

Photoacoustic measurement of liquid viscosity

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(Received 20 January 2010; accepted 28 April 2010; published online 24 May 2010)

In this letter, we report on the use of photoacoustic method to measure the viscosity of viscous liquids. The theoretical and experimental study was performed on the influence of viscosity effects on photoacoustic generation. We provide evidence that the frequency spectrum of photoacoustic signal is precisely related to the viscosity. Measurements are validated on different water–glycerol mixtures. Good agreement between theoretical and experimental results is obtained. This present method provides an insight into *in situ* viscosity measurements, which has potential for noninvasive detection of blood viscosity. © 2010 American Institute of Physics. [doi:10.1063/1.3435462]

The viscosity of liquid is one of the most important thermophysical properties in medical and industrial fields. The use of viscosity measurements for testing oils, paints, and various substances of technical interest has been given rise to a series of investigations. The correlation between viscosity of marrow fluid, blood, and diseases has also been reported. Consequently, measuring the viscosity of body fluid might serve as a useful diagnostic tool. Several different methods have been developed for the determination of liquid viscosity.^{1–3} However, these viscosity measurement techniques require both large sample volumes and complex calculations.

In this letter, we report a method for *in situ* measurement of liquid viscosity by photoacoustic (PA) effect.^{4–7} Viscosity provides the damping force to restore a dynamic system to equilibrium, hence, the PA wave not only carries information regarding the absorbed laser energy⁸ but also contains details associated with the thermal properties of the sample. In this study, viscous influences were taken into account through a damping term in the differential equation, which enables the PA technique to indirectly monitor viscosity.

For certain materials irradiated with laser pulses, a small time-dependent temperature variation can be observed, the acoustic wave is simultaneously excited by thermoelastic expansion.⁹ Under the conditions that the sample is optically opaque and thermally thick, the Navier–Stokes equation can be written as^{10–13}

$$\rho \frac{\partial u}{\partial t} + \rho(u \cdot \nabla)u = -\nabla P + \eta \nabla^2 u + (\xi + \eta/3) \nabla(\nabla \cdot u), \quad (1)$$

where ρ is the mass density, P is the PA pressure, u is the displacement of the particles in the medium, η is the shear viscosity, and ξ is the bulk viscosity. When the condition of stress confinement is met by laser irradiation, we can obtain the wave equation for the PA pressure¹⁴

$$\frac{1}{c_s^2} \frac{\partial^2 P}{\partial t^2} = \nabla^2 P + \frac{\xi + \frac{4}{3}\eta}{\rho_0 c_s^2} \frac{\partial}{\partial t} \nabla^2 P - \frac{\xi + \frac{4}{3}\eta}{\rho_0 c_s^2} \Gamma \nabla^2 H + \frac{\Gamma}{c_s^2} \frac{\partial H}{\partial t}, \quad (2)$$

where c_s is the speed of sound and c_p is the constant pressure heat capacity per unit mass. Here Γ is the Gruneisen constant

which is equal to $\beta c_s^2 / c_p$, where β means the thermal expansion coefficient. H is the heating function defined as the optical energy deposited per unit time and per unit volume, it can be expressed as $H(r, t) = I_0 A(r) f(t)$, where I_0 is a factor proportional to the incident optical energy density, $A(r)$ is the spatially distributed optical energy deposition, and $f(t)$ is the temporal irradiation of the laser that can be regarded as a Dirac delta function $\delta(t)$ for a short light pulse.

For the sake of simplicity, our present analysis is limited to the plane wave model. In such a case, the acoustic waves vary in time and with one of the three Cartesian coordinates, i.e., x , then the Laplacian operator takes the form $\nabla^2 = \partial^2 / \partial x^2$. Thus Eq. (2) can be simplified as follows:

$$\frac{\partial^2}{\partial t^2} P(x, t) = c_s^2 \frac{\partial^2}{\partial x^2} P(x, t) + \frac{\xi + \frac{4}{3}\eta}{\rho_0} \frac{\partial^3}{\partial t \partial x^2} P(x, t) + S, \quad (3)$$

where S is the source term which can be expressed by

$$S = \Gamma I_0 \left[A(x) \frac{\partial f(t)}{\partial t} - \frac{\xi + \frac{4}{3}\eta}{\rho_0} f(t) \frac{\partial^2}{\partial x^2} A(x) \right]. \quad (4)$$

Assume a PA pressure with the form of $P(x, t) = e^{i\alpha x} P(t)$ was generated at the at position x in a homogeneous medium, the variable of x can be eliminated and the second order differential equation with variable t was obtained. The source term can be first assumed to be $\delta(t)$, which, when integrated, yields the equation:

$$\frac{\partial^2}{\partial t^2} P(t) + a^2 c_s^2 P(t) + a^2 \frac{\xi + \frac{4}{3}\eta}{\rho_0} \frac{\partial}{\partial t} P(t) = \delta(t). \quad (5)$$

The Fourier transform of $\delta(t)$ and $P(t)$ is $\bar{\delta}(\omega')$ and $P(\omega')$, respectively. Replacing the variables with inverse Fourier transform yielding the frequency domain expressions of PA pressure

$$P(\omega') \left[-\omega'^2 - i \frac{\xi + \frac{4}{3}\eta}{\rho_0} a^2 \omega' + a^2 c_s^2 \right] = \frac{1}{2\pi}. \quad (6)$$

Two poles of the equation can be obtained after a Fourier inversion of both sides, and the time domain solution can be obtained by the residue theorem. For optical pulses with an intensity of the form $f(t)$, the PA pressure can be rewritten as a convolution between the source term S and the acoustic wave form that is excited by an infinitely short pulse $\delta(t)$.

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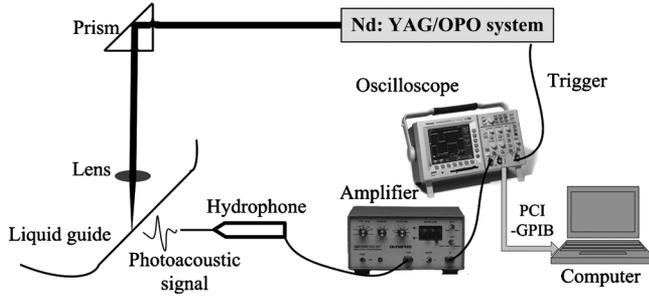


FIG. 1. Diagram of the experimental setup for PA measurement of viscosity. The PA coupling medium is water.

Thus, the following PA expression can be derived:

$$P(x,t) = \frac{1}{ac_s \sqrt{4 - a^2 l_v^2}} e^{i\alpha x} e^{-At} \sin(\omega t - \alpha x) S, \quad (7)$$

where $A = \alpha^2 c_s l_v$ is the attenuation coefficient, $\omega = 2ac_s \sqrt{1 - a^2 (l_v/2)^2}$ is the angular frequency, and $l_v = (\xi + 4\eta/3)/\rho c_s$ is the viscosity length. In the letter, we further assume that the absorber is a point source, and then the PA pressure can be simply expressed as

$$P(t) = \frac{\Gamma I_0}{ac_s \sqrt{4 - a^2 l_v^2}} e^{-At} \sin[2ac_s \sqrt{1 - a^2 (l_v/2)^2} \times t] \times \left[A(x) \frac{\partial f(t)}{\partial t} - c_s l_v f(t) \frac{\partial^2}{\partial x^2} A(x) \right]. \quad (8)$$

The PA frequency spectrum will have a reverse shift with viscosity increase, the PA signal amplitude is also inversely proportional to the coefficient of viscosity, as predicted by the formula.

The excited PA signal has a wide frequency range, while the viscosity will result in loss of high-frequency components. For liquid with different viscosity coefficient, the following equation can be given

$$\frac{(k_n^2 - k_1^2)}{l_{v1}^2 - l_{v_n}^2} = a^4, \quad \text{and} \quad k = \frac{\omega}{c_s}, \quad (9)$$

where k is the wave vector which denotes the wave number per unit length, the subscript 1 and n stands for the marker of samples with different viscosity. In the following, the change in half-power point width was used to characterize the frequency shift. The variable parameter of a can be defined by measuring liquid with a known viscosity, and the parameter of k_n can be obtained from fast Fourier transforms (FFTs) of the detected PA signals. Hence, the actual viscosity coefficient can be calculated by the following equation:

$$l_{v_n}^2 = l_{v1}^2 + (k_1^2 - k_n^2)/a^4, \quad (10)$$

where l_{v1} and l_{v_n} are the viscosity length of liquids with different viscosities. For simplicity, the variable of $\xi + (4/3)\eta$ was replaced with μ , hence the viscosity coefficient can be calculated by $\mu_n = l_{v_n} \rho c_s$.

The experimental setup is shown in Fig. 1. A Nd:YAG laser (LS-2134, LOTIS TH) operating at 532 nm with a pulse duration of 10 ns was used for optical illumination. The pulse repetition rate was 15 Hz. A silicone tube with an internal diameter of 0.5 mm was used to hold the liquid of interest. The laser beam was focused by a focusing lens and irradiated onto the sample. A needle hydrophone (HPM05/3,

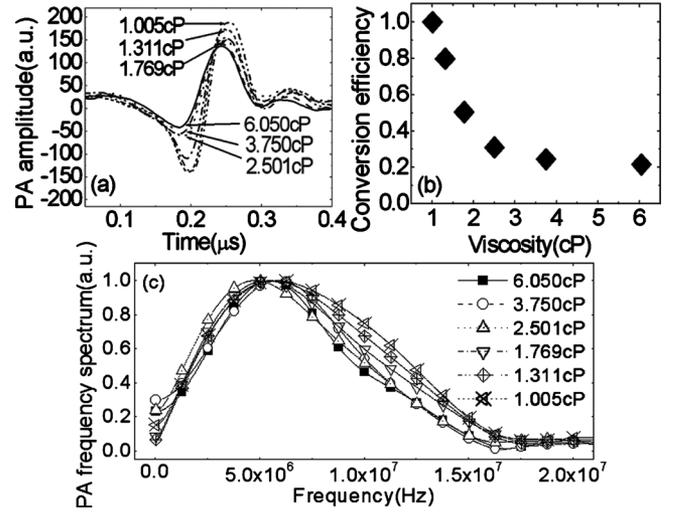


FIG. 2. Time-domain PA signals (a) and corresponding normalized frequency spectrum (c) for glycerin-water solution with different viscosity coefficients. Figure (b) shows a change in normalized PA conversion efficiency depending on the viscosity.

Precision Acoustics Ltd.) with a flat spectrum from 200 KHz to 15 MHz was used for PA signal detection, which is then amplified and recorded with a digital oscilloscope (TDS3032, Tektronix) at a sampling rate of 500 Msamples/s. For each sampling, the signal was averaged 64 times to improve the signal-to-noise ratio.

To validate the proposed method for viscosity measurements, glycerol-water mixtures of varying concentrations were used as viscous model fluids. Viscosity values of the mixtures were taken from the CRC Handbook of Chemistry and Physics.¹⁵ The PA signal was excited from the illuminated point, traveled through inviscid coupling medium and then captured by the transducer. Measurements were all carried out under controlled temperature (20 ± 0.1 °C) conditions. The absorption spectrum was measured and found that the solutions give nearly the same results.

Figure 2 displays the time-domain and the normalized frequency spectrum of PA signals emitted from glycerol-water solutions. From this graph, the expected decreases in the amplitude of PA signals with increasingly viscous fluids are observed. The Gruneisen coefficient increases with increasing concentration of the solution,¹⁵ while the PA amplitude exhibits an opposite trend. The normalized PA conversion efficiency which has the form of $1/a\sqrt{4 - a^2 l_v^2}$ can be calculated from the relationship between PA amplitude and Gruneisen coefficient, the result is shown in the Fig. 2(b). The results indicate that the conversion efficiency decrease obviously with increasing viscosity while the change ratio becomes weaker. As shown in Eq. (8), the viscosity will bring down the PA generation efficiency due to the momentum is eventually dissipated to thermal energy. In perfect elastic medium, where only elastic deformation occurs, no mechanical energy dissipation in the process of thermal expansion, and higher PA conversion efficiency will be obtained.

Figure 2(c) shows the normalized FFT of PA signals corresponding to Fig. 2(a). The result is consistent with the basic theoretical prediction. The central frequency is 7 MHz, which exhibit a significant shift toward lower frequencies with increasing viscosity. The loss of high frequency components also leads to bandwidth reduction. In addition, with the increase in glycerin concentration, the shape of PA frequency

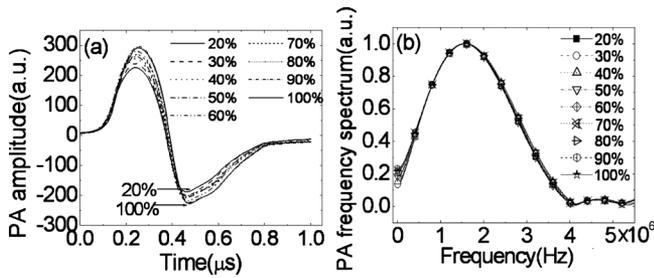


FIG. 3. The time-domain (a) and frequency spectrum (b) of PA signals that generated in ink solution with different absorption coefficients. The label in the figure shows the concentration of ink.

spectrum changes asymmetrically. The results indicate that liquids with higher viscosity will lead to higher attenuation of the sound energy, and show a loss of high-frequency features that cannot be ascribed to variations in the optical absorption coefficient, as will be shown next.

In the subsequent experiment, red ink was diluted to different concentrations to investigate the effect of absorption coefficient on PA signals. The ink diluted by water has scarcely any change in viscosity but a remarkable decrease on the absorption coefficient can be observed. The time-domain results of red ink with various absorption coefficients are shown in Fig. 3(a), and the corresponding frequency spectrum are given in Fig. 3(b). As shown in Fig. 3, the PA amplitude increases linearly with the absorption coefficients, whereas the dominant frequency and the bandwidth of the frequency spectrum keep constant. Previous researches have reported that the bandwidth increases with absorption coefficient¹⁶ but it is obvious only when the absorption coefficient had a difference of almost ten times. In current experiment, the maximum absorption difference is only 40%. Therefore, it was not surprising that the frequency spectrum kept the same with no obvious shift in the main frequency. The result indicated that the frequency shift in Fig. 2(c) is attributed to the effect of viscosity rather than the change in absorption coefficient.

The relationship between liquid viscosity and PA generation has been verified by theoretical and experimental research. The viscosity can be calculated from the PA frequency spectrum by Eqs. (9) and (10). Values obtained by the PA method are plotted against the literature value in Fig. 4. The obtained results were found to be in close agreement with the data obtained from the literature. The deviations between them might be attributed to the finite bandwidth of the transducer, the thermal conduction, and the irregular increase in relaxation time.

The accuracy of the measurement system is limited by the frequency spectrum precision, which is mainly affected by the response characteristic of the transducer. A higher response rate and more sensitive transducer will improve the performance. With a frequency resolution of 100 Hz which can be obtained in stable experiments, the viscosity resolution can thus be obtained to $\Delta\mu = \rho c_s \Delta(\omega/c_s)/a^2 \approx 3.624 \times 10^{-4}$ cP. The PA signal excited by amplitude-modulated continuous-wave laser has the same frequency as the modulation frequency. It is expected that the accuracy could be further improved by employing continuous-wave laser.

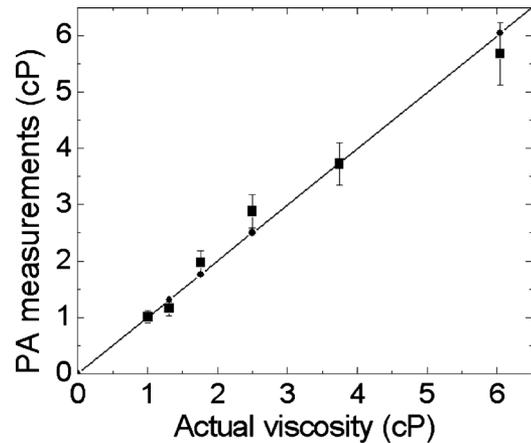


FIG. 4. The viscosity coefficient determined from the measured PA frequency (solid squares) against the actual viscosity coefficient (solid circles). The solid line represents the reference line and the error bars indicate the standard error on the mean.

In conclusion, we have predicted and verified that the amplitude and frequency spectrum of the PA wave is quite sensitive to liquid viscosity. The technique allows rapid, non-invasive, and *in situ* measurement of viscous liquids, which could be used for the viscosity measurement of human blood. It is also a potential alternative method for the routine determination of materials viscosity in chemical industry.

This research is supported by the National Basic Research Program of China (Grant No. 2010CB732602), the Program for Changjiang Scholars and Innovative Research Team in University (Grant No. IRT0829), the National Natural Science Foundation of China (Grant Nos. 30627003 and 30870676), and the Natural Science Foundation of Guangdong Province (Grant No. 7117865).

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