# Oil Viscosity Measurement by Ultrasonic Reflectance

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**ABSTRACT:** Pulses of shear-mode ultrasound (center frequency 10 MHz) are reflected from the surface of a series of vegetable and synthetic high-viscosity calibration oils at a range of temperatures (5–50°C). For all samples and temperatures there is a single negative correlation between the magnitude of the echo from the interface between the delay-line and the sample and the viscosity of the sample. Similar experiments with longitudinal ultrasonic waves show the amount of sound reflected decreased with increasing viscosity, but there is no single correlation for all samples.

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**KEY WORDS:** Longitudinal waves, oils, reflection coefficient, sensors, shear waves, ultrasound, viscosity.

Ultrasound is qualitatively similar to audible sound, but the vibrations occur at much higher frequencies (>20 kHz). Lowpowered ultrasound is ideal for many sensing applications as the material oscillations that constitute the wave are dependent on but nondestructive of the bonds of the material through which it passes (1). In most ultrasonic applications the material vibrations occur as a series of compressions and expansions in the direction of sound propagation (longitudinal ultrasound), but it is also possible to develop oscillations normal to the direction of wave motion (shear ultrasound) (1-3). The fundamental linear ultrasonic properties of a material are velocity and attenuation, which are related to the real and imaginary parts of the complex propagation coefficient:  $k = \omega/c + i\alpha$ , where c is the ultrasonic velocity,  $\omega$  the angular frequency  $(=2\pi f)$ , f is frequency,  $i = \sqrt{-1}$ , and  $\alpha$  is the attenuation coefficient (1,3). The ultrasonic parameter k is related to material properties *via* the following equation:

$$\left(\frac{k}{\omega}\right)^2 = \frac{\rho}{E}$$
[1]

where  $\rho$  is the density and *E* is the adiabatic elastic modulus of the material (1,3). For longitudinal waves, *E* is equivalent to K + 4/3G for a fluid (*K* is the bulk modulus, and *G* is the shear modulus). This relationship gives longitudinal ultrasonic measurements some sensitivity to material shear properties, but as typically K >> G, they are hard to measure. Measurement of the speed of longitudinal sound waves has been used to investigate the composition, solid fat content, and quality of oils and fats (4,5). For shear waves, E is equal to G in Equation 1, so the relationship between ultrasonic and material shear properties is direct. The frequency-independent form of Equation 1 for shear waves is:

$$c^2 = \frac{G}{\rho}$$
[2]

It should be possible to use Equation 2 to relate measured fluid properties (density and speed of shear waves) to calculate a unique value for the shear modulus. Unfortunately, shear ultrasound is so strongly attenuated in liquid media that it is impossible to transmit a wave finite distance for a velocity measurement. One solution to this problem is to generate a shear ultrasonic wave in a solid, then measure the proportion reflected from the interface between the solid and the material under investigation. The amount of sound reflected is a measure of the shear-acoustic properties of the liquid and should in principle relate to its shear-rheological properties. Variations of this approach have been used intermittently over the past several decades (2,6-10) but its advantages, disadvantages, and potential are not widely understood.

### MATERIALS AND METHODS

*Materials*. A series of hydrocarbon oils (designated by the manufacturer as S20, S60, S200, S600, and S2000), designed for the calibration of rheological measurement equipment, were obtained from the Cannon Instrument Company (State College, PA). Vegetable oils (olive, corn, and cottonseed) were purchased from a local supermarket and used without further purification.

Ultrasonic measurements. Ultrasonic pulses were transmitted along a Plexiglas delay line, and the peak-to-peak amplitude of the signal reflecting from the sample surface was recorded. A more detailed description of the method is provided by Saggin and Coupland (11). Shear (10 MHz) and longitudinal (2.25 MHz) wave transducers were used in these analyses (Panametrics V221BA and V606, Waltham MA). The temperature of the samples was maintained with a computer-controlled circulator ( $\pm 0.01^{\circ}$ C). All samples were thermally equilibrated for at least 20 min prior to analysis. A typical measurement took less than 10 s to acquire.

The peak-to-peak amplitude of the returning echo depends on the magnitude of the generated pulse (a property of the apparatus) and the acoustic dissimilarity between sample and delay line (a property of the material under investigation). Because the magnitude of the generated pulse could not be re-

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lied upon as a constant on a day-to-day basis, the measured value was normalized to a similar measurement made against a calibration material (distilled water).

Oil viscosity was measured in a controlled strain rheometer (RFS II; Rheometrics, Piscataway, NJ), which was operated with concentric cylinder geometry (inner diameter 16.5 mm, outer diameter 17 mm).

## **RESULTS AND DISCUSSION**

The viscosity of all samples was independent of shear rate  $(0-1000 \text{ s}^{-1})$  at all temperatures studied (data not reported). This is characteristic of Newtonian fluids that can be described with a single viscosity. Consequently, the viscosity at 100 s<sup>-1</sup> was used exclusively in subsequent analyses. Viscosity always decreased in an apparently Arhennius manner with temperature  $(5-50^{\circ}C)$ , as is typical of many oils (12). When normalized shear ultrasonic reflectance is plotted against viscosity for all samples, temperatures, and oil types (Fig. 1), we see a single monotonic decrease in acoustic signal with increasing viscosity. Despite clear differences in chemical composition between and within the groups of lubricating and vegetable oils, there is a single relationship between the ultrasonic and viscous properties of all samples. From this observation we conclude that shear ultrasonic reflectance is sensitive only to oil viscosity and not to composition. With this master curve, it should be possible to relate an ultrasonic measurement with oil viscosity, regardless of the sample's composition and temperature.

The amount of sound returning from a partial reflection depends on the impedance mismatch (acoustic dissimilarity) between the two materials that constitute the interface. Explicitly, when a wave traveling through medium 1 is normally incident upon a plane interface with medium 2, some of the sound energy is reflected and some transmitted. The proportion of energy reflected is the reflection coefficient R and is related to the impedance (z) of the sample as (1,3,11):

$$R = \frac{A_r}{A_i} = \frac{|z_d - z_s|}{|z_d + z_s|}$$
[3]

 $A_r$  and  $A_i$  are the amplitudes of the reflected and incident sound, respectively, and the subscripts *s* and *d* refer to the sample and delay line. Assuming frequency independence, the ultrasonic impedance is the product of sample density and speed of sound (1,3). In Equations 2 and 3 we have established a relationship between a measured parameter, *R*, and a material parameter, *G*. This relationship is the basis for the viscosity dependence seen in Figure 1. In qualitative terms, increasing the viscosity makes the sample more similar to the delay line and therefore less sound is reflected at the interface. It is notable that a 100 fold increase in viscosity causes only a 3% decrease in ultrasonic signal (Fig. 1), which may limit the precision of the technique as a sensor.

The fact that shear-mode waves are essential for viscosity measurement is demonstrated by repeating the ultrasonic reflectance measurements with a longitudinal wave transducer.



**FIG. 1.** Normalized shear reflectance vs. viscosity for all oils (the shear reflectance of a sample was normalized to the shear reflectance of water at the same temperature): S20 ( $\bullet$ ), Corn (X), Cottonseed (+), Olive (\*), S60 ( $\bigcirc$ ), S200 ( $\bigtriangledown$ ), S600 ( $\bigtriangledown$ ), S2000 ( $\blacksquare$ ).

The normalized longitudinal reflectance is plotted against sample viscosity for all oil types in Figure 2. The ultrasonic signal decreased with increasing sample viscosity and decreasing temperature, but there was no single relationship encompassing all samples. The changing response is probably due to changes in sample density or bulk modulus (Eq. 1) with temperature. For a given sample the density (or bulk modulus) may vary with viscosity, but there is no direct mechanistic basis for a general relationship and hence the series of sample-specific relations shown in Figure 2.

The reflectance of shear ultrasonic waves at the surface of a range of oil samples is directly sensitive to bulk viscosity and not to temperature or composition. Despite its obvious potential, this method has not found wide application in prac-



**FIG. 2.** Normalized longitudinal reflectance against viscosity for all oils (the longitudinal reflectance of a sample was normalized to the longitudinal reflectance of water at the same temperature): S20 ( $\bullet$ ), S60 ( $\bigcirc$ ), S200 ( $\bigtriangledown$ ), S600 ( $\bigtriangledown$ ).

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tical fluids characterization either as an on-line sensor or a laboratory instrument. One possible reason may be that the shear wave only propagates about one-half wavelength into a material on reflection at its surface (~1 mm). Therefore, if the near-surface properties of the liquid are not representative of the bulk then the measurement will not give useful information. A second limitation may be that the frequencies at which commercial shear transducers operate (~MHz) are much faster than most practical deformations involving the fluid. Many materials undergo a series of relaxations and can show different behaviors at different frequencies. Therefore, the properties measured by the shear ultrasound, whilst characteristic of the material structure, may be different from the properties observed in practical use. However, at least in this case, the correlation shown in Figure 1 demonstrates the potential of shear ultrasonic reflection measurement of oil viscosity.

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#### REFERENCES

1. McClements, D.J., Ultrasonic Characterization of Foods and Drinks: Principles, Methods, and Applications, *Crit. Rev. Food Sci. Nutr.* 37:1–46 (1997).

- Sheen, S., H. Chien, and A. Raptis, An In-line Ultrasonic Viscometer, *Rev. Prog. Quant. Nondestructive Eval.* 14A:1151–1158 (1995).
- 3. Blitz, J., *Fundamentals of Ultrasonics*, Butterworth & Co., London, 1963.
- McClements, D.J., and M.J.W. Povey, Ultrasonic Velocity Measurements in Some Liquid Triglycerides and Vegetable Oils, J. Am. Oil Chem. Soc. 65:1787–1790 (1988).
- 5. McClements, D.J., and M.J.W. Povey, Ultrasonic Analysis of Edible Fats and Oils, *Ultrasonics 30*:383–387 (1992).
- Mason, W., W. Baker, H. McSkimin, and J. Heiss, Mechanical Properties of Long Chain Molecule Liquids at Ultrasonic Frequencies, *Phys. Rev.* 73:1074–1091 (1948).
- Piccirelli, R., and T. Litovitz, Ultrasonic Shear and Compressional Relaxation in Liquid Glycerol, J. Acoust. Soc. Am. 29:1009–1020 (1957).
- Harrison, G., and A. Barlow, Dynamic Viscosity Measurements, *Meth. Expt. Phys.* 19:137–178 (1981).
- Sheen, S.H., H.T. Chien, and A. Raptis, *Measurement of Shear Impedances of Viscoelastic Fluids*, IEEE Ultrason. Symp. Proc.: 453–457 (1996).
- Kulmyrzaev, A., and D.J. McClements, High Frequency Dynamic Shear Rheology of Honey, *J. Food Eng.* 45:219–224 (2000).
- 11. Saggin, R., and J.N. Coupland, Concentration Measurement by Acoustic Reflectance, *J. Food Sci.*; in press (2001).
- Coupland, J.N., and D.J. McClements, Physical Properties of Liquid Edible Oils, J. Am. Oil Chem. Soc. 74:1559–1564 (1997).

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