Influence of a soluble surfactant on the wave characteristics of a liquid film flow

V V Guzanov, A V Bobylev, A Z Kvon and S M Kharlamov

Kutateladze Institute of Thermophysics, SB RAS, 1 Lavrentyev Ave., Novosibirsk, 630090, Russia

E-mail: guzanov@itp.nsc.ru

Abstract. In this paper we present results of experimental investigation of the influence of soluble surfactant (Triton X-100) on wave characteristics of vertically falling liquid film in the range of film flow Reynolds number $25 < Re < 95$. Shadowgraph technique is used to analyze wave patterns on the whole length of the test section (140 cm). Film thickness fields obtained with the help of high-speed Laser Induced Fluorescence (LIF) technique in two areas in the bottom part of test section allow analyzing statistical characteristics of the film flow. Expectedly, adding surfactant suppresses wave motion. For some surfactant concentrations the absence of waves is observed in the whole test section. For other concentrations the wave repetition rate decreases with distance from the liquid inlet. Analysis of spectral characteristics reveals that for water with some concentrations of surfactant, as for pure liquids, the steady-state three-dimensional wave regimes are observed. The characteristics in such flow regimes differ from the characteristics of pure water flow.

1. Introduction

At present, the effect of surfactants on the wave characteristics of liquid film flows is considered in a small number of works. This is primarily due to the complexity of the coupled problem of hydrodynamics of flows with a free surface and the mass transfer of surfactants. Therefore, research works are mainly limited to considering two-dimensional cases. For example, the linear stability of the film flow in the presence of surfactants in a two-dimensional formulation is studied in [1 – 4]. In these works, it is demonstrated that the hydrodynamic wave mode is suppressed in comparison with the flow of a pure liquid and that surfactant concentration waves move with the film velocity. The influence of surfactants on the wave characteristics is shown to be due to both the dependence of the surface tension on the surfactant concentration and the Marangoni effect, in which tangential stresses arise on the free surface of the film caused by the variability of the surface tension along the surface.

A few experimental works concerning the influence of surfactant concentration on the wave formation are mainly considered at the initial stage of the film flow in two-dimensional approach [5 – 7]. The results of these experiments are qualitatively consistent with the results of theoretical analysis. The stabilization effect of surfactants on the wave regimes is considered in [8], where appearance of some three-dimensional structures in the initial stage of the film flow was mentioned. Recent experimental study of the influence of soluble surfactant on the initial development of waves on vertically falling liquid films [9] have revealed formation of rivulets and appearance of three-dimensional waves in the initial part of the flow. The study [9] was limited to the initial stage of the flow. Therefore, because of the long evolution processes, the developed steady-state 3D regimes of wave motion could not be observed. As it is known, the developed steady-state 3D regimes are observed for
pure liquids [10] and are characterized by the absence of changes of spectral and statistical characteristics of the film thickness.

Thus, the question of whether the developed steady-state three-dimensional wave regimes of film flow can exist in the presence of surfactant concentration gradients arising on the free surface of the film and leading to a change in the local coefficient of surface tension (Marangoni effect) remains open. Therefore, the main goal of this work is experimental investigation of the influence of soluble surfactant Triton X-100 on three-dimensional wave regimes of falling liquid films.

2. Experimental conditions and measurement techniques

A detailed description of the experimental setup and measurement method is given in [10], so we will only briefly describe them. The experiments are carried out on a vertical transparent glass plate with the dimensions of 50 cm in transversal and 140 cm in vertical directions in the range of of film flow Reynolds number 25 < Re < 95. Here Re=q/ν, where q is the specific flow rate and ν is the kinematic viscosity of the working fluid. The film flow is formed by a slot distributor. Before entering the slot distributor, the fluid flow rate could be modulated with a given frequency in order to create regular waves in the initial part of the flow. The wave structure over the whole area of the plate is monitored by shadowgraph technique, for which the plate is illuminated from the side covered by the flowing film. The shadow image is formed on a white screen behind the plate and recorded by a CCD camera. Distance between the screen and the plate is adjusted to register the elements of wave structure with both small (the main hump of the wave) and higher curvature (capillary precursor) (for details see [10]).

The method of laser induced fluorescence (LIF) is used to measure instantaneous local film thickness h on the measuring areas situated in the bottom part of test section at two different distances from the film inlet. The LIF method is based on the reconstruction of local film thickness in accordance with local light intensity emitted by a small amount of fluorescent dye dissolved in working liquid. In our experiments working liquids are doped with small amounts, about 15–30 mg/l, of fluorescent dye Rhodamine 6G. The prepared solutions of the dye absorb green and emit orange light. To reconstruct 3D form of liquid film we use a high-speed LIF system composed of a continuous green laser with the wavelength of 532 nm and high-speed camera with orange filter. The LIF images are recorded with the frame rate of 1000 fps and exposure time of 0.99 ms. The length of every record is equal to 2 s. The dimensions of the measurement areas are 17 × 17 cm, which results in the resolution of 0.17 mm per pixel. The total error in determining film thickness for all investigated regimes of film flow is estimated to be not more than 5% (for details see e.g. [11]).

The above-mentioned recording parameters allow estimating statistical characteristics of film flow evolution with appropriate accuracy. As in [10], the following characteristics are calculated: the film thickness probability density function (PDF), variance (dispersion) D and power spectral density Y(f) of film thickness temporal variation. Since the film flow due to 2D-3D wave transition acquires a rivulet nature, for evaluation of downstream wave evolution we use statistical and spectral characteristics averaged in transverse direction over the full width of measurement area.

The choice of the surfactant working concentrations is made using scale considerations, according to which surfactant concentrations are selected based on a comparison of the characteristic time of the wave process Tw and the characteristic relaxation time of the surface tension of the liquid film. The period of wave repetition rate and the period of the capillary precursor of the waves are considered as maximal and minimal characteristic times of the wave process, so the approximate time interval Tw lays between 0.1 s and 0.01 s for the conditions of our experiments. The diffusion scale Td is chosen as the characteristic relaxation time of the surface tension. As it is shown in [12], the surface tension is balanced within 1 − 10 Td even for surfactants with strong interaction in a wide range of concentrations. Maximum effect of surfactant can be expected when Td ~ Tw. When Td >> Tw, the surfactant can be considered as an insoluble layer on the surface of liquid and when Td << Tw it can be expected that the waves will develop in the same way as for a pure liquid with physical properties modified by the surfactant.
In our experiments, we use a nonionic surfactant Triton X-100, the concentration $C$ of which in water is in the range of $10 < C < 2000$ mg/l, corresponding to that calculated according to [12]. The diffusion scale $T_d$ lies in the range of $115 - 0.003$ sec (for more details see [9]). These data agree with data presented in literature ([12 - 16]) on dynamic surface tension.

3. Results

The results of experiments show that, as it is mentioned in [9], the addition of surfactant stabilizes the flow in a wide range of concentrations. For concentration of Triton X-100 in water inside the range of $50 \leq C \leq 200$ mg/l complete suppression of the wave motion on the whole film surface (up to 140 cm) is observed for all investigated Re. For concentrations, which are close to this range, the waves on the surface appear not for all investigated Re. At low Re no waves at the surface are observed. The waves become distinguishable on the test section only for Reynolds numbers exceeding some threshold value. Examples of wave patterns for maximal investigated Re = 95 and different surfactant concentrations are shown in figure 1.

![Figure 1](image-url)

**Figure 1.** Shadowgraph images of the flow at Re=95 and different surfactant concentrations. (a) pure water, (b) $C = 10$ mg/l, (c) $C = 20$ mg/l, (d) $C = 500$ mg/l, (e) $C = 2000$ mg/l.

The instant film thickness fields $h$, obtained by the LIF technique in the bottom part of the flow and presented in figure 2, show the discrepancy in the wave patterns for different concentrations of surfactant in more detail. In particular, one can see the difference in the amount of waves on the unit area and in the wave shapes. The wave structures are elongated in comparison with the case of a pure liquid flow, and the waves have a shape more like a horseshoe: two elongated tails are observed after the main hump. For surfactant concentrations close to those at which complete suppression of wave motion occurs, a high amplitude capillary precursor is observed in the wave front. This precursor is indistinguishable for other surfactant concentrations and for pure water. Another feature of the flow of water loaded with surfactant is the formation of more pronounced rivulet structures, observed not only on the film thickness distributions averaged in time but even on instant film thickness fields.
The discrepancy in the wave patterns between the film flow of pure water and water with a surfactant becomes less pronounced as the surfactant concentration moves away from the mentioned range of complete suppression of wave motion. This behaviour can also be observed when analyzing statistical and spectral characteristics of the film thickness (figures 3 – 5). Except for the case of $C = 2000$ mg/l for all investigated concentrations, the variance and power spectral density values are below the values for pure liquid. And PDFs become symmetric about the maximum value which at the same time becomes closer to the Nusselt’s film thickness $h_N$ and larger than for the case of pure water. It can be noted that minimal values of film thicknesses are observed for pure liquid. All these differences indicate the suppression of wave motion. The discrepancy in the power spectrum of density distributions consists not only in lower values, but in the displacement of the maximum value towards the lower frequencies. This leads to a decrease in the slope of the distribution. At the same time, for some surfactant concentrations in the power spectral density, there are frequencies that are higher than for pure water.

The analysis of statistical and spectral characteristics reveals that fully developed steady-state 3D regimes are observed only for pure water and for water with Triton X-100 concentration $C = 2000$ mg/l at Re = 45 and 95. For each Re the values of variance are close for both liquids (lines 1 and 5 in figure 3), whereas PDF and power spectral density are quite different (lines 1 and 5 in figures 4 and 5). For these liquids, in contrast to others, the PDF distribution has asymmetric right-tailed shape (figure 4).

For other surfactant concentrations, despite weak or absent changes in variance with distance for most of the investigated wave regimes, changes in other characteristics (PDF and power spectral density) are observed. Moreover, in most cases, evolution is directed towards damping of the waves.
Figure 3. Film thickness variance normalized to $h_N$ for different concentrations of surfactant: 1 – pure water, 2 – $C = 10$ mg/l, 3 – $C = 20$ mg/l, 4 – $C = 500$ mg/l, 5 – $C = 2000$ mg/l.
(a) $Re = 45$, (b) $Re = 95$.

Figure 4. PDFs at $Re = 95$ and distance $X = 130$ cm for different concentrations of surfactant: 1 – pure water, 2 – $C = 10$ mg/l, 3 – $C = 20$ mg/l, 4 – $C = 500$ mg/l, 5 – $C = 2000$ mg/l.

Figure 5. Power spectral density for different concentrations of surfactant: 1 – pure water, 2 – $C = 10$ mg/l, 3 – $C = 20$ mg/l, 4 – $C = 500$ mg/l, 5 – $C = 2000$ mg/l. $Re = 95$, $X = 130$ cm.

In conclusion, it should be noted that for water with a Triton X-100 concentration $C = 2000$ mg/l the characteristic frequency of the surface tension relaxation (about 330 Hz) is higher than almost all
frequencies of the film thickness change (line 5 in figure 5), and this liquid in such case can be considered as having physical properties modified by a surfactant.

**Conclusion**

With the help of shadowgraph and LIF techniques with high spatial and temporal resolution the influence of soluble surfactant (Triton X-100) on wave characteristics of vertically falling liquid film has been experimentally investigated in the range of film flow Reynolds number $25 < \text{Re} < 95$. The addition of surfactant stabilizes the flow in a wide range of concentrations. For concentration of Triton X-100 in water inside the range of $50 \leq C \leq 200 \text{ mg/l}$ complete suppression of the wave motion is observed on the whole film surface (up to 140 cm) for all investigated Re. For solutions with surfactant concentrations outside this range the obtained wave patterns and statistical characteristics differ from those for pure water and indicate the suppression of wave motion. In comparison with the case of pure water, the power spectral density distribution has a smaller slope, and its maximum value shifts towards lower values.

Analysis of statistical and spectral characteristics reveals that fully developed steady-state 3D regimes are observed only for pure water and for water with Triton X-100 concentration $C = 2000 \text{ mg/l}$ at $\text{Re} = 45$ and 95. For water with this concentration of surfactant, the characteristic surface tension relaxation time is shorter than most observed times of film thickness changes. This liquid can be considered as a liquid with physical properties modified by a surfactant. So, the question about the existence of fully developed stationary three-dimensional regimes in the presence of surfactant concentration gradients, arising on the free surface of the film and leading to a change in the local coefficient of surface tension, cannot be fully answered by present investigation.

**Acknowledgements**

This work was supported by Russian Foundation for Basic Research Grant No. 18-01-00682 and under state contract with IT SB RAS.

**References**

[1] Velarde M G, Shkadov V Y and Shkadova V P 2000 *Fluid Dynamics* **35**(4) 515

[2] Shkadov V Y, Velarde M G and Shkadova V P 2004 *Phys. Rev. E* **69**(5) 056310

[3] Karapetsas G and Bontozoglou V 2013 *J. Fluid Mech.* **729** 123

[4] Pereira A and Kalliadasis S 2008 *Phys. Rev. E* **78**(3) 036312

[5] Strobel W J and Whitaker S 1969 *AIChE J.* **15**(4) 527

[6] Georganaki A, Vlachogiannis M and Bontozoglou V 2012 *J. Phys.: Conf. Series* **395**(1) 012165

[7] Georganaki A, Vlachogiannis M and Bontozoglou V 2016 *Int. J. Multiphase Flow* **86** 28

[8] Tailby S R and Portalski S 1961 *Trans. Inst. Chem. Eng.* **39** 328

[9] Bobylev A V, Guzunov V V, Kvon A Z and Kharlamov S M 2019 *J. Phys.: Conf. Ser.* **1382** 012073

[10] Guzunov V V, Bobylev A V, Heinz O M, Kharlamov S M, Kvon A Z and Markovich D M 2018 *Int. J. Multiphase Flow* **99** 474

[11] Kharlamov S M, Guzunov V V, Bobylev A V, Alekseenko S V and Markovich D M 2015 *Phys. Fluids* **27**(11) 114106

[12] Ferri J K and Stebe K J 2000 *Adv. Colloid Interface Sci.* **85**(1) 61

[13] Brazee R D, Bukovac M J, Cooper J A, Zhu H, Reichard D L and Fox R D 1994 *Transactions of the ASAE* **37**(1) 51

[14] Miller R, Joos P and Fainerman V B 1994 *Adv. Colloid Interface Sci.* **49** 249

[15] Horozov T and Arnaudov L 1999 *J. Colloid Interface Sci.* **219**(1) 99

[16] Fainerman V B, Mys V D, Makievski A V, Petkov J T and Miller R 2006 *J. Colloid Interface Sci.* **302**(1) 40