

# Enhancement of pool boiling heat transfer in water and ethanol/water mixtures with surface-active agent

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## Abstract

Surface tension of ethanol/water mixtures is measured over the whole ethanol fraction range and the effect of the surface-active agent on surface tension is also measured in the mixtures, in order to gain basic data related to enhancement of the heat transfer coefficient in water and the mixtures. The boiling heat transfer coefficient, the onset of boiling and the critical heat flux in water and ethanol/water mixtures, with and without the surface-active agent, have been measured on a horizontal fine heated wire at a pressure of 0.1 MPa. The experiment was carried out in the whole range of the ethanol fraction and in a surfactant concentration of 0–5000 ppm. The experiment shows that the coefficients were enhanced in a lower ethanol fraction ( $C \leq 0.5$ ) and in low heat flux which is slightly higher than heat flux at the onset of boiling. It is also found that the enhancement due to the surfactant disappears over 1000 ppm. Finally, we clarify that depression of the surface tension by the surfactant remarkably enhances the heat transfer coefficients in the nucleate pool boiling.

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## 1. Introduction

Many studies have been performed on the enhancement of boiling heat transfer in pure substances using passive and active methods such as additives, micro-channels and grooves. The effects of surface-active agents on the boiling heat transfer were reported [1–11] as a means of heat transfer enhancement. For example, Frost and Kippenhan [1] reported that the heat transfer coefficient is improved by an increase in nuclea-

tion sites using the surfactant additive for a forced convection boiling heat transfer of water in a vertical tube. Yang and Maa [2] and Tzan and Yang [3] reported that an anionic surfactant improves the coefficient in a pool boiling of water. Wang and Hartnett [4] also reported that both anionic and nonionic surfactants improved the boiling heat transfer coefficient in the low heat flux region.

Recently, the usage of binary mixtures instead of pure substances has been tried as a working fluid in a heat engine cycle operated in small temperature differences between heat sources and in a heat pump cycle to improve the thermal efficiency of the cycles. In this recent situation, for example, Inoue and Monde [12] and Fujita and Tsutsui [13] measured the boiling heat

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### Nomenclature

$C$	mass fraction of ethanol	$q_{\text{CHF}}$	critical heat flux
$C_s$	mass concentration of surfactant	$q_{\text{ob}}$	heat flux at onset of boiling
$g$	gravitational acceleration	$r$	inner radius of fine pipe
$h$	heat transfer coefficient (or head difference of the manometer only in Fig. 1)	$T$	temperature
$L$	depth of fine pipe	$\rho_1, \rho_2$	density of manometer and test liquid, respectively
$P$	pressure	$\sigma$	surface tension
$q$	heat flux		

transfer coefficients in wide ranges of pressure and heat flux for fluorocarbon mixtures and alcohol/water mixtures, respectively. They reported that the coefficients in binary mixtures are decreased as compared with those in pure substances and proposed general correlations to predict the boiling heat transfer coefficients of binary mixtures. Furthermore, Inoue et al. [14] measured the heat transfer coefficients of ammonia/water mixtures and reported that it is difficult to predict the coefficients over the whole fraction range with the existing correlations.

As mentioned above, the characteristics of deterioration in the heat transfer of mixtures have recently been clarified, but there are few reports on the heat transfer enhancement of binary mixtures. Here, the effects of surface-active agents on the boiling heat transfer were reported as a means of the heat transfer enhancement. This is the reason that surface tension is one of the important factors governing the boiling heat transfer.

In this study, first, the effect of the surfactant concentration on surface tension was clarified in water and ethanol/water mixtures with a cationic surfactant as the fundamental study on boiling heat transfer enhancement in binary mixtures. Second, we measured the heat transfer coefficient, the onset of boiling and the critical heat flux (CHF) in the mixtures with the surfactant at a pressure of 0.1 MPa and in overall range of the ethanol fraction and in the range of 0–5000 ppm of the surfactant concentration. We clarified experimentally the effect of the surfactant on surface tension, the heat transfer coefficient, the onset of boiling and the CHF. Here, the present surfactant was employed, because its surface tension is very small and its adsorption to a metal surface is excellent.

## 2. Experimental apparatus and procedure

### 2.1. Measurement of surface tension

Surface tension was measured using the commonly utilized maximum bubble pressure method. Fig. 1 shows a measurement apparatus for surface tension. A test

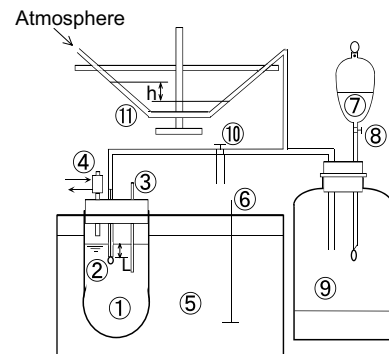


Fig. 1. Schematic diagram of experimental apparatus for surface tension measurement. (1) Test vessel, (2) fine pipe, (3) thermometer, (4) condenser, (5) thermostat bath, (6) agitator, (7) oil, (8) valve, (9) pressure vessel, (10) valve, (11) manometer.

liquid is supplied to test vessel (1). Thermometer (3) and condenser (4) are attached to the vessel. Fine pipe (2) (0.10-mm inside diameter) on the market without any process of the tip surface is inserted in the test liquid and is led into pressure vessel (9) and manometer (11). The test vessel is immersed in thermostat bath (5) to be isolated from the surroundings. Temperature, the head of the test liquid and the fraction of test liquid mixtures in the test vessel are kept constant by controlling the supply of cooling liquid to the condenser during the experiment.

The reference head of the manometer was measured after the test liquid temperature became constant. Oil (7) was poured gradually into the pressure vessel by opening valve (8) after atmospheric pressure was intercepted by valve (10). The head difference,  $h$  in Fig. 1, of the manometer was measured when bubbles began emitting from the fine pipe at a constant frequency. The surface tension was calculated from the measured value of  $h$  in Eq. (1)

$$\sigma = \frac{r}{2}(\rho_1 h - \rho_2 L)g, \quad (1)$$

where, densities of mixtures were given by [15] and the change in density with addition of the surfactant was ignored.

## 2.2. Measurement of heat transfer coefficient, onset of boiling and critical heat flux

Fig. 2 shows an experimental apparatus to measure the heat transfer coefficient. Platinum heated wire (2) (diameter  $d = 0.30$  mm, length  $l = 88.0$  mm) employed as a heated surface is horizontally hanged in pressure vessel (1) and is heated by a direct electric current. The wire diameter of 0.3 mm was chosen from the following reason. Boiling on the heated wire of 0.3 mm is the same as that on an infinite surface. Authors [14] confirmed that the boiling heat transfer on the wire can be predicted by Nishikawa and Fujita [16], Kutateladze [17] and Stephan and Abdelsalam [18].

Wall superheat,  $\Delta T_{\text{sat}}$ , is calculated using a temperature–electric resistance characteristic of the platinum wire given in [19]. The characteristic curve is verified by the previous experiment [12] to be in agreement with author's data within a mean error of 6%. The pressure vessel is immersed in thermostat bath (8), which is isolated from its surroundings. The thermostat liquid is circulated with thermostat with pump (9) around the pressure vessel. Boiling features can be observed and photographed with a camera and a high-speed stroboscope through view window (3).

Heat flux to the heated wire was stepwise increased up to CHF at which the experiment was stopped. The onset of boiling and the CHF were measured within an accuracy of 5% because the heat flux was increased at an increment of 5% in a previous heat flux. In every experiment, the heated wire and the pressure vessel were

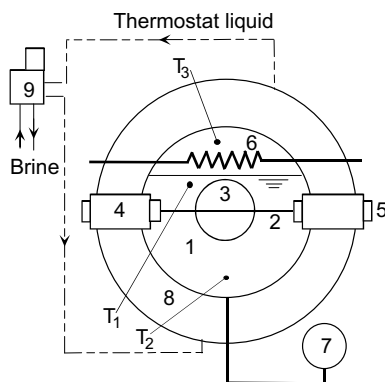


Fig. 2. Schematic diagram of experimental apparatus for heat transfer coefficient measurement. (1) Pressure vessel, (2) heated wire, (3) view window, (4) insulator, (5) electrode, (6) condenser, (7) pressure gauge, (8) thermostat bath, (9) thermostat with pump  $T_1$ ,  $T_2$ ,  $T_3$  thermocouples.

Table 1  
Experimental condition

Test substances	Ethanol/water
Surfactant	Perfluoroalkyl compound (cationic surfactant)
Pressure	0.1 MPa
Mass fraction of ethanol	0, 0.1, 0.3, 0.5, 0.7, 0.9, 1
Mass concentration of surfactant	0–5000 ppm
Surface tension of surfactant	17 mN/m at 25 °C

carefully cleaned with acetone prior to the experiment. Additional details of the experimental apparatus and the procedure as well as an accuracy of the measurement are described in [12], from which uncertainties in the measurement also are less than 0.1 K for temperature and 1% for heat flux.

## 2.3. Experimental condition

Table 1 shows the present experimental condition. The surface-active agent employed is a cationic surfactant and the basis of the surfactant is perfluoroalkyl compound. It is commercial and called Surfion S-121 produced by Seimi Chemical Co. It contains 30% perfluoroalkyl compound, 25% *iso*-propanol and 45% water. The surface tension of the surfactant is 17 mN/m at 25 °C. The surfactant concentration is for the mass of water in the binary mixtures in Table 1. Ethanol, ion-exchanged and distilled water and their mixtures were employed as the test liquid.

## 3. Experimental result of surface tension

### 3.1. Effect of ethanol fraction and temperature on surface tension

Fig. 3 shows the effect of the ethanol fraction and temperature on the surface tension at a pressure of 0.1 MPa. A broken line is an experimental equation for water proposed in [15]. The measurement values of the surface tension for water are smaller than those proposed in [15] by 3–6%. Those for benzene are also smaller than the values in [15] by 3–4%. It is inferred from this result that the measurement values for other different kinds of liquid have also the same accuracy. Solid lines in Fig. 3 were gained by the least-mean square method. Jasper [20] reported that the surface tension for pure liquids linearly varies with a temperature in the range of 40–200 °C, that is, the values of  $a$  and  $b$  can be given for different kinds of pure liquid as

$$\sigma = a + bT. \quad (2)$$

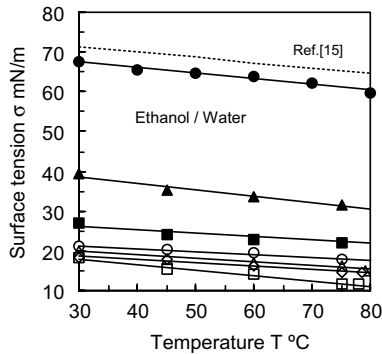


Fig. 3. Effect of temperature and ethanol fraction on surface tension: ●  $C = 0$ , ▲  $C = 0.1$ , ■  $C = 0.3$ , ○  $C = 0.5$ , △  $C = 0.7$ , ◆  $C = 0.9$ , □  $C = 1.0$ .

The surface tension in binary mixtures has the same trend as that in pure liquid. The values of  $a$  and  $b$  for mixtures are indicated in Table 2. It was found from the value of  $b$  in Table 2 that the effect of temperature on the surface tension is smaller for mixtures than for pure liquids. Here, change in the surface tension with change in temperature is only 10% in the range of 30–80 °C.

Fig. 4 shows the effect of the ethanol fraction on the surface tension. The surface tension dramatically decreases with an increase in the ethanol fraction in the range of  $C \leq 0.5$ , whereas the surface tension of the mixtures is almost the same as that of the pure ethanol in  $C \geq 0.5$ .

### 3.2. Effect of surfactant concentration on surface tension

Fig. 5 shows the effect of the surfactant concentration on the surface tension at 0.1 MPa and 30 °C. The change in critical miselization concentration, CMC, with the ethanol fraction is also shown. Arrows, ←, in Fig. 5 express the values of the surface tension at  $C_S = 0$  ppm. The surface tension in the pure liquids and the mixtures decreases with the increase in the surfactant concentration and finally approaches a limiting value at a concentration, which is called the critical miselization concentration as shown in Fig. 5. The influence of the surfactant on the surface tension rapidly disappears with an increase in the ethanol fraction. This fact is attributed to the small difference between the surface tensions of

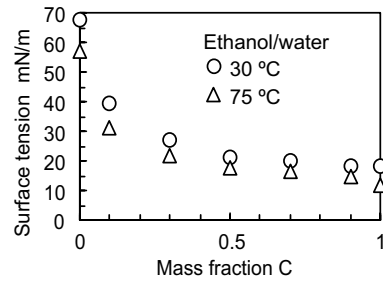


Fig. 4. Effect of alcohol fraction on surface tension.

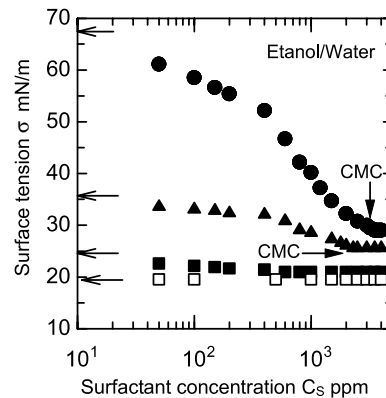


Fig. 5. Effect of surfactant on surface tension at 30 °C: ●  $C = 0$ , ▲  $C = 0.1$ , ■  $C = 0.3$ , □  $C = 1.0$ .

the ethanol (25 mN/m at 25 °C) and the surfactant (17 mN/m at 25 °C).

The point of CMC moves towards lower concentration of the surfactant with the increase in the ethanol fraction and is hardly identical in  $C \geq 0.3$ . The effect of the surfactant concentration almost disappears in  $C \geq 0.3$ . Considering that the effect of the temperature on the surface tension is small (cf. Table 2 or Fig. 3) in the range of 30–80 °C and the  $\sigma$ – $C$  characteristic at 30 °C is similar with that at 80 °C (cf. Fig. 4), one can mention that the present characteristic of  $\sigma$ – $C_S$  at 30 °C is almost the same with the characteristic in the

Table 2  
Values of  $a$  and  $b$  in Eq. (1)

Ethanol/water							
$C$	0	0.1	0.3	0.5	0.7	0.9	1.0
$a$	71.5	43.4	28.5	23.6	22.5	21.2	221
$b$	–0.138	–0.145	–0.083	–0.073	–0.091	–0.086	–0.137

present experimental condition, 78–100 °C, for boiling heat transfer.

#### 4. Experimental result of boiling heat transfer

##### 4.1. Visual observation of boiling aspect

Fig. 6 shows the change of the boiling aspect with the surfactant concentration in water. It is observed that a bubble departure diameter becomes small, and both a bubble departure frequency and a number of nucleation site increases with an increase in the surfactant concentration. Wasekar and Manglik [10] also observed the same phenomena for sodium dodecyl or lauryl sulfate, which is an anionic surfactant. The present boiling aspect is markedly changed up to 1000 ppm with the surfactant concentration. It is clearly found from the observation at  $q = 600 \text{ kW/m}^2$  that the surfactant makes bubble coalescence difficult. Hetsroni et al. [11] also reported the same result with the present observation for a cationic surfactant.

Fig. 7 shows the effect of the surfactant concentration and the ethanol fraction on the boiling aspect of the ethanol/water mixtures. At a heat flux of  $q = 200 \text{ kW/m}^2$ , the number of nucleation sites in mixtures are few at  $C_S = 0 \text{ ppm}$ . This fact is attributed to the decrease in the effective wall superheat for the nucleation because

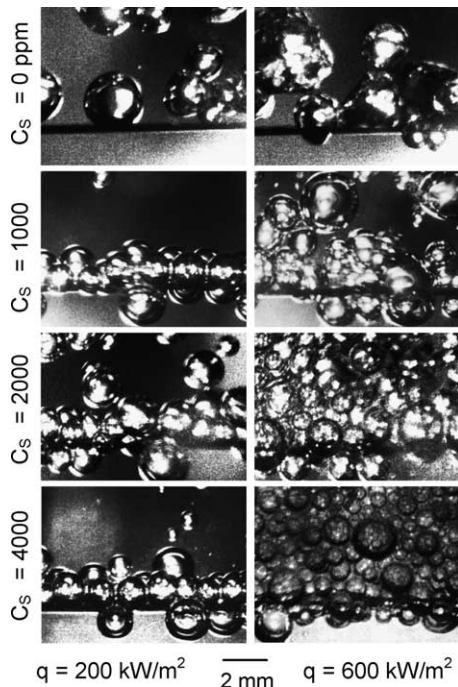


Fig. 6. Change of boiling aspect with surfactant concentration.

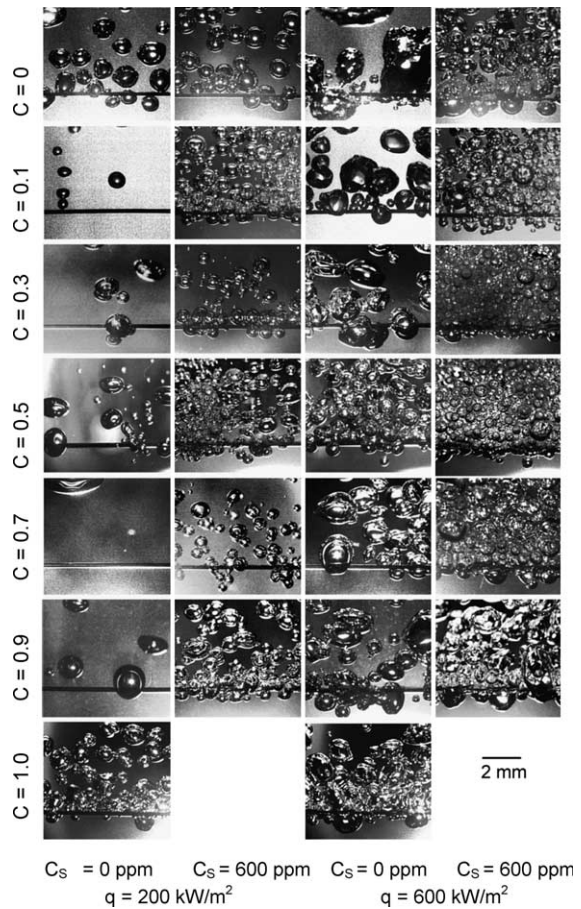


Fig. 7. Change of boiling aspect with surfactant concentration and ethanol fraction in ethanol/water mixtures.

of a rise in the bubble point temperature, since the ethanol fraction is locally decreased by preferential evaporation of the more volatile liquid in the vicinity of the heated surface. A mechanism of the rise in the local bubble point temperature was explained in detail in [21]. The bubbles emit throughout the wire and the departure frequency increases at  $C_S = 600 \text{ ppm}$ . Next, at a heat flux of  $q = 600 \text{ kW/m}^2$ , the bubbles easily coalesce with each other in  $C \leq 0.3$ , while they coalesce with difficulty in  $C \geq 0.5$  at  $C_S = 0 \text{ ppm}$ . The number of nucleation sites increase, the departure diameter becomes smaller and bubbles coalesce with difficulty throughout the ethanol fraction range at  $C_S = 600 \text{ ppm}$ .

##### 4.2. $h$ - $q$ relation

Fig. 8 shows the effect of the surfactant on the  $h$ - $q$  relation in ethanol/water mixtures. The value of  $C$  in Fig. 8 is the mass fraction of ethanol. Arrows,  $\searrow$ , and  $\downarrow$ , show the onset of boiling and a transition to the CHF, respectively. The solid lines at  $C = 0$  and 1 in

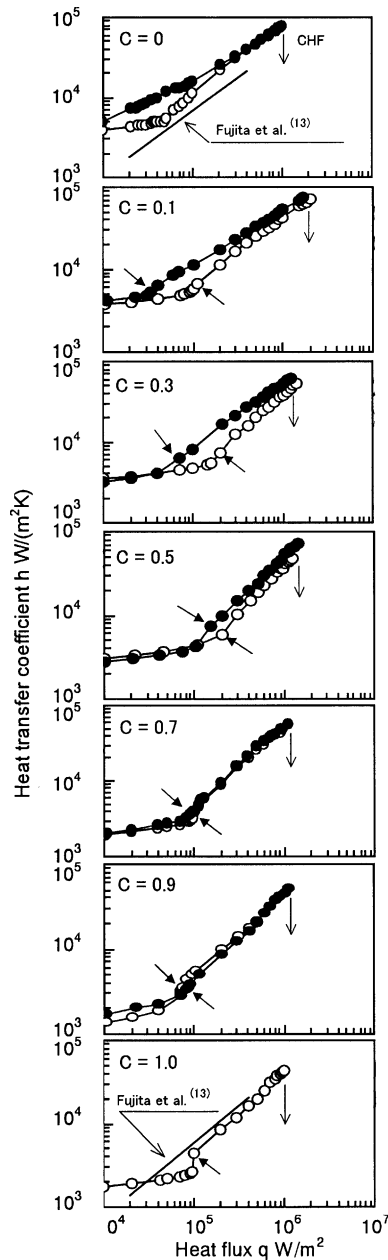


Fig. 8. Effect of mass fraction and surfactant on  $h$ - $q$  relation in ethanol/water mixtures  $\circ C_S = 0$  ppm;  $\bullet C_S = 600$  ppm.

Fig. 8 are heat transfer data on a horizontal heated plate measured by Fujita and Tsutsui [13]. Their data are different from the present data due to a different in shape and the roughness of the heated surface. The effect of the surfactant on the  $h$ - $q$  relation becomes smaller with increases in the ethanol fraction. This is attributed to the action of the surfactant on the surface tension, which becomes weaker with an increase in the fraction, since the surface tension decreases with an increase in the fraction

as shown in Fig. 4. Fig. 8 also shows that the surfactant hardly affects the  $h$ - $q$  relation in  $C \geq 0.7$ , in which the ethanol fraction hardly affects the surface tension in Fig. 4. In other words, the boiling heat transfer in  $C \leq 0.5$  is extremely influenced by the surface tension. The boiling heat transfer in  $C \leq 0.5$  increases remarkably in low heat flux immediately after boiling started by adding the surfactant because the heat flux at the onset of boiling moves toward a lower heat flux. Wasekar and Manglik [10] also reported that boiling starts at a lower heat flux by adding the surfactant. Wang and Hartnett [4] also reported that the boiling heat transfer increases in low heat flux immediately after the boiling started by the surfactant. However, the effect of the surfactant on the boiling heat transfer becomes small with increase in both heat flux and the ethanol fraction. It was found from this result that an inclination of a linear line in the  $h$ - $q$  relation, i.e. a value of  $n$  in the relation of  $h \propto q^n$ , becomes small by adding the surfactant in  $C \leq 0.5$ . Thus, both the decrease of the inclination and the advance of the onset of boiling enhance the boiling heat transfer in low heat flux and weaken the effect of the surfactant in high heat flux.

We surmise a distribution of surfactant molecules with change in heat flux as shown in Fig. 9. It is well known that a surface-active agent is easy to adsorb on solid-liquid and vapor-liquid interfaces. First, in the low heat flux range, an adsorption force to the heated surface is weak because the surfactant molecules are concentrated near the heated surface and consequently the molecule density becomes denser on the surface as shown in Fig. 9(a). Thus, the bubbles depart easily from the heated surface, the departure diameter becomes smaller and the departure period becomes shorter by adding the surfactant as shown in the boiling aspect of Figs. 6 and 7. Furthermore, an agitation effect by the bubbles increases because the nucleation site increases due to the depression of the surface tension. It is considered that the heat transfer coefficient is enhanced because of the above-mentioned reason in low heat flux.

Second, in middle heat flux range, the surfactant molecules per interface area decrease because the nucleation site increases, that leads to an increase in the number of bubbles as shown in Fig. 9(b). The effect of the surfac-

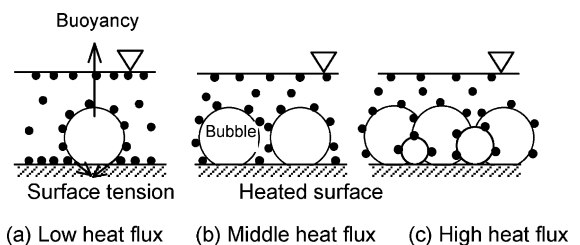


Fig. 9. Model of distribution of surfactant molecule near the heated surface ( $\bullet$ Surfactant molecule).

tant becomes weak because the molecules near the interface decrease.

Last, in high heat flux range, it is considered that the surfactant effect deteriorates extremely because the surfactant molecules cannot reach the heated surface as shown in Fig. 9(c), since the bubbles are emitted vigorously over all the heated surface and the agitation by growth and departure of bubbles is vigorous near the surface.

As a result, the depression of the surface tension by the surfactant plays a dramatic role in the heat transfer enhancement in the low heat flux range, and the effect of the surface tension would be replaced by both the effects of the agitation by the bubbles and a latent heat transport with increase in the heat flux. Then the heat is transformed only by both the effects of the agitation by the bubbles and the latent heat transport in high heat flux range.

4.3. Heat transfer coefficient and onset of boiling

Fig. 10 shows the effect of the ethanol fraction,  $C$ , and the surfactant concentration,  $C_S$ , on the onset of boiling. The heat flux,  $q_{ob}$ , at which boiling starts is lower for the pure liquid than for the mixtures and becomes maximum near  $C = 0.5$ . This reason is the same reason that nucleation becomes more difficult in the mixtures than in pure substances as described in Section 4.1. Boiling starts in lower heat flux by adding the surfactant in  $C \leq 0.5$ . This fact is attributed to the effect that cavities become active for smaller cavity sizes and that the bubbles become easier to depart from the cavities at a lower bubble pressure because of a depression of the surface tension. While, the surfactant does not affect the  $q_{ob}$  in  $C \geq 0.7$ . This fact is attributed to the effect of the surfactant on the depression of the surface tension, which disappears in  $C \geq 0.7$  because of the small difference between the surface tensions of the surfactant and ethanol.

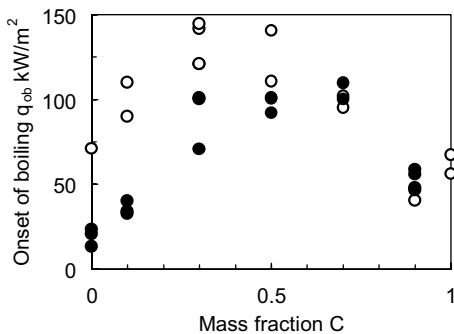


Fig. 10. Effect of ethanol fraction and surfactant concentration on onset of boiling in ethanol/water mixtures  $\circ C_S = 0$  ppm,  $\bullet C_S = 600$  ppm.

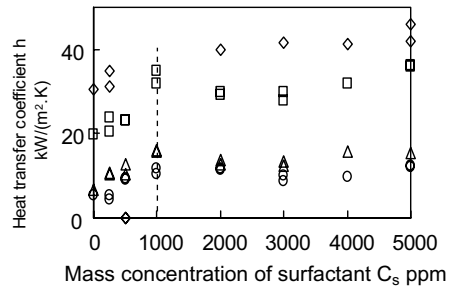


Fig. 11. Effect of surfactant on heat transfer coefficient  $\circ q = 70$  kW/m<sup>2</sup>,  $\triangle q = 100$  kW/m<sup>2</sup>  $\square q = 400$  kW/m<sup>2</sup>,  $\blacklozenge q = 700$  kW/m<sup>2</sup>.

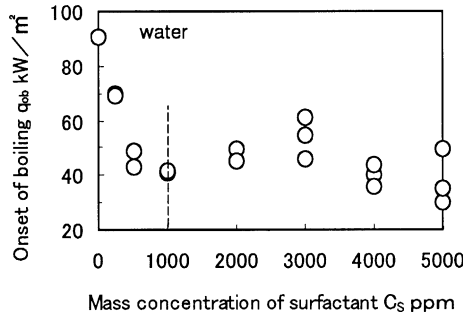


Fig. 12. Effect of surfactant on onset of boiling.

Fig. 11 shows the effect of the surfactant concentration on the boiling heat transfer coefficient in water. The coefficient is dramatically enhanced up to 1000 ppm in all heat fluxes. However, the coefficient hardly changes with the concentration in  $C_S \geq 1000$  ppm. This is attributed to be dramatically affected by the surface tension that is sharply depressed up to 1000 ppm of the surfactant as shown in Fig. 5.

Fig. 12 shows the effect of the surfactant concentration on the onset of boiling in water. The  $q_{ob}$  decreases dramatically up to 1000 ppm. However, the  $q_{ob}$  is hardly affected by a surfactant concentration over 1000 ppm.

Consequently, it is found that the results gained from Figs. 11 and 12 are closely connected with both the depression characteristics of the surface tension as shown in Fig. 5 and the boiling aspect as shown in Fig. 6.

4.4. Critical heat flux

Fig. 13 shows the effect of the ethanol fraction and the surfactant concentration on the CHF. The CHF in the mixtures is larger than those in the pure liquids and is maximum at  $C = 0.3$ . The surfactant does not affect the CHF all over the ethanol fraction. Near the CHF, huge amount of vapor is generated, so that the surfactant may be diffused on the bubble surface and may be absent

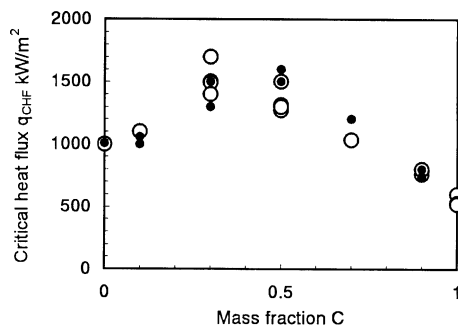


Fig. 13. Effect of mass fraction and surfactant on critical heat flux  $\circ C_S = 0$  ppm,  $\bullet C_S = 600$  ppm.

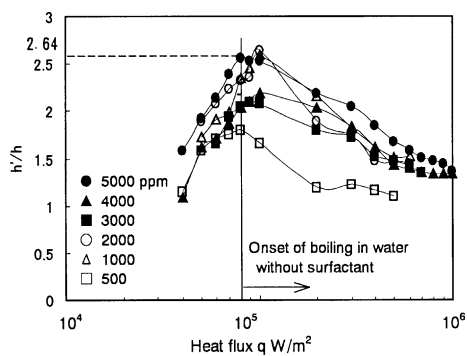


Fig. 14. Enhancement of heat transfer coefficient by surfactant:  $h$ , heat transfer coefficient in water without surfactant;  $h'$ , heat transfer coefficient in water with surfactant.

from the vicinity of the heated wire as shown in Fig. 9(c). As a result, the surfactant would hardly influence the CHF. Remembering that the heat transfer coefficient was hardly influenced by the surfactant near the CHF as shown in Fig. 8, one may consider that the surfactant has little effect on them near the CHF.

#### 4.5. Enhancement of heat transfer coefficient

Fig. 14 shows the enhancement rate,  $h/h'$ , of the heat transfer coefficient by the surfactant. The coefficient increases exceedingly in the vicinity of a low heat flux of  $q = 8 \times 10^4$  W/m<sup>2</sup> and is enhanced 2.64 times. This is attributed to the fact that the boiling starts in a lower heat flux by the surfactant. However, the enhancement rate of the coefficient decreases with an increase in heat flux.

## 5. Conclusions

We have measured the surface tension and the boiling heat transfer in water and the ethanol/water mixtures with the surface-active agent. The obtained results are summarized as follows:

1. The rate of decrease in the surface tension with an increase in the surfactant concentration becomes larger with decreases in the ethanol fraction and the surfactant hardly affects the surface tension in high ethanol fraction range.
2. The boiling heat transfer coefficient is enhanced in a low ethanol fraction range by the surfactant.
3. The coefficient is enhanced in low heat flux range immediately after the onset of boiling by the surfactant and the surfactant effect becomes weak with an increase in heat flux.
4. The surface tension decreases exceedingly, the nucleation site increases exceedingly, the heat flux at the onset of boiling becomes exceedingly low and the heat transfer is exceedingly enhanced, up to a surfactant concentration of 1000 ppm. However, they are hardly influenced by a surfactant concentration over 1000 ppm.

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