Instability of a Planar Liquid Sheet Formed with Surfactant Aqueous Solution

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Abstract

In this study, we clarified the growth characteristics of the instability waves of a planar liquid sheet formed with a surfactant solution. It has been widely reported that a liquid sheet jet oscillates in a sinuous shape because of Kelvin–Helmholtz instability, and that the growth rate of the instability wave depends on surface tension. Therefore, if the liquid is a surfactant solution, the growth characteristics depend on time-dependent surface tension, referred to as dynamic surface tension, which originates from the time delay required for the adsorption of surfactant molecules into a newly formed surface. In this study, wave growth affected by dynamic surface tension was investigated. First, the relation between the growth rate of the wave and the time-invariant surface tension was determined by using ethanol aqueous solutions of different concentrations. Second, the dynamic surface tension of the surfactant solution was measured, and surface tension varying in accordance to the distance from the nozzle was estimated. Third, the spatially varying amplitude of the wave was calculated from the results obtained in the above two steps. Finally, the calculated wave amplitude was compared with the measured amplitude of the surfactant solution. The good agreement obtained between the calculated amplitude and experimental amplitude proved that the growth characteristics for a surfactant solution depend on dynamic surface tension.

Introduction

When a new liquid surface of a surfactant solution is formed, the surface tension varies temporally because surfactant molecules adsorb the new liquid surface and then reach equilibrium. Therefore, initially, the surface tension reflects the same value as that of the solvent, and then decreases to equilibrium surface tension (static surface tension). The time-variable surface tension and the equilibrium surface tension are referred to as dynamic surface tension and static surface tension, respectively[1].

It is well-known that, in general, a liquid sheet is disintegrated by the development of flapping waves, and that the growth characteristics of the waves depend on surface tension. Several investigations have been conducted into the growth characteristics of a planar or fan liquid sheet based on Kelvin–Helmholtz instability [2]-[4]. In recent years, nonlinear development or three-dimensional deformation of these waves has been analyzed theoretically and experimentally[5-8]. In addition, recent research has reported the influence of the velocity profile of the liquid sheet and ambient gas on wave growth[9]-[11]. However, to our knowledge, few studies have been carried out on wave growth in a case where surface tension varies during liquid flow as a result of dynamic surface tension. In consideration of the fact that surfactants are added to paint or ink to improve pigment dispersion, a clarification of wave growth for a surfactant solution is important from the viewpoint of industrial application. In this study, we ascertained the influence of dynamic surface tension on wave growth characteristics by using a planar liquid sheet.

Experimental apparatus and procedure

Figures 1(a) and (b) show the experimental setup and a detailed drawing of a nozzle. Pressurized test liquid supplied by a pump passes through a surge tank and emerges from a slit nozzle measuring 150 mm in width with a 0.5 mm gap as shown in Fig. 1(b). To form a smooth planar liquid sheet, we adjusted the injection pressure while monitoring a pressure sensor mounted on the surge tank. We maintained a liquid sheet velocity of 8 m/s because too slow a velocity would induce shrinkage of the liquid sheet owing to surface tension, and too fast a velocity would create turbulence. To introduce a precise periodical perturbation into the liquid sheet jet, pieces of 0.1-mm stainless steel foil were placed on the opposite sidewalls of the surge tank and were oscillated at the same phase by vibrators (ONKYO, Model DU2506H). The voltage and frequency applied to the vibrator were set at 15 V and 250 Hz, respectively, so that growth process of the wave could be observed clearly. To measure wave amplitudes, we visualized the wavy cross section of a liquid sheet with a laser sheet (Melles Griot, Model 85GHS305, Wavelength: 532 nm, Output power 700 mW) using test liquids containing dissolved fluorescence dye.

The experimental procedures consisted of the following four steps. First, the dependency of the spatial growth rate of the wave on surface tension was determined experimentally using ethanol aqueous solutions of
different concentrations, which had a time-invariant surface tension. Second, the dynamic surface tensions of the surfactant solutions were measured at different concentrations, and the surface tension, which varied locally, of the liquid sheets was estimated. We used polyoxyeylene(10)octylphenyl ether as a typical surfactant. Third, the wave amplitudes of the surfactant solutions were predicted using the results of these two steps. The dependency of the spatial growth rate on surface tension and the measured dynamic surface tension gave a spatially varied growth rate. Wave amplitude was calculated by the integration of the growth rate. Finally, the calculated wave amplitude was compared with the measured amplitude of the surfactant solution.

Measurement of dynamic surface tension

We measured the dynamic surface tension using the oscillating jet method[1] and the capillary jet method[12]. Figure 2 is a schematic drawing of the setup of the oscillating jet method. Test liquid is fed to an overflow tank and ejected from an oval orifice at a constant head. The oval liquid jet flows downward in an oscillating manner because of the restoring force caused by surface tension. The dynamic surface tension $\sigma_d$ was calculated from the following equation using the wavelength $\lambda$ measured at the downward distance $x$ from the orifice by photography,

$$\sigma_d = \frac{4 \rho Q^2}{6a \lambda^2} \left[ 1 + \frac{37}{24} \left( \frac{a^2}{a^2} \right)^2 \right] \left[ 1 + 2 \left( \frac{\lambda}{Q} \right)^{3/2} + 3 \left( \frac{\lambda}{Q} \right)^2 \right], \quad (1)$$
where \( Q \) is the flow quantity, \( \rho \) is the density of the liquid, \( \nu \) is the kinetic viscosity, \( a \) is the average radius of the liquid jet, and \( b \) is the amplitude of the oscillation\[1\]. The distance \( x \) and jet velocity \( V = Q/(\pi a^2) \) give elapsed time \( t_s = x/V \) from the creation of a new liquid surface at the nozzle, which we refer to as “surface elapsed time.”

The setup of the capillary jet method is shown in Fig. 3. In general, when a thin liquid round jet (a capillary jet) is ejected horizontally at low speed, it exhibits a trace of downward curve. This trace depends on not only gravity but also on surface tension because surface tension becomes comparative to inertial and gravity forces. In the case of a surfactant solution, the trace of the jet depends on dynamic surface tension because the liquid surface created at the orifice continues to age during the flow. We derived a theoretical model expressing the jet trace from a balance equation of inertial force, surface tension, pressure, and gravity force based on the consideration stated above. This method measures dynamic surface tension using this theoretical model. In general, the variation of dynamic surface tension \( \sigma_d \) with surface elapsed time \( t_s \) is described by the following equation\[1\],

\[
\sigma_d(t_s) = \sigma_s + \frac{\sigma_0 - \sigma_s}{1 + \left(\frac{t_s}{t_r}\right)^n},
\]

where \( \sigma_0 \) is the surface tension of the solvent (water), and \( \sigma_s \) is the static surface tension of the solution. Constant numbers \( t_r \) and \( n \) represent parameters determining the relaxation time of the dynamic surface tension and the function form of the relaxation, respectively. Hence we expressed the dynamic surface tension by Eq (2) for a theoretical calculation of the jet trace. In the capillary jet method, jet traces corresponding to various \( t_r \) and \( n \) are calculated theoretically. A set of \( t_r \) and \( n \) provides the closest approximation of the jet trace obtained photographically. This set provides a measurement value of dynamic surface tension \( \sigma_d \). The details of the theoretical model are stated in Reference\[12\]. In this study, we used capillary jet ejected from a 1.95-mm orifice at a velocity of 0.43–0.50 m/s for measurement.

### Physical properties of test liquids

In this study, the dependency of spatial wave growth rate on surface tension was determined using ethanol aqueous solutions of six different concentrations which had time-invariant surface tension. Next, the influence of the dynamic surface tension on wave growth was considered using surfactant solutions.

The concentrations of the surfactant solutions were selected as follows. The static surface tension of the surfactant solution used in this experiment indicates a constant value of 32 mN/m at concentrations >200 ppm, and \( t_r \) only decreases with an increase in concentration. In addition, the surface elapsed time becomes \( t_{sf} = 5–15 \text{ ms} \) at 40–120 mm downstream from the slit nozzle, where the wave amplitude of the liquid sheet is measured. Therefore, surface tension of a dilute solution almost coincides with that of solvent in the measurement region because of \( t_r \gg t_{sf} \), while that of a dense solution agrees well with the static surface tension of the solution owing to \( t_r \ll t_{sf} \). If the concentration is moderate, that is \( t_r \approx t_{sf} \), the surface tension varies in the measurement region. Accord-

### Table 1 Physical properties of test liquids.

<table>
<thead>
<tr>
<th>Type of test liquid</th>
<th>Ethanol solution</th>
<th>Surfactant solution</th>
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<tbody>
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<td></td>
<td>( C_e ) mass %</td>
<td>( C_s ) mass ppm</td>
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<tr>
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ingly, we selected surfactant concentrations of 250, 1500, and 5000 ppm to realize three time relations of $t_r >> t_{sf}$, $t_r \approx t_{sf}$ and $t_r << t_{sf}$. The dynamic surface tensions of these surfactant solutions decrease from the surface tension of the solvent ($\approx 72$ mN/m) to the static surface tension of the solution ($\approx 32$ mN/m). Therefore, we prepared aqueous ethanol solutions of 0 to 18 mass% to determine the wave growth rate in the range between these surface tensions. Glycerine and sodium bromide were added to the nine test liquids so that their physical properties—except surface tension—were the same among the test liquids.

Table 1 shows the composition of the test liquid, density $\rho$, kinematic viscosity $\nu$, and static surface tension $\sigma_s$. The mass concentrations of ethanol, surfactant, glycerine and sodium bromide are represented by $C_e$, $C_s$, $C_g$, and $C_{sb}$, respectively. In addition, 50 ppm of phloxin B was added as a fluorescence dye. The densities and kinematic viscosities among all the test liquids were adjusted to be almost the same, and the static surface tensions of the three surfactant solution became identical. In addition, the static surface tensions of an ethanol solution decreased with an increase in $C_e$ from 71.9 to 37.4 mN/m. We determined the dependency of the wave growth rate on surface tension using this variation.

The dynamic surface tensions $\sigma_d$ of the surfactant solutions are plotted against surface elapsed time $t_s$ in Fig. 4. The plots and solid lines express $\sigma_d$ measured by the oscillating jet method and the capillary jet method, respectively, and the horizontal broken line and the dotted line indicate the static surface tensions of solvent and solution, respectively. The measurement values obtained by both methods agree well, and decrease to the static surface tension of the solution asymptotically. The relaxation time for the decrease becomes shorter at greater concentrations $C_s$. The relaxation time $t_r$ obtained from equation (2) are 99, 9.6 and 0.13 ms at $C_s = 250$, 1500, and 5000 ppm, respectively. Comparing $t_r$ with a surface elapsed time $t_s = 5$–15 ms for measurement of wave amplitude, time relations of $t_r >> t_{sf}$, $t_r \approx t_{sf}$ and $t_r << t_{sf}$ are actualized.

Dependency of wave growth rate on surface tension

Several investigations have been conducted on the growth characteristics of a planar liquid sheet in a quiescent gas. The simplest linear stability theory provides the following relation between temporal wave growth rate $\beta$ and surface tension $\sigma$, assuming gas and liquid to be inviscid[2][3]:

$$\beta = \sqrt{\frac{k^2 U^2 \rho_g}{\rho} - \frac{k^2 \sigma}{\tan\left(\frac{kh}{2}\right)}}$$

where $k$ is the wave number, $\rho_g$ is the density of the gas, and $h$ is the thickness of the liquid sheet. The above theory assumes quiescent ambient gas and velocity slip on the liquid surface, that is, equation (3) provides the growth rate for a gaseous boundary layer of infinitesimal thickness. However, the gaseous boundary layer is actually developed because the gas near the liquid surface is accelerated by viscosity. Lozano et al. and Tammisola et al. recently reported that a gaseous boundary layer thicker than the liquid sheet reduces wave growth rate[9][10]. These reports suggest that the theoretical prediction of growth rate is difficult for an
individual experimental setup because the gas stream around a liquid sheet depends on the configuration of each setup. Therefore, we determined the relation between the spatial wave growth rate and the surface tension experimentally, which is affected by a gaseous boundary layer, using an ethanol aqueous solutions.

Figures 5(a)–(c) show visualized images of the cross sections of a wavy liquid sheet at ethanol solutions of $C_e = 0$, 5.5, 18%, respectively. The Weber number $We = \rho U^2 h / \sigma_s$ and the Reynolds number $Re = U h / \nu$ in Figs. 5(a)–5(c) were 371–717 and 1230–1300, respectively, where $h$ is the thickness of the liquid sheet, estimated to be $h = Q_s / (w U) = 0.39$ mm from flow quantity $Q_s = 468$ ml/s, slit width $w = 150$ mm and flow velocity $U = 8$ m/s. The images are enlarged in the horizontal direction to enhance the displacement of the liquid sheet. The scale beside the images indicates the distance $x$ from the nozzle. The images show that periodic waves develop downstream preserving constant wavelength $\lambda = 32$ mm regardless of the concentration $C_e$, and that the amplitude of the wave is amplified at a greater concentration. Although the waveforms in the figures are nearly sinusoidal at $x \leq 130$ mm, they become similar to triangle waves as the wave amplitude increases.

We took 10 visualized images similar to Fig 5 at each ethanol concentration $C_e$, and measured the wave amplitude by determining the positions of the peaks and valleys of the waves. The accuracy of this amplitude measurement was about 50 $\mu$m. The measured amplitudes $A$ are plotted against $x$ in Fig. 6. The original point of the plots is shifted upward for every increment of the concentration $C_e$. Measurement values increase at $x \leq 130$ mm exponentially, regardless of $C_e$, and then decline at a growth rate at $x > 130$ mm. Considering the remarkably large amplitude and triangle-shaped deformation of the waveform at $x > 130$ mm, the decline in the growth rate is probably because of the enhancement of nonlinearity.

Fitting curves as calculated by $A = A_0 \exp(\alpha x)$ are also shown in Fig. 6, where $A_0$ and $\alpha$ are the initial amplitude and the spatial growth rate, respectively, evaluated by application of the least squares method to the experimental data at $x \leq 130$ mm. The evaluated initial amplitude $A_0$ became 0.14 mm regardless of the concentration, and the spatial growth rates $\alpha$ varied from 0.0246 mm$^{-1}$ to 0.0267 mm$^{-1}$ depending on $\sigma_s$, as shown in Fig. 6. This variation in $\alpha$ with $\sigma_s$ shows a significant relation because the error of $\alpha$, which originates from the accuracy of amplitude measurement, is estimated to be about 0.0001 mm$^{-1}$.

Figure 7 shows the relation between the wave growth rate $\alpha$ and surface tension $\sigma(=\sigma_s)$. A straight solid line

![Figure 5 Visualized cross sections of wavy liquid sheets.](image-url)
fitted to the experimental points indicates that growth rate $\alpha$ decreases with an increase in surface tension $\sigma$ linearly. In addition, it is noteworthy to compare the measured growth rate with the theoretical growth rate. The substitution of wave number $k = 2\pi/\lambda = 0.196$ mm$^{-1}$ into Eq. (3) gives hypothetical spatial growth rate $\alpha = \beta/U$ as denoted by a dotted line in Fig. 7. The experimental growth rates are less than the hypothetical values, which range from 0.0302 to 0.0318 mm$^{-1}$, by about 20%. This difference is probably because of the gaseous boundary layer formed on the liquid surface because the viscosity of the liquid rarely results in a decay of the wave under this experimental condition[4].

**Characteristics of wave growth for a surfactant solution**

First, we predict the wave growth rate for a surfactant solution. Summarizing the results in Fig. 4 and Fig. 7, it is concluded that dynamic surface tension $\sigma_d$ decreases with an increase in surface elapsed time $t_s$, and that the growth rate $\alpha$ decreases with an increase in surface tension $\sigma$ linearly. In addition, surface elapsed time $t_s$ and distance $x$ from the nozzle are related by $x = U t_s$ using flow velocity $U$ because the surface of the liquid sheet jet created at the nozzle exit moves downstream with time. Therefore, organizing this relation between $x$ and $t_s$, dynamic surface tension $\sigma_d$ measured by the capillary jet method, and the linear relation between $\alpha$ and $\sigma$, we can
obtain spatial growth rate $\alpha$ at distance $x$ as shown in Fig. 8. The figure indicates that growth rate $\alpha$ decreases as distance $x$ or surfactant concentration $C_s$ increases.

Applying $\alpha(x)$ of Fig 8 to the following equation, we can obtain wave amplitude $A$ of the wave.

$$A = A_0 \exp\left(\int_0^x \alpha(x')dx'\right)$$  \hspace{1cm} (4)

Figure 9 shows the calculated amplitude obtained by the substitution of $A_0 = 0.14 \text{ mm}$ in equation (4) and the measured amplitude, which are represented by the solid lines and plots, respectively. The broken and dotted lines denote the referential amplitudes calculated by equation (4) assuming constant static surface tensions of the solvent and solution, respectively. Both the calculated and the measured amplitudes have larger values at higher concentrations. This dependency of the amplitude on the concentration is due to dynamic surface tension because the static surface tensions of the solutions are the same. Figure 10, an enlargement from Fig. 9 in the range $x = 70–120 \text{ mm}$, demonstrates that the measured values agree well with the calculated values (solid lines). It is noteworthy that the measured and the calculated amplitudes at $C_s = 250 \text{ ppm}$, denoted by circles and a blue solid line, respectively, approach the referential amplitude based on the assumption of a constant surface tension of solvent, denoted by the broken line. Conversely, the measured and calculated values at $C_s = 5000 \text{ ppm}$ represented by triangles and an orange solid line, respectively, are very similar to the referential amplitude for static surface tension of the solution, denoted by the dotted line. This asymptotic behavior is explained by the surface tension at the measurement position for wave amplitude, that is, the dynamic surface tensions at $C_s = 250 \text{ ppm}$ and 5000 ppm become similar to the static surface tensions of the solvent and solution at $x = 40–120 \text{ mm}$, respectively. In addition, the amplitudes at $C_s = 1500 \text{ ppm}$ lie between the two referential amplitudes based on both static surface tensions because the dynamic surface tension varies between both static surface tension at $x = 40–120 \text{ mm}$.

Summary and Conclusions

We investigated the influence of time-variable surface tension (dynamic surface tension) on the growth characteristics of instability waves in a planer liquid sheet jet. The relation between the spatial growth rate of the wave and time-invariant surface tension was determined using ethanol aqueous solutions of six different concentrations. This relation and the measured dynamic surface tension of the surfactant solution provided the estimated value of the spatial wave growth rate in liquid sheet flow of a surfactant solution. The amplitudes calculated...
by the integration of the estimated growth rate agreed well with the measured amplitudes. This implies that development of the wave depends on dynamic surface tension. In addition, it was clarified that the estimation of wave growth on the basis of the static surface tension of a solvent/solution gives an excellent approximation for a dilute/dense solution, which has a relaxation time for dynamic surface tension, $t_r$, much longer/shorter than the time required for flow in a given field.

References