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Heat transfer in nucleate pool boiling of binary mixtures

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Abstract-Heat transfer coefficients for nucleate pool boiling of binary mixtures were measured on a circular copper plate facing upwards. Tested mixtures were methanol/water, ethanol/water, methanol/ethanol, ethanol/n-butanol, and methanol/benzene, each in the saturated state at atmospheric pressure. Special stress was laid on elucidating the dependence of heat transfer reduction on mixture composition, physical properties, phase equilibrium diagram, and heat flux. Experimental data were compared with available correlating methods developed for binary mixtures by Stephan and Körner, and by others. The comparison showed that among the compared correlations, Stephan and KGmer's correlation and Thome's correlation give fairly good results while their accuracy varies considerably with mixtures and heat flux levels. A modification of Thome's correlation so as to include the effect of heat flux succeeded in correlating the present data within $\pm 20\%$ accuracy.

INTRODUCTION

BOILING heat transfer has been intensively investigated and our knowledge of the physical processes involved in nucleate boiling is broad. But it is not yet possible to predict heat transfer coefficients with the accuracy necessary for reliable design of evaporators. In particular, this holds true for the boiling of mixtures.

A significant feature of mixtures is the variable temperature for phase change at constant pressure. This phenomenon can be utilized to reduce the thermodynamic irreversibility in counter-flow heat exchangers, resulting in an increase of heat exchanger efficiency. Thus mixtures are very promising working fluids for heat pumps and for power plants operated between heat sources of relatively small temperature difference. This advantage expected from a thermodynamic point of view is more or less offset by the lower heat transfer coefficients during phase change compared with pure fluids of the same physical properties as the mixtures. To make the use of mixtures popular, more accurate information about boiling heat transfer is required.

Many previous experimental investigations [1-10] have revealed a reduction of heat transfer coefficients in nucleate boiling of mixtures, compared with those for a single component substance of the same physical properties as the mixture, or compared with the linearly interpolated values between the two pure components comprising the mixture. Some physical explanations for the reduction were suggested [l, 4, 5, 6, $11-18$] and reviewed $[19-21]$. The reduction is attributed to any one of such mixture effects: a local temperature rise adjacent to the heating surface caused by preferential evaporation of the volatile component, mass transfer resistance for the volatile component to move to a bubble interface, much more energy required in forming a bubble nucleus for mixtures and a decrease in nucleation sites, nonlinear

variations of mixture properties with composition, and so forth. But there is no theoretical method of predicting the heat transfer coefficients with satisfactory accuracy. Hence, empirical or semi-empirical correlations [2,3,5,6, 12, 15, 17, 18,22-251 have been proposed to correlate the heat transfer coefficients. Most correlations represent quite well the experimental data they were developed from, but large discrepancy occurs when they are applied to other data. This fact results partly from a lack of sophisticated heat transfer models which include properly the mixture effect. What is more, lack of data for a variety of mixtures prevents verification of the proposed models and in addition, there usually exist large deviations between the heat transfer data for the same mixtures from different sources.

The object of this experimental work is to clarify the pool boiling heat transfer of five binary mixtures at atmospheric pressure. These five combinations were selected so as to represent different types of mixtures : aqueous and non-aqueous mixtures, azeotropeforming and non-azeotropic mixtures, mixtures with phase diagrams widely spreading and closely adjacent, and mixtures showing non-linear and approximately linear variations of physical properties with composition. The measurements cover the whole concentration range including pure components and a wide range of heat fluxes. Typical correlations of different structures on the mixture effects are applied to correlate the measured heat transfer coefficients. A modification is proposed to improve the accuracy of correlation.

EXPERIMENTAL APPARATUS AND PROCEDURE

Figure 1 shows the schematic of the experimental apparatus which consists of a boiling vessel and a temperature-controlled air bath. The boiling vessel is

NOMENCLATURE

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-
- constant defined in equation (1) ΔT
mass diffusion coefficient ΔT_{bp}
-
- h_{fg} latent heat of evaporation, defined as the minus bubble point ten
enthalpy difference between vapor and constant mole fraction enthalpy difference between vapor and
-
- parameter defined in equation (5) λ_L thermal conductive exponent defined in equation (1) μ_L viscosity of liquid
- n exponent defined in equation (1) μ_L
-
-
- Prandtl number
- q heat flux
 T temperat
-
- T_s saturation temperature of pure 1 more volatile component
component 2 less volatile component
- X mole fraction in the liquid phase az azeotrope
 Y mole fraction in the vapor phase. $\begin{array}{ccc} c & c \end{array}$ calculation
- Y mole fraction in the vapor phase. **C**

 α heat transfer coefficient α vapor.

- *Ao* parameter defined in equation (2) α_{id} ideal heat transfer coefficient defined *Bo* parameter defined in equation (4) by equation (7)
- C_{PL} specific heat of liquid
 C constant defined in equation (1) ΔT wall superheat
	-
- *D* mass diffusion coefficient ΔT_{bp} boiling range defined as the dew point latent heat of evaporation, defined as the minus bubble point temperatures at
	- liquid at equilibrium ΔT_{id} ideal wall superheat defined by
modification factor equation (6)
- K_{md} modification factor equation (5) equation (6)
 K_{ST} parameter defined in equation (5) λ_L thermal conductivity of liquid
	-
- *P* pressure ρ_L liquid density
	-
- P_c critical pressure P_v vapor density
 P_r Prandtl number σ surface tension.

The Experimental Experimental Subscripts

-
- less volatile component
-
-
- id ideal
- m measurement
-
-

of 104 mm and a height of 395 mm. Glass windows mm below the liquid surface. Heat flux is supplied by are open on opposite sides for visual observations of Joule heating of Nichrome heaters inserted in slits at are open on opposite sides for visual observations of the boiling phenomena. The boiling surface shown in cross section in Fig. 2, is set at the bottom of the vessel. Boiling takes place on the upper end of a copper

6 Leads for power supply

FIG. 1. Experimental apparatus. The state of FIG. 2. Test surface.

-
- Greek symbols and a liquid liquid liquid liquid
	-
- a hollow tube of stainless steel with an inner diameter cylinder, 40 mm in diameter, installed at a depth 100

the flared base of the cylinder. A circular fin, 0.2 mm thick, completely forms the base of the boiling surface and is a one unit construction with the cylindrical portion. This is to minimize the conductive heat losses in the radial direction and to avoid the preferential boiling at low heat fluxes, otherwise inevitable on the edge of the heating surface. Along the vertical axis of the heated cylinder, three thermocouples were embedded at different depths from the boiling surface, as shown in Fig. 2, to determine the boiling surface temperature and heat flux assuming one dimensional heat conduction.

The boiling surface was polished by a No. O/4 emery paper, resulting in a surface roughness of 0.125 μ m on average. To maintain the surface quality, this treatment was repeated before each series of measurements. After the polished boiling surface is installed in the boiling vessel, there follows the successive procedure of degassing, pouring the test mixture up to a 100 mm level above the boiling surface, and heating the mixture to the saturation temperature by auxiliary heaters. Thereafter, the auxiliary heater is kept at constant output, the boiling surface is powered on and the boiling experiment starts. The vapor formed is condensed in the water-cooled condenser and returned to the vessel, thus maintaining a constant level of mixture in the vessel and a constant composition of the mixture during the measurement. The saturation state is always confirmed by monitoring the vapor pressure, and the temperatures of vapor and liquid measured at five vertical points within the boiling vessel.

Tested mixtures are methanol/water, ethanol/ water, methanol/ethanol, ethanol/n-butanol, and methanol/benzene. Twice distilled water and other organic components of super-high reagent grade were used to make up the liquid mixture on a weight basis to correspond to the mole fraction desired. The resulting mole fraction was accurate to within ± 0.01 . Experimental pressure is atmospheric. After boiling at the highest heat flux more than one hour to stabilize the heating surface temperature, the heat flux was lowered stepwise. Thus heat transfer measurement was performed in the direction of decreasing the heat flux to avoid boiling hysteresis. This procedure provided reproducible boiling curves. The surface temperature and heat flux are determined from the measured temperatures in the heated cylinder by assuming one dimensional conduction. Table 1 indicates the mixture composition and heat flux covered in the measurements. Table 2 shows the physical properties of the pure components under the saturated conditions at atmospheric pressure taken from ref. [26]. Figure 3 shows the phase equilibrium diagrams determined by applying the Wilson equation [27] to the phase equilibrium data [28] for the mixtures used in this experiment. The discrepancy of the measured vapor and liquid temperatures in the boiling vessel with the phase equilibrium diagrams shown in Fig. 3 was less than 0.5 K at worst.

FIG. 3. Phase equilibrium diagrams: (a) methanol/water; (b) ethanol/water; (c) methanol/ethanol; **(d)** ethanol/ n-butanol ; (e) methanol/benzene.

EXPERIMENTAL RESULTS AND DISCUSSION

Presentation of data

A total of 765 data points were obtained for five mixtures, methanol/water, ethanol/water, methanol/ ethanol, ethanol/n-butanol, and methanol/benzene, and their pure components. Owing to limited

Table 1. Experimental conditions : MVC : more volatile component : LVC : less volatile component: AZ : azeotrope

Mixture MVC/LVC	Mole fraction of MVC X,	Heat flux q (kW m ⁻²)
methanol/ water	0, 0.10, 0.20, 0.30, 0.40, 0.60, 0.80, 0.90, 1.0	$5.0 - 610$
ethanol/ water	0, 0.07, 0.17, 0.20, 0.31, 0.40, 0.55, 0.60, 0.80, 0.88(AZ), 1.0	$4.5 - 610$
methanol/ ethanol	0, 0.20, 0.40, 0.60, 0.80, 1.0	$5.0 - 510$
ethanol/ n -butanol	0, 0.20, 0.40, 0.60, 0.80, 0.90, 1.0	$4.7 - 500$
methanol/ benzene	0, 0.10, 0.20, 0.30, 0.40, 0.50, $0.60(AZ)$, 0.70 , 0.80 , 0.90 , 1.0	$3.9 - 510$

space for giving a graphic representation of all the data, the boiling curves are presented in Table 3 in the form of the following equation describing the heat transfer coefficient, α , as a function of heat flux, q :

$$
\alpha = Cq^n \tag{1}
$$

where constant, C , and exponent, n , were determined by fitting equation (1) to the data points in the fully developed nucleate boiling region. Thus equation (1) underestimates one to three data points from the lowest heat flux.

The values of exponent n in Table 3 are a little high compared to the ordinarily obtained values of $0.6-0.8$. This may be due to the fact that the present heating surface is very smooth and provides a very narrow range of cavity size distribution.

Heat tramfer coefJicient jbr single components

Measured nucleate boiling data for five single components of water, methanol, ethanol, n-butanol, and benzene, were compared with the predictions from the correlations by Kutateladze [29], Nishikawa and Fujita [30], and Stephan and Abdelsalam [3l]. For water these three correlations gave satisfactory predictions to similar degrees but for the other organic fluids the former two correlations underestimated the data by more than 20%. Among these three correlations the Stephan and Abdelsalam correlation can adequately predict the pool boiling heat transfer coefficients of the five pure components used in this investigation. Figure 4 shows a comparison of the present measured heat transfer coefficients with the prediction of the Stephan and Abdelsalam correlation as the ratio of both coefficients. As the exponent *n* in equation (1) is 0.67 in the Stephan and Abdelsalam correlation and less than the present results ranging from 0.82 to 0.96, the ratio of the measured to calculated heat transfer coefficients increases with heat flux.

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Variation of heat transfer coefficient of mixtures with *composition*

Variations of heat transfer coefficient with mole fraction of the more volatile component in mixtures are shown for several heat fluxes in Fig. 5 along with the variations of ΔT_{bp} and $|Y_1 - X_1|$ for reference. Data points are not the measured ones but the ones read from fitting curves to the measured set of data. It is clear that the heat transfer coefficient of mixtures is reduced below the interpolated values between their pure components, or between one of their pure components and the azeotrope in case of azeotrope-forming mixtures. A distinct minimum in heat transfer coefficient is observed at an intermediate concentration. As the heat flux increases the reduction becomes larger and the minima are more pronounced at higher heat fluxes. Detailed comparison between variations of heat transfer coefficient and ΔT_{bp} and $|Y_1 - X_1|$ with composition indicates that the minima of heat transfer coefficient do not necessarily occur at the mole fractions at which ΔT_{bp} or $|Y_1 - X_1|$ is a maximum. These facts may be more clearly seen when the mixture heat transfer coefficients are expressed as the ratio to the ideal ones, as defined and illustrated in Fig. 8 in the later section.

Table 3. Data presentation by an equation $\alpha = Cq^n$, where α in W m⁻² K⁻¹ and q in W m⁻²:

 X_1 : mole fraction of the more volatile component;

NOD: number of data points;

A *VD :* average deviation in % ;

 ROH : range of heat flux in kW m⁻²

(b) ethanol/water

Composition dependency predicted from single component correlations

Physical and transport properties have a large influence on boiling heat transfer. Sternling and Tichacek [l] and Stephan and Preusser [13] attributed the reduction in boiling heat transfer coefficient of mixtures to a significant change in the mixture physical properties with shifts in composition. Figure 6 illustrates two typical examples of the concentration dependency of some properties important in boiling heat transfer. The properties are expressed in a normalized form by division with the values of the less volatile component. For the calculation of these properties of the mixtures, the mixing rules recommended by Reid *et al.* [32] were adopted along with the necessary data for pure components shown in Table 2.

Mixture properties are calculated for equilibrium conditions, thus they reflect the double effect of composition and variation in the bubble point temperature with composition. As evident in Fig. 6, the nonlinear variations are significant for the aqueous mixture of ethanol and water, while the methanol and ethanol mixture behaves approximately as a fluid with property variations idealized with regards to composition.

FIG. 4. Comparison of heat transfer coefficients for pure components with the predictions by the Stephan and Abdelsalam correlation.

As proved in Fig. 4 the Stephan and Abdelsalam taking into account the property variation of mixtures correlation can satisfactorily predict the heat transfer with composition. Contact angles of 45° for water and

with composition. Contact angles of 45° for water and coefficient for five pure single components whose of 35° for hydrocarbon should be used in employing physical properties are considerably different. Figure the Stephan and Abdelsalam correlation. But its effect physical properties are considerably different. Figure the Stephan and Abdelsalam correlation. But its effect 7 shows the measured heat transfer coefficients at a is very small and accurate data for contact angle of 7 shows the measured heat transfer coefficients at a is very small and accurate data for contact angle of heat flux of 10^5 W m⁻² and predictions from the mixtures are not available. Thus a contact angle of heat flux of 10⁵ W m⁻² and predictions from the mixtures are not available. Thus a contact angle of Stephan and Abdelsalam correlation and others by 45[°] was used for all mixtures. Although the ethanol 45° was used for all mixtures. Although the ethanol

FIG. 5. (a) and (b).

FIG. 5. Variations of measured heat transfer coefficients with mixture composition and heat flux, along with variations of ΔT_{bp} and $|Y_1 - X_1|$: (a) methanol/water; (b) ethanol/water; (c) methanol/ethanol; (d) ethanol/ n -butanol ; (e) methanol/benze

FIG. **6.** Physical property variations with mixture composition : (a) ethanol/water; (b) methanol/ethanol.

and water mixture shows the nonlinear variations of its physical properties with composition, the three referred correlations which are developed for single components are unable to predict the variation of measured heat transfer coefficient with composition. The Stephan and Abdelsalam correlation predicts, differently from the trend of the experimental data, a higher heat transfer coefficient than the linearly interpolated value between the two pure components. As exemplified here, and as pointed out often in many studies, nucleate boiling heat transfer coefficients for mixtures cannot be satisfactorily predicted using correlations developed for single components even when they are qualified to give an adequate prediction for single pure components.

Prediction from correlations developed jbr mixtures

Predictive methods or correlations to estimate the reduction of heat transfer coefficient of mixtures have been developed by Stephan and Körner [12], Calus and Rice [5], Calus and Leonidopoulos [6], Happel [22], Jungnickel et al. [23], Thome [15, 18], Schlünder [17], Thome and Shakir [25], and Unal [24]. These correlations account for the mixture effects by applying certain correction factors related to phase diag-

FIG. *7.* Composition dependency of heat transfer coefficients predicted by correlations developed for single components $(\nabla:$ measured data): (a) ethanol/water; (b) methanol/ ethanol.

rams or physical properties. The details of their development, models and features are discussed in review articles [19-21].

Among the above mentioned correlations, the following typical ones are employed to compare with the present results. The correlations proposed by Calus and Rice [5], Calus and Leonidopoulos [6], and Thome [15] are unique in that they include the effect of mass diffusivity in the form of the Lewis number. But the variation of mass diffusivity with composition is often difficult to determine or predict. And the correction factors in terms of mass diffusivity show a very complex change with composition, compared to the variation of heat transfer coefficient with composition. Thus these three correlations are excluded from the present comparison

Stephan and Kiirner [121:

$$
\frac{\alpha}{\alpha_{\rm id}} = \frac{1}{1 + Ao|Y_1 - X_1|(0.88 + 0.12P)}; \quad P \text{ in bar} \quad (2)
$$

Thome [18]:

$$
\frac{\alpha}{\alpha_{\rm id}} = \frac{1}{1 + \Delta T_{\rm bp}/\Delta T_{\rm Id}}\tag{3}
$$

(4)

Schlünder [17]:

$$
\frac{\alpha}{\alpha_{\rm id}} = \frac{1}{1 + \frac{(T_{\rm s2} - T_{\rm s1})|Y_1 - X_1|}{\Delta T_{\rm id}} \left[1 - \exp\left(\frac{-Bog}{\beta_1 \rho_1 h_{\rm fg}}\right)\right]}
$$

$$
\frac{\alpha}{\alpha_{\rm id}} = \frac{1}{1 + K_{\rm ST} |Y_1 - X_1| (\rho_{\rm v}/\rho_{\rm L}) q^{0.48 + 0.1X_1}}.
$$
 (5)

In the above correlations, the ideal wall superheat, ΔT_{id} , is defined from a linear molar interpolation between the wall superheats for the two pure components, ΔT_1 and ΔT_2 , at the same heat flux as the mixture. Then ΔT_{id} is given by

$$
\Delta T_{\rm id} = X_1 \Delta T_1 + (1 - X_1) \Delta T_2. \tag{6}
$$

The ideal heat transfer coefficient, α_{1d} , can then be determined for the same heat flux as

$$
\alpha_{\rm id} = \frac{q}{\Delta T_{\rm id}} = \frac{1}{X_1/\alpha_1 + (1 - X_1)/\alpha_2} \tag{7}
$$

where α_1 and α_2 are heat transfer coefficients for the two pure components.

Ao in equation (2) is an empirical constant. Stephan and Körner [12] determined the value of *Ao*, to range from 0.42 to 3.56, for seventeen mixtures by fitting equation (2) to the measured data. Their average value of 1.53 is recommended for mixtures whose data are not available. ΔT_{bp} in equation (3) is the temperature difference between the dew point and bubble point curves as shown in Fig. 3. Thus its value is determined from phase equilibrium data. In equation (4) , T_s is the saturation temperature of pure components at the same pressure as the mixture and Bo is a scaling factor that accounts for the fraction of heat consumed for evaporation vs the total heat input from the boiling surface. Schlünder [17] assumed $Bo = 1.0$ and set the liquid mass transfer coefficient, β_L , equal to a constant value of 0.0002 m s⁻¹ in correlating other author's data. K_{ST} in equation (5) is an empirical constant. Jungnickel *et al.* [23] determined the value of K_{ST} to five freon mixtures and gave a smooth curve of K_{ST} as a function of the temperature difference between the normal boiling points of the two pure components. Equations (4) and (5) include the heat flux explicitly but the predicted influence of the heat flux on the reduction of heat transfer coefficient is very small in these two correlations.

Figure 8 gives a comparison of the data with the predictions. The values of ΔT_1 and ΔT_2 , or α_1 and α_2 , for the two pure components comprising the mixture are obtained from the present experimental data. In the case of azeotrope-forming mixtures, the values for the azeotrope are used instead of ΔT_i and α_i for compositions to the left of the azeotrope, or ΔT_2 and α_2 for compositions to the right of the azeotrope. It is found from all the comparisons with the measured data that no single correlation can predict heat transfer coefficients within a similar degree of accuracy for all the mixtures. As for the variation of heat transfer coefficient with composition and for the more significant reduction of heat transfer coefficient with heat flux, there exist large deviations between the data and the correlations.

Jungnickel et al. [23] : *Modification for correlations*

The reduction of heat transfer coefficient for mixtures relative to the ideal heat transfer coefficient is compensated by the second term in the denominator on the right hand side of correlating equations (2) – (5) . To modify this term, a modification factor K_{md} is multiplied as an adjustable constant, the values of K_{md} being determined from fitting the modified form of correlations to the data. The results are shown in Fig. 9. The case of $K_{\text{md}} = 1$ refers to the original correlation. And a large deviation of K_{md} from unity means an increased difference between the original correlation and the data. It is found that as the heat flux is lowered the greater the modification required.

The modification factor K_{md} for the Thome correlation decreases from about unity at higher heat flux with a decrease of heat flux. Thome [18] noted the need to modify his correlation to improve the accuracy at low heat fluxes far below the peak nucleate boiling heat flux because his model holds true at the peak heat flux in the strict sense. Since the difference of K_{md} between the five mixtures is smallest in the case of the Thome correlation, a variation of K_{md} with heat flux is approximated by a solid **curve** in Fig. 9(b) irrespective of the mixtures. Then the Thome correlation is modified into the following form so as to account for the effect of heat flux :

$$
\frac{\alpha}{\alpha_{\rm id}} = \frac{1}{1 + [1 - 0.8 \exp(-q/10^5)] (\Delta T_{\rm bp} / \Delta T_{\rm id})} \quad (8)
$$

where the unit of heat flux, q , is W m⁻². The modified Thome correlation (8) predicts the heat transfer coefficients for the five present mixtures within $\pm 20\%$ accuracy as shown in Fig. 10. The modification factor in equation (8) should be expressed in a dimensionless form, for instance, just like as the heat flux term in equation (4). To perform such refinement future work is needed.

CONCLUSION

Heat transfer coefficients during nucleate pool boiling were measured for five saturated mixtures at atmospheric pressure. Significant influences of the concentration and the heat flux on the heat transfer reduction were found. The available empirical and semi-empirical correlations developed for mixtures were compared with the measured data. Among them two correlations proposed by Stephan and Abdelsalam and by Thome gave fairly good results on the whole but, going into details, a certain degree of deviation is found concerning the composition for the maximum reduction of heat transfer coefficient and

FIG. 8. Comparison of the measured heat transfer coefficients with the predictions by correlations developed Fig. 8. Comparison of the measured heat transfer coefficients with the predictions by correlations developed
for mixtures : (a) methanol/water ; (b) ethanol/water ; (c) methanol/ethanol ; (d) ethanol/n-butanol ;
(e) metha

FIG. 9. Modification factors: M/W: methanol/water; E/W: ethanol/water; M/E: methanol/ethanol; E/Bu : ethanol/n-butanol; M/Bz : methanol/benzene. (a) Stephan and Körner correlation; (b) Thome correlation ; (c) Schliinder correlation ; (d) Jungnickel et *al.* correlation.

the heat flux dependency of the heat transfer reduction. The heat transfer data are well correlated by the Thome correlation modified by multiplying to the term expressing the mixture effect a modification factor, which was determined as a function of heat flux irrespective of mixtures.

FIG. 10. Comparison of the measured data with the predictions by a modified Thome correlation: M/W: methanol/water; E/W: ethanol/water; M/E: methanol/ ethanol; E/Bu : ethanol/n-butanol; M/Bz : methanol/ benzene.

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