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**POOL BOILING HEAT TRANSFER OF ETHANOL-WATER MIXTURES
AT SUB-ATMOSPHERIC PRESSURES**

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ABSTRACT

Pool boiling has been carried out on circular aluminum surfaces ($D = 30$ mm) with two surface roughness values (0.8 μm and 20 μm , respectively) for operating vapor temperatures of 30°C to 70°C . The heat transfer coefficient has been estimated as a function of applied heat flux in the range of $0.01 - 0.15$ MW/m^2 for 2.0%, 25.0% and 80.0% molar ethanol-water mixtures, and 100% pure forms of both the liquids, respectively. In addition, bubble dynamics is observed by high speed camera at 2000 fps. The heat transfer measurement on both surfaces shows degradation even with the presence of small amount of ethanol in water. At higher concentrations of ethanol, the heat transfer first reaches minimum and then attains a maximum at 80.0% molar ethanol concentration. A comparison of heat transfer coefficient with predictions obtained from recent mixture boiling correlations indicates marked difference for all compositions studied for the smooth surface. However, in case of rough surface, the values are closer to predictions. Analysis of high speed images of bubble growth indicate bubble growth on both surfaces is significantly hampered in case of mixtures, which is attributed to mass transport limitation of ethanol from bulk to interface and non-linear variation of thermo-physical properties with composition. A unique bubble departure behavior at sub-atmospheric pressure is observed wherein a strong liquid reflux takes place into the base of the very large sized bubbles.

NOMENCLATURE

A_0	Empirical constant (-)
C_{sf}	Surface constant (-)
h	Heat transfer coefficient ($\text{W}/\text{m}^2\text{K}$)
K	Correction factor (-)
k	Thermal conductivity (W/mK)
p	Pressure (Pa)
q''	Heat flux (W/m^2)
Ra	Average roughness number (μm)
T	Temperature (K)
x	Liquid mole fraction (-)
y	Vapor mole fraction (-)
δ	Thickness (m)
ΔT	Temperature difference (K)

Subscripts

1,2	Component
bp	Boiling range
id	Ideal
mix	Mixture
s	Surface
c	Thermocouple

INTRODUCTION

Pool boiling of binary mixtures has been a subject of study since the time when binary refrigerants became popular. Besides this, such mixture systems exist in a variety of industrial applications like petroleum distillation, avionics cooling, and hydrogen production with ethanol/methanol water systems, etc., to name a few. It is known in the literature that the heat transfer coefficient (HTC) for binary mixtures deteriorates compared to that of the respective pure components [1-4]. The reasons cited for the same include, amongst other, rise in the local interfacial temperature due to preferential evaporation of the more volatile component, and the consequent mass transfer limitation for transport from bulk to the interface and non-linear variation of mixture properties with composition.

A variety of refrigerant mixtures, organic to aqueous have been studied and the general trend in variation of HTC with composition is known for both zeotropic and azeotropic type refrigerant mixtures [1-3, 5]. In addition, the general trend in the heat transfer coefficient is known for a range of binary mixtures that have been studied at atmospheric conditions. For zeotropic mixtures, the HTC for any mixture composition is always less than that of the pure components. However, for azeotropic mixtures, the mixture HTC shows a minimum corresponding to the maximum boiling point range [6] and a maximum corresponding to the azeotropic point [5].

Researchers have also tried to predict the heat transfer characteristics of mixtures by formulating correlations based on pure fluid predictions. Prediction of binary mixture HTC is based on two possible approaches [6]. One can directly employ existing pure fluid correlations and make appropriate use of binary mixture properties in the same. In such an approach the binary mixture is approximated as a pseudo pure

fluid having same properties as that of binary mixture. While, this approach may take care of the non-linear variation of mixtures properties, other profound effects related to transport mechanisms cannot be addressed.

A more popular method of prediction is to first estimate the binary mixture HTC as a mole fraction average of the individual component HTC. Hence for a given heat flux q'' ,

$$\Delta T_{id} = x_1 \Delta T_1 + (1 - x_1) \Delta T_2 \quad (1)$$

where, x_1 is the mole fraction of more volatile component and ΔT_1 and ΔT_2 are wall superheats for pure components. Hence,

$$\frac{1}{h_{id}} = \frac{x_1}{h_1} + \frac{1 - x_1}{h_2} \quad (2)$$

The final binary HTC is then calculated by applying a correction factor that is based on any one, or a combination of the various other factors responsible for the deterioration of heat transfer, as indicated earlier. Hence, the heat transfer coefficient for binary mixture is then given by,

$$h_{mix} = h_{id} \times K \quad (3)$$

The expression for K has been proposed in varied manner in the literature, depending on the methodology adopted for including the effect of degradation. Following is a very brief account of such studies: The correlation by Körner [7] includes the effect of vapor liquid mole fraction difference and mixture density. Jungnickel et al. [8] added a term to account for the heat flux dependence. Stephan and Körner [5] included the effect of pressure in the Körner relation. Thome [9] has formulated a simple expression for binary mixture HTC only to include the effect of boiling temperature range of a mixture. Fujita and Tsutsui [6] modified the Thome correlation to include the effect of heat flux on heat transfer. These different K factors available are indicated below:

Stephan and Körner [5],

$$K = \frac{1}{1 + A_0(0.88 + 0.12p)|y_1 - x_1|} \quad (4)$$

Thome [10],

$$K = \frac{1}{1 + \Delta T_{bp} / \Delta T_{id}} \quad (5)$$

Fujita and Tsutsui [6],

$$K = \frac{1}{1 + (1 - 0.8 \exp(-q''/10^5))(\Delta T_{bp} / \Delta T_{id})} \quad (6)$$

However, looking at the literature it is noted that the explicit effect of operating pressure and temperature has not been extensively investigated and the relation with bubble dynamics is not correlated accordingly.

EXPERIMENTAL SETUP AND PROCEDURE

The schematic of the setup is shown in figure 1, while figure 2 shows the details of the heater surface and instrumentation. Pool boiling was carried out on 30 mm diameter flat surfaces of aluminum ($k = 170 \text{ W/mK}$), having two surface roughness values of $0.8 \mu\text{m}$ and $20.0 \mu\text{m}$, respectively. This surface was mounted in a cylindrical heater block assembly from where the heat was transferred to the surface by conduction. The heater block was fitted with four cartridge heaters (150 W each). The entire heater block and test surface was surrounded radially and from the bottom by Teflon insulation ($k = 0.25 \text{ W/mK}$).

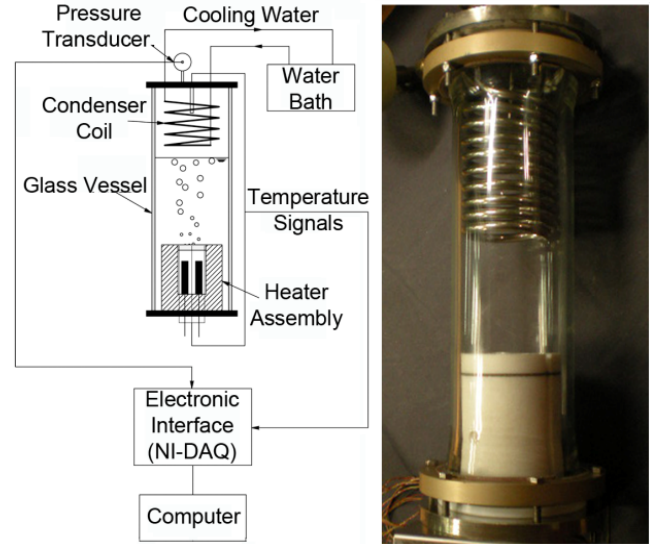


Figure 1. SCHEMATIC DIAGRAM OF THE EXPERIMENTAL SETUP AND PHOTOGRAPH OF THE PRESSURE VESSEL.

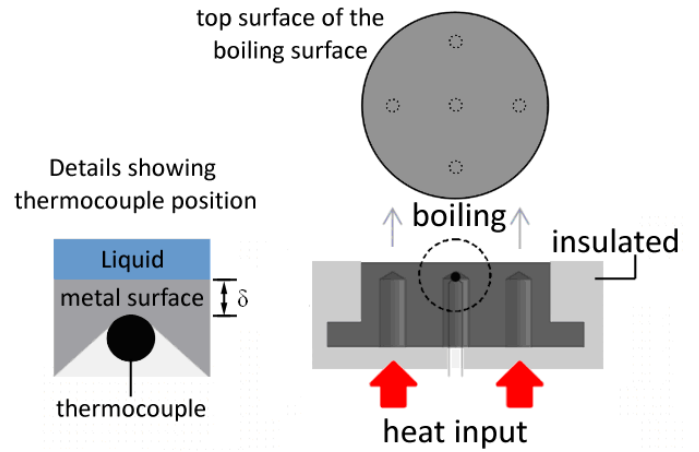


Figure 2. SCHEMATIC OF THE BOILING TEST SURFACE AND POSITIONS OF THE EMBEDDED THERMOCOUPLES.

The heater and the helical tube condenser assembly were mounted on the two sides of a cylindrical glass pressure vessel, capable of maintaining vacuum better than 10^{-4} mPa . The glass vessel itself was insulated by a 15 mm Polyurethane insulation ($k = 0.025 \text{ W/mK}$), which had a small window for visualization.

Five thermocouples were mounted 0.8 mm below the surface of the test piece – one at the centre and remaining four in each quarter of the surface, as shown in figure 2. One thermocouple each was employed for measuring the bulk liquid pool and vapor temperature, respectively. Additionally, a thermocouple was mounted at the base of the heater block to measure maximum safe temperature of the system and also to facilitate estimation of heat flux to the surface by conduction approximation. The temperature on the boiling surface was estimated by considering 1-D conduction through the small $\delta = 0.8 \text{ mm}$ aluminum material that was present above the 5 surface thermocouples. Hence, if T_c is the average thermocouple reading then the actual surface temperature (T_s) was taken as in eq. 7.

$$T_s = T_c - (q'' \delta) / k \quad (7)$$

The operating pressure was controlled by circulating cooling water through the condenser coil mounted in the boiling pressure vessel at the desired temperature by a cryostat

(Haake[®]). In addition, the bubble dynamics were captured by high speed camera (Photron[®]) at a frame rate of 2000 fps. An absolute pressure transducer was mounted on the vessel to measure system vapor pressure (Honeywell[®], 0-3 bar abs.).

The heat flux at the boiling surface was taken from the electrical power supplied and the boiling surface area (0.0707 m²). This estimation was found to conform to heat flux estimate using a one-dimensional conduction approximation through the heater block within 5%-7%.

RESULTS AND DISCUSSION

The pool boiling experiments were carried out under operating temperatures in the range of 30°C to 70°C. The heat flux was controlled from 0.01 MW/m² to 0.15 MW/m². Actual wall temperature was estimated from the sub-surface thermocouples by 1-D conduction approximation. At each heat flux level, the average wall temperature was noted at steady state (the average of five surface mounted thermocouples). The wall superheat (ΔT) was then obtained as the difference between the average surface temperature and the bulk saturation temperature (measured by the thermocouple placed in the vapor space). Depending on the applied heat flux, the vapor space temperature was controlled by the temperature of the coolant water passing through the condenser.

The pool boiling characteristics for water were first benchmarked with existing popular correlations for pure fluids, i.e. Rohsenow correlation [10] and Forster-Zuber correlation [11]. Figure 3 shows comparison of the obtained experimental data with these two correlations, respectively (For the Rohsenow correlation the surface constant C_{sf} has been taken to be equal to 0.022). The experimental results were found to conform to correlations with a maximum deviation of 16 %.

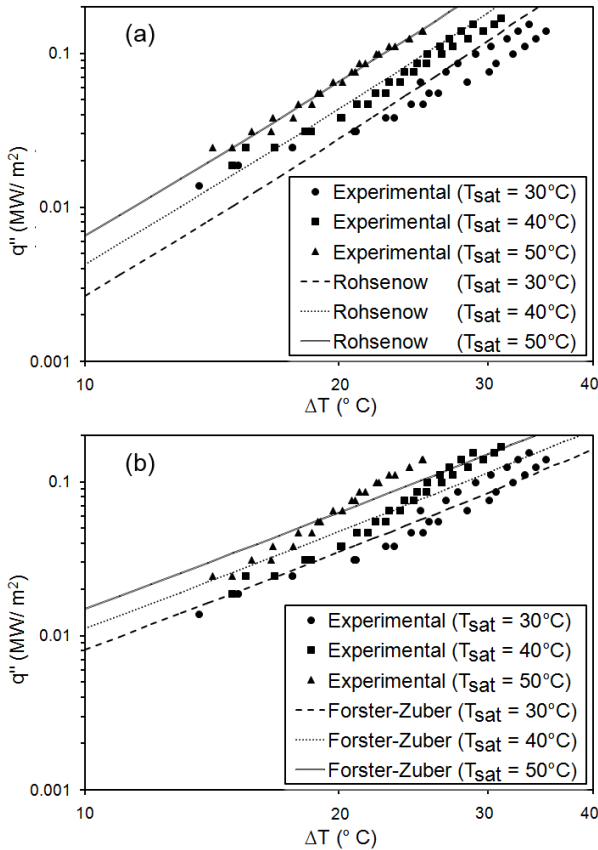


Figure 3: BENCHMARKING DATA SET: COMPARISON OF HEAT FLUX VERSUS WALL SUPERHEAT FOR PURE WATER ON ALUMINUM SURFACE ($R_a = 0.8$ MICRONS) AT SATURATION TEMPERATURES FOR 30°C, 40°C AND 50°C, WITH TWO POPULAR POOL BOILING CORRELATIONS.

Subsequently, experiments were performed for 2.0% and 25.0% ethanol-water mixtures as well as pure ethanol, to estimate the heat transfer coefficient at different operating temperatures and heat fluxes for the two test surfaces having different roughness. Figure 4 shows the results for the smooth surface ($R_a = 0.8 \mu m$).

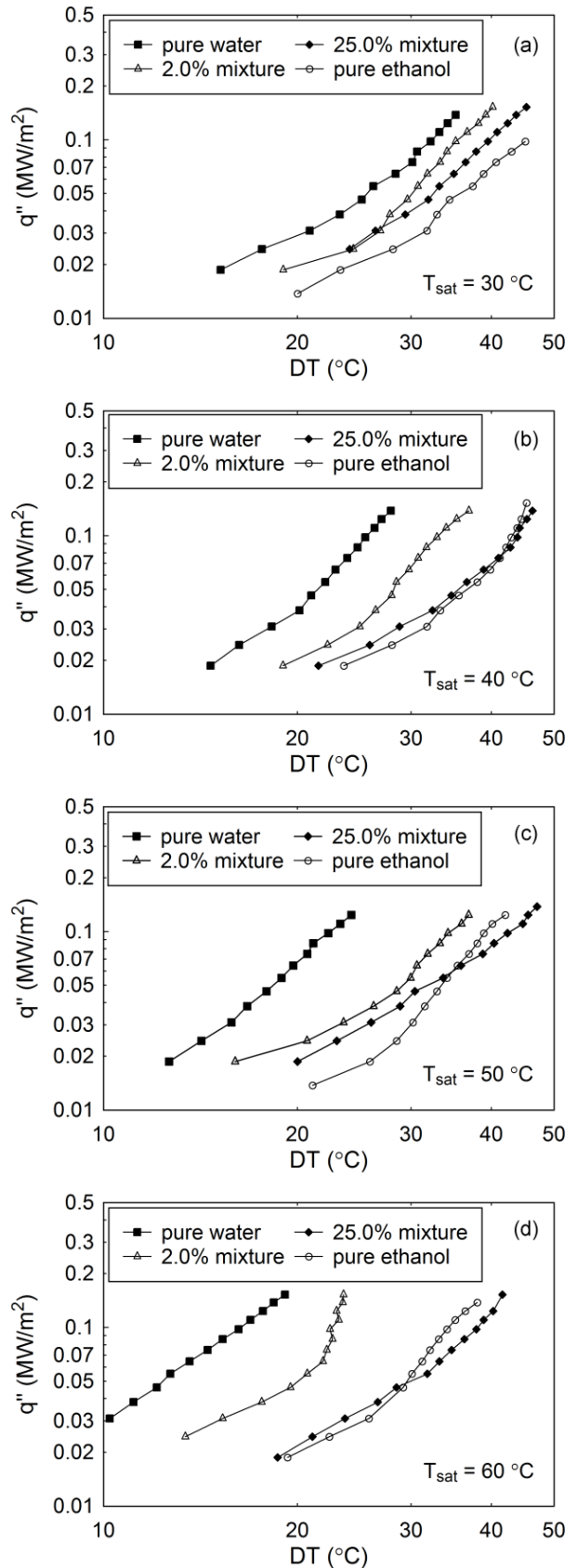


Figure 4. HEAT TRANSFER COEFFICIENT ON THE ALUMINUM SURFACE ($R_a = 0.8 \mu m$) FOR 2.0%, 25.0% ETHANOL-WATER MIXTURES AND PURE COMPONENTS FOR OPERATING TEMPERATURES OF (a) 30°C, (b) 40°C, (c) 50°C, (d) 60°C.

As a general observation it is noted that, addition of even very small amount of ethanol in water (2.0% molar), the heat transfer coefficient reduced significantly for all operating temperatures. Such a general behavior has also been noted in the available literature with two possible reasons: (i) change in thermophysical properties of the mixture [2,3], and, (ii) mass transfer limitation which prevents water molecules to come closer to the growing bubble interface as preferential evaporation of more volatile component (ethanol) takes place [1]. It must also be noted that the boiling point of the mixture is not unique.

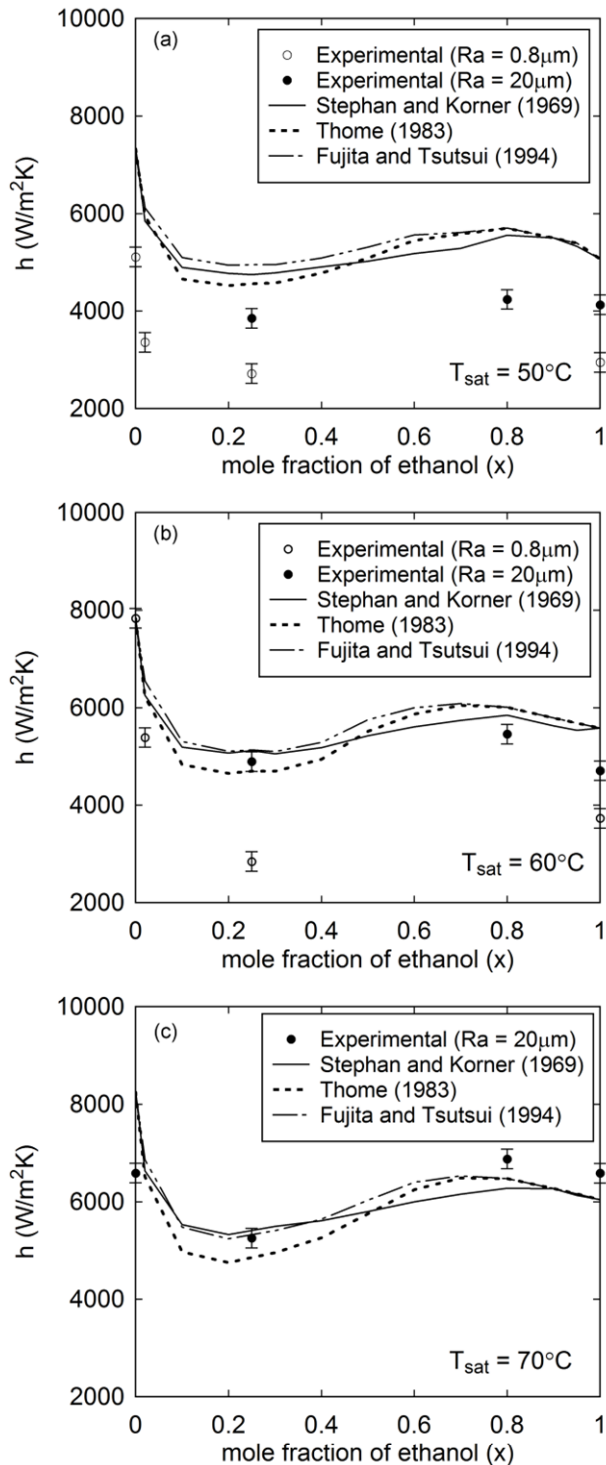


Figure 5. COMPARISON OF THE EXPERIMENTAL HEAT TRANSFER COEFFICIENT AT DIFFERENT ETHANOL-WATER MIXTURE CONCENTRATIONS WITH AVAILABLE CORRELATIONS, FOR 0.8 μm AND 20.0 μm ROUGH SURFACES, RESPECTIVELY, AT (a) $T_{\text{sat}} = 50^\circ\text{C}$, (b) $T_{\text{sat}} = 60^\circ\text{C}$, (c) $T_{\text{sat}} = 70^\circ\text{C}$. (For All CASES $q'' = 0.123 \text{ MW/m}^2$)

As the ethanol concentration is increased from 2.0% to 25.0%, further reduction in heat transfer coefficient slows down. Comparatively, slower reduction in heat transfer at higher concentration is related to the fact that the boiling temperature ranges (i. e. the range of temperature over which boiling takes place at a concentration is maximum at very low concentrations whereas it decreases at higher concentrations [6]. The heat transfer coefficient of pure ethanol was observed to be approximately close to that of 25.0% mixture but the value was higher or lower than the later depending on the operating temperature and heat flux. Similar experiments were performed on the rough surface, for operating temperatures of 50°C , 60°C and 70°C . Figure 5, shows comparison of the results for both the surfaces with predictions from binary mixtures HTC correlations from eq. 3 to eq. 6. The results from figure 5 in general, indicate that the measured values show qualitative similarity in the trend predicted by correlations for variation of heat transfer coefficient with composition for both roughness values alike. However, there exists a striking difference in the actual experimentally observed magnitude at each composition. The values for pure fluids are closer to predictions; however, for mixtures, there is a remarkable difference in case of smooth surface ($Ra = 0.8 \mu\text{m}$) with values for rough surface being closer to predictions.

As the operating temperature is increased, the percentage deviation in the observed and predicted values decreases. As all of the correlations for binary mixtures are formulated from data gathered at atmospheric pressure, they do not incorporate the effect of pressure on heat transfer. Hence, they tend to over predict the experimental data reported here for mixture boiling at sub-atmospheric pressures. However, as can be seen from the previous observation, the experimentally observed values seem to converge towards the predicted values from the correlations, as operating temperature is increased. In fact, the heat transfer characteristics actually coincides well with the predictions, for the rough surface at $T_{\text{sat}} = 70^\circ\text{C}$. Also, all binary mixture heat transfer correlations used are based on Stephan-Abdelsalam relation, where the surface roughness is assumed to be $1.0 \mu\text{m}$ [12].

In addition to heat transfer measurements, high speed videographic images of individual bubbles were captured at 2000 fps, for binary mixtures, as well as pure water at low pressures. Figure 6 shows representative isolated bubble growth images for pure water on smooth surface ($Ra = 0.8 \mu\text{m}$). As expected from classical bubble growth mechanisms, initially the growth is rapid, during the inertia controlled regime, wherein the bubble remained nearly hemispherical in shape. Eventually, the bubble spread on the surface stops as the buoyant forces overcome the surface tension. As buoyant forces begin to dominate, the bulk of bubble shifts upwards even as the base of the bubble sticks to the surface causing necking. Images post detachment indicated considerable reflux of liquid into the vapor bubble from the bottom as the interface that was stretched just before detachment, rebounded into the bubble, due to the surrounding liquid gushing from below, as seen in the last two images (after bubble departure).

A comparison of bubble departure diameters, as shown in Figure 7, shows considerable reduction from pure water to mixture (2.0% and 25.0% ethanol-water mixture) as well as pure ethanol. From the figure, the bubble departure diameter reduces from 38.0 mm for water to 27.4 mm for 2.0% mixture (27.9% decrease compared to water) whereas for 25.0% mixture the diameter is 17.6 mm (53.7% decrease compared to water). For ethanol, the bubble departure size is in the range of 4 mm to 8 mm.

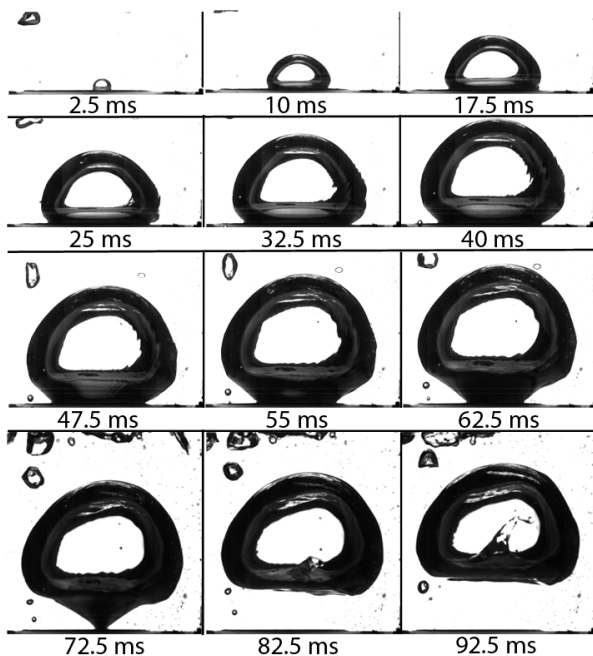


Figure 6. OBSERVED BUBBLE GROWTH BEHAVIOUR FOR $Ra = 0.8 \mu\text{m}$ SURFACE IN PURE WATER AT $T_{\text{sat}} = 50^\circ\text{C}$, $q'' = 0.046 \text{ MW/m}^2$. AFTER THE DEPARTURE THE LIQUID REFLUX IS CLEARLY SEEN.

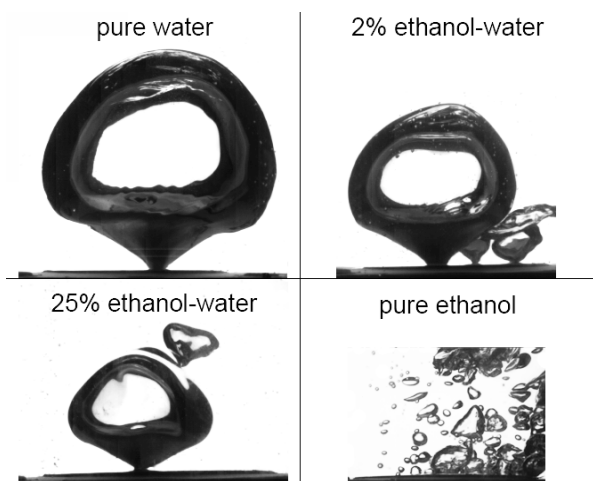


Figure 7. RELATIVE BUBBLE DEPARTURE SIZES FOR $T_{\text{sat}} = 50^\circ\text{C}$, $q'' = 0.046 \text{ MW/m}^2$ ON SMOOTH SURFACE. THE BUBBLE DEPARTURE SIZE OF PURE WATER is 38.0 mm.

Here, the departure diameter is taken as the diameter of a sphere having the same volume as that of the actual bubble. Hence, considerable reduction in departure diameter takes place for 2.0% mixture as compared to 25.0% mixture.

CONCLUSIONS

Following conclusions can be drawn from this study:

There is a significant reduction in heat transfer coefficient for ethanol-water mixture compared to pure water even for very low concentration of ethanol. This phenomenon is observed at all operating pressures. At higher concentrations of ethanol, the heat transfer coefficient first decreases to a minimum, then achieves a maximum, close to the azeotropic composition. In addition, the change in heat transfer is intensified with increase in operating vapor pressure which is attributed to reduction in the boiling temperature range (difference between the bubble point and the dew point temperature) with pressure.

Heat transfer coefficient for rough surface ($20.0 \mu\text{m}$) is ~30-40 % higher than the smooth surface ($0.8 \mu\text{m}$) which is attributed to higher nucleation site density and large number of smaller bubble sizes in case of rough surface. Experimental data at sub-atmospheric pressures, in the entire range of ethanol concentration, only qualitatively match the trend predicted by the available correlations for binary mixtures. However, quantitatively, heat transfer coefficients for rough surface are closer to the predictions as compared to the smooth surface. With increase in operating temperature (and therefore the operating pressure), experimentally obtained heat transfer coefficient values for both the surfaces tend to be more closer to the predictions.

A significant reduction in bubble departure diameters due to bubble growth deterioration in mixtures was observed from high speed images of bubble growth. A unique phenomenon of liquid reflux into the bubble base was observed as large sized bubbles caused change in relative importance of inertia, buoyancy and surface tension forces.

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