

BOILING HEAT TRANSFER CHARACTERISTICS OF IMMISCIBLE LIQUID MIXTURES

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ABSTRACT

Cooling technology becomes more important because of the increase in heat generation density of semiconductor chips. Boiling and two-phase flow is expected as one of the most effective methods to cool the electronic devices because of its superior heat transfer characteristics. Because the boiling heat transfer depends on the selection of coolant, the development of liquid mixtures and their components optimized for the individual cooling systems becomes very important. Most of the existing researches on nucleate boiling of binary mixtures clarify the heat transfer characteristics peculiar to the miscible ones, while the studies for immiscible mixtures were very limited. Pool boiling experiments on nucleate boiling of immiscible mixture FC72/Water are performed in a closed vessel at 0.1MPa. The experimental data is acquired under the steady-state conditions at heat flux ranging from those for the boiling incipience to CHF or to the highest possible value under the restriction of the available cooling. Before the experiments, the height of liquid layers on the horizontal heating surface is kept at a constant value of 100mm, and the thickness of FC72 layer above the heating surface is varied at three different values of 5mm, 10mm and 50mm.

A new phenomena of “intermediate burnout” due to the evaporation of FC72 is observed provided that the layer thickness of FC72 is not large. At heat flux higher than that for intermediate burnout, the immiscible mixture decreases the surface temperature from that for pure water and simultaneously increases CHF. The immiscible mixture has a potential to realize high performance heat exchange systems by the self-sustaining subcooling of components under the constant system pressure.

NOMENCLATURE

c_p isobaric specific heat, J/kg K
 H height, m
 h_{fg} latent heat of vaporization, J/kg
 P pressure, N/m²

q heat flux, W/m²
 R gas constant, J/kg K
 T temperature, °C or K
 x mole fraction of liquid, -
 y mole fraction of vapor, -

Greek symbols

α heat transfer coefficient, W/m²K
 ΔT surface superheat, K
 ρ density, kg/m³

Subscripts

1 more volatile component
2 less volatile component
CHF critical heat flux
g gas
l liquid
sat saturation
sub subcooled
b at boiling point of pure liquid

INTRODUCTION

In the recent development of electronic devices, the heat generation density from electric semiconductors is growing. On the other hand, the electronic devices are often composed of Si semiconductors with a low upper limit of operating temperature, because SiC or GaN semiconductors with toughness against higher temperature are still under development. So, it requires the improvement of the existing cooling system if high heat generation density is concerned.

Liquid cooling systems using the latent heat during boiling heat transfer have more advantages than the conventional ones using air or single-phase liquid. By the application of boiling, the cooling at high heat flux becomes possible minimizing the temperature difference between the cooling surface and the coolant because of inherent high heat transfer coefficient. The cooling systems using boiling heat transfer, however, have an

unavoidable disadvantage additionally to the advantage. Once incondensable gasses are mixed into the vapor phase, the condensation process is seriously interfered because the local vapor saturation temperature is decreased in the vicinity of condensation interface due to the local reduction of vapor partial pressure. To avoid the mixing of incondensable gasses, the cooling systems are designed to perform at the pressure larger than the atmospheric. However, the elevation of operating pressure increases the saturation temperature, which in turn implies the increase in the surface temperature to be cooled.

Although the use of water for the cooling of electronic devices is sometimes avoided because of its potential of short-circuit by the leakage, large values of critical heat flux are expected for water compared to those for organic coolants. Additionally, the saturation temperature for pure water is higher than the upper limit temperature of Si semiconductors, if the cooling system is operated at the pressure larger than the atmospheric. The application of immiscible liquid mixtures to the cooling systems can decrease its saturation temperature, because subcooled boiling occurs by the summation of the partial pressures for both components. In such systems, the volumetric ratio of immiscible liquids accumulated on the heating surface plays an important role on the heat transfer. Under gravitational conditions, the immiscible liquids are separated to form the layers with individual thicknesses on the heating surface at least before the starting of boiling. The thicknesses are determined by the volumes of components.

There are a limited number of existing researches on the pool boiling of immiscible liquids. The heaters employed so far are horizontal flat plates or thin wires, and the pressure range investigated is limited in the vicinity of the atmospheric.

There exist a number of experimental and theoretical researches for the boiling heat transfer using miscible binary mixtures. The heat transfer coefficients due to nucleate boiling of mixtures are usually lower than those of the "ideal" values obtained by the interpolation of the coefficients for the component pure liquids, or more precisely, are lower than the hypothetical single components with the same thermophysical properties as the relevant mixtures. Van Wijk et al. [1] explained the reason for the lower heat transfer coefficient of binary mixtures. Stephan-Körner [2] proposed a simple correlation to predict the heat transfer coefficient in nucleate boiling of miscible mixtures, which was followed by many modified predictions for the deteriorated heat transfer. However,

the characteristics for immiscible liquid mixtures are quite different from the miscible ones, and the investigation on the boiling phenomena peculiar to the immiscible liquids is required. Bragg-Westwater [3] classified the boiling modes observed for different immiscible mixtures as shown in Table 1. The present experiment is categorized as Cases 2 and 6. Nucleate boiling of immiscible mixtures in a closed vessel was investigated by Bonilla-Eisenberg [4], where the experimental data show new characteristics. A jump of surface superheat was observed on the boiling curve of water/styrene immiscible mixture at an intermediate heat flux under a certain condition of volumetric ratio. The authors, however, gave little explanation on this trend. Sump-Westwater [5] investigated the heat transfer due to film boiling, and showed the enhanced heat transfer for immiscible mixture of R113/water compared to that for pure R113.

Although the heat transfer characteristics in boiling of immiscible liquid mixtures are completely different from those for miscible mixtures, no coherent trend or conclusion was derived so far. The objective of the present investigation is to clarify the boiling heat transfer for immiscible liquids, and to discuss about the unusual trends not observed in the past.

EXPERIMENTAL APPARATUS AND PROCEDURE

The experimental apparatus shown in Fig. 1 is composed of a boiling vessel, condensers and a heating section. The boiling vessel is made from a stainless steel pipe with an inner diameter of 200mm and a height of 410mm. Liquid and vapor temperatures are measured by four thermocouples placed in the vessel. The condensers, hanged from the top of the boiling vessel, control the total pressure and then the liquid equilibrium temperature. In the heated block made of copper, eight thermocouples are inserted at the depth of 1, 7, 13, 19mm from the heating surface along two axes at the center and 14mm from the center. The upper surface of the cylindrical copper block is operated as the heating surface, and is heated by the cartridge heaters inserted in the bottom of the block. The heating surface has a horizontal circular area with a diameter of 40mm facing upwards, which is polished by the sandpaper (No.600) to keep the same nucleation characteristics independent of the experimental runs. Additionally, the heating surface is surrounded by a circular thin fin cut out in one unit body to prevent the preferential bubble generation at the edge of heating block, and to minimize the heat loss from the periphery.

Pool boiling experiments were conducted. After the enough

Table 1 Heat transfer modes observed in boiling of immiscible mixtures classified by Bragg and Westwater[3] arranged by the present authors.

Bottom phase		Top phase			Case	Combination	Case	Combination
		Free boiling	Nucleate boiling	Film boiling				
Free convection	Free boiling				1	Water/mercury	6	Water/R112
	Nucleate boiling	Case2	Case6	Case3	2	Hexane/water	7	Hexane/water
	Film boiling	Case4	Case11	Case9	3	Octadecene/water	8	Butane/water
Film boiling	Case5	Case10	Case8	4	Pentane/mercury	9	Water/mercury	
				5	Water/perchlor-ethylene	10	Hexane/water	
								11

time is elapsed to confirm the steady state of indicated temperatures by all thermocouples and of the pressure, the data is acquired. The condition of critical heat flux is detected by the excursion of temperature in the copper heating block. The heat flux, which is one step before the value resulting temperature excursion, is defined as the critical heat flux. The increment of heat flux near CHF is 10% of the previous heat flux and the increment determines the resolution or the accuracy of CHF measured.

The experiments are performed at 0.1MPa, and FC72 and water are selected as components of the test mixture. Figure 2 shows the images of liquid layers for the immiscible liquid mixtures, where the heights H_1 and H_2 are varied as one of important parameters. Once nucleate boiling occurs, the liquid-liquid interface cannot be smooth but the complex mixing of immiscible liquids occurs. The density of FC72 is larger than that of water, which realizes the layer of FC72 contacting directly to the heating surface at least before the heating. At the same time, FC72 is a more volatile component, and the subcooled nucleate boiling is started at a small surface superheat if the temperature of FC72 contacting the heating surface approaches the saturation temperature, i.e. 56°C, corresponding to the total pressure 0.1MPa. In case less volatile component is accumulated lower position and contact to the heating surface, subcooled boiling cannot be initiated until the temperature of less volatile component approaches its saturation temperature, i.e. 100°C. Because the surface temperature larger than 100°C does not meet the requirement for the cooling of the existing semiconductors, more volatile component should be selected so that it has higher density than that of less volatile component. Actually, the equilibrium temperature of the mixture composed of FC72 and water becomes 51.6°C, where the summation of the corresponding partial pressures for both components equals to the total pressure. This means only subcooled boiling is possible in the immiscible mixture system.

The boiling behavior for very small thickness H_1 of FC72 layer is expected to be quite different from that for liquid pool because the liquid layer on the heating surface is easily consumed by the evaporation. The thickness of liquid layers is varied as an experimental parameter keeping the total thickness at the fixed value of 100mm from the heating surface. The thicknesses of both layers are denoted here by FC72/Water [H_1/H_2]. Three conditions of [5mm/95mm], [10mm/90mm] and [50mm/50mm] are tested.

PHASE EQUILIBRIUM DIAGRAM

Figure 3 shows the phase equilibrium diagram at 0.1MPa for the immiscible liquid mixture FC72/Water. Below the bubble point curve which is represented by a solid line, the mixtures keep the liquid state. Above the dew point curve shown by a broken line the mixture is at the vapor state. The point where the two curves merge represents the azeotropic point, which is observed for all of immiscible liquid mixtures.

The dew point curve in Figs.3 (a) and (b) is calculated from the following equations [6] using the partial pressures of both components corresponding to an equilibrium temperature T .

$$\ln y_1 = -\frac{h_{fg,1}}{R} \left(\frac{1}{T} - \frac{1}{T_{b,1}} \right) \quad (1)$$

$$\ln y_2 = -\frac{h_{fg,2}}{R} \left(\frac{1}{T} - \frac{1}{T_{b,2}} \right) \quad (2)$$

EXPERIMENTAL RESULTS AND DISCUSSION

Figures 4-6 shows the experimental results for immiscible liquid mixtures FC72/Water. Each figure includes also the data for pure FC72 and pure water. The levels for obtained CHF values are also represented by the broken lines in these figures. Both of CHF values for pure FC72 and pure water in the present experiment are higher than those predicted by Zuber correlation [7]. For FC72/Water [5mm/95mm], CHF cannot be observed even at the upper limit of heat flux which is determined by the capacity of cooling coils installed in the boiling vessel.

Pool boiling curve

Figure 4 shows the boiling curves for FC72/water, where the surface superheat is defined by using the equilibrium temperature of 51.6°C. In the case of small thickness of FC72 layer, i.e. FC72/Water [10mm/90mm] and [5mm/95mm], the local burnout and small temperature excursion occur at the heat

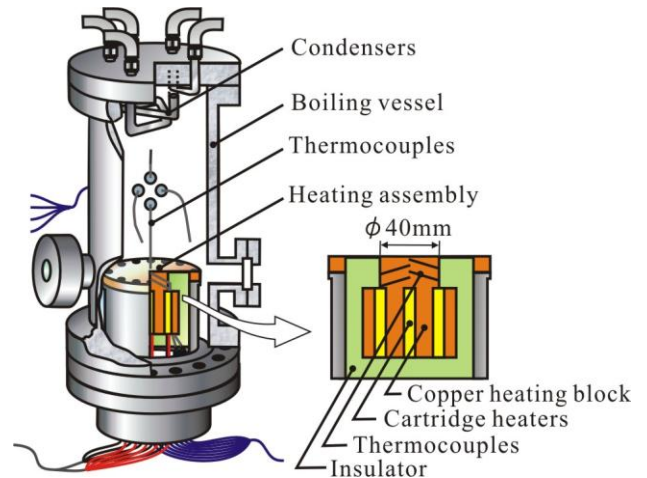


Fig. 1 Schematic of experimental apparatus

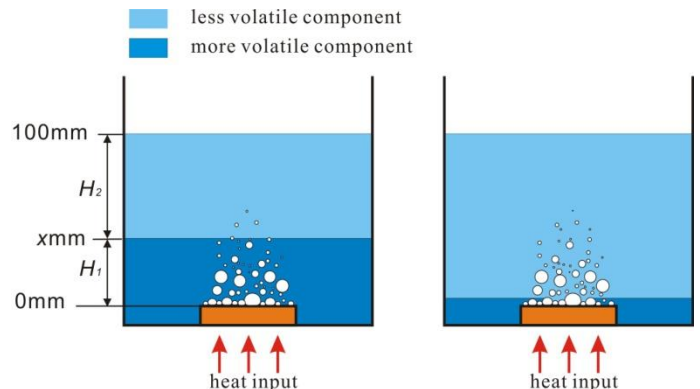


Fig. 2 Image of liquid-liquid interface with different height in nucleate boiling of immiscible mixtures on the horizontal heating surface

flux of around $2 \times 10^5 \text{ W/m}^2$ which is lower than that of pure FC72. The lower CHF than pure FC72 at the same total pressure is curious because subcooled boiling of FC72 occurs for the immiscible mixture. Just before the local dryout, a part of FC72 contacting to the heating surface is replaced by water, where the heat transfer to water by natural convection increases excessively the surface temperature. The evaporation of FC72 and the extension of local dry patches are promoted resulting the burnout at lower heat flux than that for pure FC72. After the increase of wall superheat ΔT_{sat} by around 20K, the gradient of pool boiling curve for FC72/Water mixture becomes similar to that for pure water. The interruption of further increase of surface superheat due to the local dryout is caused by the existence of water located at the top part of the liquid layers, i.e. the descending water cools down the heating surface. This small temperature excursion is one of important phenomena inherent in the immiscible mixtures, and is referred to as "intermediate burnout" in the present paper. On the other hand, for a large thickness of FC72 at FC72/Water [50mm/50mm], the heat transfer characteristics are almost the same as those for pure FC72. These results imply the existence of a boundary dominated, for example, by the liquid layer thickness, where the heat transfer characteristic for immiscible mixtures changes from those for pure liquid of more volatile component.

Heat transfer coefficient

Figure 5 shows heat transfer coefficients. The heat transfer coefficient is lower than that of pure liquids, except the case of FC72/Water [50mm/50mm] which is almost equivalent to pure FC72 as mentioned before. The heat transfer coefficient is defined by the surface superheat ΔT_{sat} , where the measured equilibrium temperature independent of the volumetric ratio is used. At low heat flux before the occurrence of the intermediate burnout, the deteriorated heat transfer from that of pure FC72 is caused by the non-negligible contribution of natural convection by descending water. The situation is fundamentally different from the case of miscible mixtures, where the deterioration is caused mainly by the diffusion resistance of a component. On the other hand, at high heat flux beyond the intermediate

burnout, the heat transfer deterioration is again observed. This is caused mainly by the definition of the heat transfer coefficient by using the equilibrium temperature at around 52°C measured for mixtures instead of around 100°C for pure water.

Critical heat flux

In Fig. 4, the levels of critical heat flux are represented by the broken lines. As mentioned before, the actual CHF value accompanied by the serious temperature excursion was not measured for FC72/Water [5mm/95mm] because of the limited cooling capacity of the apparatus. However, it is noteworthy that the CHF value is at least 36% higher than that of pure water. On the other hand, CHF for FC72/Water [10mm/90mm] is quite lower than that of pure water. This difference in the trend can never be explained by the variation of thermophysical properties evaluated by using the volume ratios for assumed "miscible" mixtures. The difference seems to be caused by the different amount of subcooled water which can be supplied to the heating surface even under such high heat flux conditions. Ivey-Morris [8] proposed the following equation;

$$\frac{q_{CHF,sub}}{q_{CHF,sat}} = 1 + 0.1 \left(\frac{\rho_g}{\rho_l} \right)^{1/4} \frac{c_{pl} \rho_l \Delta T_{sub}}{\Delta h_{fg} \rho_g} \quad (3)$$

The CHF estimated by this equation is $4.3 \times 10^6 \text{ W/m}^2$ for pure subcooled water. In the evaluation, the liquid subcooling of 48K corresponding to the measured equilibrium temperature around 52°C at 0.1MPa is substituted into Eq.(3). It is obvious that the measured CHF value at least for FC72/Water [10mm/90mm] can never be explained only by the subcooling of pure water. The thick layer of FC72 becomes a barrier for the subcooled water to descend and to contact the heating surface.

SUMMARY ON NUCLEATE BOILING OF IMMISCIBLE LIQUID MIXTURES

Figure 6 shows the heat flux versus heating surface temperature for FC72/Water. In Fig. 7, the trends of immiscible liquid mixtures are represented by the boiling curve with

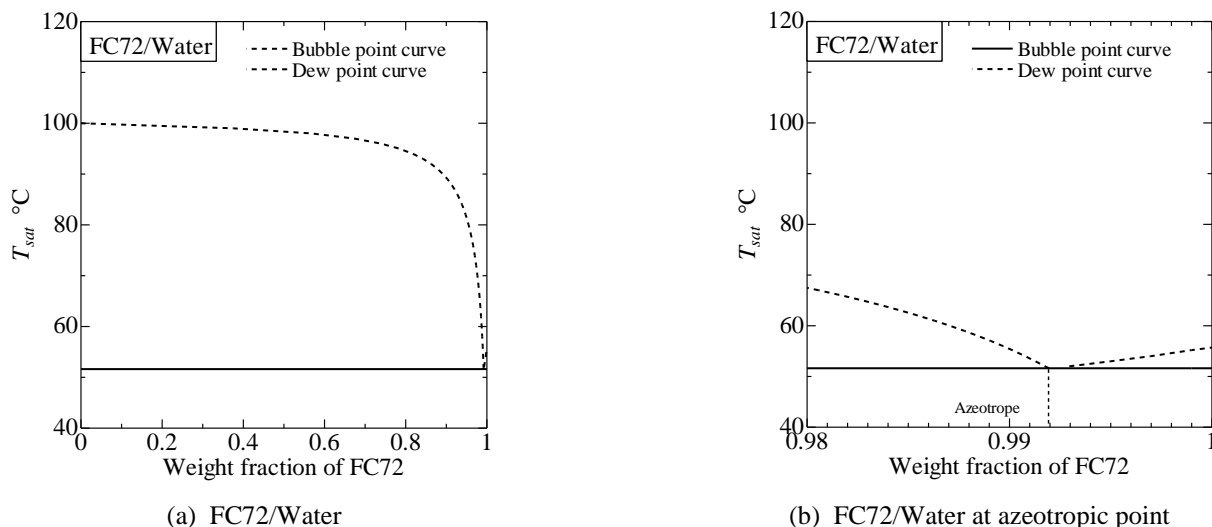


Fig. 3 Phase equilibrium diagram for immiscible mixture FC72/Water at 0.1MPa

reference to the data for FC72/Water [5mm/95mm]. In a number of existing studies, the surface superheat ΔT_{sat} is used as an abscissa of boiling curve [9]. To understand the boiling of immiscible binary mixtures for the application to the cooling of semiconductors, the plot of heat flux versus the surface temperature is more useful.

Pool boiling characteristics for immiscible mixtures, where the layer of more volatile component with higher density is contacting the heating surface at least before the incipience of boiling, can be classified into two regimes; more volatile dominated regime and less volatile dominated regime which are connected by the temperature jump due to the intermediate burnout. In the more volatile dominated regime, the boiling curve is almost the same as that for pure liquid, because the influence of the layer thickness for less volatile liquid is not large even at the minimum layer thickness 5mm of more volatile liquid tested in this experiment. On the other hand, the boiling curves in the less volatile dominated regime depend on the layer thickness of more volatile liquid located at the bottom, where the penetration of less volatile liquid from the top is seriously influenced by the thickness of more volatile component.

In the less volatile dominated regime, the deterioration of heat transfer coefficient for immiscible mixtures is clear as already shown in Fig.5. This is mainly due to the excessive surface superheat defined by using the low equilibrium temperature. However, the surface temperatures for the immiscible mixtures are lower than that of pure water, because subcooled water descends toward the heating surface and FC72 is replaced partly by water. In such a situation, the deteriorated heat transfer of the immiscible mixture is superficial and it has larger cooling ability provided that its CHF value is also larger than that of pure water.

The value of CHF, not the heat flux of intermediate burnout for the immiscible mixtures, is seriously influenced by the thickness of liquid layer for more volatile component. The decrease of the surface superheat is directly related to the amount of less volatile liquid supplied from the top part, and it is, in turn, influenced by the thickness of more volatile liquid occupied on the heating surface. The thin layer of more volatile liquid allows the penetration of less volatile liquid and vice versa. As a result, a thin layer of FC72 increases markedly the CHF value from that for pure water, while a very thick layer of FC72 prevents the penetration of subcooled water and behaves as if it is pure FC72.

Between the more volatile dominated regime and the less volatile dominated regime, the small temperature jump due to "intermediate burnout" occurs because of low CHF values inherent in more volatile liquid. The heat flux at the intermediate burnout seems to be quite insensitive to the thickness of more volatile liquid and is lower than that for pure more volatile liquid by the reason above mentioned. However, the value is the same as that of pure more volatile liquid for the extremely large thickness of the relevant component.

It is noteworthy that, under optimized conditions, the boiling of immiscible mixtures reduces the surface temperature and simultaneously increases CHF from the values for pure liquid of less volatile component. The situation is quite

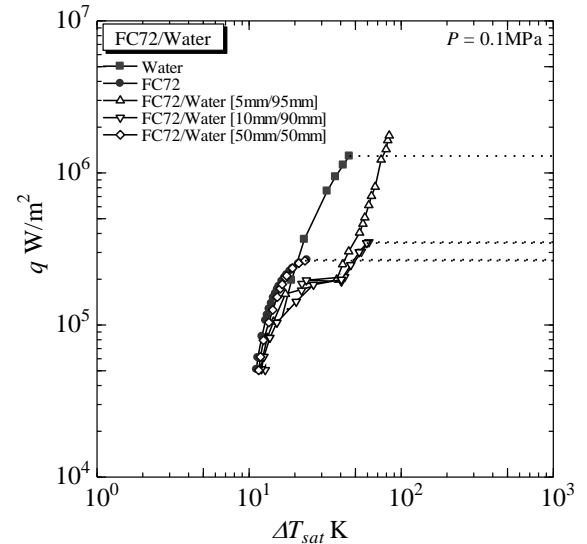


Fig. 4 Pool boiling curve for FC72/Water

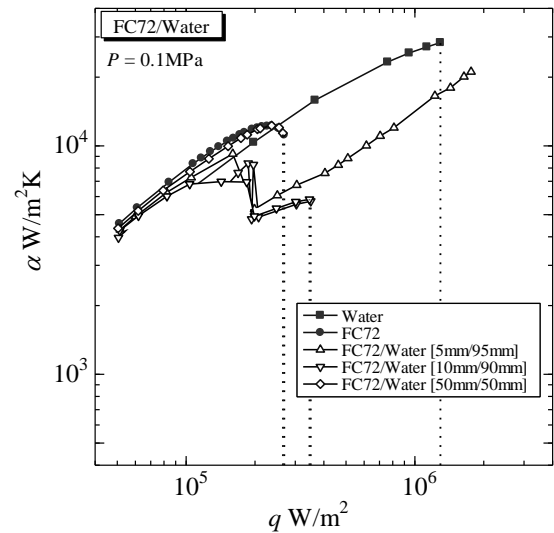


Fig. 5 Heat transfer coefficient for FC72/Water

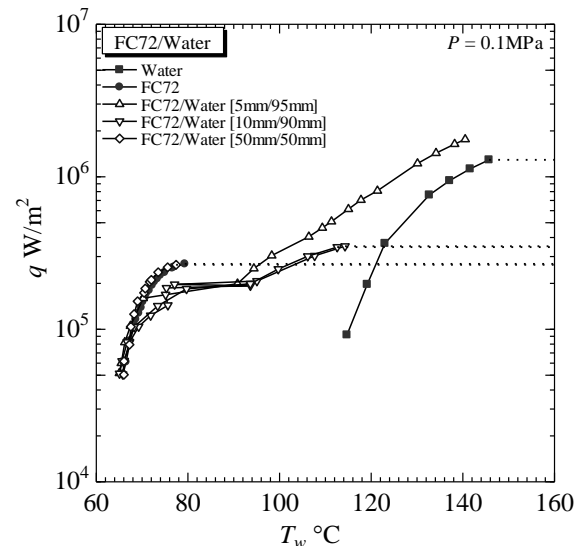


Fig. 6 Heat flux versus heating surface temperature for FC72/Water

different from that for non-azeotropic miscible mixtures, where the addition of more volatile component decreases the surface temperature and simultaneously CHF value as far as the boiling on the flat plate is concerned. In short, the boiling for immiscible liquid mixtures has a potential to be applied to the high-performance heat exchange.

CONCLUSIONS

The boiling heat transfer and the critical heat flux of immiscible liquid mixtures were investigated using FC72/Water at 0.1MPa. The components of the mixture were stratified horizontally on the flat heating surface. The thickness of liquid layer before the heating was varied keeping the total thickness unchanged. In the case of thin layer of more volatile component with higher density, i.e. FC72, contacting directly to the heating surface, the following characteristics inherent in the immiscible mixture were observed.

i) A new phenomena “intermediate burnout” occurs accompanied by the small jump of surface temperature at the heat flux lower than the CHF for pure FC72. The penetration of subcooled water from the top increases the surface temperature and promotes the burnout of FC72.

ii) When heat flux is further increased, the heat transfer becomes “less volatile liquid, i.e. water, dominated”, where the surface temperature is kept at lower than that for pure water and simultaneously the actual CHF accompanied by the serious temperature excursion becomes large.

The self-sustaining subcooling of less volatile component brings the benefits in ii). Furthermore, the decrease of surface temperature is possible even at the pressure higher than the

atmospheric. This prevents the mixing of incondensable gases from the outside. By these features, the immiscible mixtures have a potential to become a promising working medium for the high-performance cooling system.

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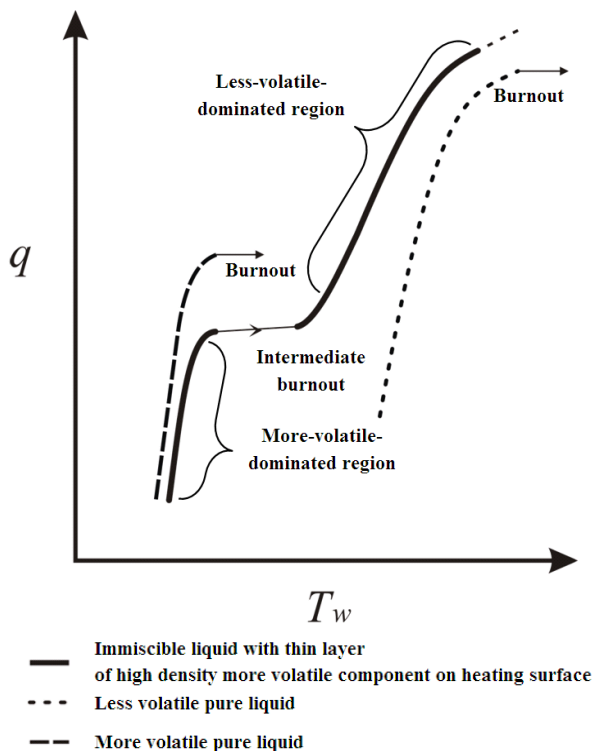


Fig. 7 Pool boiling characteristics of immiscible liquid mixtures with thin layer of more volatile component contacting the heating surface