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On the mechanism of subcooled flow boiling of binary mixtures

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Abstract—Correlations have been developed for forced convection subcooled boiling of binary mixtures using data on acetone–water, isopropanol–water and *n*-butanol–water systems. Separate correlations for the partial boiling region and for the fully developed boiling region incorporating the effects of degree of subcooling, liquid velocity and binary composition have been proposed.

INTRODUCTION

NUCLEATE boiling is an important mechanism of heat transfer from a surface to a fluid adjacent to it. The liquid may be either subcooled or saturated at the prevailing pressure. In saturated boiling, the vapor generated at the surface drifts into the bulk of the liquid to result in a two-phase system. On the other hand, when the liquid is below its saturation temperature, as it is with subcooled boiling, the vapor bubbles formed at the heating surface condense in the cold liquid in the bulk with no net generation of vapor leading to an advantage in the design of many process equipment.

In subcooled boiling, a higher heat transfer rate coincides with low vapor generation. With sufficient subcooling, a condition may be attained where there is negligible vapor generation. This important effect considerably increases the possibility of using binary mixtures as cooling media in heat exchangers, due to the fact that the evaporation of the more volatile component in the neighborhood of the surface can be avoided with subcooled boiling.

Very high heat fluxes can be realized by combining subcooled boiling with forced convection. At moderate wall superheat, the heat transfer is governed by the combined effects of forced convection and surface boiling, whereas at high wall superheat, the effect of forced convection ceases and fully developed subcooled boiling prevails (Sivagnanam and Varma [1]). The engineering applications of forced convection subcooled boiling include the cooling of rocket motors, nuclear accelerator targets, high power electromagnets, electronic tubes and high temperature probes.

Forced convection subcooled boiling of single component liquids and subcooled pool boiling of binary mixtures have been the topics of several investigations in the past. These have been reviewed elsewhere [2]. In addition there is a lot of information on saturated boiling of binary mixtures [3–5]. However, the literature in respect of forced convection subcooled boiling of binary mixtures is very limited (Table 1). Rammohan *et al.* [6] studied subcooled flow boiling of binary mixtures and reported that the heat transfer coefficient was dependent on the liquid velocity. However, at constant liquid velocity, the heat flux and the heat transfer coefficient increased with a decrease in the degree of subcooling for the same temperature driving force, ΔT_1. Sivagnanam and Varma [1] identified (i) a partial boiling region at low and moderate wall superheat where the heat transfer is governed by the combined effect of forced convection and surface boiling, and (ii) a fully developed boiling region where the effect of forced convection ceases to influence heat transfer. The liquid velocity and the degree of subcooling were reported to have significant effect on the heat flux in the former region, and they cease to influence the heat transfer coefficient or the heat flux in the fully developed boiling region. Sivagnanam *et al.* [7] proposed a model, based on an additive mechanism of heat transfer, for forced convection subcooled boiling of binary mixtures. The contributing modes are (i) the heat transferred as latent heat by the rising bubbles, (ii) the heat transferred through the superheated layer through its periodical removal from the heating surface in the wake of the rising bubbles and (iii) the single phase convective heat transfer from the heating surface, uninfluenced by the rising bubbles. Rammohan *et al.* [6] and Sivagnanam and Varma [1] presented correlations for the boiling heat transfer coefficient, essentially modifying the correlation due to Moles and Shaw [8] given for subcooled boiling of pure liquids, by introducing the heat and mass transfer correction factor to account for the concentration of the more volatile component of the mixture.

Wenzel and Steinhagen [9] reported that the heat transfer coefficient decreased with a corresponding

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NOMENCLATURE

C_p	specific heat [J kg^{-1} K^{-1}]
d	bubble diameter at departure, $(\sigma/g\Delta\rho)^{1/2}$ [m]; diameter of heating element in equations (1) and (2) [m]
D	mass diffusivity [m^2 s^{-1}]
E	voltage [V]
F	convective boiling factor
g	acceleration due to gravity [m s^{-2}]
h	heat transfer coefficient [W m^{-2} K^{-1}]
I	current [A]
k	thermal conductivity [W m^{-1} K^{-1}]
Nu	Nusselt number
Pe	Peclet number
q	heat flux [W m^{-2}]
R	resistance of the wire [Ohms]
S	boiling suppression factor
T	temperature [K]
ΔT_l	temperature driving force, $T_w - T_l$ [K]
ΔT_s	wall superheat, $T_w - T_s$ [K]
ΔT_{sub}	degree of subcooling, $T_s - T_l$ [K]
v	liquid velocity [m s^{-1}]
x	liquid concentration (mole fraction)
y^*	vapour concentration (mole fraction) in equilibrium with liquid
Z	length of heating element [m].

Greek symbols

α	thermal diffusivity [m^2 s^{-1}]
λ	latent heat [J kg^{-1}]
μ	viscosity [Pa s]
ρ	density [kg m^{-3}]
σ	surface tension [N m^{-1}].

Subscripts

b	boiling
eff	effective
l	liquid
ls	saturation temperature of pure more volatile component
nb	non boiling
o	at 0°C
pt	platinum
s	saturation
t	temperature
v	vapor
w	wall
ws	saturation temperature of water.

increase in wall temperature for a subcooling range of 5 to 20 K, beyond which both remained constant. The authors attributed the phenomenon to two opposing effects caused by an increase in subcooling, i.e. the reduction in wall temperature due to convective cooling by the colder liquid, and the increase in wall temperature because of decreasing amount of boiling which acts as a cooling mechanism for the surface. The authors [9] noted that the heat transfer coefficient for fully developed subcooled boiling is independent of liquid velocity at all binary compositions, suggesting that the mechanism of heat transfer is by bubble growth and its departure, rather than by convective heat and mass transfer. The authors reported good agreement of their data with the correlation based on the superposition of convective and boiling heat transfer, following the formulation due to Chen [10] for saturated single component flow boiling.

Sivagnanam and Varma [11], analysing the experimental data (Sivagnanam and Varma [1]), noted that the heat transfer coefficient and the wall temperature attain a constant value for $\Delta T_{sub} \geqslant 25$ K. They observed that the heat transfer coefficient rapidly decreases with increase in high volatile component in the binary and attains a minimum where $|y^* - x|$ is maximum.

EXPERIMENTAL

Experiments were conducted with a platinum wire as heating surface and with water, acetone–water, isopropanol–water and n-butanol–water as fluids (Sivagnanam and Varma [1]). The test loop used is shown in Fig. 1. The heating element was a 0.3 mm diameter and 495 mm long platinum wire held taut in a vertical glass tube 47 mm i.d. and 590 mm long through which the liquid flows upward. Direct current (d.c.) was applied across the heating element. The experiments were conducted at atmospheric pressure and in the increasing mode of heat flux for different liquid velocities, degrees of subcooling and compositions of the binary mixtures.

The test loop was dried with hot air at the beginning of each run and the liquid mixture introduced into the test loop. The liquid was circulated and the pre-heaters adjusted to obtain the desired liquid temperature. About 45 min were needed to attain steady state before the d.c. supply could be applied across the heating element in the test section to commence the experiments.

The heat flux density q of the wire was calculated from the measured values of voltage and current using

$$q = \frac{EI}{\pi d Z}. \tag{1}$$

The temperature of the wire was calculated from the equation of Hahne and Feurstein [12]

$$T_w = 3383.33 - 1304.1\left\{7.7308 - \frac{R_t}{R_0}\right\}^{0.5} - \frac{qd}{8k_{pt}} \tag{2}$$

Table 1

Rammohan *et al.* [6]:

System	Heating surface	v (m s^{-1})	$q \times 10^{-5}$ (W m^{-2})	ΔT_{sub} (K)
glycerol–water isopropanol–water	platinum	0.16–0.54	7.9–24	40–64

Correlation:

$$\frac{h_{\mathrm{eff}}}{h_{\mathrm{nb}}} = 34.8(1-0.73x^{0.58})\left(\frac{C_{\mathrm{p}}\mu}{K}\right)_{\mathrm{l}}^{0.56}\left(\frac{q}{\lambda\rho_{\mathrm{v}}v}\right)_{\mathrm{s}}^{0.67}\left(\frac{\lambda\rho_{\mathrm{v}}}{C_{\mathrm{p}}\Delta T_{\mathrm{sub}}\rho_{\mathrm{l}}}\right)_{\mathrm{s}}^{0.6}$$

Sivagnanam and Varma [1]:

System	Heating surface	v (m s^{-1})	$q \times 10^{-5}$ (W m^{-2})	ΔT_{sub} (K)
acetone (5–25%)–water isopropanol (5–20%)–water butanol (2–8%)–water	platinum	0.16–1	1.9–28	10–40

Correlation:

$$\frac{h_{\mathrm{eff}}}{h_{\mathrm{nb}}} = 55[1+|y^{*}-x|(\alpha/D)^{0.5}]^{-0.7}\left[\frac{T_{\mathrm{ws}}}{T_{\mathrm{ls}}}\right]^{4}\left[\frac{C_{\mathrm{p}}\mu}{K}\right]_{\mathrm{l}}^{0.5}\left[\frac{\lambda\rho_{\mathrm{v}}}{C_{\mathrm{p}}\Delta T_{\mathrm{sub}}}\right]_{\mathrm{s}}^{0.76}\left[\frac{q}{\lambda\rho_{\mathrm{v}}v}\right]_{\mathrm{s}}^{0.84}$$

Wenzel and Steinhagen [9]:

System	Heating surface	v (m s^{-1})	$q \times 10^{-5}$ (W m^{-2})	ΔT_{sub} (K)
isopropanol (0–67.5%)–water	S.S. annular section	0.1–0.9	0.1–4	5–25

Correlation:

$h = h_{\mathrm{nb}}F + h_{\mathrm{b}}S$; F = convective boiling factor

S = nucleate boiling suppression factor

$$\frac{h_{\mathrm{b}}}{h_{\mathrm{id}}} = 1\left\{1+\frac{h_{\mathrm{id}}}{q}(T_{\mathrm{S1}}-T_{\mathrm{S2}})(y^{*}-x)[1-\exp(-q/(1\times10^{-4}\rho_{\mathrm{l}}\lambda))]\right\}$$

where $\dfrac{1}{h_{\mathrm{id}}} = \dfrac{x_1}{h_1} + \dfrac{x_2}{h_2}$

and subscripts 1 and 2 refer to water and component 2, respectively.

where T_{w} is in °C, R_{t} is the resistance of the wire at any temperature T °C, R_0 is the resistance at 0°C and k_{pt} is the thermal conductivity of platinum in W m^{-2} K^{-1}. Equation (2) is based on the temperature dependence of the electrical resistance of pure platinum. The driving force ΔT_{l} $(=T_{\mathrm{w}}-T_{\mathrm{l}})$ can be obtained from the mean T_{l} measured and the heat transfer coefficient defined as

$$h = q/(T_{\mathrm{w}}-T_{\mathrm{l}}). \tag{3}$$

The common source of error in finding the heat transfer coefficient h, is in the estimation of the wall temperature T_{w} using equation (2). Thus for a given heat flux, the error in the determination of h will be high when the liquid temperature T_{l} is close to the saturation temperature T_{s}. In the present study, a minimum value of $T_{\mathrm{s}}-T_{\mathrm{l}}$ of 10°C was maintained. The measured minimum heat flux was 0.2×10^{6} W m^{-2}. These values are beyond the range specified for possible inaccuracies in the determination of h [8]. The range of ΔT_{s} $(=T_{\mathrm{w}}-T_{\mathrm{s}})$ in the present study was maintained above 20°C, limiting the error in the estimation of the heat transfer coefficient using the above equations to less than 5% (Sivagnanam and Varma [1]).

RESULTS AND DISCUSSION

Effect of subcooling

Figure 2 is a typical plot of heat flux shown against wall superheat, ΔT_{s} $(=T_{\mathrm{w}}-T_{\mathrm{s}})$ with the degree of subcooling as parameter for the binary mixtures of the study. T_{s}, the saturation temperature of the binary mixture is defined as the bubble point of the mixture at the given liquid composition and is obtained from Vapor Liquid Equilibria. It is noticed that at low x, the degree of subcooling influences the heat flux, for a given ΔT_{s}, in the partial boiling region as well as in

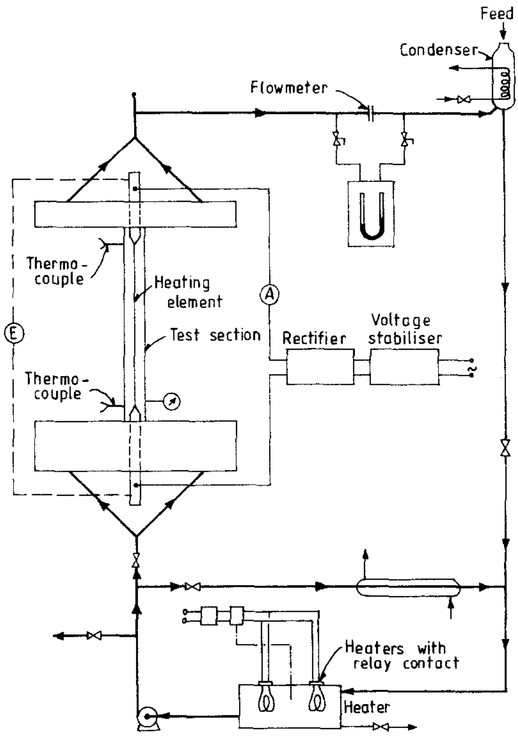

FIG. 1. Experimental test loop.

fully developed boiling region, namely for a given heat flux, a higher ΔT_s is necessary as ΔT_{sub} is increased. However, at high x, the ΔT_{sub} appears to have no influence on q in the fully developed boiling region; a smaller wall superheat is, however, required in the partial boiling region for a given heat flux, with an increase in the degree of subcooling. These observations with respect to the binary mixtures are in qualitative agreement with the observation due to Hasan *et al.* [13] reported for single component forced convection subcooled boiling that the wall superheat remains lower at high subcoolings in the partial boiling region followed by an asymptotic approach to fully developed boiling where the subcooling influence becomes minimal.

The variation in wall heat transfer coefficient with the degree of subcooling for water is shown in Fig. 3 and compared with typical data reported in literature (Hasan *et al.* [13]), confirming the trend and accuracy of the present data.

Figure 4 is a typical plot of the heat transfer coefficient shown against the driving force, ΔT_l $(=T_w-T_l)$ for the different subcoolings and compositions of binary mixtures. Since ΔT_l is smaller for lower subcoolings for any given heat flux in the partial as well as fully developed boiling regions, the heat transfer coefficient is higher at lower subcoolings. This agrees with the observations due to Sivagnanam and Varma [11] and Wenzel and Steinhagen [9] that the heat transfer coefficient rapidly decreases with increase in ΔT_{sub} till $\Delta T_{sub} \leqslant 25$ K, beyond which it is independent of the degree of subcooling (Fig. 5).

Effect of binary composition

Figure 6 shows the variation in the heat transfer coefficient with the degree of subcooling for different compositions of binary mixtures. It is noticed from Figs. 6(a) and (b) that at low x, the heat transfer coefficient rapidly decreases with increase in ΔT_{sub} till $\Delta T_{sub} < 25$ K, beyond which it approaches an asymptotic value. An increase in x lowers the heat transfer coefficient for a given ΔT_{sub} for acetone–water and isopropanol–water mixtures. Further, the variation in the heat transfer coefficient with increase in x depends on the magnitude of x itself; it is rapid at low values of x as compared to its decrease at high values of x. With butanol–water mixture, however, the heat transfer coefficient increases with increase in butanol concentration in the binary mixture (Fig. 6(c)). This behavior with butanol–water mixture may be explained on the basis that maximum in $|y^*-x|$ occurs for butanol–water mixture at 1.5 wt.% butanol, while for acetone–water and isopropanol–water mixtures the maximum in $|y^*-x|$ is at 21 and 27 wt.%, respectively, in acetone and isopropanol. Thus, for a given subcooling, as the concentration of the second component in water increases, the heat transfer coefficient decreases till $|y^*-x|$ is maximum, beyond which the coefficient increases with an increase in x in the binary mixture, namely h is minimum for a given ΔT_{sub} when $|y^*-x|$ is maximum for the binary mixture.

The decrease in the heat transfer coefficient using the binary mixture may be postulated as follows. Firstly, the addition of even a small quantity of organic liquid to water decreases the contact angle. The contact angle determines the nucleating ability of a cavity. A small contact angle gives vapor trapping in cavities, thus requiring a larger temperature difference to initiate nucleate boiling.

On boiling, a binary liquid mixture gives vapor richer in the more volatile component. The liquid, adjacent to the heating surface where bubbles are generated at the specific nucleation sites, is thus depleted of high volatile component, as compared to the composition of the bulk liquid. This sets in concentration gradient from bulk to the wall with the high volatile component diffusing from bulk to the liquid layer adjacent to the heating surface. The generated bubble, being richer in the high volatile component, sheds the low volatile component through condensation into the bulk liquid. Thus, during its growth, the bubble continues to become richer in the high volatile component. A diffusion cycle thus sets in with subcooled boiling of binary mixtures, whereby the more volatile component diffuses from bulk to the layer adjacent to the surface, gets more of it vaporized to make the bubble richer in the more volatile component, while simultaneously depriving the bubble of its low volatile component through con-

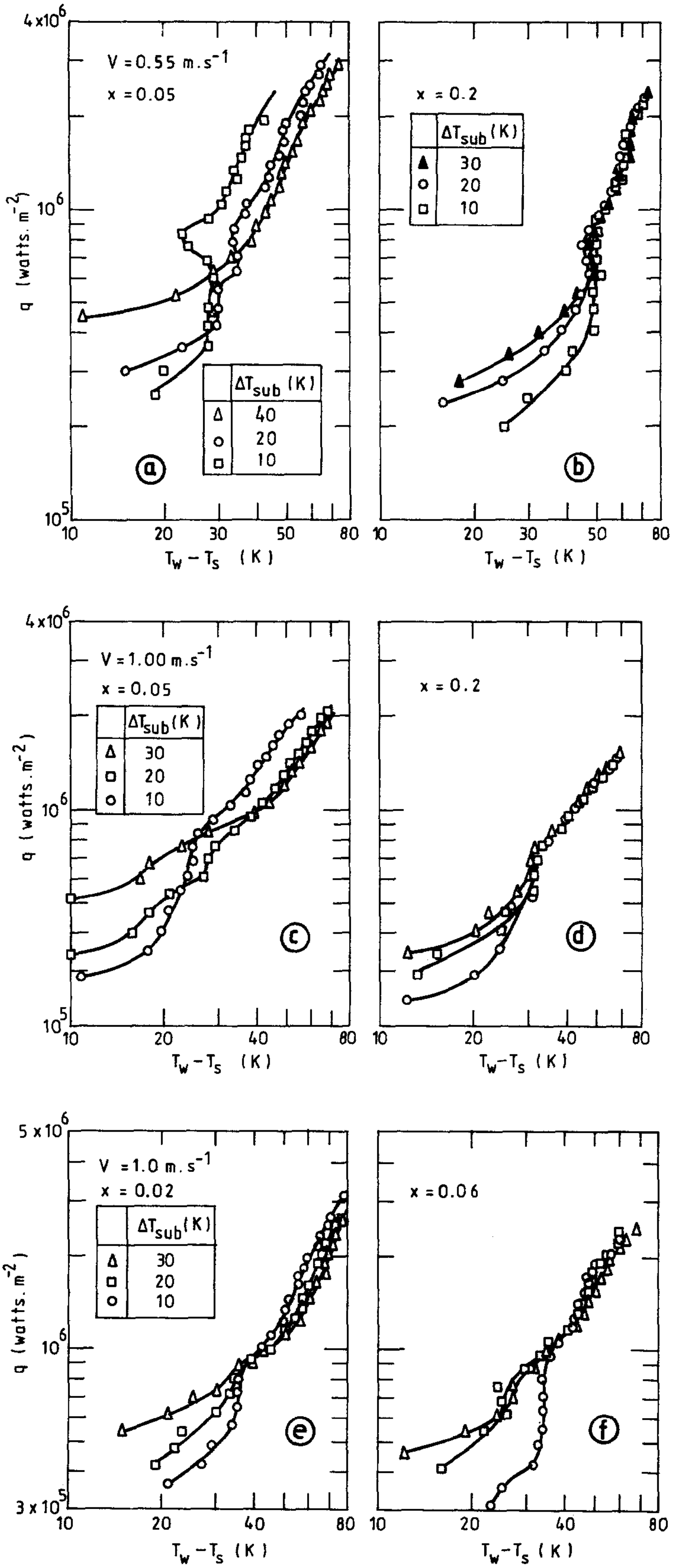

FIG. 2. Heat flux against wall superheat ΔT_s; (a) and (b): acetone–water system; (c) and (d): isopropanol–water system; (e) and (f): *n*-butanol–water system.

densation into the bulk liquid (Fig. 7). The process of enrichment in a more volatile component during the bubble growth is similar to that in distillation operation; the difference, however, is that in distillation the vapor bubble is continuously enriched with the more volatile component during its ascent through the liquid column, while with subcooled boiling of binary mixture, the enrichment is limited to the bubble growth period as the bubble condenses later in the bulk liquid releasing its contents into the bulk.

When $|y^* - x|$ is maximum, the concentration difference between the bulk and the liquid layer adjacent to the wall is maximum to make the system essentially a mass transfer controlling process. This

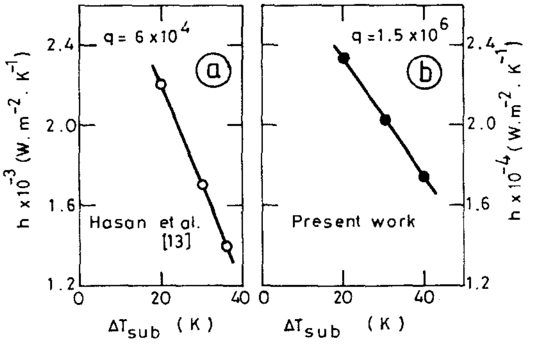

FIG. 3. Heat transfer coefficient against degree of subcooling ΔT_{sub}.

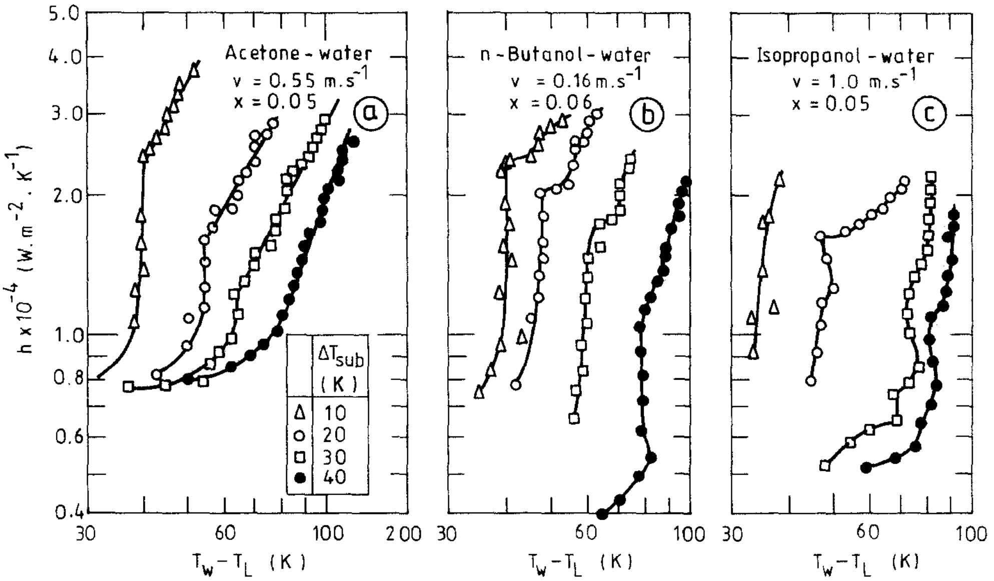

FIG. 4. Heat transfer coefficient against driving force ΔT_l; (a): acetone–water; (b): *n*-butanol–water; (c): isopropanol–water.

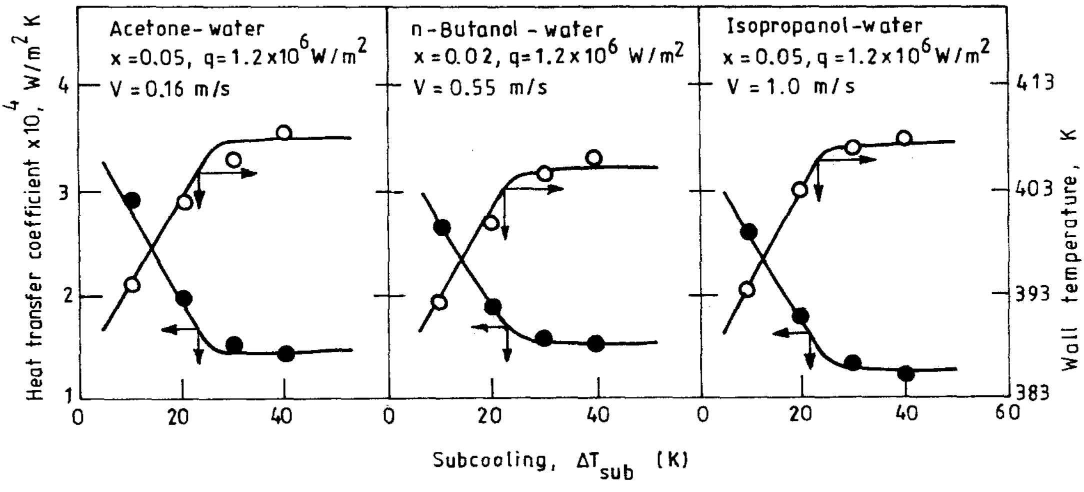

FIG. 5. Influence of subcooling on heat transfer coefficients and wall temperature.

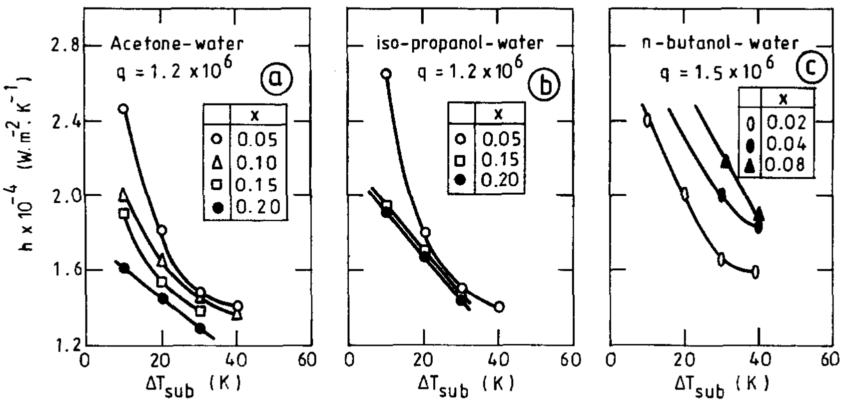

FIG. 6. Variation of heat transfer coefficient with degree of subcooling for different binary compositions; (a): acetone–water; (b): isopropanol–water; (c): *n*-butanol–water.

is further accentuated because of the fact that the latent heat of vaporization of organic liquids is in general considerably smaller than that of water, to make the process less thermally dependent.

As the bubble becomes richer in the more volatile component, the bubble diameter (at departure) decreases, the frequency of bubble generation approaches maximum value, and the bubble growth rate is minimum (Tolubinsky and Ostrovsky [14]). The bubble rise velocity from the thermal boundary layer, adjacent to the heating surface, is small due to its smaller size. Thus, with the addition of organic liquid to water under conditions of subcooled boiling, the slow bubble growth rate is coupled with slow rise velocity of bubbles to give higher concentration of bubbles in the liquid adjacent to the heating surface and thus decrease the heat transfer coefficient to a minimum when $|y^*-x|$ is maximum.

The phenomenon of boiling of binary mixture is thus similar and complementary to that of degree of subcooling. An increase in subcooling too, decreases the bubble diameter (at departure) and its growth time. The bubble rise velocity decreases due to its smaller size so that the bubbles are held for a longer time near the heating surface to give a lower heat transfer coefficient. In fact, an increase in fluid velocity at a given heat flux and subcooling further lowers the bubble size and its growth rate.

The heat transfer coefficient, however, cannot decrease beyond a value, which may be assumed to correspond to the maximum resistance offered by a close-packed array of vapor bubbles in the liquid adjacent to the heating surface. With ΔT_{sub} small, x influences the heat transfer coefficient provided its variation leads to a maximum in $|y^*-x|$. Likewise, with $|y^*-x|$ being small, ΔT_{sub} significantly influences the heat transfer coefficient till $\Delta T_{sub} \leqslant 25$ K.

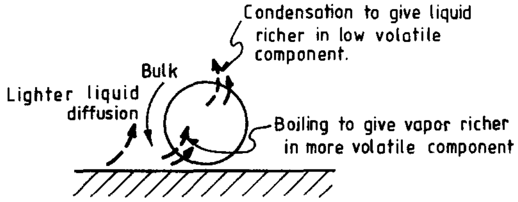

FIG. 7. Mechanism of bubble growth during boiling of binary mixtures.

CORRELATION OF THE DATA

The experimental data of Sivagnanam and Varma [1] as well as those reported in the literature are satisfactorily correlated taking into consideration the effects of subcooling, composition of the binary mix-

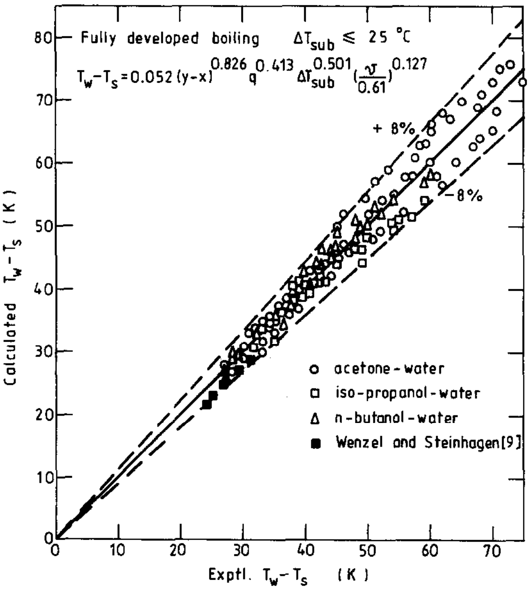

FIG. 8. Performance of correlation for fully developed boiling region; $\Delta T_{sub} \leqslant 25$ K.

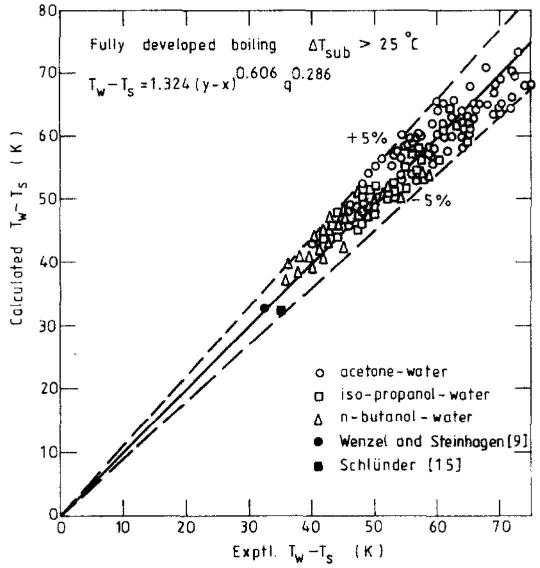

FIG. 9. Performance of correlation for fully developed boiling region; $\Delta T_{sub} > 25$ K.

ture and the liquid velocity on the heat transfer coefficient. The correlations are given separately for the partial boiling region and for the fully developed boiling region.

Fully developed boiling region:

$$T_w - T_s = 0.052|y^* - x|^{0.826} q^{0.413} \Delta T_{sub}^{0.5} (v/0.61)^{0.127}, \quad \Delta T_{sub} \leqslant 25 \text{ K} \qquad (4)$$

$$T_w - T_s = 1.324|y^* - x|^{0.606} q^{0.286}, \quad \Delta T_{sub} > 25 \text{ K}. \qquad (5)$$

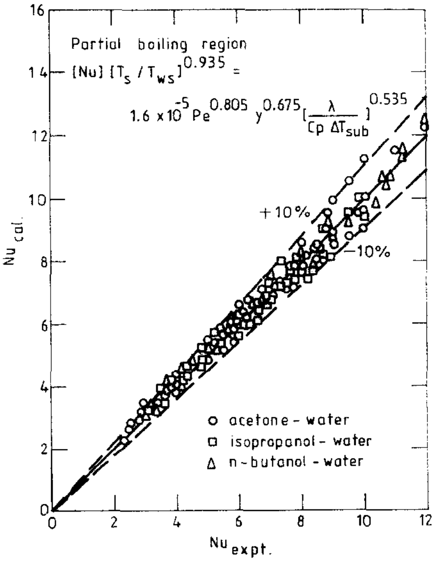

FIG. 10. Performance of correlation for partial boiling region.

Partial boiling region:

$$Nu\left(\frac{T_s}{T_{ws}}\right)^{0.935} = 1.6 \times 10^{-5}\, Pe^{0.805} y^{0.675} \times \left(\frac{\lambda}{C_p \Delta T_{sub}}\right)^{0.535} \qquad (6)$$

where $Nu \equiv (hd/k)$; $Pe \equiv (qd/\alpha\rho_v\lambda)$ and all physical properties are evaluated at T_s.

Figures 8–10 compare the experimental data and the predicted values using equations (4)–(6). It is seen from the correlations that the liquid velocity and the degree of subcooling cease to influence the heat transfer coefficient in fully developed boiling region for $\Delta T_{sub} > 25$ K. Both variables, however, influence the coefficient when $\Delta T_{sub} \leqslant 25$ K. The binary composition, on the other hand, affects the heat transfer coefficient in the fully developed boiling region. These represent the significant observations to be noted in the design of equipment. Equation (6) shows that the heat transfer coefficient is influenced in the partial boiling region by the liquid velocity in addition to the binary composition and the degree of subcooling. The equations are valid for liquid compositions other than azeotropic composition (i.e. constant boiling mixture).

CONCLUSIONS

From the above discussion the following conclusions can be drawn. The degree of subcooling ΔT_{sub} and the equilibrium composition in the vapor and liquid of the binary mixture $|y^* - x|$ influence the heat transfer coefficient, h. h is a minimum when $|y^* - x|$ is a maximum or when ΔT_{sub} is approximately 25 K. It therefore follows that there is no advantage in using subcooling levels greater than about 25 K. Binary mixtures are beneficial compared to single component systems due to the $|y^* - x|$ effect with respect to permissible heat flux limits. The choice of a binary has to be made judiciously so that $|y^* - x|$ is a maximum and at low concentrations to take maximum advantage of this effect. For example, a butanol–water system has the maximum $|y^* - x|$ at 1.5 wt.% butanol while for acetone–water system it is 21 wt.% acetone and for isopropanol–water system it is 27 wt.% isopropanol. Velocity or degree of subcooling have no influence on h when they are above certain values.

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