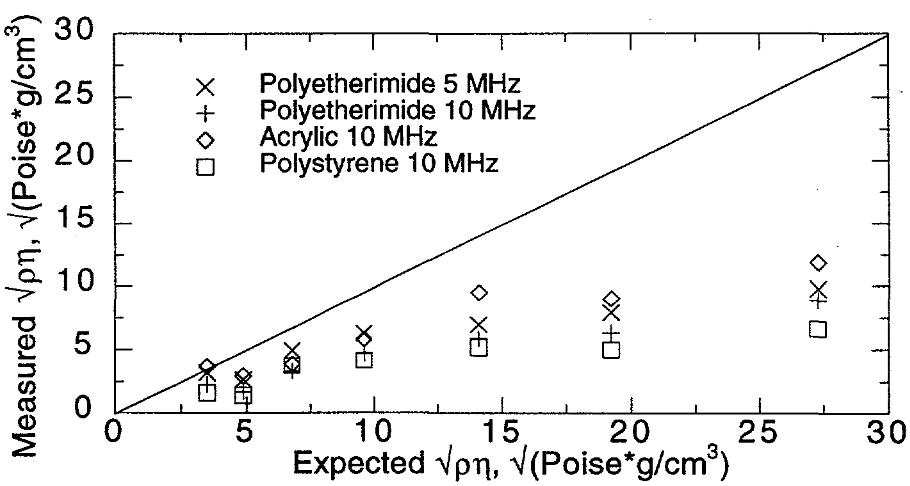

Figure 8. Viscosity calibration data for various wedge materials.

shows the reflection coefficients as a function of viscosity for the polyetherimide wedge at two frequencies. In the high-viscosity range, better sensitivity is obtained at the higher frequency (10 MHz). We performed the calibration tests with three low-impedance wedge materials, polyetherimide, acrylic (Lucite), and polystyrene. Table 3 lists the liquids used for the calibration tests; note that the density of all of the liquids is similar but the viscosity varies. Figure 8 shows the calibration results. Lucite is the best of the three as a wedge

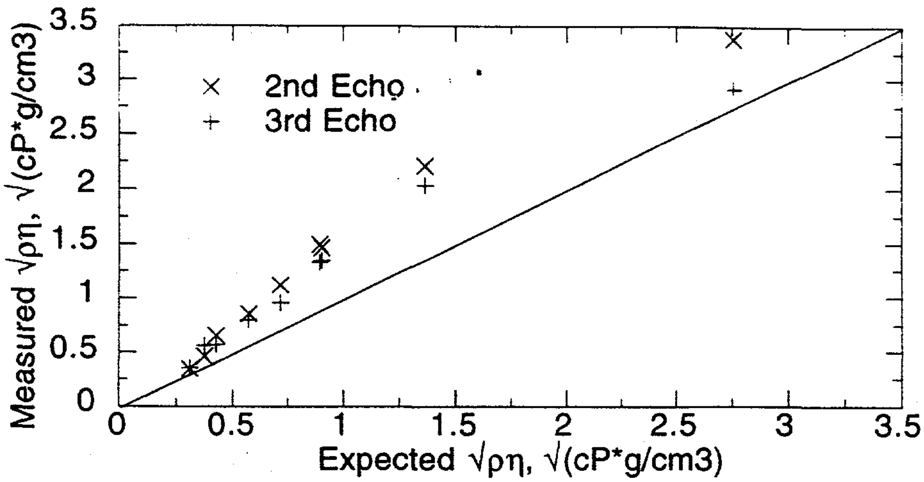

Figure 9. Viscosity calibration data in low-viscosity range, obtained with polyetherimide wedge and multiple reflection technique.

material for viscosity measurement. However, all of the measured viscosities are lower than their expected values. The discrepancy may be attributed to non-Newtonian fluid behavior [7], surface wetting, and poor sensitivity.

For low-viscosity liquids, we must improve the detection sensitivity of the technique based on measurement of impedance (or reflection coefficient). One approach is to monitor multiple reflections, because each echo represents one interaction at the wedge/liquid boundary. To obtain multiple echoes, the wedge design must be modified. Two design factors must be considered, echo interference and signal attenuation. Because the simplest design is to use a thin-plate configuration, we fabricated a thin polyetherimide plate and tested it with glycerol-water solutions. In Fig. 9, we show the measured results derived from the second and third echoes over a viscosity range of 1 to ≈600cP. The derived viscosities in Fig. 9 are calculated from the measured reflection coefficients by solving Eq. 5, in which R_S is replaced by $(R_S)^{1/n}$, where n is the echo number. It is evident that multiple reflections improve measurement sensitivity; thus, low-viscosity liquids can be monitored with this technique.

CONCLUSIONS

In this paper, we describe a nonintrusive in-line ultrasonic instrument for measuring liquid density and viscosity. The instrument is based on a patented ANL ultrasonic viscometer that derives liquid density and viscosity by measuring the acoustic impedances of the liquid and the speed of sound in the liquid. The liquid density is determined from the sound velocity and the longitudinal acoustic impedance. The liquid viscosity is determined from the measured density and the shear impedance of the liquid. We developed a thin-wedge design that allows one to use higher order reflections to determine the shear-wave reflection coefficient and thus improve the sensitivity in the low-viscosity range. The instrument will be used to monitor nuclear waste in a typical nuclear-waste transport line. However, many practical questions require further investigation before the instrument can be put into service. For example, how do temperature variation, presence of solids, and flow rate affect the measurements ?

ACKNOWLEDGMENT

Work supported by U. S. Department of Energy, Office of Environmental Management Technology Development, Characterization, Monitoring, and Sensor Technology Crosscutting Program.

REFERENCES

1. S. H. Sheen, W. P. Lawrence, H. T. Chien, and A. C. Raptis, *Method for Measuring Liquid Viscosity and Ultrasonic Viscometer*, U. S. Patent 5,365,778 (1994).
2. R. S. Moore and H. J. McSkimin, Dynamic shear properties of solvents and polystyrene solutions from 20 to 300 mhz, in: *Physical Acoustics*, W. P. Mason and R. N. Thurston, ed., VI:167 (1970).
3. F. Cohen-Tenoudji, et al., A Shear Wave Rheology Sensor," IEEE Trans. on Ultrasonics, Ferroelectrics, and Frequency Control, IEEE-UFFC, 34(2):263 (1987).
4. G. Harrison and A. J. Barlow, Dynamic Viscosity Measurement, *Meth. of Exp. Phy.*, 19:137 (1981).
5. S. H. Sheen and A. C. Raptis, A Feasibility Study on Advanced Techniques for On-line Monitoring of Coal Slurry Viscosity, *Argonne National Laboratory Report*, ANL/FE-87-8 (1987).
6. S. H. Sheen, H. T. Chien, and A. C. Raptis, An In-line Ultrasonic Viscometer, *Review of Prog. in QNDE*, 14A:1151 (1995).
7. S. H. Sheen, H. T. Chien, and A. C. Raptis, Measurement of Shear Impedances of Viscoelastic Fluids, *1996 IEEE Int'l Ultrasonics Symp.*, to be published in 1997.