IMPROVEMENT OF COOLING PERFORMANCE FOR ELECTRONIC DEVICES BY NUCLEATE BOILING OF IMMISIBLE MIXTURES

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Abstract. The employment of immiscible mixtures in nucleate boiling improves the cooling performance drastically. Increase of CHF is possible by high subcooling of less-volatile liquid compressed by high-vapor pressure of more-volatile component. In addition, the reduction of surface temperature from that of pure less-volatile liquid is resulted from the co-existed vapor of more-volatile component. And the increase of pressure above the atmospheric keeping low liquid temperature is possible to prevent the mixing of incondensable gases. Furthermore, boiling can be initiated at lower surface temperature, which is required for the cooling of e.g. automobile inverters accompanied by large variation of thermal load. The performance of immiscible mixtures in nucleate boiling are summarized with reference to new data for Novec7100/water and FC72/ethanol and existing data obtained by the present authors.

1. Introduction

The heat generation density from semiconductors is increasing by the development of electronics technology. Under such a situation, the latent heat transportation during the boiling and evaporation attracts much attention. There are a number of researches on the boiling heat transfer for the cooling of semiconductor chips, where micro or mini-structures are devised for flow boiling systems and on the surface for pool boiling systems. On the other hand, the use of devised liquids are concentrated on the development of nano-fluids which are not verified for the long-term operation under the conditions of boiling.

In non-azetropic miscible mixtures, the heat transfer deterioration cannot be avoidable. The preferential evaporation of more-volatile component results in the increase of interfacial temperature and in turn the decrease of the effective surface superheat. The existence of Marangoni effect has a possibility to enhance the heat transfer and increase critical heat flux (CHF). The increase of CHF were reported for a heated wire in [1][2] and for heat pipes [3]. According to the experimental results by using a heating surface of flat plate [4], the heat transfer enhancement by 30 to 40% was observed at very low concentration of alcohol for i-propanol and n-propanol aqueous solutions. On the other hand, only the decrease of CHF was observed. Marangoni effect promotes the evaporation from the microlayer of primary bubbles underneath a coalesced bubble and the consumption of liquid in macrolayer was promoted locally before the supply of liquid by the detachment of a coalesced bubble.

It was clarified from a series of pool boiling experiments by the present authors that the immiscible mixtures have superior heat transfer characteristics for the cooling of semiconductors, which are summarized as follows [5-10] ; i) The increase of critical heat flux due to the self-sustaining high subcooling of less-volatile liquid, ii) the reduction of surface temperatures due to the heat transfer

enhancement caused by the generation of bubbles of more-volatile component, iii) the system pressure higher than the atmospheric keeping low liquid equilibrium temperature, iv) the low surface temperature at the boiling initiation if more-volatile liquid with larger density is heated preferentially.

In despite of the superior heat transfer characteristics above mentioned, there are very limited numbers of existing studies on the boiling of immiscible mixtures [11-13], where no general information about the heat transfer characteristics were derived. There was almost no attempt to use immiscible mixtures for the purpose of cooling.

In this study, we add the new experimental data for two different combinations of immiscible mixtures, Novec7100/water and FC72/ethanol, and the heat transfer performance is compared with the existing data obtained by the present authors.

2. Immiscible mixtures from thermodynamic point of view

For immiscible mixtures at the thermal equilibrium state of liquid and vapor, the total pressure P_{total} is obtained by the summation of saturated vapor pressures $P_{sat,1}$ and $P_{sat,2}$ for more-volatile and less-volatile components, respectively, at the equilibrium temperature T_e.

$$P_{sat,1}(T_e) + P_{sat,2}(T_e) = P_{total} \qquad (1)$$

Figure 1 shows the vapor pressure curves for more-volatile and less-volatile components, where an equilibrium state is also indicated. Because the more-volatile component has a larger vapor pressure at the equilibrium temperature T_e, the less-volatile liquid is compressed largely from the saturation pressure $P_{sat,2}(T_e)$ to the total pressure P_{total}. As a result, high subcooling $\Delta T_{sub,2}$ is imposed to less-volatile liquid. On the other hand, the more-volatile liquid is compressed slightly from the saturation pressure $P_{sat,1}(T_e)$ to the total pressure P_{total}. Then, small subcooling $\Delta T_{sub,1}$ is imposed to more-volatile liquid. As a consequence, self-sustaining subcooled conditions are realized for both of component liquids. Especially, the large subcooling, imposed to the less-volatile liquid, is expected to increase CHF value provided that the heat transfer to the less-volatile liquid is dominated at high heat flux.

Figure 2 shows phase equilibrium diagram obtained from the Clasius-Clapeyron equations and ideal gas approximation [14]. Independent of the liquid concentration, the concentration of vapor is fixed at the value similar to the azeotropic point of miscible mixtures. Quite different from the non-azeotropic miscible mixtures, the interfacial temperature is not changed by the preferential evaporation of more-volatile component, provided that both of components have liquid phases. This implies the free of heat transfer deterioration for immiscible mixtures in both of nucleate boiling and condensation.

3. Experimental apparatus and procedure

Pool boiling experiments are conducted using the apparatus shown in Figure 3. A circular flat heating surface of 40 mm in diameter is installed horizontally facing upwards. The flat surface has no micro-structures but is finished by the polishing using the emery paper 0/4 (#600). The heating surface is located at the upper end of a cylindrical heated block, where cartridge heaters are inserted in the bottom. The maximum heat flux at the surface is 4.5MW/m^2. The heat flux and the temperature of the heating surface are evaluated from the measured temperature distributions in the copper cylindrical block by the thermal conduction analysis. Eight thermocouples are inserted at different depths of 1mm, 7mm, 13mm and 19mm at the center and at the radial distance of 17mm from the center. To prevent the preferential nucleation from the periphery of the circular heating surface, a thin fin structure surrounding it is cut out in one unit body as shown in the figure. The bulk temperatures are measured by three thermocouples at the position of 2mm, 80mm and 160mm above the center of heating surface. The first two are used for the measurement of liquid temperatures, while the last one is for vapor temperature. The heat transfer coefficients are defined by using the liquid temperature at the position of 80mm above the heating surface. The generated vapor is condensed at the surface of cooling coils hanged from the top flange, which keeps the concentration of bulk liquid almost constant. The condensate is returned to the periphery
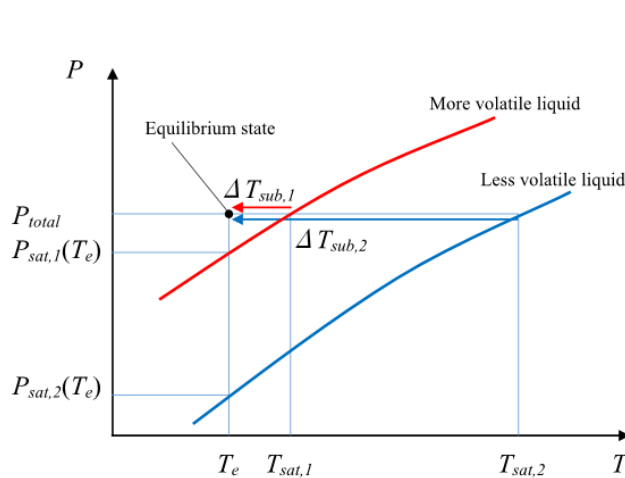
Figure 1. Degree of subcoolings imposed to immiscible mixtures under the equilibrium state.
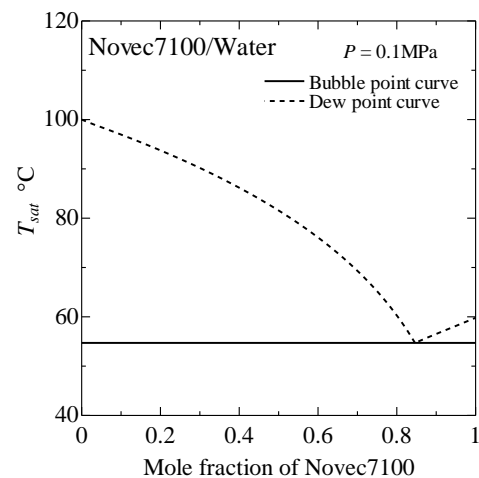

Figure 2. Phase equilibrium diagrams at 0.1 MPa for Novec7100/Water.

of heating surface assembly by the aid of a conical guide plate located at the bottom of the condenser coils. Before the starts of measurements, liquids are boiled to degas the dissolved air.

Considering the error of 0.2 mm for the thermocouple locations in the heating block and the error of 0.025 K for the temperature measurement by them, the uncertainties in the surface temperature evaluated by the extrapolation of temperatures in the heating block are 0.075K and 0.53 K at heat fluxes of 10^5 and 10^6 W/m^2. The corresponding uncertainties of temperature difference between the heating surface and bulk liquid are 0.10 and 0.55 K, resulting in the error of 1.0% and 1.4% for the measured temperature differences of 10 and 40 K, respectively, for the relevant heat fluxes. The errors of heat flux evaluated from the gradient of thermocouples are 3.4 and 24 kW/m^2 reflecting the uncertainties for the thermocouple locations and temperature measurement above mentioned, which result in the errors of 3.4% and 2.4% for the relevant heat fluxes, respectively. Therefore, the errors of heat transfer coefficient are 4.4% and 3.8% at heat fluxes of 10^5 and 10^6 W/m^2, respectively.

Two immiscible mixtures of Novec7100/water and FC72/ethanol are tested at pressure of 0.1MPa, where Novec7100 and FC72 are more-volatile components with higher liquid densities. As references, the results of FC72/water [10] and FC72/n-propanol [8] are cited in the following section to compare the heat transfer characteristics by the change of one component from the mixtures tested here. In both reference immiscible mixtures, FC72 is the more-volatile component with higher liquid density. Table 1 summarizes the equilibrium temperatures and corresponding self-sustaining subcoolings of the component liquids at 0.1MPa. The properties of component pure fluids are cited from the published data [15] and [16].

The composition of component liquids are varied as an important experimental parameter which is adjusted by the height of liquid layers as shown in Figure 4. The value H_1 indicates a liquid level from the heating surface of more-volatile component with higher density before heating, and H_2 is a liquid layer thickness of less-volatile component with lower density. The total of liquid heights is kept at a constant $H_1 + H_2 = 100$ mm. The case of $H_1 = 0$ implies that less volatile liquid is accumulated only in the gap between the heating block assembly and inner wall of the vessel before the heating. The combination of liquid thicknesses before heating is represented by [H_1/H_2] in the figures of experimental results, where H_1 is varies as 0, 5, 10, 50mm in a series of the experiments. The value of H_1 is directly measured across the window of the vessel. The difficulty to improve the accuracy of the measurement and the sensitivity of heat transfer characteristics to the layer thickness cause data scattering when it is correlated by the value of H_1. The liquid layer thickness H_2 has a secondary effect on the heat transfer characteristics.
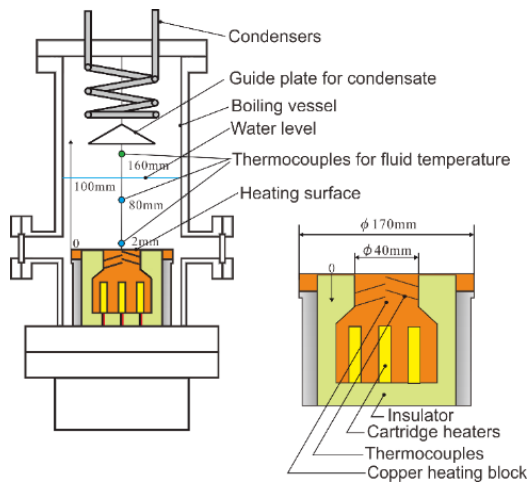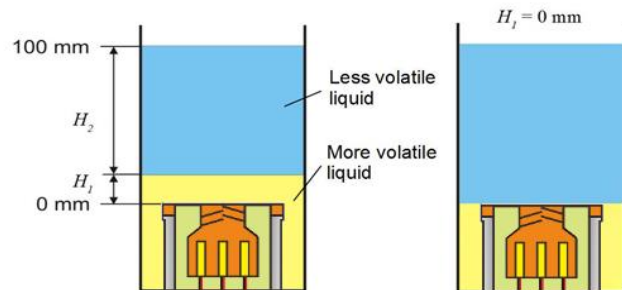
Figure 3. Experimental apparatus.

Figure 4. Definition of liquid layer thicknesses H_1 and H_2 for both component liquids before heating.

Table 1. Equilibrium temperature and self-sustaining subcoolings of component liquids at 0.1MPa.

More-volatile component ($T_{sat,1}$)	Less-volatile component ($T_{sat,2}$)	Mixture T_e [°C]	$\Delta T_{sub,1}$ [K]	$\Delta T_{sub,2}$ [K]
Novec7100 (59.8°C)	Water (100°C)	54.7	5.1	45.3
FC72 (55.9°C)	Ethanol(78.3°C)	47.3	8.7	31.0
FC72 (55.9°C)	Water (100°C)	51.6	4.1	48.4
FC72 (55.9°C)	n-Propanol(97.3°C)	51.7	4.2	45.6

4. Experimental results

4.1. Heat transfer characteristics of immiscible mixtures with water

Figures 5 and 6 show experimental results for Novec7100/water and FC72/water [10], respectively. The latter is given as a reference to evaluate the data obtained here. Figures 5(a) and 6(a) show the relation between heat flux q and the temperature difference ΔT_b between the heating surface and bulk liquid measured at 80mm above the heating surface for the immiscible mixtures. The figures also include the relation between heat flux and surface superheat ΔT_{sat} for the pure component liquids. Because both liquids are subcooled under the equilibrium condition, the data at saturated conditions for pure liquids and the subcooled conditions for mixtures are compared in the same figure. In general, the temperature difference ΔT_b under subcooled conditions is larger as much as degree of subcooling from the temperature difference ΔT_{sat} under the saturated conditions. Therefore, the comparison of both temperature differences between the subcooled and saturated conditions to evaluate the heat transfer performance is not appropriate. However, it is impossible for the immiscible mixtures to define the liquid temperature corresponding to the saturation temperature of pure liquids. The same is true for the evaluation of the performance by using heat transfer coefficients α if the temperature difference ΔT_b is used for the immiscible mixtures. As shown in Figures 5(b) and 6(b), the heat transfer coefficients for the immiscible mixtures are deteriorated superficially, except the case of H_1=50mm (Figure 6(b)). In this case, the heat transfer to more-volatile component FC72 occurs under the small subcooling of 4.1 K as shown in Table 1, and the heat transfer coefficients are almost the same as those for pure FC72. The similar trend is confirmed for H_1=10mm at low heat flux.

Both of pure Novec7100 and FC72 have low CHF values of 2.5×10^5 W/m² to 2.7×10^5 W/m², respectively. If the amount of more-volatile liquid is increased as H_1=5mm and H_1=10mm, the properties
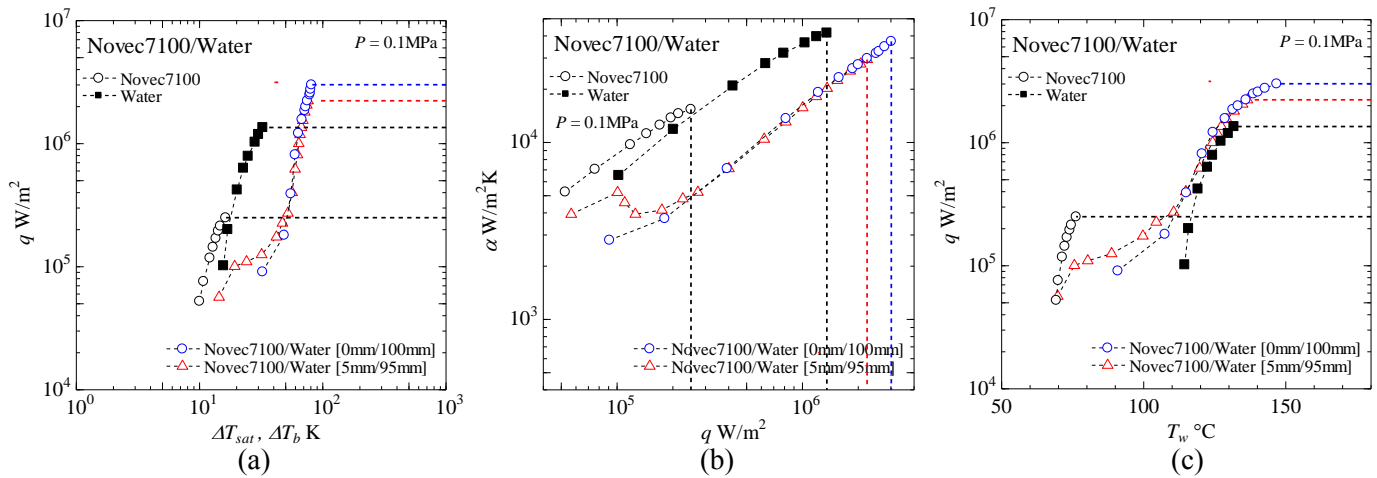
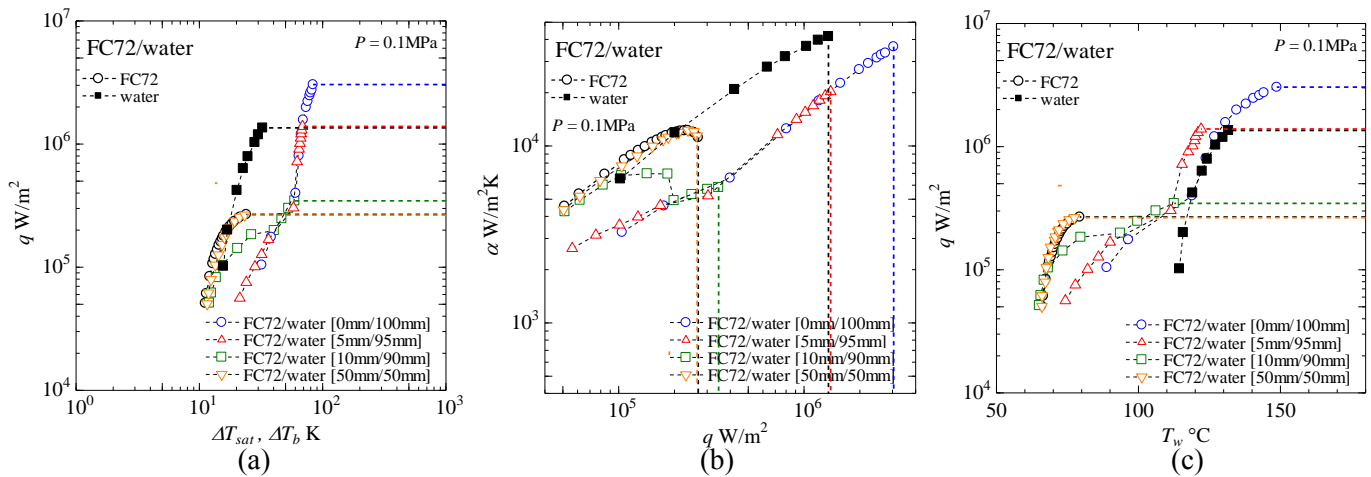**Figure 5.** Heat transfer characteristic for Novec7100/water

Figure 6. Heat transfer characteristic for FC72/water [10]

of more-volatile component reduce CHF from that for H_I=0mm. Furthermore, for FC72/water at H_I=10mm, small jump of temperature difference, i.e. jump of surface temperature, is observed at around 2.0×10^5 W/m². which is caused by the burnout-like phenomena of heating surface immersed in the liquid of FC72. The surface temperature is gradually shifted by the mixing of water into FC72. The phenomena is referred to as "Intermediate heat flux burnout" by the present authors [8]. Two mechanisms seems to be concerned for the phenomena. The major mechanism for the increase of surface temperature is caused by the lateral coalescence of FC72 bubbles under the liquid-liquid interface acting as barrier for the penetration of rising bubbles of FC72. As a consequence, the lateral size of flattened bubbles exceeds the wave length of Taylor instability and liquid water starts to contact the heating surface. Another mechanism of the mixing is due to the penetration of FC72 bubbles into the liquid water across the liquid-liquid interface. In the former case, the temperature of heating surface jumps abruptly with small increment of heat flux and the heat transfer to FC72 is suddenly replaced by the heat transfer to water, while the gradual change of surface temperature with increasing heat flux, as shown in Figure 6(a), is resulted from the latter mechanism.

The situation of "Intermediate heat flux burnout" is well recognized by the plot of heat flux q versus heating surface temperature T_w in Figures 5(c) and 6(c). Also, the performance of heat transfer becomes clear in these figures. The surface temperature of immiscible mixtures are located between those of pure more-volatile component and pure less-volatile component. The variations of surface temperature with composition in despite of the same equilibrium temperature of liquid is caused by the difference in the
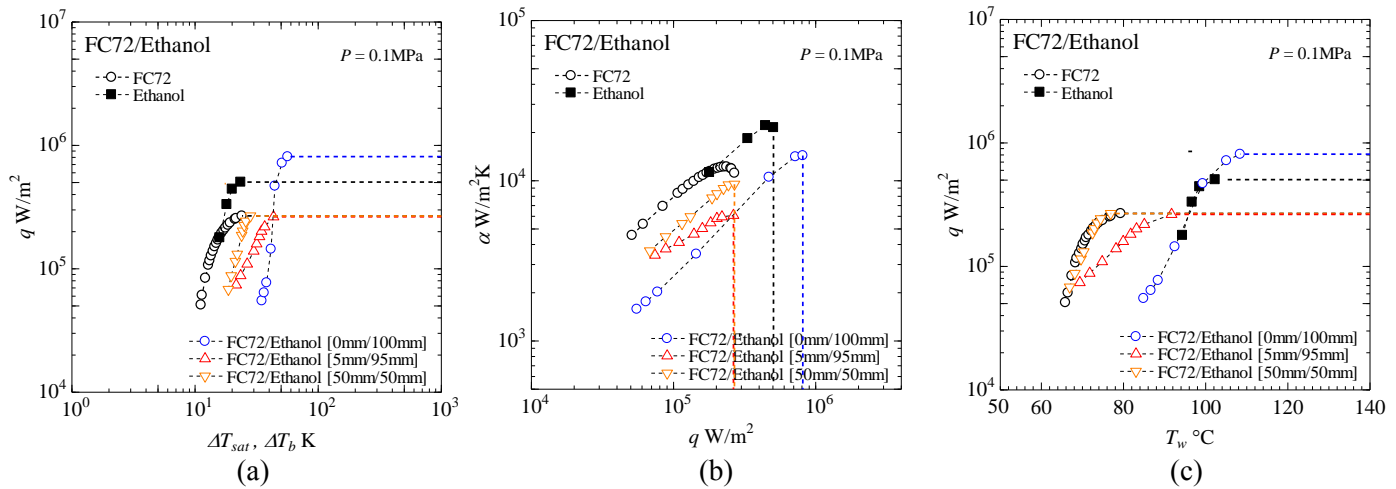
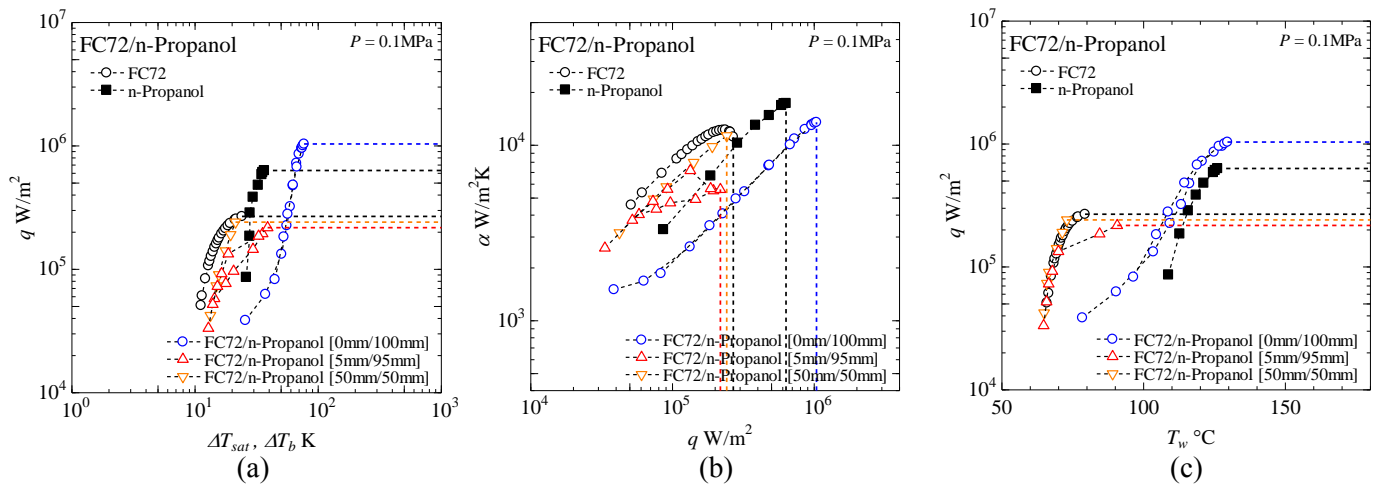**Figure 7.** Heat transfer characteristics for FC72/ethanol

Figure 8. Heat transfer characteristics for FC72/n-propanol [8]

component liquid to which heat is directly transferred, and the difference in the heat transfer mode and heat transfer enhancement.

For Novec7100/water at H_1=0mm, mixing of the component liquids occurs even at low heat flux. The droplets of Novec7100 are carried on the heating surface by the disturbance of liquid-liquid interface at the same horizontal plane as that of heating surface. The heat transfer to Novec7100/water is shifted to the heat transfer to water as the increase of heat flux. The change of liquid contacting the heating surface is confirmed clearly for H_1=5mm. At lowest heat flux the heat transfer mode is nucleate boiling of Novec7100, and the surface temperature increases gradually with increase of heat flux by the mixing of water. The heat transfer to water is realized at high heat flux. The increase of gradient is caused by the nucleate boiling of water in addition to the nucleate boiling of Novec7100. It is clear that the surface temperature for the incipience of boiling seems to be far larger than 120°C for pure water because of the large overshoot of the surface temperature. On the other hand, nucleate boiling starts at 70°C by the mixing of Novec7100. The prevention of the excessive overshoot of surface temperature is one of the important characteristics for the nucleate boiling of immiscible mixtures. It is also an essential requirement for the cooling of automobile inverters where the highest power dissipation is unavoidable by the increase in the revolution of engine.

For Novec7100/water, the reduction of surface temperature at high heat flux is not large compared to FC72/water at H_1=5mm where the nucleate boiling of water is enhanced by the generation of FC72

bubbles. The mechanism of the heat transfer enhancement for FC72/water seems to be caused by the increased agitation of liquid-vapor interface on the heating surface and/or the enhancement of evaporation from microlayers of water extended widely by the aid of FC72 bubbles.

4.2. Heat transfer characteristics of immiscible mixtures composed of FC72 and alcohol
Figures 7 and 8 show the heat transfer characteristic for FC72/ethanol and FC72/n-propanol [5]. By the preceding discussion, it is appropriate to evaluate the heat transfer performance by using the relation between heat flux q versus the surface temperature T_w given in Figures.7(c) and 8(c). Values of CHF for ethanol and n-propanol is increased from 0.50×10^5 W/m^2 to 0.81×10^5 W/m^2 and from 0.63×10^5 W/m^2 to 1.04×10^6 W/m^2, respectively, by the addition of small amount, i.e. H_1=0mm, of FC72. From Table 1, the degree of self-sustaining subcooling is 31.0K and 45.6K for FC72/ethanol and FC72/n-propanol, respectively. The increment of CHF is similar in despite of larger subcooling for FC72/n-propanol. This might be due to the high sensitivity of heat transfer characteristics to the liquid layer thickness H_1 of more-volatile liquid. The adjustment of H_1 with high accuracy is quite difficult because of the structure of the boiling vessel.

For FC72/ethanol at H_1=5mm, nucleate boiling of FC72 started at low surface temperature and the mixing of liquid water starts even at low heat flux. As a consequence, the surface temperature gradually shifts towards that of pure ethanol. It is clear that the surface temperature at the boiling incipience for H_1=0mm is higher than the value for H_1=5mm because of the shortage of FC72 as a starter of nucleate boiling.

For FC72/n-propanol, the reduction of surface temperature from that of less-volatile component, n-propanol, is observed at H_1=0mm by the same reason as described in the last part of the preceding section. The existence of such substantial enhancement of heat transfer might be true also for FC72/ethanol under the condition of H_1=10mm to be tested in further studies.

5. Conclusions
To improve cooling performance for electronic devices, nucleate boiling of immiscible liquid mixtures were studied. New data of two immiscible mixtures of Novec7100/water and FC72/ethanol was obtained in this study. The following was concluded.
1. The critical heat flux as much as 3.0×10^6 W/m^2 is realized by the addition of small amount of Novec7100 to water. This is caused by the self-sustaining high liquid subcooling of water compressed by high partial pressure of a more-volatile component, Novec7100.
2. The surface temperature at the boiling incipience is drastically reduced from that of pure water by 50K at least, when the small amount of Novec7100 is added to water. On the other hand, the reduction of surface temperature from that of ethanol is more than 20K by the addition of small amount of FC72. The reduction of surface temperature at the boiling incipience is an important requirement for the cooling of automobile inverters with high fluctuation of thermal dissipation rate.

Nomenclature

H	thickness of liquid layer	(m)		the surface and liquid	(K)
P	pressure	(N/m^2)			
P_{total}	total pressure	(N/m^2)	Subscripts		
q	heat flux	(W/m^2)	1	more volatile component	
q_{CHF}	critical heat flux	(W/m^2)	2	less volatile component	
T	temperature	(°C)	b	bulk liquid	
T_w	heating surface temperature	(°C)	e	equilibrium	
			sat	saturation	
Greek Symbols			sub	subcooled	
α	heat transfer coefficient (W/m$^2 \cdot$K)				
ΔT	temperature difference between				

Acknowledgment

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