Research on Spontaneous Diffusion and Fragmentation of Liquid Droplets Caused by Marangoni Effect

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Abstract. The Marangoni effect is important to drying silicon wafers, the fields of welding and improving engine liquid fuel efficiency. In this paper, we investigate the Marangoni flow caused by the evaporation of droplets of alcohol solution, which eventually causes the droplet "atomization" phenomenon. The Marangoni convection phenomenon was studied in terms of temperature and droplet concentration, and the changes of droplet diffusion and the degree of droplet "atomization" were investigated after droplets of different volume concentrations of Isopropyl Alcohol (IPA) were added to different solutions.

Keywords: Diffusion, Marangoni, alcohol, atomization.

1. Introduction

When a drop of a water mixture such as water-alcohol is deposited on the surface of a hydrophobic liquid (e.g. vegetable oil), the resulting drop may sometimes fragment into smaller droplets. This phenomenon is identified to be the result of the Marangoni effect[1,2]. The general effect was first noticed in the so-called "tears of wine" by physicist James Thomson[1] in 1855. Researchers believe that this effect is due to the evaporation of the liquid droplets making the result of changes in the temperature and concentration of the liquid. These two types of factors lead to local variations in the surface tension of the liquid, resulting in large surface tension gradients between different phases. The evolution of Marangoni flow[3] of a liquid in another liquid that does not mix with it did not receive attention until the early 22nd century, and the study is of great significance for the chemical field which can even be involved up to drying silicon wafers, the fields of welding,crystal growth and improving engine liquid fuel efficiency[4,5].

Our main focus is on spontaneous Marangoni flow caused by the evaporation of alcohol substances in a short time, and this article will rely on the natural volatilization of alcohols, explore the parameters of Marangoni convection phenomenon, and combine the actual exploration of the effect of alcohol concentration on the degree of atomization of small droplets.

2. Theory and Experiment

During the process, the droplet appears to spread and then shrinks.The droplet diffusion occurs because the surface tension of the droplet decreases due to the diffusion of the alcohol from both temperature and concentration, creating a surface tension gradient with the oil phase, resulting in a Marangoni effect that causes the droplet to "explode".

1. It is assumed that water is not evaporated and the evaporation rate of alcohol is constant for solution evaporation dissipation;

2. In the flow process, most of the viscous dissipation occurs in the oil layer. Assuming that droplets experience advective flow, the effect of viscous stress in the oil layer and droplet viscous stress is analyzed;

3. Since the thickness of the droplet is small and not easily measured, we assume that the thickness is equal everywhere and that the Marangoni "ridge" is considered as a regular blob.

Due to the deformation of the droplet, the edge of the droplet is subjected to surface tension from each of the three phases acting on the edge of the droplet in Figure 1(a).

2.1 Droplets explode

Figure 1(b) shows that due to the natural evaporation of droplets, under the action of surface tension gradient, the formation of Marangoni flow diffusion will inevitably form a radial outward pull droplet tangential stress, and we call it "Marangoni stress".
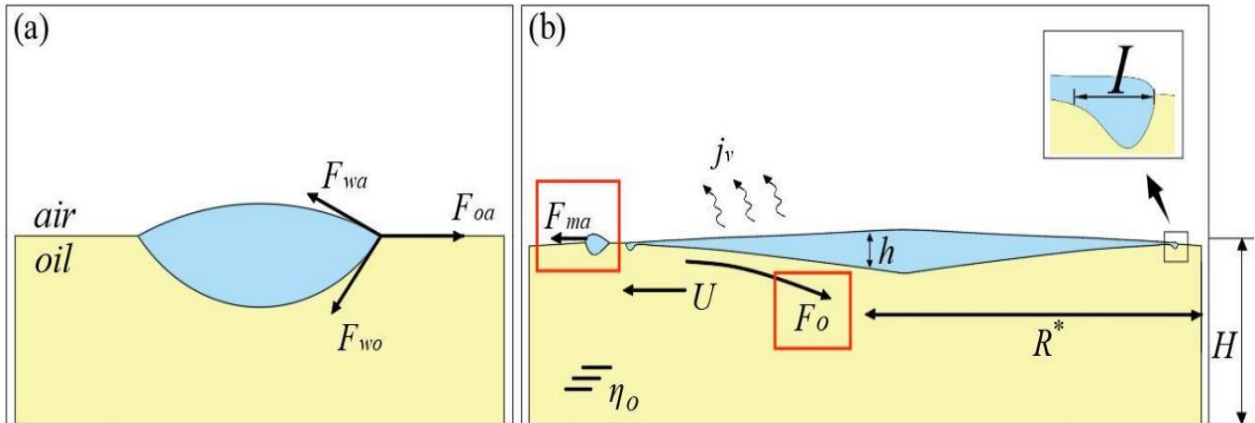

Figure 1(a). F_{wa} is the surface tension created between the air and the droplet, F_{wo} is the surface tension created between the oil and the droplet, F_{oa} is the surface tension created between the oil and the air; Figure 1(b). F_{ma} is the force that spreads the droplet, F_0 is The viscous resistance created by the oil, R^* is the maximum outer diameter during droplet diffusion, U is the rate of motion at the edge of the droplet, η_0 is viscous resistance coefficient, j_v is Evaporation rate, H is oil thickness;h is droplet thickness, I is small droplet diameter.

The power series γ varies with diameter as follows:

$$\gamma = \gamma_0 + \sum_{i=1}^{n} a_i \frac{\gamma_c - \gamma_0}{R^{*i}} r^i (\sum_{n=1}^{\infty} a_i = 0) \tag{1}$$

At γ , the surface tension coefficient gradient:

$$F_{ma} \sim grad = \sum_{i=1}^{n} i a_i \frac{\gamma_c - \gamma_0}{R^*} \sim \frac{\Delta\gamma}{R^*} \tag{2}$$

R^* is the maximum outer diameter during droplet diffusion and γ is the surface tension coefficient.

For the action of F_{ma}, the rate of outward diffusion of droplets U increases, resulting in an increase in viscous resistance.

$$F_0 \sim F_{ma} \qquad \eta_0 U/H \sim \Delta\gamma /R^* \tag{3}$$

The evaporation dissipation of droplets is analyzed from two angles: solvent volume conservation.

$$(\phi_0 V_0 - \phi_c V_c) = j_v R^{*2} t \tag{4}$$

$$(1 - \phi_0)V_0 = (1 - \phi_c)V_c \tag{5}$$

where ϕ_0 and V_0 are the concentration and volume of droplets at the initial moment, respectively. ϕ_c and V_c are the concentration and volume of droplets at a certain moment during the droplet explosion.

Obtained by (3)(4)(5):

$$R^* = (\frac{\Delta\gamma H}{\eta_0}t)^{1/2} \tag{6}$$

$$U \sim \sqrt{\Delta\gamma H}/\sqrt{\eta_0 t} \qquad (7)$$

2.2 Small droplet size

"Marangoni ridge" theory: because the larger the droplet thickness, the greater the width I of the "Marangoni ridge", and the two have a simple linear relationship.

$$d = I \sim h = U/\pi R^{*2} = U\eta_0/\pi\Delta\gamma H t \qquad (8)$$

2.3 Simulation

According to the above theoretical formula, it can be known that the magnitude of the concentration is proportional to the strength of the Marangoni flow to characterize the effect of the surface tension gradient on the Marangoni flow.
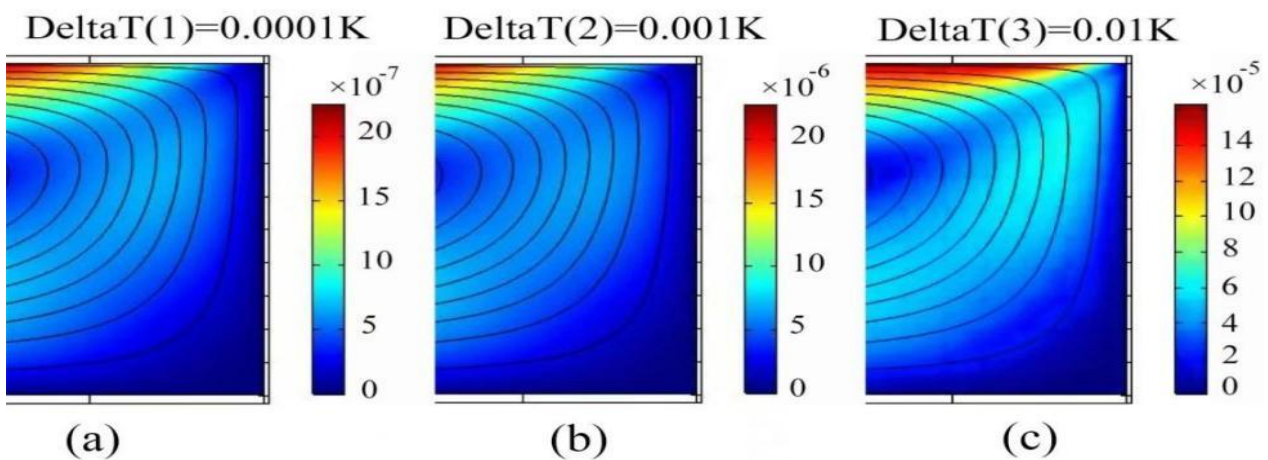

Figure 2. Simulation results for different temperature gradientsSet different temperature gradients.

According to the comparison of the flow field values among the Figure 2(a), (b), (c), the higher the temperature DeltaT, the greater the flow field velocity.

In experimental study, The control variable method was used to study the diffusion of isopropyl alcohol (IPA) droplets, and IPA solutions with different volume concentrations (CBM%) were dropped in the center of the Petri dish containing vegetable oil, and the data were recorded with software.
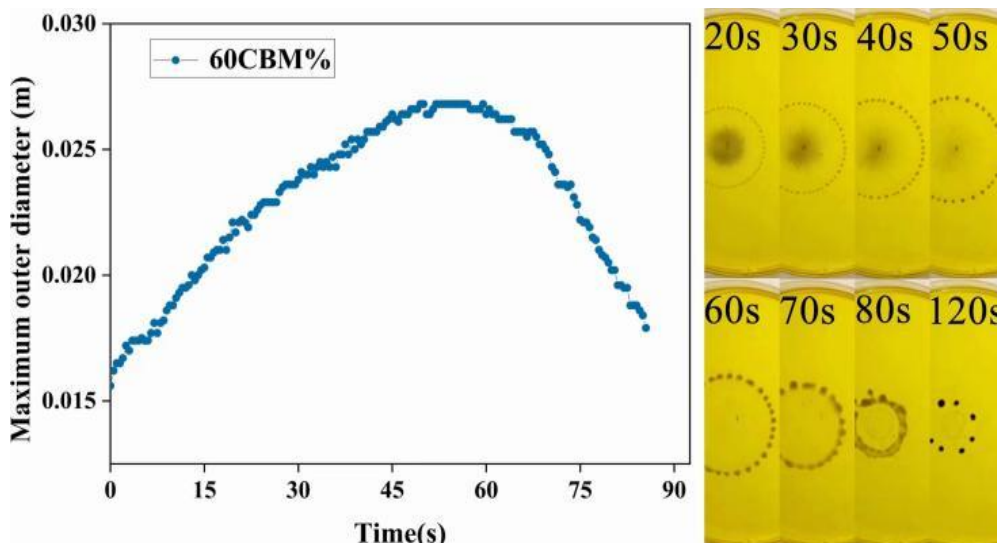

Figure 3. Data acquisition at 60 CBM% maximum outer diameter

As shown in Figure 3, the maximu outer diameter of the first half of the droplet process of 60 CBM% is slowly increasing, and the second half shrinks sharply, mainly studying the time when the Marangoni stress dominates, and the data are fitted in formula (6), and the results obtained are shown in Figure 4(a).
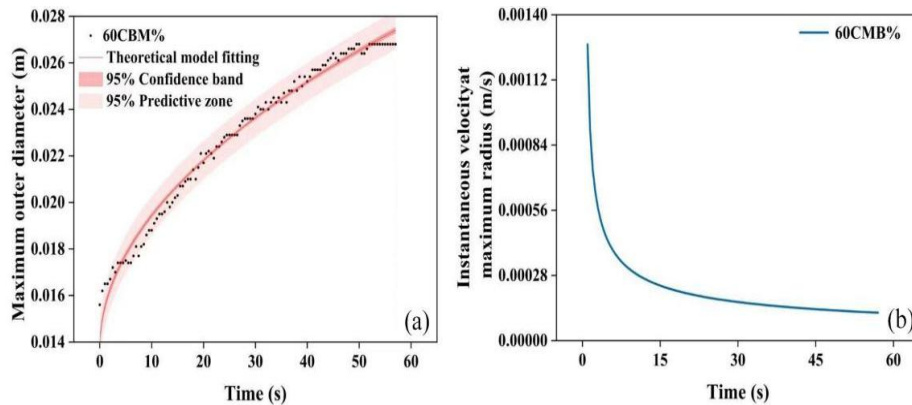

Figure 4(a). It shows the experimental data shown in the relationship between R^* and diffusion time of the 60 CBM% droplet fits well with Equation (6); Figure 4(b). It shows the relationship between the U of the 60 CBM% droplet and the diffusion time of the experimental data shown is consistent with the experimental phenomenon: the droplet spreads quickly first, and then diffuses slowly.
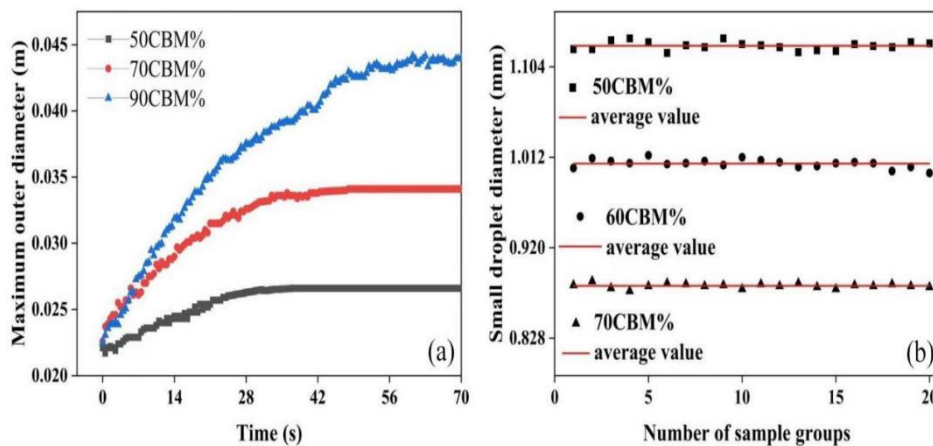

Figure 5(a). It shows the R of different CBM% over time; Figure 5(b). It shows the size of small droplets that diffuse into different CBM% of droplets.

In the experiment, the droplet does not explode below 37~40 CBM%, which indicates that the surface tension gradient formed by the dynamics can not overcome the resistance of the oil. When the CBM% is higher than 50% , droplet explosion occurs significantly. At the same time, the higher the initial CBM% of droplets, the larger the outer diameter of droplet diffusion, as shown in Figure 5(a). The same experimental conditions, ethanol CBM% up to 85~90% before the explosion, and fragmentation is not obvious enough. The use of silicone oil (viscosity coefficient is much larger than vegetable oil) found that no matter how high the CBM% of IPA solution will not appear droplet explosion phenomenon, "Ethanol" and "silicone oil" respectively from the reduction of Marangoni stress and strengthen the viscous resistance of two aspects to prove: CBM% and viscous resistance coefficient is an important condition for the droplet to diffuse autonomously.

The droplets were divided into eight equal parts, a total of 20~22 final summations were taken and averaged, and the data statistics results were shown in Figure 5(b).As can be seen from Figure 5 （b）, the larger the CBM%, the smaller the diameter of the small droplets. That is, as evaporation proceeds, the surface tension of the small droplets gradually converges to that of water. The size of the small droplets after fragmentation depends mainly, on the strength of the Marangoni flow. If the degree of flow is stronger, the viscous drag will tear the droplets in the travel path and form the

fragmented droplets. Conversely, the weaker the flow, the less likely the droplets will be torn apart. Therefore, when the initial droplet concentration is increased, the diameter of the "torn" droplets will inevitably be smaller.This phenomenon conforms to the theoretical formula (8).

3. Conclusion

From the above, it can be concluded as follows: The cooling caused by alcohol evaporation, the diffusion time is squared with the instantaneous maximum diffusion radius. The CBM% and viscous resistance coefficient is an important condition for the droplet to diffuse autonomously. The size of the small droplets atomized by the droplets is maximum when the droplets spread, and the maximum value is positively correlated with the concentration in the concentration range where small droplets can be formed. In the standard condition, $H/\eta_0 \approx 0.17 \text{mm} \cdot \text{Pa} \cdot \text{s}$, the diffusion of isopropanol solution CBM% reaches 37~40 CBM%, and the volume concentration exceeds 70~75CBM, and the spontaneous "atomization" of small droplets is not obvious. Diffusion occurs when the volume concentration of the ethanol solution reaches 85~90 CBM%.

Acknowledgement

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