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Pool boiling of a non-azeotropic binary mixture under microgravity

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Abstract—Pool boiling experiments with non-azeotropic water–ethanol mixtures were conducted under 10-second microgravity conditions available from a drop shaft facility with a 490 m free fall. Comparing with the terrestrial condition, the boiling heat transfer was enhanced under microgravity. The observation from the back side of a transparent glass plate heater suggests that the enhancement could be ascribed to the Marangoni flow induced by an ethanol concentration gradient due to preferential evaporation of ethanol along the surface of enlarged vapor bubbles detached from, but still kept close to, the heater surface under microgravity.

INTRODUCTION

Boiling heat transfer will play a dominant role in future thermal management in space. In addition, information on boiling phenomena under microgravity should also be important in the processing of many materials in space where distillation, extraction, electrolysis, etc., are involved. The early studies on microgravity pool boiling up to the mid 1960s are described by Siegel [1], and more recently Merte [2] summarized boiling heat transfer characteristics both in elevated and reduced gravity conditions. Straub and his co-workers have extensively carried out microgravity pool boiling experiments for the last 15 years or so [3], and they currently propose a 'micro-wedge model' for describing the primary mechanism of boiling heat transfer under microgravity [4].

The present authors have also conducted a series of experiments of pool boiling on plate heaters for single-component liquids, namely *n*-pentane, CFC-113 and water, under reduced gravity conditions, using parabolic flight of an aircraft [5–7], ballistic trajectory of a sounding rocket [8], and free-fall in a drop shaft [9]. Our understanding from the experiments is, in general, summarized as follows.

Organic fluids

(1) Bubbles generated on the heater surface in subcooled liquid are enlarged to the size at which condensation and evaporation mass transfers are balanced, while continuously attaching to the heater surface with a large contact area. As the heat flux at the surface increases, the bubbles begin to sweep across the surface and coalesce with each other.
(2) Gravity reduction does not have an appreciable influence on heat transfer in a low heat flux region (the isolated bubbles regime), since the latent heat transport at the base of enlarged bubbles attaching to the heater surface and/or at the liquid/bubble/heater interline region is considered to work efficiently under reduced gravity [5].
(3) The heat transfer in a high heat flux region (the coalesced bubbles regime and the vapor slugs regime) deteriorates increasingly with a reduction in gravity. This can be explained by the fact observed with a transparent heater that a reduction in gravity diminishes the liquid supply to the heater surface beneath coalesced bubbles and/or vapor slugs. In the aircraft experiments, in which the gravity was reduced to the order of $10^{-2}g$ (g denotes terrestrial gravity), a network of liquid canals was formed on the heater surface covered by each coalesced bubble or slug [6, 7], but the bases of coalesced bubbles were completely dry in the sounding rocket experiments in which the gravity was reduced to the order of $10^{-4}g$ [8]. The influence of gravity reduction on the heat transfer deterioration becomes more significant with a reduction in the subcooling in the bulk of the liquid.
(4) The transition from the isolated bubbles regime to the coalesced bubbles regime occurs at lower heat flux as gravity reduces. This tendency is more pronounced at smaller subcoolings. As a result,

NOMENCLATURE

F	fraction of bubble/heater contact area on the heater surface
g	terrestrial gravitational acceleration
q	heat flux.

Greek symbols

α	boiling heat transfer coefficient
ΔT_{sub}	subcooling of bulk liquid
ΔT_{sat}	heater surface superheat.

Subscripts

μg	under microgravity
$1g$	under terrestrial gravity.

stable saturated nucleate pool boiling can be maintained only in a very restricted heat flux region under a good quality microgravity environment [8, 9].

(5) The CHFs (critical heat fluxes) in subcooled boiling were about 40% of the corresponding terrestrial CHFs in either a $\sim 10^{-2}g$ or $\sim 10^{-5}g$ environment, which is definitely inconsistent with the well-known one-fourth power relationship for gravity dependence of CHF predicted by the conventional Taylor–Helmholtz instability models.

Water

(1) The boiling heat transfer is appreciably deteriorated with a reduction in gravity in the whole nucleate boiling regime irrespective of the material of the heater [6, 7, 9]. This gravity dependence, which is considerably different from that in the boiling of organic fluids, is considered to come from a smaller contribution of the latent heat transport in water boiling in which bubbles generated on the heater surface either continue to attach to the surface with very small contact areas at small subcoolings or immediately detach from the surface and condense in the bulk at large subcoolings.

(2) Because of the large surface tension of water, bubbles are much more spherical and less deformable than those in the organic fluids, and they rarely coalesce with each other while attaching to the heater surface. As a result, little motion of the liquid over the surface is induced in water. This fact contrasts with the frequent coalescence of attaching bubbles in organic fluids, which is considered to be an effective mechanism for maintaining liquid motion in microgravity environments [5].

(3) Instead of inactivity in the coalescence of bubbles laterally distributed on the heater surface, the coalescence of bubbles generated at each particular site on the heater surface occurs so frequently that large bubbles are formed above the layer of small primary bubbles covering the heater surface. This particular behavior of bubbles is reasonably ascribed to the large surface tension of water and also to a relatively poor wettability of the heater surface with water.

From the space thermal engineering viewpoint, the next step in our microgravity pool boiling studies is to find out an appropriate means to effectively enhance the boiling heat transfer under microgravity. In this context, our basic philosophy is not to utilize some external forces such as electrohydrodynamic forces or ultrasonic waves but to devise a somewhat passive and spontaneous method of enhancement. Here we focus our attention upon the Marangoni effect possibly induced by a concentration gradient along the surface of bubbles generated in the so-called 'positive' non-azeotropic binary mixture, which is characterized by the combination of a more volatile component with a lower surface tension and a less volatile component with a higher surface tension, e.g. mixtures of some lower alcohol or ammonia and water. Because of the preferential evaporation of the more volatile component in the mixture, we can expect the formation of a Marangoni-force-driven flow directed from the top to the bottom of each bubble, which may contribute to enhancing the heat transfer even under microgravity conditions.

It is also expected that the study of the role of the Marangoni effect on boiling in the absence of buoyancy could provide some new insight into our fundamental understanding of the role of the Marangoni effect in the boiling heat transfer mechanism of binary mixtures in the terrestrial conditions claimed by Hovestreijdt [10].

The present study is, in fact, not the very first one to deal with microgravity pool boiling of a binary mixture. In the literature we find that Cochran and Aydelott [11] observed bubble motion in the microgravity boiling of a water–ethanol mixture with the aid of a 25.9 m drop tower in the mid 1960s, but they gave no argument concerning the heat transfer of the boiling mixture during the free-fall period. The Marangoni effect was entirely out of the scope of their study.

EXPERIMENTAL

The experiments were carried out at JAMIC (Japan Microgravity Center) in Hokkaido, Japan where a drop-shaft facility of 490 m free fall is operated. This facility provides a good quality microgravity of the order of $10^{-5}g$ for 10 s with the aid of a double-capsule structure and a precise air-thruster control. An optical communication system in the facility enables the telecontrol of experiments even in the free-fall period. The 200 m deceleration zone allows 'soft landing' within $8g$ by both aerodynamic and mechanical braking systems. The detailed technical information about this facility is given elsewhere [12].

The experimental hardware consisted of a boiling chamber holding a transparent glass plate heater inside, video cameras with lights, and control units. The hardware was essentially the same as that used in our former parabolic flight experiments [6, 7]. As Fig. 1 illustrates, the heater was made of a 30 mm thick Pyrex plate, one side of which was coated, over a 40×40 mm area, with a transparent ITO (indium-oxide with a trace of tin oxide) film to be Joule-heated, and permitted us to observe the boiling on it simultaneously from two directions—from its side and from its rear at a proper angle. Two 8 mm video cameras (30 fps) were employed for the observations. The spatial variation in solid/liquid/vapor contact on the heater surface could be recognized from the brightness in the rear view obtained with one of the cameras. An area on the heater surface, either covered with a thin liquid film (the so-called microlayer) or dried-out, was bright to look at due to the specular reflection of the light at the smooth liquid/vapor and solid/vapor interfaces, while an area in contact with a thick liquid layer (the so-called macrolayer) or the liquid bulk was dark due to the diffuse reflection at the wavy liquid/vapor interface or the turning aside of the specularly reflected light from the camera axis.

A water–ethanol binary mixture system was used in the present experiments. Previous studies on terrestrial pool boiling on a plate heater have reported that the minimum and maximum of the critical heat flux can be obtained at particular compositions—about 12 and 30 wt% of ethanol, respectively [13, 14]. In the present microgravity experiments, we intended to employ such mixtures to correspond to these two particular compositions. For the preparation of the mixtures, water distilled and degassed in advance and reagent-grade ethanol prepared in the same manner were separately introduced into the boiling chamber at a prescribed volume ratio. The accurate determination of the concentration of ethanol in the mixtures was made by a chemical analysis after the boiling experiments; it gave 11.3 wt% and 27.3 wt%, respectively.

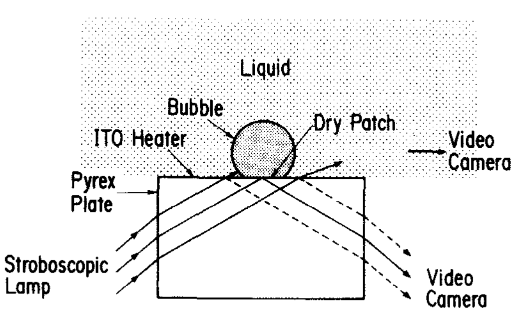

FIG. 1. Principle of observation with a transparent glass heater.

The bulk temperature in the chamber was adjusted close to the bubble point corresponding to the standard atmosphere. In practice, however, slight temperature and pressure fluctuations yielded 1–4 K subcooling in the bulk of the liquid, and the temperatures and pressures in the experiments of 11.3 wt% and 27.3 wt% mixtures were 92.1–94.8°C, 104–114 kPa and 85.9–86.9°C, 101–105 kPa, respectively. The ITO film on the heater plate was stepwise heated up immediately after dropping the capsule in which the whole experimental hardware was integrated. In most cases, the temperature of the film changed asymptotically within the first 4 s, then remained almost constant during the rest of the 10 s free fall, as long as the nucleate boiling was maintained on the heater. When boiling did not occur or when a dry-out area (or areas) formed on the heater, however, the temperature continued to increase throughout the free-fall period.

The heat flux directed to the liquid was calculated from the voltage imposed spanwise on and the resultant current through the ITO film, but account was taken of the backward heat loss which was estimated on the assumption of one-dimensional transient heat conduction into the glass plate. The instantaneous average heater surface temperature was determined by the calibrated resistance versus temperature relationship of the ITO film. The accuracy of the temperature thus determined was estimated to be 1 K.

The phase equilibria for the water–ethanol mixtures at the experimental conditions were calculated by means of the Antoine and the Wilson equations based on the relevant phase equilibrium data [15, 16].

RESULTS AND DISCUSSION

Boiling behavior

No significant difference was found in the boiling behavior between the two mixtures with different concentrations. Every bubble generated on the heater surface first grew into a hemispherical shape (or a segment-of-sphere shape) occupying a large circular 'contact area' on the heater surface. This observation bears an apparent resemblance to our previous observation of bubbles growing in organic fluids [6]. The bubbles thus grown in the present mixtures, however, detached immediately from the surface, and stayed near the surface afterwards as long as the heat flux was relatively low. This behavior is considerably different from that observed with CFC-113 and water in saturated microgravity boiling [9]; the bubbles of CFC-113 continuously attached to the heater surface, each occupying a large contact area, and often coalesced with each other on the heater surface even at low heat fluxes, while in water some bubbles remained in contact with the heater surface with very small contact spots, and some detached from, and stayed above, the surface.

At higher heat fluxes, 'primary bubbles' grown individually on the heater surface coalesced with succeeding 'primary bubbles', and thus formed large 'secondary bubbles' which were lifted onto the array of smaller primary bubbles packing on the heater surface (Fig. 2). (Note that the planar layer composed of the array of primary bubbles and the liquid filling the residual space in the array is referred to as the 'macrolayer' in this paper.) The formation of such dual bubble layers—one made of primary bubbles and the other of secondary bubbles—interacting with each other is essentially the same as what is observed in the microgravity boiling for pure water [7, 9]. In the experiments with the 11.3 wt% mixture, the heat flux could be raised up to the CHF, at which point a local breakdown of the dual bubble-layer structure accompanied by the development of a dry-out area on the heater surface was clearly observed from the rear side (Fig. 3).

A very remarkable bubble behavior was observed in the rear view of most of the bubbles before their detachment from the heater surface—a 'dark patch' appeared in the center of each bubble/heater contact area (see Fig. 4(b)). Such dark patches were generally clearer in the 27.3 wt% mixture than in the 11.3 wt% mixture. These dark patches were definitely caused by diffusive reflection of light at the undulating vapor/liquid interface close to the heater surface, and from this observation it is reasonably presumed that the bubble/heater contact area is not dry but is covered with a thin liquid layer. The above finding has been taken into account in considering the mechanism of bubble detachment in the mixtures, particularly at low heat fluxes (in the isolated bubbles regime), as illustrated in Fig. 5.

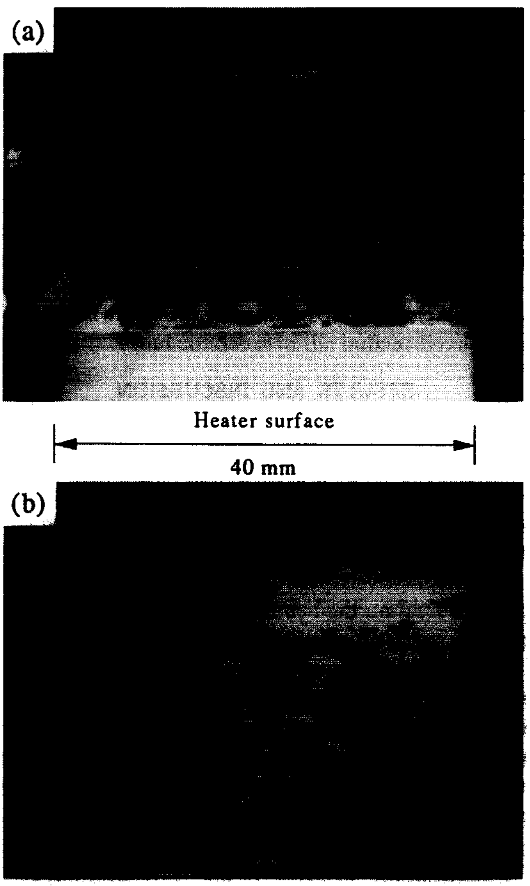

FIG. 2. Microgravity boiling (11.3 wt% ethanol, ΔT_{sub} = 1.8 K, q = 146 kW m^{-2}). (a) Side view (large coalesced bubbles are formed over the layer of primary bubbles). (b) Rear view (bright spots show bubble/heater contact area of primary bubbles).

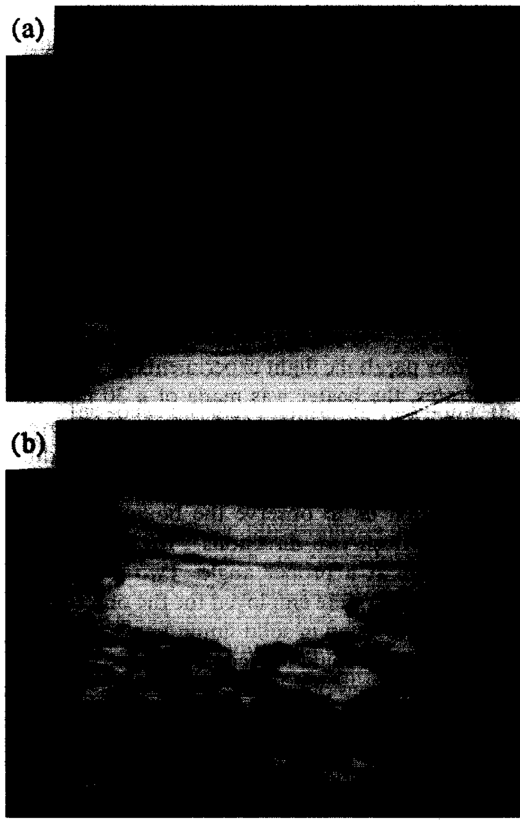

FIG. 3. Critical heat flux of microgravity boiling (11.3 wt% ethanol, ΔT_{sub} = 3.8 K, q = 384 kW m^{-2}). (a) Side view (large coalesced bubbles overhang the heater surface). (b) Rear view (stationary dry-out area develops on the heater surface).

Since the preferential evaporation of ethanol into a bubble from its surface is more intensive at positions closer to the heater surface, the bubble surface must experience a Marangoni force directed from the ethanol-rich bubble top to the ethanol-poor bubble bottom (Fig. 5(a)). The liquid adjacent to the bubble surface is then dragged by this force down to the bottom of the bubble and thereby lifts it up from the heater surface (Fig. 5(b)). The radially converging liquid flow thus established beneath the bubble makes the vapor/liquid interface plunge into the bubble at the central part of its base, causing a boundary undulation detected as a 'dark patch' (Fig. 5(c) and (d)).

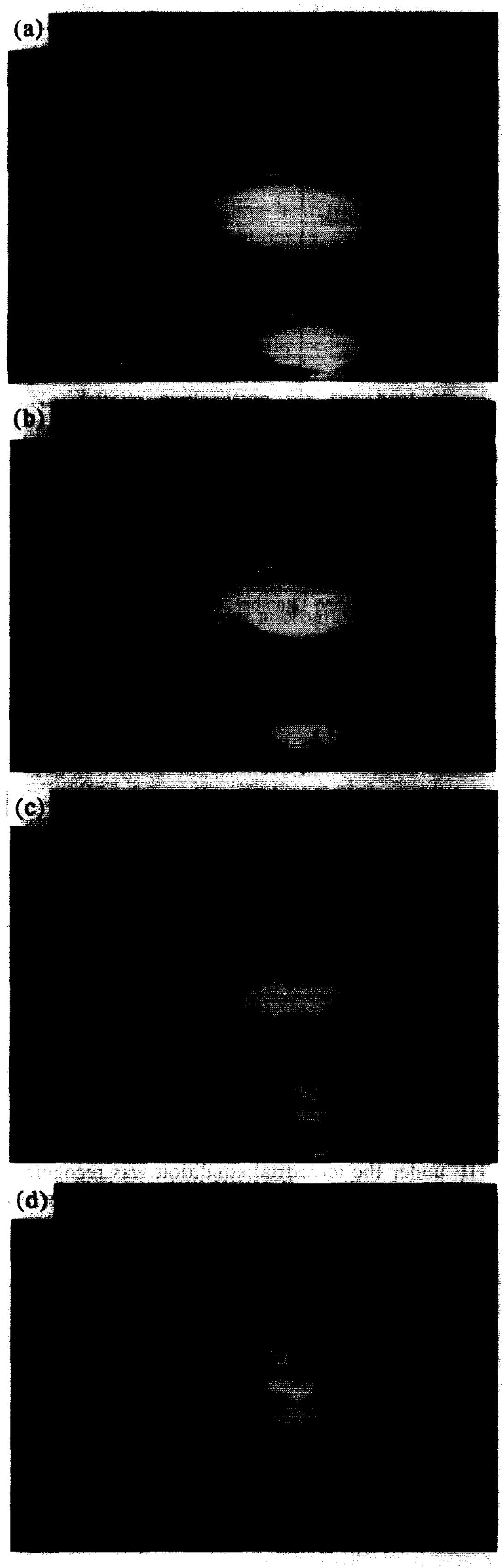

FIG. 4. Successive (1/30 s time interval) rear views of growth and departure of isolated bubble under microgravity (27.3 wt% ethanol, $\Delta T_{sub} = 3.8$ K, $q = 23$ kW m^{-2}).

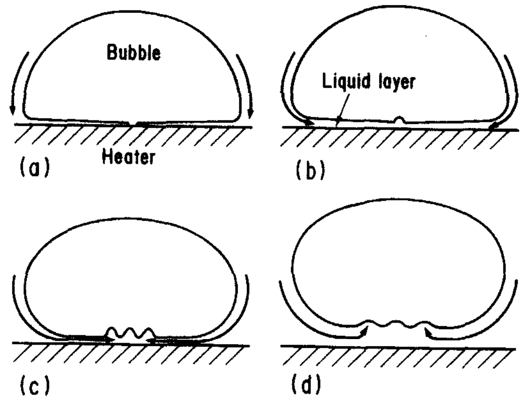

FIG. 5. Schematic illustration of bubble detachment mechanism.

Also under terrestrial conditions, we noted that the 'dark patch' appeared in the center of bubble/heater contact area and that the bottom of each bubble just detaching was concaved (see Fig. 6(a)–(c)). This observation suggests that in the pool boiling of water–ethanol mixtures the Marangoni-force-assisted bubble detachment mechanism plays some role irrespective of the level of gravity, though the role under terrestrial gravity is rather masked by the liquid motion (wake) induced by rising bubbles.

In pursuit of more experimental evidence about the behavior of the liquid layer at the bubble/heater interface described above, supplemental terrestrial tests were made using the same transparent heater facing downward and the Fizeau interferometer employed in a previous study [8]. The experimental set-up is illustrated in Fig. 7. The saturation conditions at reduced pressures were held in the boiling chamber, and very low heat fluxes were imposed on the heater so that isolated bubbles formed intermittently. Typical interferograms observed with water, CFC-113 and a 27 wt% water–ethanol mixture are shown in Figs. 8(a), (b) and (c), respectively.

Figure 8(a) shows the development of a dry-out area in the center of a bubble/heater contact area and concentric interferometric fringes due to a liquid microlayer covering the rest of the contact area. Figure 8(b) shows interferometric fringe, indicating the presence of a thin liquid layer spread over a part of the area beneath a bubble sliding on the slightly inclined heater surface. (In the case of CFC-113, it was not possible to observe, using 30 fps video camera, interferometric fringes in the area of contact of each bubble onto the heater surface set horizontal, because the depletion of the microlayer once formed beneath the bubble was too fast. The heater surface was therefore slightly inclined so that the bubbles as they developed continuously slid over the surface.)

In Fig. 8(c) we can see irregular interference patterns in the whole bubble/heater contact area. They are quite irregular in shape at each instant and wriggling without interruption. This observation indicates again that the water–ethanol liquid layer formed

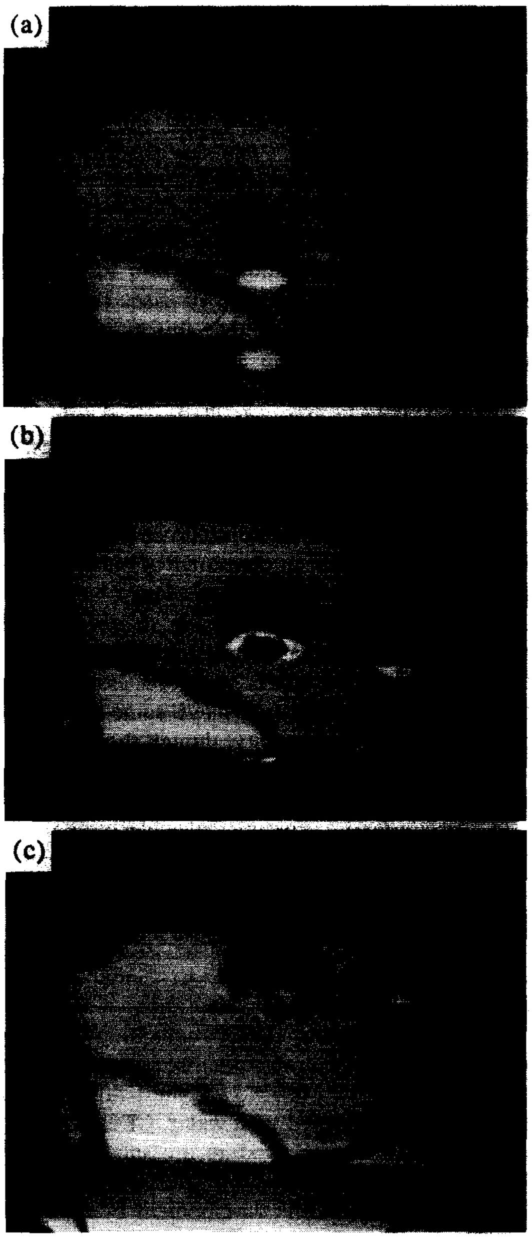

FIG. 6. Successive (1/30 s time interval) rear views of growth and departure of an isolated bubble under $1g$ (27.3 wt% ethanol, $\Delta T_{sub} = 0.5$ K, $q = 34$ kW m^{-2}).

beneath as-yet undetached bubbles is so thick, compared with the 'microlayer' detected as parallel interference fringes, as to permit some fluctuating flow inside. Presumably the liquid layer is prevented from a significant thinning both by an increasing concentration of water component in the layer during the period of bubble growth and by the liquid supply to the bottom of the bubble, which is promoted by the Marangoni effect.

Bubble/heater contact area

The size of contact area for each primary bubble was similar to that in the terrestrial boiling particularly at high heat fluxes, though the coalesced bub-

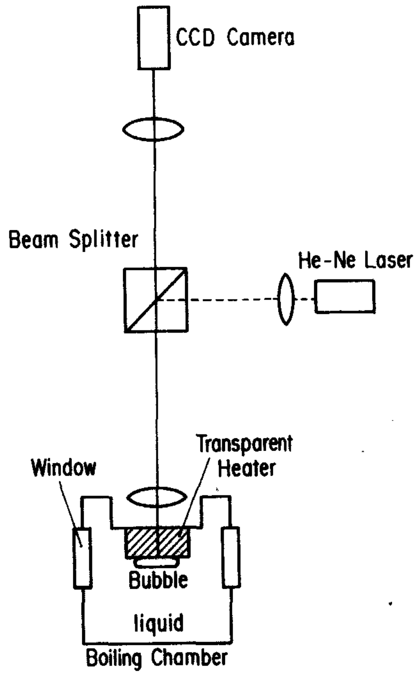

FIG. 7. Experimental set-up for interferometric observations.

ble itself was much more enlarged under microgravity. It should be noted here again that the bubble/heater contact area is not dry but is covered with a thin liquid layer. In comparison with the terrestrial condition, the shape of contact areas was generally more disturbed, and the spatial distribution of contact areas over the heater surface varied more irregularly with time under microgravity. Such differences in shape and distribution of contact area suggest that the lateral liquid motion on the heater surface is possibly more vigorous in microgravity.

Figures 9(a) and (b) plot the fraction of bubble/heater contact area on the heater surface, F, against the heat flux imposed on the heater, q, in the boiling of 11.3 wt% and 27.3 wt% mixtures, respectively. The uncertainty in the determination of F was, in general, judged to be about 20–30%. For either mixture, the CHF under the terrestrial condition was recognized when F increased up to 25–30%. In the case of the 11.3 wt% mixture, the F values under microgravity slightly exceed the corresponding terrestrial values at relatively high heat fluxes. As for the 27.3 wt% mixture, on the other hand, some excess of microgravity F values over the terrestrial values, which is recognized in a rather low heat flux region, diminishes with an increase in the heat flux. In general, the F versus q relations observed with the water–ethanol mixtures in the present experiments do not greatly differ from those observed previously with water in the terrestrial boiling on an ITO film heater [7] similar to the one used in the present experiments.

Heat transfer characteristics

Figures 10(a) and (b) compare the boiling curves for the mixtures under microgravity and terrestrial conditions. The boiling curve obtained for saturated water on the same heater under terrestrial conditions is also given in Fig. 10(a). It is worthwhile noting here that the boiling heat transfer for either mixture is not deteriorated but rather enhanced with the reduction in gravity over the major portion of the nucleate boiling regime.

The ratio of the boiling heat transfer coefficient under microgravity conditions, $\alpha_{\mu g}$, to the terrestrial heat transfer coefficient, α_{1g}, at each level of heat flux is plotted in Fig. 11. Here we find out that some 20% increase in the boiling heat transfer coefficient is available as a result of the reduction in gravity, whichever mixture is used.

No definite explanation for the heat transfer enhancement with the reduction in gravity can be provided at present. Still, it seems reasonable to ascribe the enhancement to the interaction between the Marangoni effect and the change in bubble behavior with the reduction in gravity. The Marangoni effect intrinsic to a 'positive' mixture works effectively to detach 'grown-up' bubbles from the heater surface even under microgravity, and most of the bubbles thus detached are adrift near the heater surface for sufficiently long periods whenever the gravity is small enough. Such bubbles adrift near the heater surface provide a large interfacial area on which the Marangoni force can work to convert the adjacent liquid towards and/or along the surface (cf. Figs. 4 and 6).

For quantitative evaluation of the role of the Marangoni force in the convection close to the heater surface, microgravity experiments for closely observing both temperature and concentration profiles at the vicinity of each bubble may be useful, and a two-wavelength interferometer will be a very powerful tool for such a purpose.

In the present drop-shaft experiments, we could confirm the boiling transition and thereby determine the CHF only with the 11.3 wt% mixture. The CHF thus determined was lower than the corresponding terrestrial CHF by only 20–40%. The extent of this reduction in the CHF is considerably smaller than for saturated CFC-113 which was evaluated to be as large as 50–85% [9]. The boiling transition was not observed with the 27.3 wt% mixture even at the highest heat flux set in the present experiments, which was about 50% of the terrestrial CHF for the same mixture.

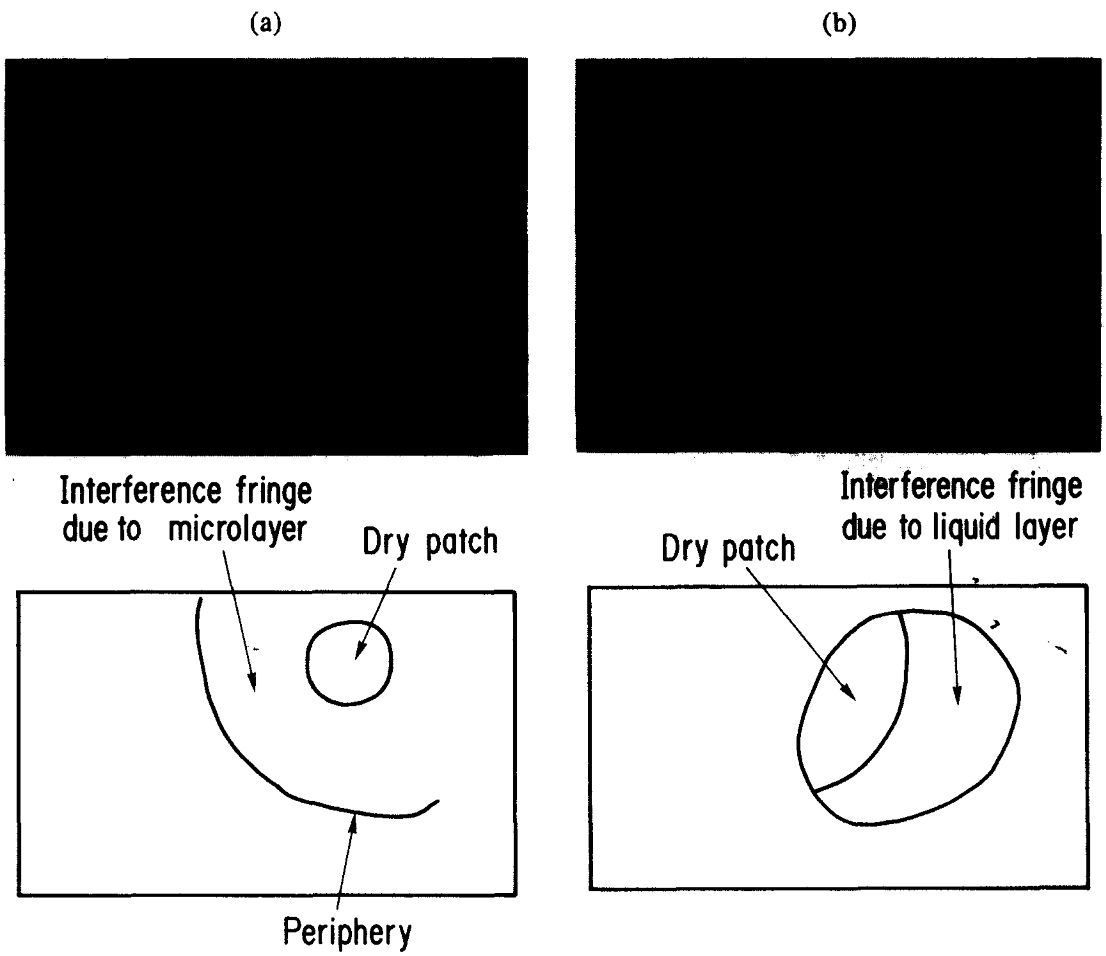

FIG. 8. Interferograms indicating liquid-layer configurations beneath bubbles. (a) Water, (b) CFC-113, (c) water ethanol mixture.

(c)

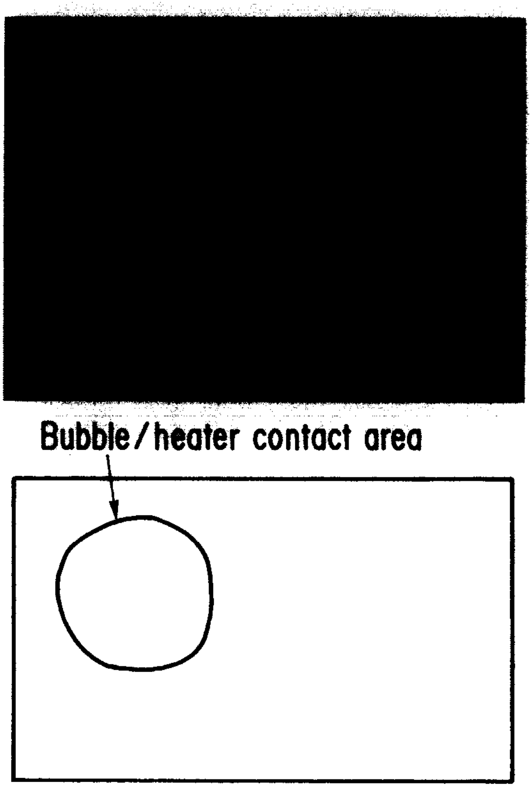

FIG. 8—*continued.*

(a)

(b)

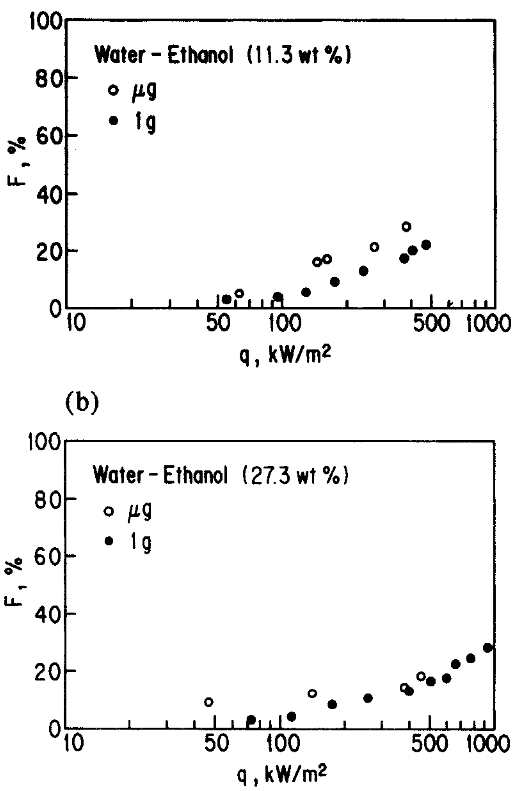

FIG. 9. Fraction of bubble/heater contact area plotted against heat flux. (a) 11.3 wt% ethanol, (b) 27.3 wt% ethanol.

(a)

(b)

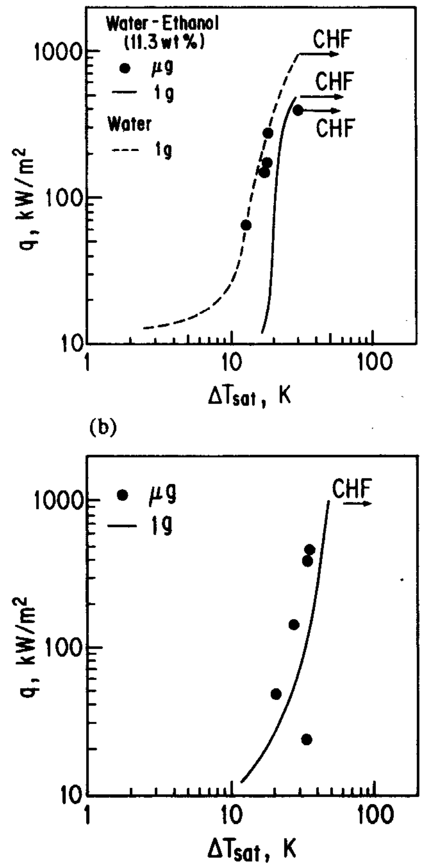

FIG. 10. Comparison of boiling curves under microgravity and $1g$. (a) 11.3 wt% ethanol, (b) 27.3 wt% ethanol.

CONCLUDING REMARKS

Pool boiling experiments with water–ethanol mixtures, 11.3 wt% and 27.3 wt% of ethanol, were performed in a $\sim 10^{-5}g$ condition. The experimental findings considered to be characteristics of microgravity boiling for such 'positive' non-azeotropic mixtures are:

(1) When bubbles develop, they immediately lift up

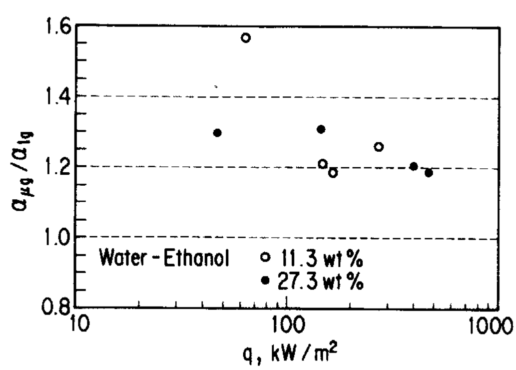

FIG. 11. Ratio of boiling heat transfer coefficient under microgravity to that under $1g$.

and detach from the heater surface and stay at a small distance away from the surface thereafter.

(2) The liquid supply to the base of each as-yet undetached bubble is very intensive, presumably because of the Marangoni effect, and thereby promotes its detachment.

(3) At relatively high heat fluxes, large coalesced bubbles are formed above the array of small primary bubbles lying on the heater surface, and continually suck up the latter.

(4) The heat transfer to the mixtures is enhanced by the reduction in gravity over the major portion of the nucleate boiling regime.

(5) The fractional decrease in the CHF for the 11.3 wt% mixture with the reduction in gravity from the terrestrial level is 20–40%, which is much more limited than for pure organic fluids.

The enhancement of heat transfer and an unexpectedly small amount of reduction of the CHF under microgravity conditions indicate that the Marangoni effect in 'positive' binary mixtures can work more effectively when vapor/liquid interfaces are apt to be adrift near the heat transfer surface because of the absence, or a significant reduction, of gravity. More detailed experimental examinations on this point are now prepared.

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