Experimental study of the effect of surfactants on the characteristics of waves in three-dimensional wave regimes

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Abstract. Effect of a surfactant Triton X-100 on three-dimensional wave regimes of the falling liquid film flow was experimentally studied. The shadow method and laser induced fluorescence measurements revealed that for surfactant concentrations at which complete suppression of wave motion is not observed, the flow characteristics at the three-dimensional wave regime differ from the characteristics of a pure water flow. Most likely, these differences are associated with differences in the structure of flows under the three-dimensional wave, which were revealed by simultaneous measurements of the thickness and volumetric measurements of the velocity in the wave at the initial section of the flow. For films with the surfactant concentration of 2000 mg/l, the capillary ripples in front of the three-dimensional wave are weakly expressed, there are no backflows, and transverse flows in the area of the capillary precursors are much weaker than in the case of pure water.

1. Introduction

Falling liquid films are an integral part of numerous technical applications. It is well known that the addition of the surfactant, even in a very small amount, can significantly change the interfacial properties and wave patterns of the films. Therefore surfactants are actively used as part of technological processes related to film flows. Sometimes the presence of the surfactants in a working fluid may be the result of the contamination. For these reasons, a lot of attention is paid to the study of the effect of surfactant on the flow. Even though surfactant doped solutions are actively studied, the influence of surfactants on the wave hydrodynamics of falling liquid film remains underinvestigated. Among few studies is the numerical simulation \cite{1} of the liquid film flow in presence of insoluble surfactant. Influence of the Marangoni stresses appeared due to gradients of the interfacial surfactant concentration and suppression of the humps of the travelling waves as well as capillary precursors were shown. Soluble surfactant Triton-X was used in experimental work \cite{2}. The initial stage of the pure liquids flow evolution when the film is covered with two-dimensional (2D) waves was not observed. At low and moderate surfactant concentrations three-dimensional (3D) wave motion was suppressed wherein at high surfactant concentrations the 3D waves start to grow again. Since there is a lack of experimental data about wave characteristics in presence of a surfactant, a more detailed study is required.

2. Experimental conditions and methods
Non-ionic surfactant Triton X-100 was used in our experiments. There are several reasons why we used this type of surfactant. The adsorption of the Triton X-100 on the water surface is well described by the Langmuir isotherm and is controlled by diffusion [3], the adsorption isotherm constants, and the diffusion coefficient for Triton X-100 in aqueous solutions are widely represented in the literature e.g. [3,4] and as a non-ionic surfactant, it has a reduced sensitivity to water quality. Our main focus was on the description of the 3D wave regimes, this determines the choice of the surfactant concentration $C = 2000 \text{mg/l}$ at which the 3D waves are not suppressed [2]. Liquid film flow at lower concentrations, at which suppression of the 3D wave motion is expected, also has been studied. Therefore water solution of non-ionic soluble surfactant Triton X–100 with the concentration $C = 10 \text{ mg/l}$, $20 \text{mg/l}$, $200 \text{mg/l}$, $500 \text{mg/l}$ and $2000 \text{ mg/l}$ were used as a working fluid. Obtained characteristics were compared with the case of the pure water flow. All experiments were performed with a working fluid temperature of $24^\circ \text{C}$.

Two types of test sections were used in this work. In order to obtain a general wave pattern over the long run, liquid film flow at the Reynolds number $\text{Re} = 44$ was formed on a vertical glass plate with the dimensions $50 \text{ cm}$ in transversal and $140 \text{ cm}$ in a longitudinal direction. The wave structure over the whole area of the plate was visualized by the shadowgraph technique, for which the plate was illuminated in such a way that the shadow image was formed on the white screen behind the plate and recorded by the CCD camera. The distance between the screen and the plate was adjustable so that the elements of wave structure with both, small (the main hump of the wave) or higher curvature (capillary precursor) were registered (for details see [5]). The method of laser-induced fluorescence (LIF) was used to measure instantaneous local film thickness $h$ in the bottom part of the 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. To reconstruct the interface of the liquid film we used a high-speed LIF system composed of a continuous green laser with a wavelength of $532 \text{ nm}$. The orange color reemitted by the fluorescent dye was recorded through an orange light filter on a digital high-speed camera. The recording frame rate was $1000 \text{ fps}$ and the exposure time was $0.99 \text{ ms}$. The achieved spatial resolution was $0.17 \text{ mm/pixel}$ with dimensions of the measurement area $17 \times 17 \text{ cm}$. The total error in the determination of film thickness for all investigated regimes of film flow was estimated to be not more than $5\%$ (for details see e.g. [6]). As in [5], the next statistical characteristics were calculated: the film thickness probability density function (PDF), variance (dispersion) $D$, and power spectral density $Y(f)$ of film thickness temporal variation. Obtained statistical and spectral characteristics were then space averaged in transverse direction over the full width of the measurement area for evaluation of downstream wave evolution. This step is required due to the rivulet nature of the film flow which takes place during the 2D-3D transition.

The second test section was similar to that described above, but with smaller dimensions of $16 \times 25 \text{ cm}$ and described more detailed in [7]. It was used to study the surface morphology of an individual three-dimensional wave and the flow structure in it. Therefore water solution with the surfactant concentration $C = 2000 \text{mg/l}$ at which the 3D waves are not suppressed was used. Regular 3D waves were artificially generated by the flow rate modulation with the frequency $F = 14 \text{ Hz}$. The light-field camera Raytrix R11m and principles of the particle tracking velocimetry were used for the volumetric velocity measurements. The light-field camera (otherwise called plenoptic camera) allows determining the position of the tracer particles (tracers) in volume. Images obtained were used also for the reconstruction of the shape of the wave by the LIF method. Fluorescent dye Rhodamine G6G and fluorescent tracers were added to the working fluid. The working area was illuminated by a pulsed laser with a wavelength of $532 \text{ nm}$. Illumination and recording were performed from the dry side of the glass plate. Achieved spatial resolution in the $X \to Z$ plane was $6.7 \text{ μm/pix}$. The position of the tracers in $Z$ direction was determined with an accuracy of $\pm 25 \text{ μm}$. A liquid film was formed at the Reynolds number $\text{Re} = 40$. Formed 3D waves were recorded in a certain position $20 \text{ cm}$ from the liquid film inlet. After that, obtained instantaneous velocity fields were phase averaged and interpolated into a regular grid.
3. Results

Obtained results show that the addition of a surfactant stabilizes the flow in a wide range of concentrations, as was mentioned in [2]. And for concentration of Triton X-100 in water within the range $20 \leq C \leq 200$ mg/l suppression of the wave motion on the whole film surface (up to 140 cm) for $Re = 44$ is observed. Wave patterns for $Re = 44$ and different surfactant concentrations are shown in figure 1. The results of experiments show that for $C = 10$ mg/l initially developed waves become indistinguishable at the bottom part of the test section and formation of the rivulets is observed (figure 1 (b)). On the contrary, for $C = 500$ mg/l, the film surface is almost all flat, and the waves become distinguishable only in the bottom part of the test section, therefore the shadowgraph images are not presented here.

The instant film thickness fields $h$, obtained by the LIF technique in the bottom part of the flow, are presented in figure 2. It can be seen in more detail the wave patterns for surfactant concentration, at which there is no strong attenuation of the wave motion in comparison with the case of the pure water flow. In particular, one can see the difference in the number of waves per 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. Another feature of the flow of water loaded with surfactant is more pronounced rivulet structures formation observed not only on the averaged in time film thickness distributions but even on instant film thickness fields.

![Figure 1](image1.png)

**Figure 1.** Shadowgraph images of flow at $Re = 44$ and different surfactant concentrations. (a) pure water, (b) $C = 10$ mg/l, (c) $C = 2000$ mg/l.

![Figure 2](image2.png)

**Figure 2.** Wave patterns at $Re = 44$. (a) pure water, (b) $C = 2000$ mg/l.

The discrepancy in the wave patterns between the flow of a film of pure water and water with a surfactant can also be observed when analyzing film thickness statistical and spectral characteristics (figures 3 – 5). For example, PDFs (figure 4) become symmetrical about the maximum value which at the same time becomes closer to 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. And for all investigated concentrations except $C = 2000$ mg/l, the variance (figure 3) is below the values for pure...
liquid. All these differences indicate the suppression of wave motion. The discrepancy in the power spectral density distributions (figure 5) consists not only in lower values but also in the displacement of the maximum value towards lower frequencies. This leads to a decrease in the slope of the distribution. At the same time, for some surfactant concentrations, there are frequencies in the power spectral density that are higher than for the case of the pure water flow.

Figure 3. Film thickness variance normalized to $h_N$ for different concentrations of surfactant: 1 – pure water, 2 – $C = 10$ mg/l, 3 – $C = 500$ mg/l, 4 – $C = 2000$ mg/l. $Re = 44$.

Figure 4. PDFs at $Re = 44$ and distance $X = 130$ cm for different concentrations of surfactant: 1 – pure water, 2 – $C = 10$ mg/l, 3 – $C = 500$ mg/l, 4 – $C = 2000$ mg/l.

Figure 5. Power spectral density for different concentrations of surfactant: 1 – pure water, 2 – $C = 10$ mg/l, 3 – $C = 500$ mg/l, 4 – $C = 2000$ mg/l. $Re = 44$, $X = 130$ cm.

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$^{-1}$ at $Re = 44$. The values of variance are close for both liquids (lines 1 and 4 in
figure 3), whereas PDF and power spectral density are quite different (lines 1 and 4 in figures 4 and 5). For these liquids, the PDF distribution has an 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 the damping of the waves. It should be also noted that for water with a concentration of Triton X-100 C = 2000 mg/l the characteristic relaxation time of the surface tension (about 0.003 sec) is less than almost all times of change in the film thickness (based on the frequency characteristic, see line 4 in figure 5), and this liquid can be considered as a liquid with physical properties modified by a surfactant.

Since the 3D wave motion was not suppressed only for the case of the surfactant concentration C = 2000 mg/l, this solution was chosen for the simultaneous velocity and film thickness measurements in the 3D wave. The reconstructed film thickness distribution of the 3D wave for the case of surfactant solution flow is presented in figure 6(a). The brightness of the background in figure 6(a) corresponds to the film thickness. The characteristic phase averaged velocity field in the 3D wave for the case of the surfactant solution film is shown in figure 6(b). The velocity field obtained for the layer with a thickness of 50 μm centred at the distance of 125 μm from the wall is shown. Backflows are not observed and flows along the capillary precursor minimum and maximum are weaker compared to the case of distilled water flow described in [7]. Differences in the flow structure may be related to the differences in waveform between the pure water flow and surfactant solution flow, in particular with the weakly expressed capillary ripple on the free surface in the case of the surfactant solution.

![Figure 6](image_url)

**Figure 6.** Reconstructed (a) surface of the 3D wave and (b) phase averaged velocity field at the distance 100 - 150 μm from the wall. Distilled water with surfactant (2000 mg/l). Re = 40; F = 14 Hz, X= 20cm

In conclusion, we note that for the fluid with the concentration of surfactant C = 2000 mg/l at Re = 44 the stable-state regime (characterized by the absence of changes of statistical and spectral characteristics) as for pure water is observed despite the depression effect of surfactant on wave motion. This is apparently due to the fact that the characteristic relaxation time of the surface tension (about 0.003 sec) is less than almost all times of change in the film thickness (based on the frequency characteristic, see line 4 in figure 5), and this liquid, in this case, can be considered as a liquid with physical properties modified by a surfactant. Despite the close values of variance for the solution with C = 2000 mg/l and pure water other statistical and spectral characteristics of the flow are different. These differences, apparently, are associated with differences (in comparison with pure water) in the flow structure and waves shapes at the initial stage of three-dimensional waves evolution, e.g., backflows is not observed and flows along the capillary precursor minimum and maximum are weaker.
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