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Evaporation progress of macroscopic-scale droplets on heated substrates

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Abstract. Evaporation progress of a macroscopic-scale sessile droplet with the pinned triple line on heated isothermal substrates has been experimentally investigated in the terrestrial gravity, in order to study the interface effect, heat and mass transfer behaviours during the phase change process. The experiments were carried out in a closed chamber in which the environment temperature and pressure were regulated. The contact radius, liquid volume and contact angle during evaporating were observed to study the influence of the gravity effect on the drop shape. The instant evaporation rate was calculated and compared with the theoretical prediction to analyse the coupling influence of diffusion and thermal convection. The effect of substrate temperature on the heat flux were also focused on. It was found that the evolution of heat flux density could be separated into four stages, began with the rapid increase and warm up, then switched to the long-time stable stage and ended up with the final rapid decrease stage.

1. Introduction

Sessile liquid droplet evaporation has attracted more and more scientific interests owing to its increasing engineering applications in industry and space thermal management system, such as spray drying, DNA mapping, direct-ink-write 3D printing. Many novel experiments were performed on ground and in microgravity environment since the important research discovery of "coffee ring" [1]. Erbil [2] and Kovalchuk et al. [3] gave thorough reviews of research on droplet evaporation.

Most previous studies pay attention to the evaporation rate, i.e., mass transfer during evaporation. Based on quasi-steady diffusion-limited evaporation, the theoretical predicted evaporation rate given by Hu and Larson [4] is widely adopted:

$$-\rho \frac{dV}{dt} = \pi R D c_{\rm sat} (0.27\theta^2 + 1.30) \tag{1}$$

where ρ , *V*, *t*, *R*, *D*, c_{sat} and θ represent the liquid density, droplet volume, time, contact radius, vapor diffusion coefficient, saturation vapor concentration and the contact angle respectively. It should be noted that this formula is based on evaporation experiments of a small water droplet on the non-heated substrate and only diffusion is considered. Here, we classify whether a droplet is small or large according to the relative magnitude of contact radius *R* and capillary length L_c which is defined by:

$$L_c = \sqrt{\frac{\sigma}{\rho g}} \tag{2}$$



where σ and g represent the surface tension coefficient and gravitational acceleration respectively. A droplet is defined to be large if $R > L_c$, otherwise it is small. An equivalent non-dimensional description is given by Bond number *Bo*:

$$Bo = \frac{\rho g R^2}{r} \tag{3}$$

As mentioned before, evaporation is a coupling process of heat and mass transfer. In the aspect of heat transfer, heat flux transferred from the substrate to the liquid droplet is commonly used experimentally. A strong increase of the heat flux density was observed during the evaporation of ethanol droplets while the microgravity duration created by parabolic flights was too short to observe a constant value [5]. Carle et al. [6] verified existence of the constant value after the beginning increase of heat flux density. Sobac [7] found that there was a constant heat flux density only in the situation of pinned triple line. A further study showed that there were four stages during droplet evaporation: warming-up, evaporation with thermal-convective instabilities, transition and rapid evaporation [8]. Chen et al. [9] found that the evaporating process of the drop on a substrate with lower conductivity was easier to have a stable heat flux density.

There are already many studies about the evaporating liquid droplet, which focus on the contact line motion, shape evolution, convection and the hydrothermal waves inside the liquid. It is noted that most previous studies on heat transfer are confined to small droplets, which means that the radius smaller than the capillary length, i.e., $R/L_c<1$. As far as the author knows, the reported maximum value of R/L_c is 2.6 [6]. Droplets with varying scales show different wetting behaviors [10], which have relatively significant influence on the evaporation process.

Undoubtedly, macroscopic-scale droplets are unique in natural life and industry application. The evaporation phenomenon of a sessile "big" drop on the heated substrate is a complex problem which involves behaviors of the triple line, heat transfer with thermal conduction and convection, mass transfer into the vapor phase. Recently, developing applications of the liquid droplets in space also demand clear knowledge and full identification of the physical mechanisms. In present work, evaporation progress of a macroscopic-scale sessile droplet on heated isothermal substrates has been experimentally investigated in the terrestrial gravity, using the qualification model onboard Chinese SJ-10 recoverable scientific satellite, we extend R/L_c to 3.5, for the framework of studying the vapor-liquid interfacial effect, heat and mass transfer behaviours during the phase transition process.

2. Experimental facility

Schematic diagram of the experimental apparatus is shown in Figure 1. Briefly, the definitive instrument with the size of 500×300×300 mm³, consists mainly of the central experimental module, liquid injection module, optical observation module, environment monitor and the electrical control module, in which the liquid drop could be injected to create as the settled volume, observed during evaporating progress at controlled conditions. Ethanol is selected as the working fluid of this experiment due to its volatility and contamination resistance. For the central experimental module, a substrate made of aluminum is used to create and sustain the evaporating drops. The substrate could be heated from the bottom by a thin-film heater and two inserted thermocouples with PID control loops to achieve a stable temperature with an accuracy of ± 0.2 °C. In present experiments, the top surface of the substrate is covered by thin polytetrafluoroethylene (PTFE) sheet with a thickness of 100 µm and a diameter of Φ12mm for the overall consideration of relatively small surface energy and thermal resistance. That is to say, a relatively big liquid drop could be created on the surface of the substrate, enough heat could be supplied quickly from the bottom of the substrate for the liquid drop evaporation. To measure the heat transfer resulted by evaporation and thermal flow, a 400µm-thick heat flux sensor with a sensitivity of better than $0.3\mu V/(W/m^2)$, an accuracy of better than 3% of the indication, is fixed just below the top PTFE surface to measure the instantaneous heat flux distribution between the liquid drop and the solid substrate during drop evaporation.

The sessile liquid drops are injected and created through a 700 μ m hole inside the substrate by the liquid injection module. Ethanol is stored in a liquid reservoir. The electrical control module could

control a step motor to regulate the injection rate and volume. In present experiments, the injecting rate is controlled ranging from 5 μ L/s to 30 μ L/s to achieve steady drop creating and maintain. The shapes of liquid drops during evaporation are acquired to calculate the evaporating rates. In order to obtain clear images of the liquid drops, a CCD camera with a resolution of 1024*768@25fps and background light are set before and behind the drops, respectively. The pictures acquired by the CCD camera are transferred to image recorders for computer analysis after the experiments.

All the experiments are carried out in a closed chamber to isolate the vapor flow around the liquid drop, also the environment temperature and pressure could be PID controlled to stay stable during the evaporation. It is noted that the environment humidity is also regulated to be quasi-steady. The present experiments are carried out in steps as follows, after creating a sessile droplet by injecting liquid with controlled volume V_0 , ranging from 30.0μ L to 120.0μ L, on the heated substrate with PID-controlled temperature T_s , the images of free evaporating process of the droplet will then be acquired from side view using a high resolution CCD camera, and geometric parameters like the liquid drop shape and its contact angle along the triple line will be record, which can be calculated the evaporating rate of the droplet. Additionally, a heat flux-meter and some thermocouples, integrated just below the evaporation. Three fans are used to refresh environment after each experiment. The delivered signals, such as temperature, heat flux, humidity and pressure are recorded synchronously by a data acquisition system.



Figure 1. Schematic diagram of the experimental apparatus.

3. Results and discussion

Experiments of the pinned ethanol droplets on the heated PTFE surface have been carried out for the framework of analyzing the mass and heat transport characteristics during the evaporation progress in the terrestrial gravity. The shape evolution, the instant evaporation rate and the heat flux evolution of the evaporating liquid droplets are focused on.

3.1. Shape evolution

After acquiring the side-view images of evaporating drops, the contact radius R, the drop volume V and the contact angle θ could be calculated by the image-analyse software automatically and accurately. The results are normalized by initial parameters (R_0 , V_0 , θ_0) as shown in Figure 2. For large sessile droplets with pinned triple line, of which the contact radius is greater than the capillary length, results reveal that the contact radius keeps relatively constant during the almost lifetime, with the $R/R_0=0.96$ corresponding

to the end of lifetime $t/t_f=0.85$, that is to say, the triple line of the droplet is pinned to appear the CCR (constant contact radius) mode. It is also found that the drop volume and the contact angle decrease linearly with time, exhibiting constant global evaporating rate (dV/dt), which has the similar tendency for small evaporating liquid drop with $R/L_c < 1$.



Figure 2. Droplet volume, contact radius and contact angle evolution with $V_0=95.5\mu$ L.

3.2. Instant evaporation rate

The evolution of droplet instant evaporation rate can directly reflect the stability of thermal management. For the small droplet, the triple line will go through the stages of pinning and depinning successively, and the volume does not decrease linearly during the whole lifetime, that is, the instant evaporation rate can't remain constant during the whole evaporation process. As mentioned above, the quasi-static diffusion model proposed by Hu and Larson underestimated the instant evaporation rate of large droplets attached to the substrate, therefore, Kelly-Zion [11] and Carle F [12] proposed modified empirical model to take both vapor diffusion and natural convection in the air into account.

The instant evaporation rate during evaporating progress is shown in Figure 3, comparisons are also performed with the theoretical values predicted by Hu's, Kelly-Zion's and Carle's model. In present experiments, it is found that the instant evaporation rate increases firstly and then keeps stable until the end of the lifetime. The present instant evaporation rate basically maintains a constant value, while the predicted values of the three theoretical models decreases gradually with the evaporation process. In present evaporation progress, the Diffusion-limited evaporation model underestimates the instant evaporation rate of large sessile droplets with the pinned triple line as big as 29%. The quasi-static diffusion model ignores the contribution of buoyancy convection, which enhances the energy transport in the droplet, thus underestimating the heat transfer from the base to the gas-liquid interface, and leading to predicting the instant evaporation rate too small. While for the empirical models considering both diffusion and natural convection deriving from typical liquid, the accuracy of predicted evaporating rate by 15% for Kelly-Zion's model, which uses the alkane for study.



Figure 3. Evolution of instant evaporation rate with V_0 =113.8µL: experiment and model prediction.

3.3. Heat Flux Density Evolution

Heat flux density evolution represents the energy transport during droplet evaporation. The typical result is presented in Figure 4. It is found that the evolution of heat flux density could be separated into four stages:

Stage 0: There is a non-zero stable heat flux density. The substrate temperature is regulated to be stable before droplet injection. Since $T_s > T_a$, the substrate provides heat to air by natural convection. Clearly, heat transfer modes include heat conduction and convection, which are directly relative to the substrate material and have a positive correlation with the value of T_s - T_a [13]. In addition, the value of heat flux lost by the natural convection in air is positively related to the gravity level. This is verified by experiments carried out on ground [14] and in hypergravity environment [5]. We can deduce that the nonzero value in microgravity will be smaller than that on ground.

Stage 1: Once droplet injection starts, droplet absorbed energy from the heated substrate, as a result, heat flux density gone up dramatically. Liquid spreads until the triple line pins at the edge of substrate, subsequently, the contact angle and droplet height increase until max volume is achieved. It is found that the heat flux density still increases after the droplet volume achieving the pre-set maximum volume. Liquid temperature in reservoir is colder than substrate temperature. Once liquid touches substrate to form a droplet, substrate will provide heat to the droplet to keep temperature continuous at solid-liquid interface. During droplet formation, not all heat supplied by the hot substrate is used to warm droplet and part is used to evaporate injected liquid. At the moment that the droplet volume reaches a maximum, the newest injected liquid is still colder than substrate temperature, resulting an inhomogeneous thermal state, which is supposed to be the reason why there is a hysteresis between max volume and max heat flux density.

Stage 2: This is the main stage in which the heat flux density keeps stable during the droplet evaporating. Because the instant evaporation rate is constant, the heat flux, which reflects the heating power of the substrate mainly used to evaporate droplet in the form of mass loss, is also basically stable, which is similar to the investigations of small droplet evaporation [4] and droplet evaporation in microgravity environment [6]. Sobac [8] observed the thermal-convective instabilities induced by the thermocapillary flow near the triple line of small droplet in microgravity, while the flow observed in present big pinned droplet belongs to buoyancy flow which derives from large contact radius.

Stage 3: Heat flux density decreases rapidly to a level nearly equal to that in Stage 0. Triple line recedes until the complete vanishing of residual liquid film. Sobac [8] observed the destabilization of

the central part of the drop flow and complete disappearance of the hydrothermal waves. It is supposed that there is a process of destabilization and successive disappearance of buoyancy flow due to the transition from pinning stage to depinning stage.



Figure 4. Heat flux density evolution (T_s =45.5 °C, V_0 =113.8µL).

3.4. Influence of substrate temperature

As mentioned above, it could be seen that the substrate temperature plays an important role in droplet evaporation process. The dependence of heat flux density evolution on the substrate temperature T_s is investigated and showed in Figure 5. As T_s increases, the curve of heat flux density becomes narrower and taller, indicating that the lifetime of droplet is shorter and heat supply is strengthened. This benefits from the enhancement of heat transfer contributing by thermocapillary flow at gas-liquid interface and buoyancy flow in gravitational environment at higher T_s . At the same time, the nonzero stable value of heat flux density in Stage 0 is larger at higher T_s . Besides, the integration of heat flow density curve at Stage2 with respect to time are roughly equal under the same initial volume V_0 even at different substrate temperature T_s , which is consistent to the fact that the total amount of heat absorbed from the substrate during the droplet evaporation process is equal for same-volume drop.



Figure 5. Dependence of heat flux density evolution on the substrate temperature ($V_0=73.0\mu$ L).

4. Summary

In present research, experimental investigations of the pinned macroscopic droplet during evaporating process were carried out using the qualification model onboard Chinese SJ-10 recoverable scientific satellite. For large sessile droplets with the pinned triple line and the contact radius greater than capillary length, results revealed that the drop volume decreased linearly with time. The triple line was pinned during most of evaporation time and no CCA (Constant Contact Angle) stage was observed. It was also found that the instant evaporation rate increased firstly and then kept stable until the end of the evaporation lifetime. In present experiments, the instant evaporation rate basically maintained a constant value, while the predicted values of former theoretical models for the instant evaporation rate gradually decreased with the evaporation. The Diffusion-limited evaporation model underestimated the evaporation rate of large sessile droplet with pinned triple line owing to ignoring the contribution of the buoyancy flow, while the accuracy of other empirical models considering both diffusion and natural convection depended on the liquid significantly. Four stages of the heat flux density evolution could be seen: an initial nonzero stable value representing heat supply from the substrate to environment, dramatical increase representing warming up the injected droplet by the hot substrate, long-time stable evaporation consistent with triple line pinning and a final rapid decrease representing evaporation with triple line shrinking.

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