Noncontact measurement technique
for wide range of viscosity of µl-order liquid sample

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ABSTRACT

We have developed a new measurement technique for liquid viscosity involving the use of the pulse carbon dioxide laser (wavelength 10.6 µm, pulse width 50 ns, power 65 mJ) as a heating source. In the present method, interfering laser beams heat a liquid surface and generate a laser-induced capillary wave (LiCW: the wavelength can be adjusted from 11 to 200 µm) caused by a spatially sinusoidal temperature distribution. The temporal behaviour of LiCW is detected by monitoring the intensity of diffracted probe beam (He-Ne laser, 15 mW) at the heating area. The dynamics of LiCW provides information regarding thermophysical properties such as viscosity and surface tension. In this paper, we have (1) measured the viscosity of several liquid samples ranging 0.1~10000 mPa·s to verify the applicability of the present method, (2) evaluated the experimental minimum limit of the sample volume, and (3) measured the dynamic behavior of the viscosity as a function of time.

KEY WORDS: dynamic measurement, food manufacturing process, laser-induced capillary wave method, surface tension, viscosity, µl-order liquid sample.
1. INTRODUCTION

In many fields of science and industry, viscosity is one of the very fundamental and important thermophysical properties in order to analyze and control liquid flow. For example, in a food processing plant, the viscosity is essential factor in the control and design of such processing, and is also useful for evaluation of food quality. Additionally, in medical field, analyzing information of the blood viscosity is important to check the health condition. However, a noncontact measurement technique applicable to such fields where the viscosity drastically changes within a short period of time, and able to examine numerous samples at a time has not been available so far. In the present study, we have developed a new apparatus to detect a laser-induced capillary wave generated by the interference of pulsed heating laser beams. In the meantime, Terazima’s group performed the first experiment of capillary wave generated by the interference of pulsed heating laser beams [1]. In addition, they revealed the mechanism of the laser-induced capillary wave by solving the phenomenological equations and compared it with the observed signals. Coincidentally, the author’s group independently started to develop a noncontact measurement technique for wide range of viscosity by observing the laser-induced capillary wave [2]. One of the unique points of the present method is to employ the carbon dioxide laser as a heating source. By employing an infrared laser beam to excite the IR absorption band of liquid sample, the capillary wave can be induced without adding any light absorbing materials to pure water. We call this technique “laser-induced capillary wave method” and this method has the characteristics applicability to wide range of science and industry fields.

2. THE PRINCIPLE OF MEASUREMENT

2.1. The Summary of the Theory for Laser-induced Capillary Wave Method

The principle of laser-induced capillary wave technique is shown schematically in Fig. 1. In this method, pulsed high-power laser beams of equal wavelength and equal intensity intersect on a sample surface under a crossing angle $\theta$. They generate an optical
interference fringe pattern as illustrated in Fig. 1, whose intensity distribution is spatially sinusoidal. The laser intensity distribution and wave number can be written as

\[
I(x) = I_h \left\{ 1 + \cos(kx) \right\},
\]

\[
k = \frac{2\pi}{\Lambda} = \frac{4\pi \sin(\theta/2)}{\lambda_h},
\]

where \(I_h\) is the heating laser beam intensity, and \(\Lambda\) the fringe space, \(\lambda_h\) the wavelength of heating laser. At the end of heating, a corresponding spatially sinusoidal temperature distribution induced by the grating pattern is expressed as

\[
T(x) = T_m + \Delta T \cos(kx),
\]

where \(T_m\) is the mean initial temperature rise, and \(\Delta T\) is the amplitude of spatially periodic temperature distribution.

The temperature distribution of the thermal grating creates capillary wave on the liquid surface driven by the thermal expansion and the temperature dependence of the surface tension. The displacement of the capillary wave along \(z\) direction can be expressed by

\[
u_z = u_m + \Delta u \cos(kx),
\]

where \(u_m\) is the spatially uniform displacement and \(\Delta u\) is the amplitude of laser-induced capillary wave. When the probing laser beam is incident on the heating area, it is diffracted because the capillary wave acts like a reflection grating. The first-order diffraction efficiency is directly proportional to the square of the surface displacement as the following equation

\[
I(t) \propto \Delta u(t)^2,
\]

where \(I(t)\) is intensity of the first-order diffracted beam. Therefore, we are able to obtain the information regarding the behavior of the laser-induced capillary wave by monitoring the intensity of the first-order diffracted beam of the probing laser.

2.2. The Motion of Laser-induced Capillary Wave

The motion of laser-induced capillary wave is described by three phenomenological equations under appropriate boundary conditions. The displacement of the Newtonian liquid
surface has been reported by Yasumoto et al. [1] and is summarized as follows.

First, the Navier Stokes equation is

$$\frac{\partial^2 \mathbf{u}}{\partial t^2} - \nu \nabla^2 \frac{\partial \mathbf{u}}{\partial t} - V_L \nabla \cdot \mathbf{u} = -\beta V_L^2 \nabla T,$$

where \( \mathbf{u} \) is the displacement, \( \beta \) the thermal expansion coefficient, \( V_L \) the sound velocity in the liquid, and \( \nu = \eta / \rho \) the kinetic viscosity (\( \eta \), the viscosity, \( \rho \), the density of the medium).

Here, in order to justify the assumption that the laser-induced capillary wave is similar to the spontaneous capillary wave, we assume that the displacement of the capillary wave on the surface is small relative to the wavelength. In addition, the convective term is neglected. Second, the continuity equation is expressed as

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho \mathbf{V}) = 0,$$

(7)
where $V$ is the velocity of the liquid. Third, the heat conduction equation can be expressed by the following equation

$$
\rho C_p \left( \frac{\partial T}{\partial t} \right) - \lambda \nabla^2 T = I_x \alpha \left[ 1 + \cos(kx) \right] \exp(\alpha z) \delta(t),
$$

(8)

where $\lambda$ is the thermal conductivity, $\alpha^{-1}$ the optical absorption length of the heating beam, $C_p$ is the heat capacity at a constant pressure, and $\delta(t)$ is the Dirac delta function.

The boundary conditions are as follows, the velocity of liquid should be zero sufficiently far from the surface, and the thermal conduction from the liquid to the gas phase is neglected. In addition, the tangential, and normal components of the stress to the surface, assumed under the conditions that the stress acting from within the liquid is exactly counterbalanced by the stress due to the surface force since no stress can exist in the gas phase, is given by

$$
\sigma \frac{\partial u}{\partial x} - 2 \frac{\partial}{\partial t} \left( \frac{\partial u}{\partial z} \right) + \rho V^2 \alpha \frac{\partial T}{\partial x} - \frac{\partial u}{\partial x} - \frac{\partial u}{\partial z} = 0,
$$

(9)

$$
\eta \frac{\partial}{\partial t} \left( \frac{\partial u}{\partial z} + \frac{\partial u}{\partial x} \right) - \left( \frac{\partial \sigma}{\partial T} \right) \frac{\partial T}{\partial x} = 0.
$$

(10)

Where $\sigma$ is the surface tension. Especially, the temperature dependence of the surface tension is considered by using Eq. (10) in which the surface tension distribution term is included.

The amplitude of the laser-induced capillary wave can be obtained by solving Eqs. (4) and (6)-(10) using the Laplace transformation. The Laplace transformed amplitude of the wave $\Delta U$ can be described as a consequence:

$$
\Delta U = F \left( s, \eta, \sigma, \lambda, d\sigma/dT, a, k, V_0, \alpha, \beta, \rho \right).
$$

(11)

The time profile of the liquid motion is numerically calculated by using the fast inverse Laplace transform algorithm [3]. Moreover, viscosity of liquid sample can be obtained by analyzing the detected signal, which is directly proportional to the square of the surface displacement, using fast inverse Laplace transform algorithm and waveform fitting.
3. EXPERIMENTAL APPARATUS

Figure 2 exhibits the present experimental apparatus. The pulsed carbon dioxide laser (Edinburgh Instruments Ltd., MTL-3; wavelength 10.6 µm, pulse width 50 ns, output energy 65 mJ) is employed as a heating source. The repetition rates are available from single shot to 100 Hz. Especially, because of the short-pulsed laser beam, it is possible to neglect the complex behavior of laser induced capillary wave in the course of heating process. The pulsed carbon dioxide laser is led to an attenuable system (AS), which consists of a half wave plate (WP) and a thin film plate (TFP). The AS was employed in order to control the power of the heating laser without damaging the optical system. The attenuated heating beam is turned vertically up by mirror 1 (M1) and divided into two beams of equal intensity by means of a beam splitter (BS). They are intersected on the sample surface by refracting in M2, M3 and M4 to produce an interference pattern. These optical devices are fixed on the vertical optical bench and the diameter of the heating beams is approximately 6 mm on the sample surface. The fringe space of the grating can be adjusted from about 11 to 200 µm. The liquid sample is filled in a petri dish (depth 1.6 mm, diameter 10 cm). The probing laser is a He-Ne laser (wavelength 632.8 nm, output power 15 mW, beam diameter 2 mm). The diffracted light signal as a function of time is detected by a photomultiplier tube (PMT; Hamamatsu, R928) through a pinhole (PH) and interference filter (IF). The output signal is sent to a digital storage oscilloscope (DSO) and is transferred to a computer.

4. EXPERIMENTAL RESULTS AND DISCUSSION

4.1. Evaluation of the Principle of Laser-induced Capillary Wave Method

As the first step, in order to evaluate the principle of the present method, we have employed pure liquids such as water, toluene, JS100, and JS14000. JS100 and JS14000 are the standard liquids for calibrating viscometer certified by the National Institute of Advanced Industrial Science and Technology (AIST) in Japan.
Fig. 2  The experimental apparatus of laser induced capillary method by using the pulsed carbon dioxide laser.

Figures 3 and 4 show the detected signal of the present technique for toluene (the viscosity 0.5 mPa·s @ 298 K) and JS14000 (the viscosity 7080 mPa·s @ 298K) and their theoretical waveform. The measurements were conducted at room temperature and atmospheric pressure, and wavelength of the capillary wave of 50 µm. The detected signal for toluene agrees well with the theoretical calculation, which takes into account the effect of the surface tension distribution.

On the other hand, the detected signal is over-dumped, when we measure a high viscosity sample such as JS14000 under the same conditions as for toluene except for a wavelength of 60 µm. The detected signal for JS14000 is also in good agreement with the theory. These remarkably different waveforms can be explained by the difference of the viscosity, i.e., the viscosity of JS14000 is over $10^4$ times that of toluene.
Fig. 3 Comparison of the detected signal with the theoretical calculation for toluene (\( \Lambda = 50 \, \mu m \)) at room temperature [4].

Fig. 4 Comparison of the detected signal with the theoretical calculation for JS14000 (\( \Lambda = 60 \, \mu m \)) at room temperature [5,6].
4.2. Attenuable System

Fig. 5 (a) shows the detected signal for water under full power of heating laser (65 mJ). At the first stage of the study, the measurements were conducted under 65 mJ heating power, because the power of the carbon dioxide laser could not be adjusted due to its specification. Since, the optical absorption length of the heating beam for water is so short ($\alpha^{-1} = 14 \text{ } \mu m @ 10.6 \text{ } \mu m[7]$), the behavior of detected signal is considerably different compared to the theoretical waveform because of excessive heating of liquid surface. Then the attenuable system was added into experimental apparatus in order to continuously change the heating power. AS was employed in order to transmit desired heating power, and the rest of the heating power is lead to a shield plate of TFP reflection in order not to damage the optical system. Fig. 5 (b) shows the detected signal under the heating power at 36 mJ. The detected signal shows the specific oscillation-damping waveform, which is a characteristic waveform of low viscous liquid such as toluene. The results verify that this method was made applicable to various samples with different optical absorption length by employing the attenuable system.

![Fig. 5](image)

**Fig. 5** The detected signal for water under the different heating power (a) 65 mJ (b) 20 mJ.
4.3. Sample Volume

Above experimental results are detected under bulk volume of sample liquid (~100 ml). Considering the needs in medical field, the desired sample volume should be as small as possible. Thus we have evaluated the experimental minimum limit of the sample volume, considering the evaporation and surface tension. Fig. 6 shows the comparison of the detected signal for water for 100 ml bulk sample and for 75 µl sample, and it is verified that they agree well.

4.4. Dynamical Measurement

One of the characteristic advantages of this method is possibility to conduct dynamical measurement under noncontact manner. The dynamical viscosity change is measured, which is conducted on JS100.
A characteristic phenomenon where the viscosity raises with temporary temperature reduction is monitored in Fig. 7. Thus, this result indicates that present method can be applied to conditions where the viscosity changes with time.

5. CONCLUDING REMARKS

Based on the above experimental results, the characteristics of laser-induced capillary wave method are the ability (1) to measure various liquids with wide range of viscosity (0.1~10000 mPa·s); (2) to monitor the dynamical change of viscosity; (3) to conduct measurement in a noncontact manner both in heating and probing; (4) to measure small sample volume, and (5) to conduct measurements without adding any light absorbing materials. From the above characteristics, it can be indicated that this measurement technique has the applicability to wide range of science and industry fields.
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