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The Effects of Surfactant on the Evolution of a Thin Film under a Moving Liquid Drop

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ABSTRACTS

The effect of surfactant on the thickness of a thin film bounded by a solid surface and a moving liquid drop was investigated. We proposed a model so that parameters from the liquid drop can be stated in a parameter that acts as normal pressure to the thin film. Using the lubrication approximation, the model was reduced to a set of nonlinear partial differential equations in terms of the film thickness and surfactant concentration. Since we were interested in the role of the surfactant in lifting up the drop, we assumed that the density of the drop is higher than the density of the thin film. Numerically, the results show that the presence of the surfactant tends to delay the decrease of the film thickness insignificantly. However, when the surfactant was added into the system, it tends to significantly increase the film thickness for a certain range value of the normal pressure.

ARTICLE INFO

1. INTRODUCTION

In this paper, the effect of surfactant on the heights of the thin film bounded by a solid surface and a moving liquid drop is examined. The dynamics of the thin film is a main issue in the wetting of the surface by a liquid drop. Under some conditions, it was possible for a drop to approach a solid surface and then remain in a stable steady state separated from the surface by an ultra-

thin film, or rupture spontaneously and permit wetting of the surface by the drop. The problem can be observed in many industrial applications. As an example, in the petroleum industry, surfactants are commonly used to increase oil production from a reservoir by reducing the interfacial tension between oil and water (Zhang *et al.,* 2006; Lai *et al.,* 2010; Schwartz *et a*l., 1995), and/or alternating the wettability of rocks (Zhang *et al.,* 2006). Oil can release water-wet rocks surface more easily than oil-wet surface. In a case where the oil drop has detached from the rocks surface, the problem is whether the drop will or will not adhere back to the surface of rocks. Understanding this issue is important because getting back the oil drop to the rock surface is undesirable due to the need of increasing the oil production.

The effect of surfactant on the evolution of liquid thin film in various situations and with various parameters has been extensively studied by previous researchers. The presence of surfactant on the thin film bounded by a solid and gas was analyzed by De Wit *et al.* (1994), Schwartz *et al.* (1995), Schwartz, *et. al.* (1996), Danov *et al.* (1998), Rubinstein & Leshansky (2000), Pereira *et al.* (2007). A thin film bounded by a thick fluid layer containing surfactant was examined by Sharma & Ruckenstein (1986). The surfactant and the thin film formed between two liquid drops were studied by Duineveld (1996), Chester & Bazhlekov (2000). In general, the surfactant can reduce the surface tension that results in the Marangoni effect. Depending on the particular physical situation, the Marangoni effect can either stabilize or destabilize the thin film.

The derivation of the governing equations of the thin film can be found in many papers, such as Oron (1997), Myers (1998), O'Brien & Schwartz (2002), and Craster & Matar (2009). The governing equations consist of the Navier-Stokes equations and the continuity equation. The lubrication theory is then applied. In general, the derived models consist of a set of the fourth order nonlinear partial differential equations. For a thin film that is bounded by a solid substrate and a second fluid phase, the dynamics of the second fluid becomes involved; thus, the Stokes Equation has to be solved in two domains (Sharma & Ruckenstein , 1986 ; Yiantsios & Higgins , 1989; Fisher & Golovin, 2005).

In the present study, we proposed an approach in order to simplify the evolution system of two-phase thin film. We considered that the liquid drop as a second-phase fluid moved downward to the substrate with much slower velocity than its translational velocity, *i.e.* the velocity of the drop in unbounded domain. We defined the velocity of the drop. The viscosity and density of the drop are accommodated in the introduced velocity. This new approach allows us to exclude the Stokes Equation in the drop domain with considering the characteristics of the drop. The Marangoni effect on the dynamics of the thin film is then investigated.

We also examined the case when a surfactant concentration is added into the system. To the best of our knowledge, previous studies that take into account the additional surfactant concentration are limited. Danov *et al.* (1998) examined the influence of adding surfactant concentration to dynamics of thin film which is bounded by a substrate and a vapour phase. Liu *et al.* (2014) experimentally simulated the effect of surfactant and its additive on the vertical gas–liquid two-phase flow.

2. METHODS

This research was conducted utilizing the computational fluid dynamics. This is as a main method in fundamental research for predicting local fluid dynamics (Sumarji *et*

al., 2019). The computation is based on governed mathematical model.

A proposed mathematical model for evolution of the thin film incorporating surfactant is as follows. The liquid thin film flowing occurs between a horizontal plane and a liquid drop. The drop is nearly spherical with density $\hat{\rho}$, and viscosity $\hat{\mu}$. Meanwhile, the thin film has density ρ and viscosity μ. Let a be the radius of the area to which the lubrication theory is applied, and let x and y denote the horizontal and the vertical coordinates, respectively. The thickness of thin film is denoted by $h(x,t)$ where t is the time. The illustration for such a situation is described in **Figure 1**.

Here are assumptions:

1. The thin film and the drop are incompressible viscous Newtonian fluids, such that the 2-D creeping flow approximation is eligible.

2. The drop is symmetrical to the normal axis.

3. The drop moves perpendicular to the horizontal smooth solid surface. When near to the solid surface, the drop's velocity is much slower than when it is far away from the solid surface (it is explained later on).

4. Surfactant is insoluble and distributed at the interface of the thin film-liquid drop by convection.

5. Surfactant concentration is sufficiently small and only affects the drop surface without any complex dynamical or rheological effects.

Let $u(x, y, t)$ and $v(x, y, t)$ be velocities of the thin film fluid in horizontal and vertical directions, respectively. Based on Laaraba and Khechekhouche (2018), the conservation of mass is reads:

$$u_x + v_y = 0. \tag{1}$$

The momentum equations in lubrication approximation read:

$$p_x = \mu(u_{xx} + u_{yy}), \tag{2}$$

$$p_y = \mu(v_{xx} + v_{yy}), \tag{3}$$

where p is the pressure in the thin film. Subscripts represent the partial derivative, except it is explicitly stated.

Equations (1) – (3) are supplemented by the following boundary conditions:

i. At y = 0, no-slip conditions hold:

$$u = v = 0 \tag{4}$$

ii. At $y = h$, shear and normal stress balances are fulfilled:

$$\mu[(u_y + v_x)(1 + h_x^2) - 4h_x u_x] = \sigma_x \sqrt{1 + h_x^2} \tag{5}$$

$$2\mu[u_x(h_x^2 - 1) - h_x(v_x + u_y)] +$$
$$(-p + \hat{p})(1 + h_x^2) = \frac{\sigma h_{xx}}{\sqrt{1 + h_x^2}} \tag{6}$$

where σ is the surface tension and $\hat{p}$ is the pressure of liquid drop. It was also assumed that the pressure variations of the drop are dominant in the vertical direction.
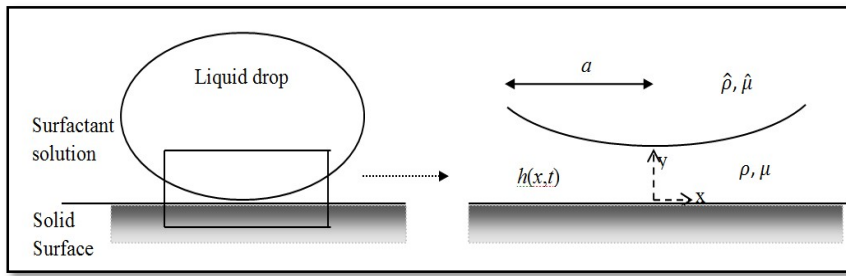
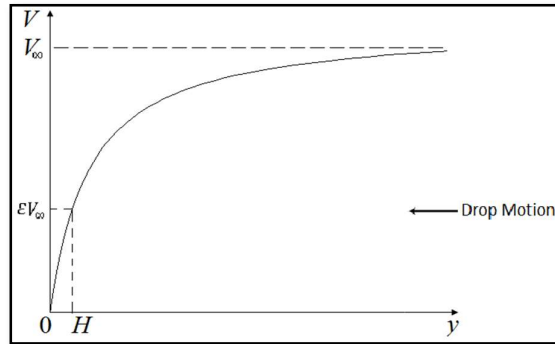**Figure 1.** The thin film between a solid surface and a liquid drop.

Figure 2. The sketch of the liquid-drop's vertical velocity.

Thus, we set $\hat{p} = -\hat{\mu}\hat{v}_y$ where $\hat{v}$ is the velocity of liquid drop that is moving normally to a solid surface. The minus sign denotes that the pressure acts downward. For the liquid drop's velocity $\hat{v}$, we proposed a model as follows. Far from the solid surface, the liquid drop, which has a density higher than the surrounding fluid, moves downward under its translational velocity. Based on Griggs *et al.* (2009), the translational velocity is defined by:

$$V_\infty = \frac{\lambda+1}{3\lambda+2}\frac{2ga^2(\rho-\hat{\rho})}{3\mu},\qquad (7)$$

where $\lambda = \dfrac{\hat{\mu}}{\mu}$ is the viscosity ratio. When the drop approaches the solid surface, the velocity decreases, and we proposed that the velocity $\hat{v}$ is given by:

$$\hat{v}(y) = \frac{V_\infty y}{H(\frac{1}{\grave{o}}-1)+y}.\qquad (8)$$

A sketch of the $\hat{v}(y)$ is described in **Figure 2**. The Equation (8) clarifies that the velocity of

the drop at infinity is V_∞ and at a certain height H is $\grave{o}V_\infty$, where $\grave{o} = \dfrac{H}{a} \square \ 1$. In other words, the velocity of the drop near the solid surface is smaller up to order $\grave{o}$ than the translational velocity. The benefit of knowing the proposed velocity of the drop is that it is not necessary to solve the momentum equation in the drop domain. Furthermore, for the liquid drop's velocity in the thin film region, we then approximate Equation (8) as:

$$\hat{v}(y) \approx \frac{\grave{o}V_\infty y}{H} + \frac{\grave{o}^2 V_\infty y}{H}(1-\frac{y}{H})+...$$

Therefore, the Eq. (6) becomes:

$$-p + \frac{2\mu(u_x(h_x^2-1)-h_x(v_x+u_y))}{(1+h_x^2)} =$$

$$\frac{\sigma h_{xx}}{(1+h_x^2)^{3/2}} - \hat{\mu}(\frac{\grave{o}V_\infty}{H} + \frac{\grave{o}^2 V_\infty}{H}(1-\frac{2y}{H})).$$

$$(9)$$

The kinematic condition at $y = h$ is stated by:

$$h_t + u\big|_s h_x = v\big|_s.\qquad (10)$$

In Equation (10), subscript s signifies that the quantity is calculated at $y = h$. The dynamics of surfactant at $y = h$ is based on the principle of mass conservation:

$$\Gamma_t + (u\mid_s \Gamma)_x = 0, \tag{11}$$

where Γ is the concentration of surfactant at interface $y = h$. In Equation (11), the diffusion term was neglected since chemical diffusivity was assumed to be small.

The surface tension variations are due to the change in surface concentration Γ, and are assumed to follow the linear law:

$$\sigma = \sigma_0 - E(\Gamma - \Gamma_0), \tag{12}$$

where σ_0 is the surface tension corresponding with concentration Γ_0 and E is the elasticity of the surfactant (Oron *et al.*, 1997). Let $\sigma_c = \sigma_0 + E\Gamma_0$ where $\sigma_c - \sigma$ is called the surface pressure. Equation (12) can be rewritten as:

$$\sigma = \sigma_c - E\Gamma. \tag{13}$$

Equations (1) - (13) are transformed into dimensionless forms by scaling: $x = ax^*, (y, h) = (Hy^*, Hh^*), (u, v) = (Uu^*, \grave{o}Uv^*),$
$t = (\dfrac{a}{U})t^*$ and $\Gamma = \Gamma_0\Gamma^*, \sigma = \sigma_c\sigma^*, p = Pp^*,$
where $P = \dfrac{a\mu U}{H^2}$. Equations (1)-(3) become (after ignoring the asterisk notations):

$$u_x + v_y = 0 \tag{14}$$

$$p_x = u_{yy} \tag{15}$$

$$p_y = 0 \tag{16}$$

For the boundary conditions, Equations (4) and (10) are still satisfied. Whereas Equations (9) and (5) become:

$$p = -h_{xx} + \frac{2}{3}\frac{\lambda^2 + \lambda}{3\lambda + 2}B_o(1 + \frac{1}{\grave{o}} - 2h), \tag{17}$$

$$u_y = -M\Gamma_x \tag{18}$$

with the Bond number $B_o = \dfrac{(\hat{\rho} - \rho)ga^2}{\sigma}$, and the Marangoni number $M = \dfrac{E\Gamma_0}{PH}$. We have used characteristic velocity $U = \dfrac{\sigma_c\grave{o}^3}{\mu}$. Note that $B_o > 0$ means the density of the liquid drop $(\hat{\rho})$ is higher than the density of the film (ρ), and vice versa for $B_o < 0$. For convenience, we write $\beta = \dfrac{4}{3}\dfrac{\lambda^2 + \lambda}{3\lambda + 2}B_o$. Since we are interested in the role that surfactant plays in lifting up the liquid drop, for the rest of discussion we will take $\beta > 0$.

The derivation for equations of evolution h and Γ is as follows. Integrating Equation (16), the pressure p is only dependent on x. Reconsidering this, Eq. (17) is valid in $[0, h]$. To find the velocity in the thin film, Equation (15) is integrated, then apply Equation (4) and condition (18). Hence:

$$u = -(h_{xxx} + \beta h_x)(\frac{1}{2}y^2 - hy) - M\Gamma_x y. \tag{19}$$

Using Leibnitz rules, Equation (11) can be written:

$$h_t + Q_x = 0 \tag{20}$$

where

$$Q = \int_0^h u\, dy. \tag{21}$$

Using Equation (11) and Equations (19) - (21), equations for the thickness of the thin liquid film and the surfactant concentration are:

$$0 = h_t - [\frac{1}{2}M\Gamma_x h^2]_x + [\frac{1}{3}h^3(h_{xxx} + \beta h_x)]_x, \tag{22}$$

$$0 = \Gamma_t - [Mh\Gamma\Gamma_x]_x + [\frac{1}{2}\Gamma h^2(h_{xxx} + \beta h_x)]_x. \tag{23}$$

We noted that for $\beta = 0$, Eqs. (22) – (23) are similar to Schwartz *et al.* (1995) and Schwartz *et al.* (1996), for the process of levelling thin liquid films that appears between a solid surface and gas, under the influence of a surfactant. Our model, $\beta = 0$ defines two parameters: first, the density of the liquid drop is equal to the density of the thin film or second, the viscosity of the liquid drop approaches zero (corresponding to gas). So, for $\beta = 0$ our model is in agreement with the works of Schwartz *et al.* (1995).

We initially approximated the thin film height as a quadratic polynomial and the surfactant concentration is uniformly distributed on the interface. The quadratic polynomial is chosen as the initial height, since we assumed that the liquid drop is nearly spherical. The initial conditions are then given by:

$$h(x,0) = K + \frac{P_1}{2}x^2,$$

$$\Gamma(x,0) = 1.$$

We also take the boundary conditions at both ends of the thin liquid film:

$$h_{xx}(\pm 1, t) = P_1, \tag{24}$$

$$(h_{xxx} + \beta h_x)(\pm 1, t) = \pm P_2, \tag{25}$$

$$\Gamma_x(\pm 1, t) = \pm C. \tag{26}$$

Parameter P_1 in Equation (24) represents the pressure magnitude at the end of the thin film, P_2 in Equation (25) describes the magnitude of pressure difference of the thin film when the surfactant is not present, and C in Equation (26) shows the magnitude of the adding rates of surfactant concentration at both ends of the domain.

3. RESULTS AND DISCUSSION

In this section, we presented numerical solution of the system (22)-(23) by the Finite Difference Method. We considered the computational domain $x \in [-1, 1]$, $t \in [0, 54]$. Based on Nurprasetyo *et al.* (2017), selection of the mesh element size should be conducted carefully in order to give an accurate result without consuming much time. Here, we selected the size of mesh $\Delta x = 0.1$ and $\Delta t = 3 \times 10^{-5}$ so that the numerical solution is stable and convergence. For initial and boundary conditions, we used parameters $K = 0.5$, $P_1 = 0.2$, and $P_2 = 0.1$. Those values can be replaced by other numbers as long as the lubrication approximation is valid (small H/a).

3.1. The Influence of the Surfactant

First, we consider the evolution of the thin film covered by surfactant and the redistribution of the surfactant concentration. In **Figure 3(a)**, we presented deformation of the thin film for $\beta = 1$, $M = 1$, and $C = 0$. We assumed that initially the curvature of the thin film is a parabola. In the initial stages of deformation, the curvature slightly changes. As time progresses, the curvature is very similar as before. The result is in accordance with those reported previously by Danov *et. al* (1999) that is due to the disjoining and capillary pressure the film drains without significant change of its radius and shape of the interfaces. It is also in agreement with Pozrikidis (1990) that there is a phase where the bottom part of the liquid drop maintains a nearly spherical shape during the motion towards a planar substrate.

The height of the thin film decreases uniformly at whole domain due to two parameters: first, the density of the liquid drop is higher than that of the thin film ($\beta = 1$), and second, a small amount of the fluid in the thin film is flowing out at the boundaries. As a result, the liquid drop directly moves toward the solid surface.

The result in **Figure 3(a)** also demonstrates that the liquid drop moves

toward the solid surface with reduced velocity. This behaviour is in a good agreement with the results of Pozrikidis (1990) indicating that as the drop approaches the wall, it slows down. The solid surface serves as a barrier to the liquid drop to move.

The redistribution of the surfactant concentration on the thin film surface is presented in **Figure 3(b)**. In this simulation, initially the surfactant concentration is uniformly distributed at the interface of the thin-film. In other words, its gradient concentration is zero. As the liquid drop moves with a slight change in shape, the surfactant convects out uniformly in the boundaries. As a result, the gradient of the surfactant concentration for $t > 0$ is almost zero.

To illustrate the effect of the presence of surfactant, in **Figure 4** we compare the dynamics of the thin film thickness at the center point $h(0)$, that is its lowest point,

for $M = 0$ and $M = 1$. Here, $M = 0$ denotes a surfactant free case, and vice versa. The solid line is for free-surfactant and the dashed-line is for covered-surfactant. The magnitude of the thin film height hardly differs from the free-surfactant case. The presence of the surfactant insignificantly delays the liquid drop in moving down. This result occurs because the gradient of the surfactant concentration almost vanishes (see **Figure 3(b)**). Thus, the stress of the Marangoni stress has a minor influence on the dynamics of the thin film.

To justify those descriptions theoretically, we considered the Equation (21). The surfactant parameter affects the dynamics of the thin film in the term $[\frac{1}{2}M\Gamma_x h^2]_x$. Thus, if $\Gamma_x \sim O(\grave{\text{o}})$ then its dynamics will be similar to the free-surfactant case, i.e:

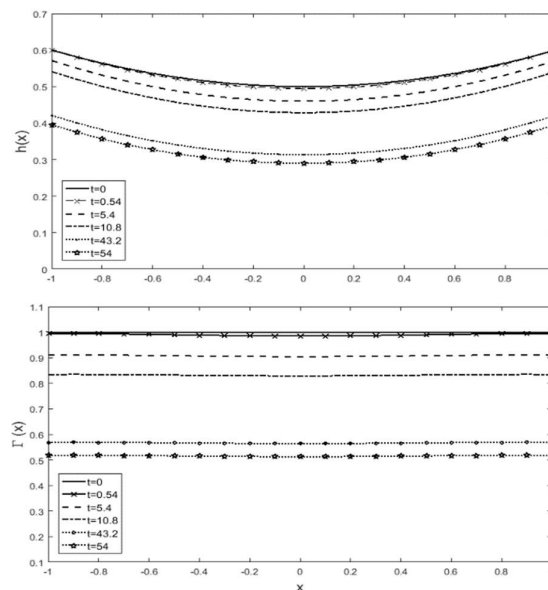$$h_t = O(\grave{\text{o}}) - [\frac{1}{3}h^3(h_{xxx} + \beta h_x)]_x.$$

Figure 3. The dynamics of (a) the height of the thin film, (b) the surfactant concentration, for $\beta = 1$,
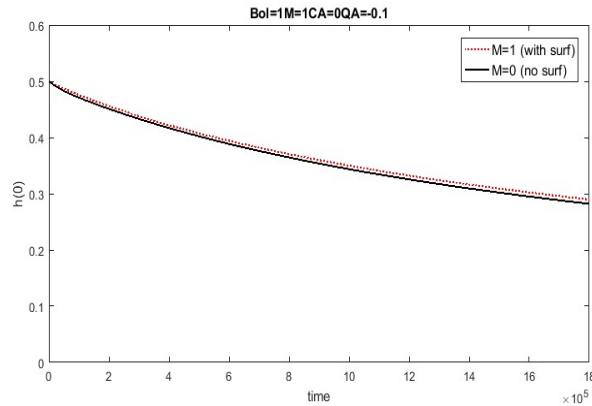
Figure 4. The dynamics of the thin film at $h(0)$ for free-surfactant (solid

This result is in a good agreement with the results of Danov *et al.* (1999) showing that when the surfactants are insoluble, they remain uniformly distributed throughout the drop surface during the film thinning, and interfacial tension gradients do not appear. As a consequence, the drainage of the film surfaces is not opposed by surface tension gradients, and the rate of film thinning is the same as in the case of pure liquid phases. The simulation is also similar to the result of Yulianti, *et al.* (2013) that the presence of insoluble surfactants does not have a significant effect on liquid drop movement.

3.2. Effect of the Surfactant Adding Rates

Based on the results in the previous subsection, we found that the presence of the surfactant with $\Gamma_x \sim O(\grave{o})$ does not have a significant influence on the thin film height. Therefore, in this subsection, we considered the addition of surfactant concentration at the boundaries in order to increase its gradient; i.e. $\Gamma_x(\pm 1, t) = \pm C \neq 0$.
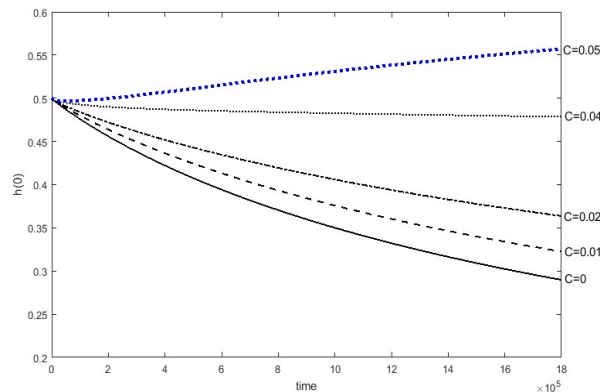

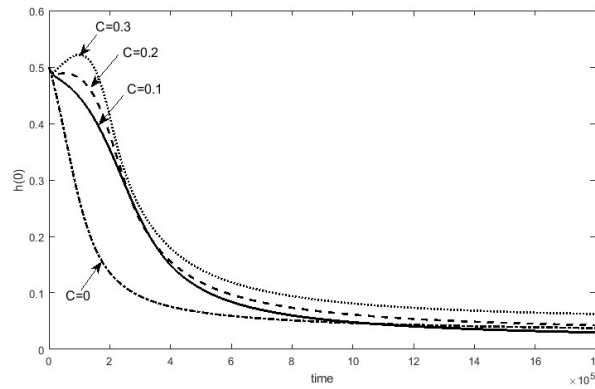**Figure 5.** The dynamics of thin film's height at $x = 0$ for $\beta = 1$ and different C.

Figure 6. The dynamics of thin film's height at $x=0$ for $\beta=3$ and different

To illustrate the effect of the surfactant adding rate to the height of the thin film, in **Figure 5,** we showed the dynamics of the thin film's height at $x=0$ for a constant $\beta=1,$ and different $C \neq 0$. For the case $C=0$, we already presented these results in Section 3.1. For the case $C \neq 0$, as the value of the adding rate increases, the height of the thin film also increases. For the case $\beta=1,$ the adding rate of surfactant can prevent the thinning of the film. This is because the adding rate of surfactant improves the gradient of the surfactant concentration, which leads to the Marangoni effect. For this case, the influence of the Marangoni effect becomes more important compared to the pressure from the Bond number.

Next, we considered the effect of adding rate surfactant to the height of the thin film with high bond number or viscosity ratio. **Figure 6** presents the dynamics of thin film height at the center point ($x=0$) for constant $\beta=3$ and some $C=P_2$. For values adding rate $C = 0, 0.1, 0.2$, the height of the thin film decreases monotonically. For $C = 0.3$, the height of the thin film increases for a moment; then, it decreases. It can be clearly seen that for $\beta=3$, even though the adding rates of surfactant is increased, the height of the thin film continues to decrease. Here, we observed that for high Bond number or high viscosity ratio (high value of β), the adding rate of surfactant cannot prevent the rupture of the thin film. This result aligns with the result of Chester & Bazhlekov (2000), indicating that by increasing the viscosity ratio, the effect of surfactant becomes weaker.

Based on results that are presented in **Figures 5 and 6**, there is a limitation of β value so that the surfactant adding rates work effectively to lift the liquid drop. This is due to the surfactant adding rates only working at the end points, whereas β, which acts according to the normal pressure from the liquid drop, works in the entire domain $x \in [-1, 1]$. So, the adding rates of the surfactant are not able to lift the heavy liquid drop.

4. CONCLUSION

This paper has presented a model and numerical explorations of the influence of the insoluble surfactant on the dynamics of thin film between a solid surface and a liquid drop. We set the density of the liquid drop higher than that of the thin film so that gravity caused the drop to move towards the solid surface. It has been numerically demonstrated that the presence of the surfactant delayed the decrease of the thin film height insignificantly. Meanwhile, the surfactant adding rate significantly increased the thin film's height, but for a certain range of bond number value and viscosity ratio.

5. ACKNOWLEDGEMENTS

6. AUTHORS' NOTE

The author(s) declare(s) that there is no conflict of interest regarding the publication of this article. Authors confirmed that the data and the paper are free of plagiarism.

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