



Heat transfer in nucleate pool boiling of multicomponent mixtures

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Abstract

Steady state pool boiling heat transfer coefficients have been obtained experimentally for acetone–isopropanol–water and acetone–MEK (methyl ethyl ketone)–water ternary systems. The data show that, for a given heat flux, the heat transfer coefficients of mixtures are lower than those obtained for pure components constituting the mixture. The measured heat transfer coefficients were compared with predictions from literature correlations for pool boiling of multicomponent mixtures. In all the cases, overestimation or underestimation of the data was observed. The literature correlations incorporate an ‘ideal’ heat transfer coefficient and a correction term for the presence of other liquids. Part of the uncertainty associated with the literature correlations on comparison with the present data appears to be due to the uncertainty in estimating the ideal heat transfer coefficient. The methods suggested by the earlier authors found to be not applicable for the present heating surface–liquid combinations. Therefore, in the present study, two different correlations were tried for the ideal heat transfer coefficient and it was found that the performance of the literature correlations improved considerably. However, the methods reported in the literature and the correlations tried in the present study to estimate ideal heat transfer coefficient did not consider heating surface–liquid interaction and the effect of heating surface micro-roughness in boiling. Therefore, a correlation to estimate the ideal heat transfer coefficient has been proposed taking into account surface–liquid interaction parameter and surface roughness group. A new correlation to estimate mixture heat transfer coefficients has been proposed in the present study in terms of an ideal heat transfer coefficient and a correction term. In general, the correction term in binary mixtures is obtained by incorporating the binary diffusivity of the mixture. In multicomponent systems, the multicomponent diffusion coefficients have to be incorporated in the expression for the correction term. The mixture heat transfer coefficient correlation along with the correlation for ideal heat transfer coefficient proposed in the present study predicts the present experimental data as well as literature data satisfactorily. The heat transfer coefficient was found to be a function of the difference between the equilibrium vapor and liquid concentration of the light component(s) and the minimum heat transfer coefficient occurs at the maximum of this value.

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1. Introduction

When evaporation occurs at a solid–liquid interface, it is termed boiling. Nucleate boiling is characterized by the formation of vapor at preferred sites (“nucleation” sites) on a heating surface that is submerged in the liquid and maintained at a temperature above the saturation temperature of the liquid. Boiling of pure components and binary mixtures has been well established, while

the boiling of mixtures with three or more components has been studied in less detail. Boiling of mixtures is of considerable practical significance. Nucleate pool boiling of liquid mixtures finds many applications in chemical and petrochemical process industries, air-separation, refrigeration and air-conditioning, and power generation. In addition it has applications in the high heat flux cooling in the electronics industry. It is found in the literature that the nucleate boiling heat transfer coefficients of mixtures can be considerably lower than those of an equivalent pure liquid with similar physical properties as the mixture. This is due to (a) reduction in temperature driving force because of increase in the boiling point of the micro-layer (the liquid layer trapped under a growing bubble) which is due

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Nomenclature

B	inverse of binary diffusion coefficient (s/m^2)	γ	surface–liquid interaction parameter, $\sqrt{(k_w \rho_w C_{p_w}) / (k_l \rho_l C_{p_l})}$ (dimensionless)
C_p	specific heat ($\text{J}/(\text{kg K})$)	λ	latent heat (J/kg)
d	diameter (m)	μ	viscosity (N s/m^2)
D	mass diffusivity (m^2/s)	ν	kinematic viscosity (m^2/s)
ΔT	wall superheat $T_w - T_s$ (K)	ρ	density (kg/m^3)
g	gravitational constant (m/s^2)	σ	surface tension (N/m)
h	heat transfer coefficient ($\text{W}/(\text{m}^2 \text{K})$)	<i>Subscripts and superscripts</i>	
k	thermal conductivity ($\text{W}/(\text{m K})$)	a	additional
Le	Lewis number, α/D (dimensionless)	b	bubble
M	molecular weight (kg/kmol)	bp	boiling range
P	pressure (N/m^2)	eff	effective
Pr	Prandtl number ($C_p \mu / k$) (dimensionless)	i, j	components
q	heat flux (W/m^2)	id	ideal
R_a	centre line average or arithmetic average roughness (μm)	l	liquid
T	temperature (K)	mix	mixture
V	molar volume at normal boiling point (m^3/kmol)	n	number of components
x	liquid mole fraction	s	saturation
y	vapor mole fraction	v	vapor
<i>Greek letters</i>		w	wall
α	thermal diffusivity (m^2/s)	1, 2, 3	more, intermediate and less volatile components
β	mass transfer coefficient (m/s)	0	reference

to the preferential evaporation of the light components during bubble growth, (b) the mass diffusion of the light components to the micro-layer (caused by the preferential evaporation) which is much slower than the heat transfer, (c) the fact that there is usually a significant and non-linear variation in the mixture physical properties with composition and (d) the effect of composition on nucleation itself.

Many industrial processes involve boiling of multi-component mixtures. However, most boiling research has been limited to the behavior of pure components or binary mixtures. One of the earliest studies on ternary mixture boiling on plain surfaces was by Grigor'ev et al. [1] who performed an experimental investigation on pool boiling of acetone–methanol–water and acetone–ethanol–water ternary mixtures. Stephan and Preusser [2] measured pool boiling heat transfer coefficients in acetone–methanol–water and methanol–ethanol–water ternary mixtures. Sardesai et al. [3] measured pool boiling heat transfer coefficients in hydrocarbon mixtures with up to five components. Bajorek et al. [4] reported experimental measurements of pool boiling heat transfer coefficients in six aqueous ternary systems. Kadhum et al. [5] carried out an investigation on acetone–isopropanol–water ternary system. Spindler et al. [6] measured heat transfer coefficients of ternary refrigerant mixtures *R404A* and *R407C*. Köster et al. [7]

investigated ternary refrigerant mixtures *R404A*, *R407C*. Krupiczka et al. [8] conducted an experimental study on methanol–isopropanol–water ternary system. Fujita and Tsutsui [9] studied the ternary refrigerant system *R134a-R142b-R123*.

There are many correlations available in the literature for pure components and binary mixtures but the boiling of mixtures is so complex that extending them to multicomponent mixtures is difficult. Only a few correlations exist in the literature on boiling of mixtures with more than two components. These are due to Palen and Small [10], Stephan and Preusser [2], Schlünder [11], Thome [12], Thome and Shakir [13], Bajorek and Lloyd [14], and Fujita and Tsutsui [9]. The correlations incorporate an 'ideal' heat transfer coefficient and a correction term for the presence of other liquids. The correction term, in general, is a function of thermo-physical and transport properties of the mixtures, the vapor–liquid equilibrium relationship and the slope of the vapor pressure curve. It apparently takes into account slower bubble growth rates, smaller departure diameters and non-linear variation of the mixture properties with composition. The ideal heat transfer coefficient h_{id} , is the heat transfer coefficient the mixture would be expected to have without any mass transfer effects. It can be estimated using the linear mixing law, which is a linear combination of pure component heat

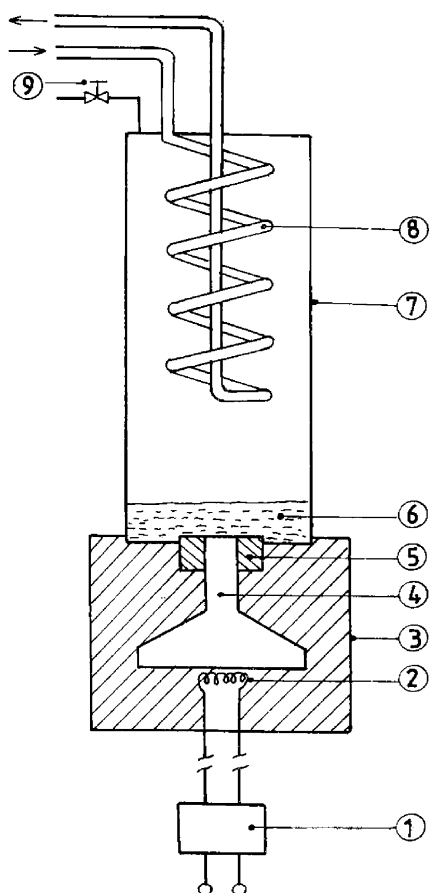
transfer coefficients or alternatively using mixture properties in empirical correlations applicable to pure liquids.

However, both the data and the correlations applicable to multicomponent mixtures are limited and cover very few mixtures so much so, the correlations for pool boiling of multicomponent liquid mixtures have not been validated conclusively as yet. This formed the motivation for the present study. Much of mixture boiling hypotheses, experimental studies and correlations have been summarized by Thome and Shock [15], and Bajorek and Lloyd [14].

2. Experimental

2.1. Experimental set up

The experimental set up [16,17] is shown in Fig. 1. It consists of a double walled glass cylinder with the annular space between the two walls under vacuum to minimize heat loss. The height of the glass column is 260 mm and



1. Dimmerstat 2. Plate heater 3. Insulation 4. Heating block
5. Teflon hollow cylinder 6. Liquid 7. Double-walled glass vessel
8. Cooling coil 9. Relief valve

Fig. 1. Schematic of the experimental set up.

its inner diameter is 93 mm. The glass cylinder is held tightly between two stainless steel slabs, which have cylindrical grooves cut in them to fit the column exactly. The top slab contains a relief valve and has a provision to allow circulation of cooling water (using cooling coils) to condense the vapor back into the bulk liquid.

To hold the cylinder tightly between the two slabs, tie rods and rubber gaskets are used. Aluminium is used as a heating block and it is fitted through the centre of a hollow Teflon cylinder in the bottom slab. The heating block is thereby separated from the slab with Teflon as the insulation material. The heating block and the Teflon cylinder are at the same level. The heating block is 29.8 mm in diameter and is heated by an electrical heating element of 3 kW capacity. The heating element is connected to a voltmeter and ammeter through a dimmerstat to read the voltage and current supplied to it. The heating block is wrapped with insulation material to ensure one-dimensional heat flow in the axial direction. Provision was made to measure temperatures at four points along the axis of the heating block using chromel–alumel thermocouples. The liquid temperature is also measured using the same type of thermocouple. The thermocouples were connected to a millivoltmeter through a selector switch to read the generated emfs.

2.2. Experimental procedure

Experiments were conducted at atmospheric pressure and standard gravity conditions on acetone(1)–isopropanol(2)–water(3), acetone(1)–MEK(2)–water(3) systems. Data obtained for respective pure components present in the system, binary mixtures of isopropanol–water and MEK–water with concentration of each component of 0.5 molefraction, and ternary mixtures covering the concentration range of acetone from 0.1 to 0.9 keeping the concentrations of other two components equal. These liquid systems were chosen for investigation as the experimental vapor–liquid equilibrium data is readily available for binary and ternary mixtures of the system. Further, they are readily miscible with each other in all proportions, do not form azeotropes and there is a significant difference in the boiling point of the pure liquids. The boiling points of binary and ternary mixtures were estimated using Aspen Plus Steady State Simulation Package, Version 10.1-0. The bubble points estimated were found to match the temperatures measured during the boiling experiments.

About 150 ml of liquid mixture is boiled in each run. After a particular voltage was applied across the heater, a steady state condition was assumed to exist when the temperature along the axis of the heating block did not vary by more than 0.1 K for about 2 min. Then the temperatures along the axis of the heating block are measured. The temperature gradient along the axis of the heating block was obtained by solving the

one-dimensional steady-state heat conduction equation with no energy generation within the conducting medium and constant thermal conductivity. Thereafter, the surface temperature can be obtained by extrapolation. Using the Fourier’s law, the heat flux was determined. The heat transfer coefficient was obtained using the Newton’s law of cooling. Before taking the first reading of each run the liquid and the heating surface are heated for nearly 2 h to ensure degassing and aging. For a given composition, boiling experiments were conducted at different excess temperatures (wall superheats). The boiled liquid is condensed back into the test section using cooling water. Therefore, the bulk liquid composition does not change during the course of the experiment. Prior to the start of each boiling experiment, the heating surface was polished with 4/0 emery paper and the surface roughness as characterized by the centre line average R_a was measured using a surface profilometer (Perth-o-meter). It was found to be $0.52 \mu\text{m}$. The R_a value has been extensively used in boiling literature to characterize the surface micro-roughness.

2.3. Experimental uncertainty

The resolution of the voltmeter used in the present study is 1 V. The resolution of the ammeter used is 0.01 A. The resolution of the millivoltmeter used is 0.01 mV. The uncertainty in the measurement of temperature is $\pm 0.2 \text{ K}$.

3. Results

The variation of heat transfer coefficient with heat flux for different compositions of the two ternary systems studied is shown in Figs. 2 and 3. The solid symbols indicate pure components or binary mixtures and the hollow symbols indicate ternary mixtures. From the figures, it is observed that for a given heat flux, the heat transfer coefficients of mixtures are lower than those obtained for the constituent pure liquids or binary mixtures. It is also noticed from Figs. 2 and 3 that the heat transfer coefficient for a given heat flux decreases

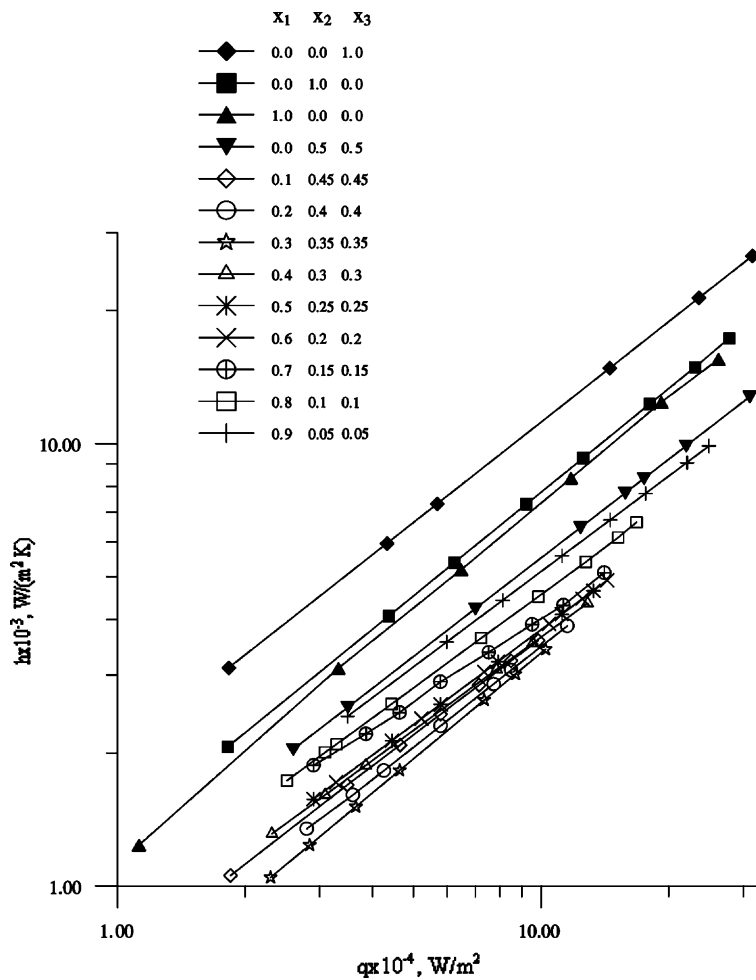


Fig. 2. Nucleate boiling curves for acetone(1)–isopropanol(2)–water(3) system (x_1 = molefraction of acetone; x_2 = molefraction of isopropanol; x_3 = molefraction of water).

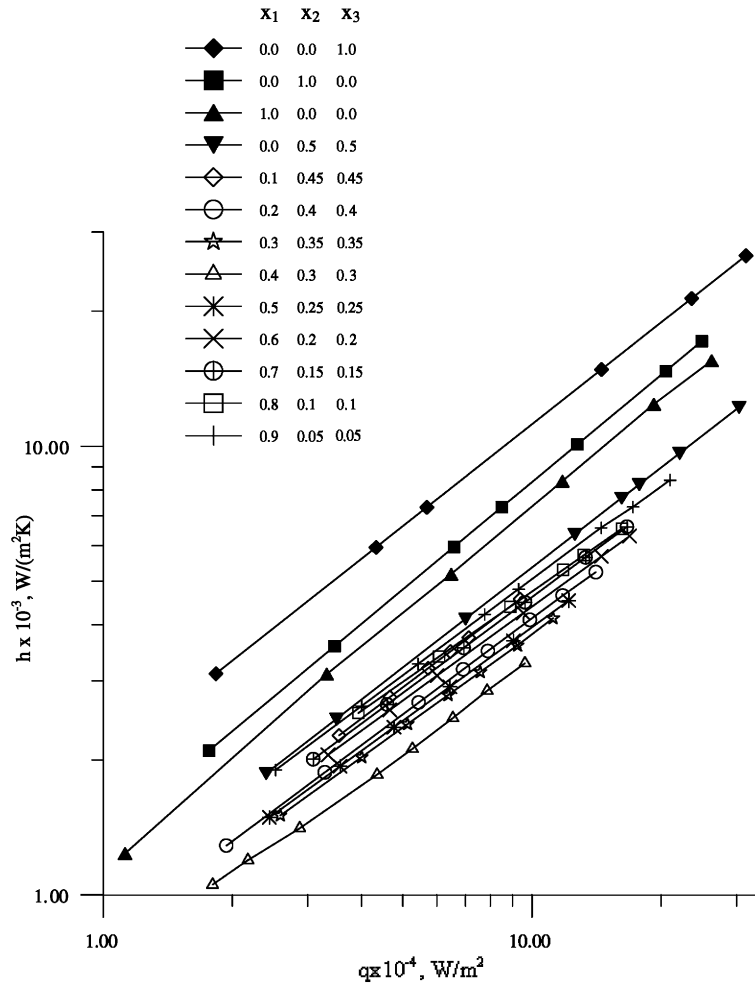


Fig. 3. Nucleate boiling curves for acetone(1)–MEK(2)–water(3) system (x_1 = molefraction of acetone; x_2 = molefraction of MEK; x_3 = molefraction of water).

and then increases with increase in the concentration of acetone. As can be seen from the figures, the heat transfer coefficient at a given heat flux decreases up to $x_1 = 0.3$, $x_2 = 0.35$ and then increases as the concentration of acetone increases in acetone(1)–isopropanol(2)–water(3) system, whereas in acetone(1)–MEK(2)–water(3) system the heat transfer coefficient decreases up to $x_1 = 0.4$, $x_2 = 0.3$ and then increases with increase in the concentration of acetone. This kind of decreasing–increasing trend in heat transfer coefficient or heat flux with increase in the concentration of the more volatile component has been observed previously in the case of binary mixtures. Hui and Thome [18] found that the heat transfer coefficient decreases and then increases with increase in ethanol concentration (for all subcoolings) for ethanol–water system. Benjamin and Balakrishnan [19] while studying acetone– CCl_4 and n -hexane– CCl_4 systems observed that heat flux (and the nucleation site density) decreases when a more volatile component is added to a high boiling liquid and then increases as the concentration of the more volatile component in the liquid mixture increases.

4. Discussion

4.1. Comparison of present experimental data with multicomponent correlations in the literature

One of the objectives of this investigation is to compare the present experimental data with existing multicomponent pool boiling correlations in the literature. The correlations chosen for comparison are those of Palen and Small [10], Schlünder [11], Thome and Shakir [13], and Bajorek and Lloyd [14]. This has been done earlier by Vinayak Rao and Balakrishnan [16] and is given in Appendix A. As can be seen from Table 3, the literature correlations either overpredicted or underpredicted the present experimental data.

4.2. Estimation of the ideal heat transfer coefficient

Part of the uncertainty associated with the correlations for pool boiling of multicomponent mixtures in the literature on comparison with the present data (Table 3) was due to the inadequacy of the methods used

in estimating the ideal heat transfer coefficient. As Bajorek et al. [20] said, “The linear mixing law assumes that the effect of surface on heat transfer in mixture is the same as for pure components but it does not take into account non-linear property variation with composition. The pure component correlations account for non-linear property variations, however they do not contain a term to explicitly model a surface effect”. Therefore, it is necessary to verify the applicability of these methods to a specific surface–liquid combination. The large scatter of present experimental data predicted by the literature correlations was probably due to the non-applicability of these methods for the present surface–liquid combinations. Therefore, in the present study two different correlations were chosen to estimate ideal heat transfer coefficient and to verify their applicability for the present surface–liquid combinations. They are the linear mixing law in conjunction with the correlation due to Gorenflo [21], and Stephan and Abdelsalam’s [22] correlation for pure components.

The linear mixing law is defined as,

$$\frac{1}{h_{id}} = \sum_{i=1}^n \frac{x_i}{h_i} \quad (1)$$

where h_i represents the heat transfer coefficient of the pure component at the same heat flux as the mixture.

The pure component heat transfer coefficients were calculated using the Gorenflo [21] correlation expressed as,

$$\frac{h_i}{h_0} = \left(\frac{q}{q_0}\right)^{n(p^*)} F(p^*) \left(\frac{R_a}{R_{a0}}\right)^{0.133} \quad (2)$$

where the pressure function $F(p^*)$ and the exponent $n(p^*)$ were calculated with the following equations.

For organic liquids,

$$F(p^*) = 1.2p^{*0.27} + \left(2.5 + \frac{1}{1-p^*}\right)p^* \quad (3)$$

$$n(p^*) = 0.9 - 0.3p^{*0.3} \quad (4)$$

For water,

$$F(p^*) = 1.73p^{*0.27} + \left(6.1 + \frac{0.68}{1-p^*}\right)p^{*2} \quad (5)$$

$$n(p^*) = 0.9 - 0.3p^{*0.15} \quad (6)$$

The reference heat transfer coefficients h_0 which were measured at a heat flux of 20,000 W/m² and at a reduced pressure of 0.1 are listed by Gorenflo [21] for numerous liquids. For the liquids used in the present study the reference heat transfer coefficients are 3700, 3000, 2500 and 5600 W/m²K for acetone, isopropanol, MEK and water respectively. The reference R_a for these heat transfer coefficients is 0.4 μm.

The Stephan and Abdelsalam [22] correlation used to estimate ideal heat transfer coefficient is,

$$h_{id} = 0.23 \cdot \frac{k_l}{d_b} \cdot \left(\frac{qd_b}{k_l T_s}\right)^{0.674} \cdot \left(\frac{\rho_v}{\rho_l}\right)^{0.297} \cdot \left(\frac{\lambda d_b^2}{\alpha_l^2}\right)^{0.371} \cdot \left(\frac{\alpha_l^2 \rho_l}{\sigma d_b}\right)^{0.350} \cdot \left(\frac{\rho_l - \rho_v}{\rho_l}\right)^{-1.73} \quad (7)$$

The bubble departure diameter was calculated using the expression [2]

$$d_b = 0.0146\theta \sqrt{\frac{2\sigma}{g(\rho_l - \rho_v)}} \quad (8)$$

The contact angle was assumed to be 35° for the mixtures. Mixture properties were used in Eq. (7) and these were obtained using selected correlations and mixing rules for the present ternary systems from Reid et al. [23] and Holland et al. [24].

Before applying these correlations for the present ternary mixtures, the correlations were compared with the pure component data of acetone to verify if the correlations are applicable for the present surface–liquid combinations. The correlations predicted the boiling data of acetone (Fig. 4) as well as the data of other pure liquids used in the present study satisfactorily.

The comparison of the present experimental data with the multicomponent correlations in the literature with these correlations for ideal heat transfer coefficient is shown in Figs. 5–8. The respective errors for all the correlations is given in Tables 1 and 2. As can be seen from Figs. 5–8 and Tables 3, 1 and 2, the Palen and Small [10] correlation overestimates acetone–isopropanol–water ternary data by about 46% and acetone–

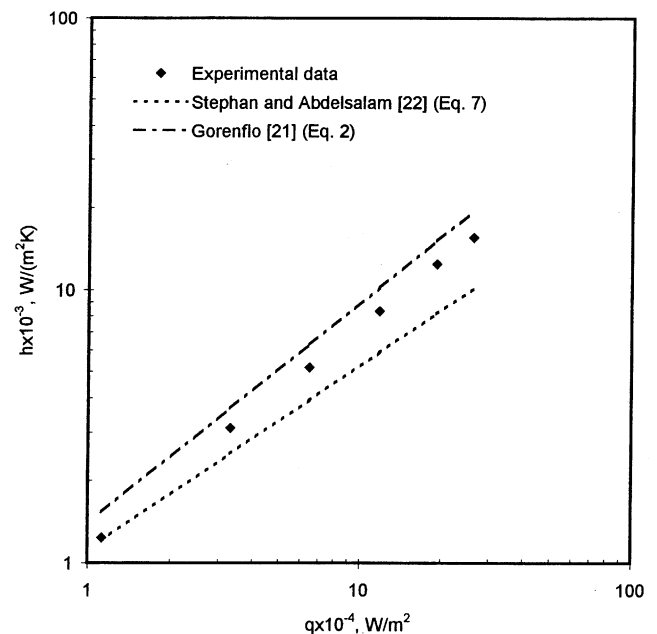


Fig. 4. Comparison of pure component data of acetone with Gorenflo [21] correlation (Eq. (2)) and Stephan and Abdelsalam [22] correlation (Eq. (7)).

MEK–water ternary data by about 42% when McNelly [25] correlation is used to estimate ideal heat transfer coefficient. When linear mixing law along with the correlation due to Gorenflo [21] is used to estimate the ideal heat transfer coefficient the average error is reduced to

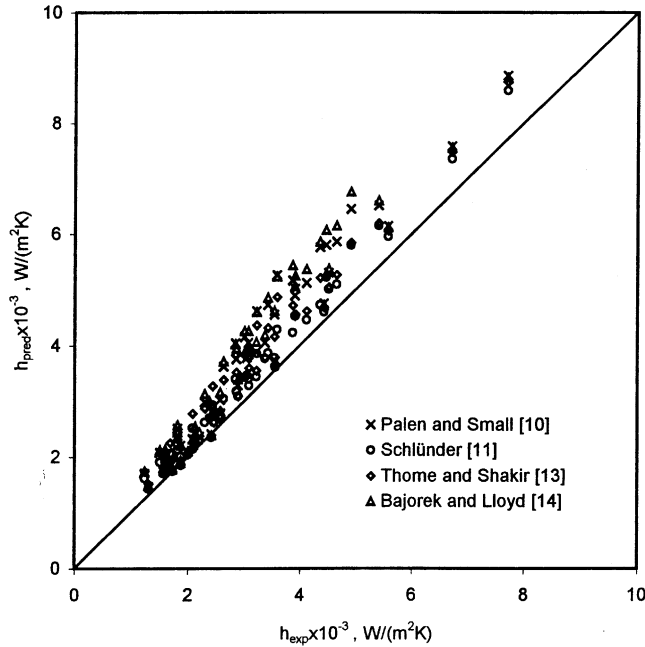


Fig. 5. Comparison of acetone–isopropanol–water ternary boiling data with existing multicomponent correlations in the literature when h_{id} is calculated using the linear mixing law along with Gorenflo [21] correlation (Eq. (2)).

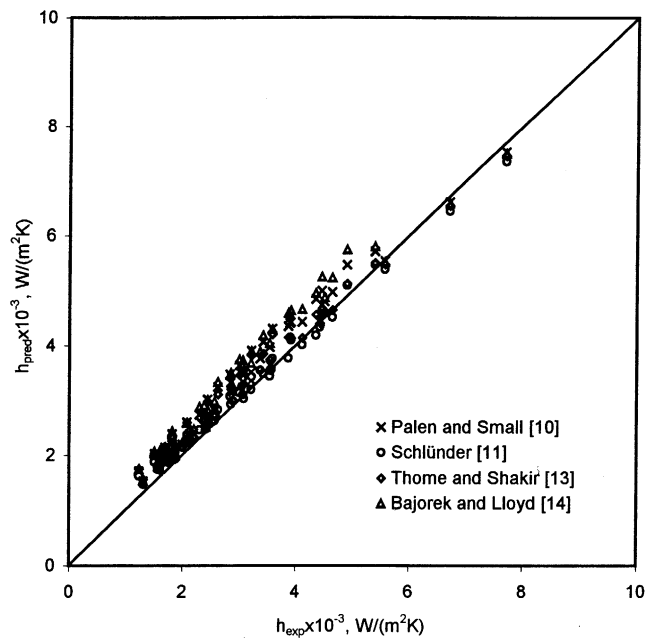


Fig. 6. Comparison of acetone–isopropanol–water ternary boiling data with existing multicomponent correlations in the literature when h_{id} is calculated using the Stephan and Abdelsalam [22] correlation (Eq. (7)).

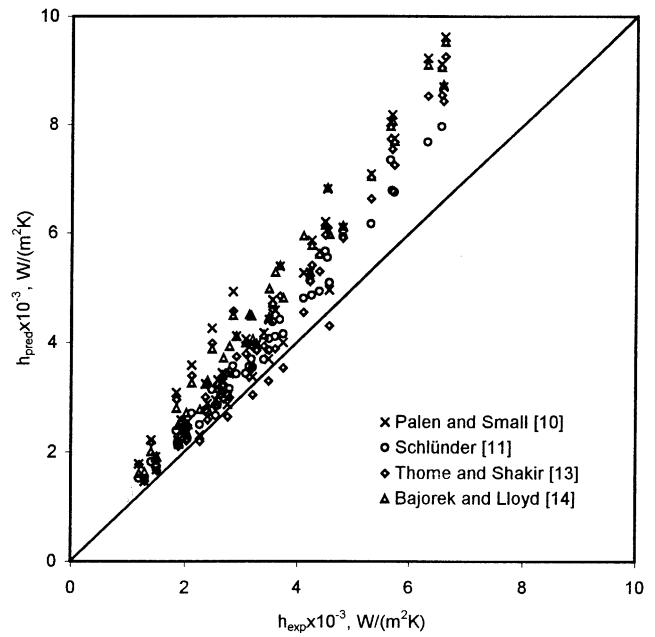


Fig. 7. Comparison of acetone–MEK–water ternary boiling data with existing multicomponent correlations in the literature when h_{id} is calculated using the linear mixing law along with Gorenflo [21] correlation (Eq. (2)).

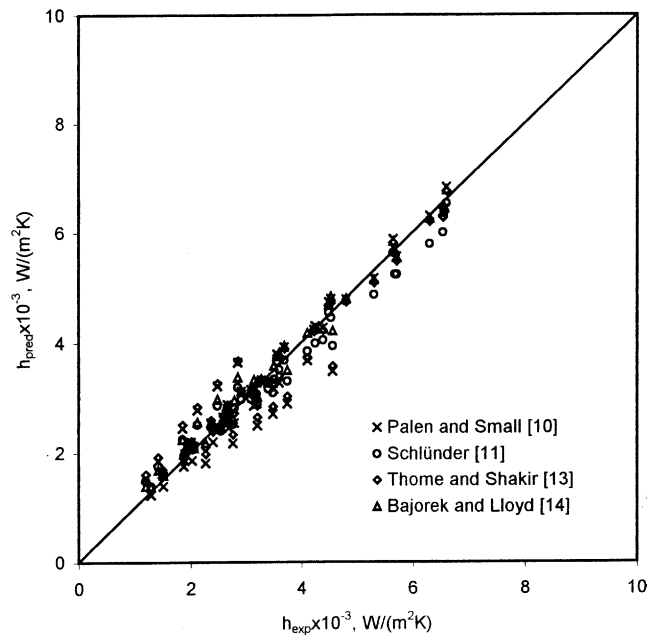


Fig. 8. Comparison of acetone–MEK–water ternary boiling data with existing multicomponent correlations in the literature when h_{id} is calculated using the Stephan and Abdelsalam [22] correlation (Eq. (7)).

21% for acetone–isopropanol–water ternary data and to 30% for acetone–MEK–water ternary data. When Stephan and Abdelsalam [22] correlation (Eq. (7)) is used to estimate the ideal heat transfer coefficient the average error further reduced to 14% for acetone–isopropanol–water ternary data and to about 0.3% for

Table 1

Performance of multicomponent correlations in the literature (h_{id} based on linear mixing law along with Gorenflo [21] correlation, Eq. (2))

Correlation	Eq.	Mixtures	Average error (%)	MAE (%)	RMS error (%)	SD (%)
Palen and Small [10]	(A.1)	Acetone–isopropanol–water	21.4	21.4	24.6	12.0
		Acetone–MEK–water	30.4	30.4	34.7	17.0
Schlünder [11]	(A.3)	Acetone–isopropanol–water	11.4	11.4	13.4	6.8
		Acetone–MEK–water	17.6	17.6	18.7	6.4
Thome and Shakir [13]	(A.4)	Acetone–isopropanol–water	17.3	17.4	20.4	10.8
		Acetone–MEK–water	22.0	23.1	27.8	15.6
Bajorek and Lloyd [14]	(A.6)	Acetone–isopropanol–water	24.5	24.5	27.7	13.1
		Acetone–MEK–water	34.5	34.5	36.0	10.3

Table 2

Performance of multicomponent correlations in the literature (h_{id} based on Stephan and Abdelsalam [22] correlation, Eq. (7))

Correlation	Eq.	Mixtures	Average error (%)	MAE (%)	RMS error (%)	SD (%)
Palen and Small [10]	(A.1)	Acetone–isopropanol–water	13.9	14.0	16.2	8.1
		Acetone–MEK–water	0.3	9.8	13.5	9.3
Schlünder [11]	(A.3)	Acetone–isopropanol–water	5.6	6.7	9.0	6.0
		Acetone–MEK–water	0.4	6.5	8.7	5.7
Thome and Shakir [13]	(A.4)	Acetone–isopropanol–water	10.7	10.9	14.3	9.3
		Acetone–MEK–water	2.6	9.1	13.4	10.0
Bajorek and Lloyd [14]	(A.6)	Acetone–isopropanol–water	16.8	17.0	19.1	8.7
		Acetone–MEK–water	3.8	6.0	8.0	5.3

acetone–MEK–water ternary data. Schlünder [11] correlation overestimates acetone–isopropanol–water ternary data by about 45% and acetone–MEK–water ternary data by about 41% when linear mixing law is used to estimate the ideal heat transfer coefficient. When linear mixing law along with the correlation due to Gorenflo [21] is used to estimate the ideal heat transfer coefficient the average error is reduced to about 11% for acetone–isopropanol–water ternary data and to about 17% for acetone–MEK–water ternary data. When Stephan and Abdelsalam [22] correlation (Eq. (7)) is used to estimate the ideal heat transfer coefficient the average error further reduced to about 6% for acetone–isopropanol–water ternary data and to about 0.4% for acetone–MEK–water ternary data. Thome and Shakir [13] correlation underestimates acetone–isopropanol–water ternary data by about 25% and acetone–MEK–water ternary data by about 32% when Stephan and Abdelsalam [22] correlation (Eq. (A.5)) is used to estimate the ideal heat transfer coefficient. When linear mixing law along with the correlation due to Gorenflo [21] is used to estimate the ideal heat transfer coefficient the predictions being higher by about 17% for acetone–isopropanol–water ternary data and by about 22% for acetone–MEK–water ternary data. When Stephan and Abdelsalam [22] correlation (Eq. (7)) is used to estimate the ideal heat transfer coefficient the average error is about 11% for acetone–isopropanol–water ternary data and about 3% for acetone–MEK–water ternary data. Bajorek and Lloyd [14] correlation underestimates acetone–isopropanol–water ternary data by about 25% and acetone–MEK–water ternary data by about 33% when

Stephan and Abdelsalam [22] correlation (Eq. (A.5)) is used to estimate the ideal heat transfer coefficient. When linear mixing law along with the correlation due to Gorenflo [21] is used to estimate the ideal heat transfer coefficient the predictions being higher by about 25% for acetone–isopropanol–water ternary data and by about 35% for acetone–MEK–water ternary data. When Stephan and Abdelsalam [22] correlation (Eq. (7)) is used to estimate the ideal heat transfer coefficient the average error is about 17% for acetone–isopropanol–water ternary data and about 4% for acetone–MEK–water ternary data. Both linear mixing law along with Gorenflo [21] correlation, and Stephan and Abdelsalam [22] correlation (Eq. (7)) improved the performance of the multicomponent correlations in the literature considerably. Of the two correlations used in the present study to estimate the ideal heat transfer coefficient, the correlation by Stephan and Abdelsalam [22] (Eq. (7)) gave better predictions. This shows that Stephan and Abdelsalam [22] correlation (Eq. (7)) is applicable for the present surface–liquid combinations and incorporation of non-linear variation of properties with composition is found to improve predictions. Similar observation has been made by Bajorek et al. [20] while comparing multicomponent correlations in the literature with their experimental data. They used linear mixing law and Stephan and Abdelsalam [22] correlation (Eq. (A.5)) to calculate the ideal heat transfer coefficient. They noticed that the performance of the existing multicomponent correlations was improved when the ideal heat transfer coefficient was calculated using Stephan and Abdelsalam [21] correlation (Eq. (A.5)).

The present study and the study of Bajorek et al. [20] indicate the importance of including the effect of mixture properties and show that the non-linear variation of properties is taken care by different correlations of Stephan and Abdelsalam [22] thereby predicted the experimental results better than other methods, which is consistent with the expectations that properties also play an important role in mixture boiling. However, as discussed previously, these correlations have to be evaluated for a specific surface–liquid combination before using them in the multicomponent correlations for pool boiling in the literature.

4.3. Correlation for heat transfer coefficient of mixtures

The Gorenflo [21] correlation with linear mixing law, and Stephan and Abdelsalam [22] (Eq. (7)) improved the performance of the literature correlations considerably. However, they do not take into account the effect of heating surface micro-roughness in boiling and the surface–liquid interactions. Further, most of the mixture boiling models/correlations are specific to the liquid mixtures and surfaces for which they were developed and hence cannot be used with confidence for other systems and surfaces. Therefore, in the present study, a new approach has been followed to develop a correlation for heat transfer coefficient of mixtures.

As discussed earlier, for a given heat flux, the heat transfer coefficients of mixtures are lower than those obtained for the equivalent pure liquids. The equivalent pure liquid is an imaginary liquid with the same physical properties as the mixture. The physical process of boiling of multicomponent mixtures is significantly different from those of boiling of a pure liquid. During the boiling of a liquid mixture, due to the vapor–liquid phase equilibrium characteristics, the vapor phase has a composition different from that of the liquid phase. Hence, as a liquid mixture evaporates on the heating surface, the vapor contains more of the light components. Therefore, the micro-layer is depleted of the light components and enriched in heavy components. This results in mass diffusion of the light components from the bulk to the micro-layer. Since the rate of mass diffusion is much slower than heat diffusion, mass transfer of the light components to the bubble interface becomes the limiting process and a portion of the driving force is utilized in overcoming the mass transfer resistance. Therefore, to obtain a given heat flux, an additional temperature driving force is required for mixtures.

If q is the obtained heat flux for a wall superheat ΔT then,

$$q = h\Delta T \quad (9)$$

The wall superheat ΔT in Eq. (9) contains an effective temperature driving force and an additional temperature driving force

$$\Delta T = \Delta T_{\text{eff}} + \Delta T_a \quad (10)$$

To obtain the same heat flux for an equivalent pure liquid with the same properties as the mixture the wall superheat required is lower as there are no mass transfer effects.

$$q = h_{\text{id}}\Delta T_{\text{eff}} \quad (11)$$

From Eqs. (9) and (11) we obtain,

$$\frac{h}{h_{\text{id}}} = \frac{\Delta T_{\text{eff}}}{\Delta T} \quad (12)$$

In an earlier study, Benjamin and Balakrishnan [17] developed a model for heat flux for binary mixtures. They related the effective and superficial temperature driving forces as,

$$\frac{\Delta T_{\text{eff}}}{\Delta T} = \left[1 - \left(|y - x| \sqrt{\frac{D_{\text{AB}}}{\alpha_{\text{mix}}}} \right)^{0.5} \right] \quad (13)$$

Here D_{AB} is the binary mass diffusivity. A binary mixture has one light component and one heavy component. For the light component, the difference between the equilibrium vapor and liquid concentration, $(y - x) > 0$ and for the heavy component $(y - x) < 0$. In the above expression (Eq. (13)) the concentration difference $(y - x)$ is the driving force for mass diffusion of the more volatile component. Since mass diffusion is the slower and rate controlling process $(D_{\text{AB}}/\alpha_{\text{mix}})^{0.5}$ is a measure of the resistance to heat transfer. The above equation therefore quantifies the effective driving force in the boiling of binary mixtures. For an n component mixture, it is possible for there to be many light components and heavy components. Therefore, the reduction in the temperature driving force depends on the diffusion of all the light components to the bubble interface in order to maintain equilibrium. Unlike binary mixtures where a single diffusivity $D_{\text{AB}} = D_{\text{BA}}$ can describe the molecular diffusion, in the case of mixtures with more than two components, the main diffusion coefficients and cross-diffusion coefficients are necessary to characterize the system and these form a matrix of diffusion coefficients. Hence, the diffusion coefficient D_{ii} represents the diffusion of a component i due to its own gradient. The cross-diffusion terms, D_{ij} represents the diffusion of component i through j in the mixture. In the present study, these multicomponent diffusion coefficients were determined using a method from Taylor and Krishna [26] as described below.

For a non-ideal system, the matrix of Fick diffusion coefficients and Maxwell–Stefan diffusion coefficients in a multicomponent mixture are related as:

$$[D] = [B]^{-1}[I] \quad (14)$$

The elements of the thermodynamic correction factor matrix $[I]$ were calculated using the NRTL model for multicomponent mixtures [26].

For an n component mixture, the Fick matrix $[D]$ is of order $n - 1$ and the elements of $[B]$ are,

$$B_{ii} = \frac{x_i}{D_{in}} + \sum_{\substack{k=1 \\ k \neq i}}^n \frac{x_k}{D_{ik}} \quad (15)$$

$$B_{ij} = -x_i \left(\frac{1}{D_{ij}} - \frac{1}{D_{in}} \right) \quad (16)$$

The Maxwell–Stefan diffusivities [26] for each binary pair in a multicomponent mixture were estimated using,

$$D_{ij} = \left(D_{ij}^0 \right)^{(1+x_j-x_i)/2} \left(D_{ji}^0 \right)^{(1+x_i-x_j)/2} \quad (17)$$

where the infinite dilution diffusivities D_{ij}^0 were calculated using the correlation given by Taylor and Krishna [26].

$$D_{ij}^0 = 1.17282 \times 10^{-16} \frac{(\varphi_j M_j)^{1/2} T}{\mu_j V_i^{0.6}} \quad (18)$$

The association factor φ for the solvent has the values 1, 1.2, 1, 2.26 for acetone, isopropanol, MEK and water, respectively.

The main and cross-diffusion coefficients evaluated as described above were then averaged and the effective temperature driving force, Eq. (13) was modified and extended to multicomponent mixtures. Thus, the modified temperature driving force due to diffusion of light components in a multicomponent mixture may be written as,

$$\frac{\Delta T_{\text{eff}}}{\Delta T} = \left[1 - \sum_{i=1}^{n-1} \left((y_i - x_i) \sqrt{\frac{(\bar{D})^{1/n-1}}{\alpha_{\text{mix}}}} \right)^{0.5} \right] \quad (19)$$

where \bar{D} is the determinant of matrix of Fickian diffusion coefficients. For a binary mixture the Fick matrix $[D]$ reduces to a scalar and becomes a single binary Fick diffusivity.

Combining Eqs. (12) and (19), we obtain the expression,

$$\frac{h}{h_{\text{id}}} = \left[1 - \sum_{i=1}^{n-1} \left((y_i - x_i) \sqrt{\frac{(\bar{D})^{1/n-1}}{\alpha_{\text{mix}}}} \right)^{0.5} \right] \quad (20)$$

The methods suggested for ideal heat transfer coefficient in the literature can be used in Eq. (20). However, they do not consider surface–liquid interactions and the effect of surface micro-roughness in boiling. Therefore, the pure component data of Kurihara and Myers [27], Zuber [28], Kadhum et al. [5], Benjamin and Balakrishnan [29], Nageswara Reddy [30] and the data obtained in the present study were used to develop a correlation for ideal heat transfer coefficient. The boiling data, liquids studied and surfaces used by Kurihara and Myers [27] and Zuber [28] have been summarized by

Benjamin and Balakrishnan [29]. Kadhum et al. [5] obtained data for acetone, isopropanol, water on a horizontal brass tube (70% Cu and 30% Zn). The authors specified 400 grade emery paper used for polishing the heating surface. Therefore, the heating surface material used by them was polished using the appropriate grade of emery paper and the centre line average was measured using the Perth-o-meter. The R_a value obtained was 0.256 μm which was then used in the boiling heat transfer coefficient correlation. Benjamin and Balakrishnan [29] obtained data for acetone, CCL_4 , n -hexane and water on aluminium and steel surfaces. Nageswara Reddy [30] obtained data for methanol, ethanol, ethyl acetate and water on an aluminium surface. He used 4/0, 3/0 and 2/0 emery papers for polishing the aluminium surface and the surface roughnesses obtained were 0.52, 0.89, 1.17 μm respectively. In the present study, boiling data was obtained for acetone, isopropanol, MEK and water on an aluminium surface. The measured surface roughness was 0.52 μm for 4/0 emery paper. Using the data obtained by these authors, the ideal heat transfer coefficient was correlated taking into account surface–liquid interaction parameter and the surface roughness group. The correlation obtained is:

$$h_{\text{id}} = 0.74 \cdot \frac{k_l}{d_b} \cdot \left(\frac{q d_b}{k_l T_s} \right)^{0.674} \cdot \left(\frac{\rho_v}{\rho_l} \right)^{0.297} \cdot \left(\frac{\lambda d_b^2}{\alpha_l^2} \right)^{0.371} \cdot \left(\frac{\alpha_l^2 \rho_l}{\sigma d_b} \right)^{0.350} \cdot \left(\frac{\rho_l - \rho_v}{\rho_l} \right)^{-1.73} \times (\gamma)^{-0.1} \cdot \left(\frac{R_a P}{\sigma} \right)^{0.133} \cdot (\text{Pr})^{-0.5} \quad (21)$$

The proposed correlation for pure components to estimate ideal heat transfer coefficient compares satisfactorily with a wide range of data from the present study and from the literature and is shown in Fig. 9. The correlation is different from the Stephan and Abdelsalam [22] correlation (Eq. (7)) only in the exponents of additional parameters introduced, which were fitted for the data used in the present study.

Eq. (21) now can be used in Eq. (20) for ideal heat transfer coefficient with mixture properties. Fig. 10 shows the comparison between the present experimental data for ternary mixtures and the predictions of Eq. (20). The correlation predicts both acetone–isopropanol–water and acetone–MEK–water ternary data satisfactorily. Further, the correlation has been compared with the binary data of Van Stralen [31], and Tzan and Yang [32]. The boiling data, liquids studied and the surfaces used by Van Stralen [31], Tzan and Yang [32] have been summarized by Benjamin and Balakrishnan [17]. The correlation also compared satisfactorily with the ternary boiling data of Kadhum et al. [5]. The correlation predicts both present and the literature data to within $\pm 16\%$.

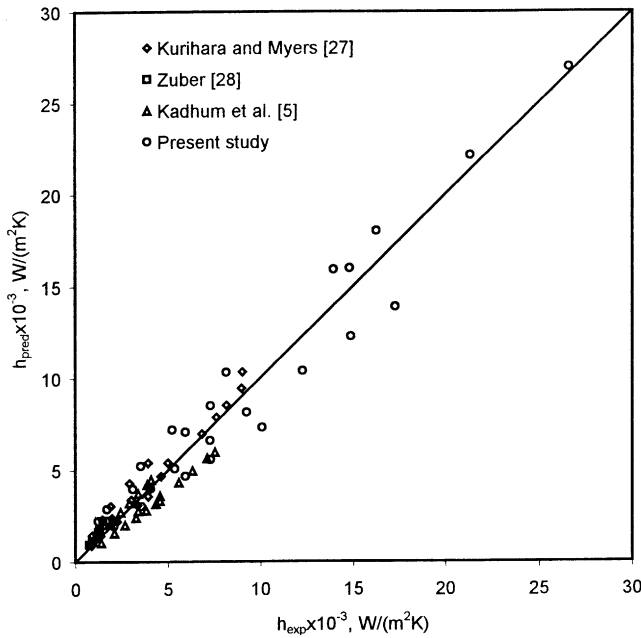


Fig. 9. Performance of the heat transfer coefficient correlation proposed for pure components.

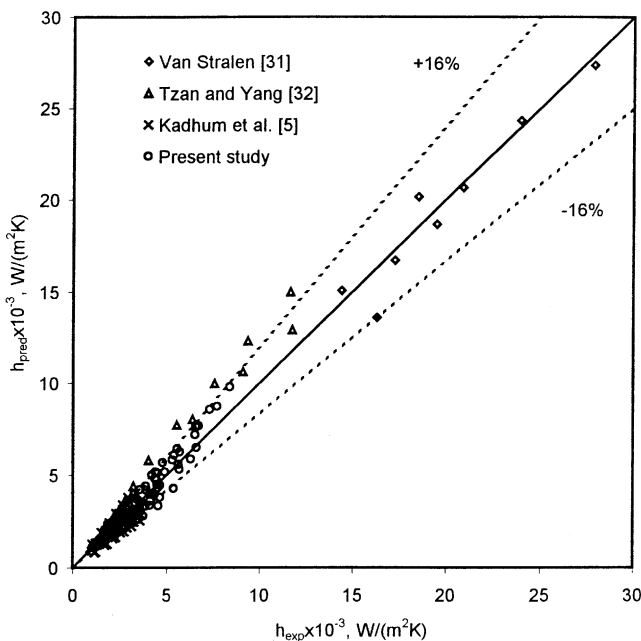


Fig. 10. Performance of the heat transfer coefficient correlation proposed for mixtures.

4.4. Uncertainty analysis

The relative uncertainties in using the proposed correlation for pure components can be estimated using the method due to Moffat [33]. In the present study the measured variables are the liquid temperature, the temperatures along the axis of the block and the centre line average R_a . The uncertainty in the measurement of

temperature is $\pm 1\%$. Therefore, the uncertainty in estimating the wall superheat is $\pm 1.4\%$. The uncertainty in heat flux is $\pm 2\%$ and the uncertainty in heat transfer coefficient is $\pm 2.4\%$. The uncertainty in the measurement of R_a is $\pm 1\%$. Therefore, the uncertainty in estimating $(R_a P / \sigma)$ is $\pm 1\%$. The uncertainty in estimating $(q d_b / k_l T_s)$ is $\pm 2.24\%$. Therefore, the relative uncertainty in the proposed heat transfer coefficient correlation for pure components is 1.5%.

4.5. Physical significance of additional parameters used in the pure component correlation for heat transfer coefficient

4.5.1. Centre line average, R_a

The surface roughness in the present study was measured using a profilometer. Both the centre line average R_a and the averaged roughness depth R_z were measured in the present study. But the R_z values measured in different directions showed a lot more variation than the R_a value which were consistently close (for a particular surface and surface finish). Therefore in the present study the centre line average R_a was used to characterize the heating surface micro-roughness.

The centre line average R_a has been extensively used in the literature to characterize the heating surface micro-roughness during boiling heat transfer studies by many investigators and are summarized by Benjamin [34]. Moreover, the centre line average R_a has been approved by the International Standards Organization (ISO) as a parameter to quantify the surface micro-roughness. The parameter R_a is defined by ISO 4287/1:1984 [35]. Therefore, the R_a value obtained from the profilometer measurements was used to characterize the surface micro-roughness in the present study.

4.5.2. The surface–liquid interaction parameter, γ

Ramilison et al. [36], while seeking to correlate the peak heat flux in terms of the surface micro-roughness and contact angle observed based on boiling data in the literature, that “smooth” (“rough”) and “wetted” (or “non-wetted”) surfaces can be discussed only in connection with a surface–liquid combination. A “smooth” surface for one liquid can serve as a “rough” pipe for another. This is analogous to a “rough” pipe being hydrodynamically “smooth”. This means that the profilometer data (R_a value) alone is not sufficient to characterize the effect of surface micro-roughness on the nucleation site density and the heat flux. The liquid and surface physical properties also play a significant role in determining the nucleation characteristics and the heat transfer rate. Therefore, in the present study, a surface liquid interaction parameter γ (the ratio of thermal conductivity, density and specific heat of the surface to those of the liquid) was used in conjunction with the

profilometer data (the R_a values for different surface finishes) in the correlation as,

$$\gamma = \sqrt{\frac{k_w \rho_w C_{p_w}}{k_l \rho_l C_{p_l}}} \quad (22)$$

The surface–liquid interaction parameter γ is the ratio of the product of the thermal diffusivity of the solid to the liquid and the heat content of the solid to the liquid. The parameter γ has been used by many investigators in the analysis of bubble growth during nucleate boiling. Further, Magrini and Nannei [37], while proposing a correlation for the boiling heat transfer coefficient, observed that a parameter, frequently referred to in the literature as thermal effusivity, $(k_w \rho_w C_{p_w})^{0.5}$ affected the heat transfer coefficient obtained. According to Kant and Weber [38], the temperature of the heating surface during bubble growth is a function of γ and γ indirectly affects the bubble growth time. Therefore in addition to the profilometer output, the surface–liquid interaction parameter γ was used in this study to correlate the experimental data.

4.5.3. The dimensionless surface roughness group, $R_a P / \sigma$

This dimensionless group is used because the centre line average R_a and the surface tension together take into account the effect of contact angle on the nucleation site density implicitly. There is some experimental contact angle data available in the literature [39]. However, the contact angle is specific to a surface material–surface finish–liquid combination. Changes in micro-roughness cause a lot of variation in the contact angle [40]. Further, the term ' $R_a P / \sigma$ ' comes from the force balance on a bubble. That is,

$$2\pi r \sigma = \pi r^2 (P_v - P_l) \quad (23)$$

Therefore,

$$P_v - P_l = 2\sigma / r \quad (24)$$

where r is the bubble radius. Therefore, in an analogous manner, " $R_a P / \sigma$ " was used to obtain a dimensionless surface roughness rather than the centre line average R_a or the contact angle alone in the present study.

4.5.4. The Prandtl number

$$Pr = \frac{C_p \mu}{k} \quad (25)$$

During bubble growth in nucleate boiling, there is convective flow of liquid away from the surface as liquid is being pushed away by growing vapor bubbles. Once the bubble departs from the heating surface, fresh liquid flows towards the heat transfer surface to take the place of departed vapor bubbles. This was envisaged as "source" and "wake" flows respectively by Zuber [28]. Therefore, although there is no bulk flow, the viscosity

of the liquid plays a significant role in pool boiling also. Han and Griffith [41] analyzed pool boiling using distilled water as the boiling liquid and also developed a "bulk convection" model. They concluded that the viscosity of the boiled liquid determines the contact angle and the bubble departure diameter. The Prandtl number of the liquid (which takes the viscosity into account) was used by Rohsenow [42] to develop his widely used correlation. He argued that although there is no net flow during pool boiling, the Prandtl number is important because the heat transfer (at a nucleation site) takes place directly from the wall of the liquid. The Prandtl number of the liquid was also used by Nishikawa and Fujita [43] to develop their bubble agitation model for pool boiling. Since the Prandtl number of the liquid is an important parameter which effects both for the nucleation site density and the heat flux in pool boiling, it is used as a correlating parameter.

4.6. Mixture boiling characteristics

Figs. 2 and 3 show the boiling curves for various compositions of the two ternary systems studied. In the boiling of a pure liquid, the liquid and vapor compositions are the same and hence the entire boiling process is heat transfer controlled. In contrast, when a multicomponent mixture is boiled, the vapor–liquid phase equilibrium characteristics of the mixture allow the vapor and liquid phases to be at different compositions. The composition is an important factor affecting the heat transfer process during the boiling of multicomponent mixtures. As the composition varies, a number of physical properties characterizing the mixture also change, such as the vapor pressure of the boiling mixture. When the concentration difference between the vapor and the liquid ($y - x$) is a maximum the relative volatility is a minimum.

Stephan and Körner [44] suggested that as ($y - x$) decreases, heat transfer to the bubble increases. They found the minimum heat transfer occurs at a maximum wall superheat corresponding to a maximum ($y - x$) of the more volatile component. Thome and Davey [45] investigated the influence of the liquid composition and ($y - x$) on bubble growth ratio of nitrogen–argon mixtures and found that the bubble growth rate varies linearly with ($y - x$) and that the bubble growth rates decrease with increase in ($y - x$). Alpay and Balkan [46] found that the decrease in boiling heat transfer rates observed with binary mixtures increases with increase in the heat flux and that the minimum heat flux is obtained at the maximum ($y - x$). These studies suggest that the heat transfer coefficient/heat flux is a function of ($y - x$) of the more volatile component and the minimum heat transfer coefficient/heat flux occurs near the composition at which ($y - x$) is a maximum in binary mixtures.

Heat transfer coefficient of ternary mixtures is a function of composition and heat flux for a given pressure and heating surface.

$$h = f(x_1, x_2, x_3, q) \tag{26}$$

In the present study, the concentrations of x_2 and x_3 were kept equal for all the concentrations of x_1 thus reducing the number of variables to two. Therefore two-dimensional representation of experimental results is used as shown in Figs. 11 and 12.

Fig. 11 shows the variation of heat transfer coefficient at different heat fluxes and the variation of difference between the equilibrium vapor and liquid concentrations for all the components with the concentration of acetone for acetone(1)–isopropanol(2)–water(3) ternary system. Fig. 12 shows similar results for the acetone(1)–MEK(2)–water(3) ternary system. It is seen from the figures that except for $x_1 = 0.1, x_2 = 0.45$ of acetone(1)–MEK(2)–water(3) ternary system where in two light components are present, all other compositions of the two ternary systems studied have only one light component (acetone). Therefore, in the present study, one can expect the heat transfer coefficient predominantly to be a function of $(y - x)$ of acetone. From Fig. 11, it can be seen that the $(y - x)$ of acetone increases, reaches a

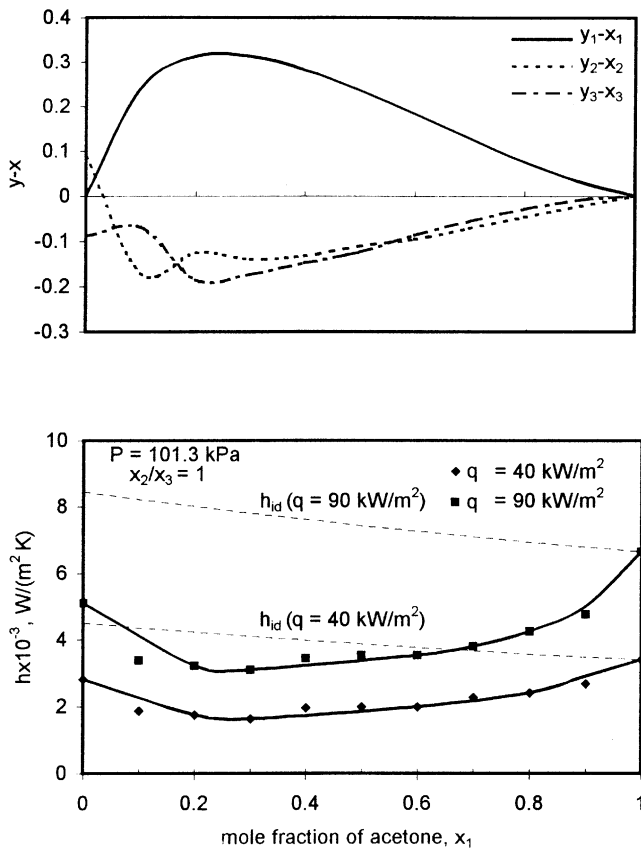


Fig. 11. Dependence of heat transfer coefficient on $(y - x)$; system: acetone(1)–isopropanol(2)–water(3).

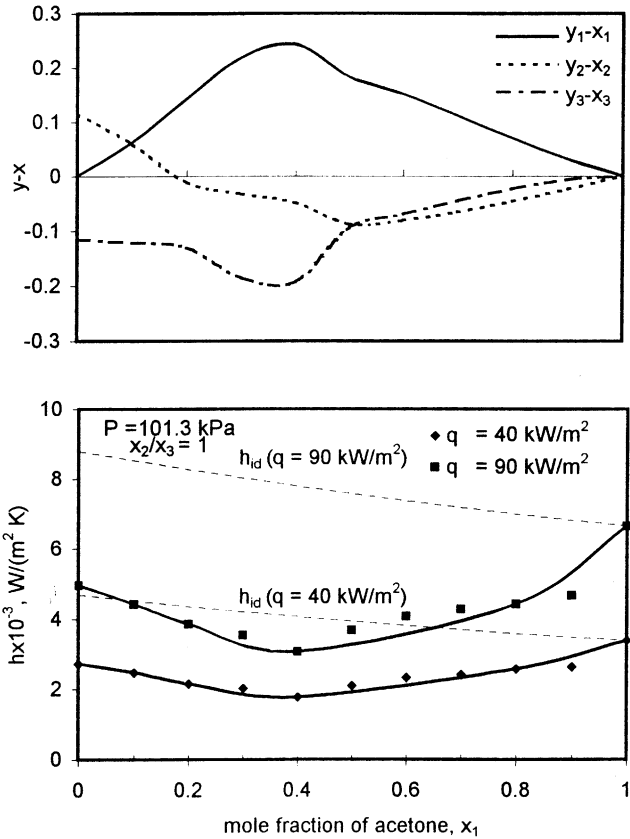


Fig. 12. Dependence of heat transfer coefficient on $(y - x)$; system: acetone(1)–MEK(2)–water(3).

maximum at $x_1 = 0.3, x_2 = 0.35$ and then decreases as the concentration of acetone increases in acetone(1)–isopropanol(2)–water(3) ternary system. Correspondingly it can be observed that heat transfer coefficient decreases, reaches a minimum at $x_1 = 0.3, x_2 = 0.35$ and then increases. Similar trend is observed for acetone(1)–MEK(2)–water(3) ternary system and is shown in Fig. 12. For this mixture the $(y - x)$ of acetone increases, reaches a maximum at $x_1 = 0.4, x_2 = 0.3$ and then decreases as the concentration of acetone increases. The heat transfer coefficient correspondingly decreases, reaches a minimum at $x_1 = 0.4, x_2 = 0.3$ and then increases. From Figs. 11 and 12, it can be seen that the heat transfer coefficient is a function of $(y - x)$ of acetone and the minimum in heat transfer coefficient occurs at the maximum $(y - x)$ of acetone. Therefore the decreasing–increasing trend of heat transfer coefficient with composition shown in Figs. 2 and 3 can be attributed to the variation in $(y - x)$ of acetone.

From Figs. 11 and 12, it can also be seen that the reduction in heat transfer coefficient increases with heat flux. A similar observation has been made by Fujita and Tsutsui [9] for ternary refrigerant mixtures. Gu and Bart [47] while analyzing binary ethanol–water data of Fujita and Tsutsui [48] observed that the effect of heat flux on heat transfer coefficient reduction was significant at low

heat fluxes ($q < 200 \text{ kW/m}^2$) but at high heat fluxes ($q > 200 \text{ kW/m}^2$) where fully developed nucleate boiling dominates, the heat flux was found to have no significant effect. They observed a similar effect with the data of Valent and Afgan [49]. According to Gu and Bart [47], the effect of heat flux is not significant under engineering conditions where developed boiling occurs. However, the present correlation does not account for the heat flux regime on heat transfer coefficient reduction. The correlation predicted satisfactorily, a wide range of data from the present study and from the literature. However, further work is recommended to examine the effect of heat flux regime (high/low) on mixture boiling.

5. Conclusions

Heat transfer coefficient data in nucleate pool boiling of acetone–isopropanol–water and acetone–MEK–water systems was obtained. The following conclusions can be made from an analysis of the experimental data.

(1) For a given heat flux, the heat transfer coefficients of mixtures are lower than the values obtained by the pure components constituting the mixture. This is due to utilization of part of the temperature driving force to overcome the mass transfer resistance caused by diffusion of the light component(s) to the bubble interface. Therefore, to obtain a given heat flux, an additional temperature driving force is required for multicomponent mixtures, hence the heat transfer coefficients are lower than those of constituent pure components.

(2) The experimental data was compared with existing correlations for pool boiling of multicomponent mixtures in the literature. The correlations either over-predicted or under-predicted the present experimental data. It was found that part of the uncertainty associated with the literature correlations in estimating the heat transfer coefficients for the present data was due to the non-applicability of the methods used to estimate the ideal heat transfer coefficient for the present surface–liquid combinations. Therefore, two different correlations have been used for ideal heat transfer coefficient and it was found that the performance of the correlations improved considerably.

(3) A correlation for pure components has been developed in the present study taking into account the surface–liquid interaction parameter and the surface roughness group. The correlation predicts the present experimental data and the literature data satisfactorily.

(4) The heat transfer coefficient correlation for multicomponent mixtures proposed in the present study predicts wide range of data obtained in the present study as well as from the literature satisfactorily. The correlation does not incorporate the heat flux regime on heat transfer coefficient reduction. It was found in the present

study and in the literature that the reduction of heat transfer coefficient increases with heat flux at low heat fluxes. However, at high heat fluxes, it was found in the literature that the heat flux has no significant effect. Hence, the present correlation may be effective for a wide range of mixtures as it was developed from a strong theoretical basis. However, this has to be further established with more experimental data.

(5) The reduction in temperature driving force in mixtures is due to the diffusion of the light component(s) to the bubble interface. The diffusion in multicomponent mixtures is different from binary mixtures. For multicomponent mixtures, main diffusion coefficients and cross-diffusion coefficients are necessary to characterize the system. Therefore, the effective temperature driving force was determined taking into account multicomponent diffusion coefficients and the vapor–liquid equilibrium data of the light component(s).

(6) It was found from the present study that the heat transfer coefficient is a function of the difference between the equilibrium vapor and liquid concentration, $(y - x)$ of the light component (acetone) and the minimum heat transfer coefficient occurs at the maximum of $(y - x)$ of the light component (acetone). This is in accordance with the binary boiling phenomena. However, for multicomponent mixtures, the driving force for mass transfer depends on all the light components present in the system and hence the $(y - x)$ of all the light components have to be considered to establish the dependence of heat transfer coefficient on $(y - x)$.

Appendix A

A.1. Palen and Small [10] correlation

This is one of the earliest correlations developed for binary mixtures with wide boiling ranges for reboilers of distillation columns

$$\frac{h}{h_{id}} = \exp[-0.027(T_{bo} - T_{bi})] \quad (\text{A.1})$$

Palen and Small [10] determined the ideal heat transfer coefficient using the McNelly [25] correlation proposed for pure liquids using average physical properties

$$h_{id} = 0.225 \left(\frac{k_l}{d_b} \right) \left(\frac{d_b q}{\lambda \mu_l} \right)^{0.69} \left(\frac{P d_b}{\sigma} \right)^{0.31} \left(\frac{\rho_l}{\rho_v} - 1 \right)^{0.33} \times \left(\frac{C_{pl} \mu_l}{k_l} \right)^{0.69} \quad (\text{A.2})$$

The term $(T_{bo} - T_{bi})$ is the temperature difference between the vapor leaving and liquid feed to a kettle reboiler and is equal to the boiling range, that is the temperature difference between the dew point and the bubble point at the liquid feed composition if all the feed is evaporated.

A.2. Schlünder [11] correlation

Based on the film theory of mass transfer, Schlünder [11] developed a semi-empirical correlation for nucleate boiling of multicomponent mixtures

$$\frac{h}{h_{id}} = \left\{ 1 + \frac{h_{id}}{q} \left[\sum_{i=1}^{n-1} (T_{sn} - T_{si})(y_i - x_i) \right] \times \left(1 - \exp \frac{-B_0 q}{\rho_1 \lambda \beta_{li}} \right) \right\}^{-1} \quad (\text{A.3})$$

Schlünder [11] used linear mixing laws to estimate the ideal heat transfer coefficient. He assumed that the dominant mass transfer resistance is in the liquid phase near the growing bubble and the vapor side mass transfer resistance can be neglected. He assumed that all the β_{li} , liquid side mass transfer coefficients, are equal and fixed a value of 0.0002 m/s. B_0 , the scaling factor, is equated to unity on the assumption that all the heat transfer from the heated surface in nucleate boiling passes into the bubble in the form of latent heat. He also assumed that vapor and liquid are in thermodynamic equilibrium at the interface and that the concentration difference of more volatile components between the liquid and the vapor at the interface is the same as that in the bulk.

A.3. Thome and Shakir [13] correlation

Thome and Shakir [13] analyzed the expression for the slope of the curve used by Schlünder [11] and concluded that the actual slope is predicted by the expression due to Schlünder [11] only at one composition. The authors then used the boiling range, which is the difference between the dew point and the bubble point at constant mole fraction to approximate the slope of the bubble point curve and obtained

$$\frac{h}{h_{id}} = \left\{ 1 + \frac{h_{id}}{q} \Delta T_{bp} \left[\left(1 - \exp \frac{-B_0 q}{\rho_1 \lambda \beta_{li}} \right) \right] \right\}^{-1} \quad (\text{A.4})$$

Thome and Shakir [13] used the Stephan and Abdelsalam [22] correlation for pure components with mixture properties to calculate the ideal heat transfer coefficient.

The Stephan and Abdelsalam [22] correlation used by them is,

$$h_{id} = 0.0546 \left(\frac{k_l}{d_b} \right) \left[\left(\frac{\rho_v}{\rho_l} \right)^{0.5} \left(\frac{q d_b}{k_l T_s} \right) \right]^{0.67} \times \left(\frac{\rho_l - \rho_v}{\rho_l} \right)^{-4.33} \left(\frac{\lambda d_b^2}{\alpha_l^2} \right)^{0.248} \quad (\text{A.5})$$

A.4. Bajorek and Lloyd [14] correlation

Bajorek and Lloyd [14] developed a mechanistic model accounting for the diffusion of the more volatile components to the interface of a growing bubble. According to them, the diffusion of multicomponent mixtures is different from binary mixtures in that cross-diffusion terms must be considered. They then incorporated cross-diffusion coefficients in their model and following Schlünder's [11] approach arrived at the following expression for a ternary mixture.

$$\frac{h}{h_{id}} = \left\{ 1 + \frac{h_{id}}{q} \left[\left(\frac{dT}{dx} \right) (y_1 - x_1) \left[1 - \phi_1 \frac{\exp(-\psi_1)}{\zeta_1 - \zeta_2} + \phi_2 \frac{\exp(-\psi_2)}{\zeta_1 - \zeta_2} \right] \right] \right\}^{-1} \quad (\text{A.6})$$

where,

$$\phi_1 = D_{22} - D_{12} \left(\frac{y_2 - x_2}{y_1 - x_1} \right) - \zeta_2 \quad (\text{A.7})$$

$$\phi_2 = D_{22} - D_{12} \left(\frac{y_2 - x_2}{y_1 - x_1} \right) - \zeta_1 \quad (\text{A.8})$$

the multicomponent transport terms are,

$$\psi_1 = \frac{k_1 q \zeta_1}{\rho_1 \lambda \det[D] h_{id} \sqrt{Le}} \quad (\text{A.9})$$

$$\psi_2 = \frac{k_1 q \zeta_2}{\rho_1 \lambda \det[D] h_{id} \sqrt{Le}} \quad (\text{A.10})$$

where ζ_1 and ζ_2 are the eigenvalues of the matrix of diffusion coefficients $[D]$. They suggested the slope (dT/dx) of the bubble point curve can be replaced with the boiling range. They used the Stephan and Abdelsalam [22] correlation (Eq. (A.5)) for pure components,

Table 3
Performance of multicomponent correlations in the literature

Correlation	Eq.	Mixtures	Average error (%)	MAE (%)	RMS error (%)	SD (%)
Palen and Small [10]	(A.1)	Acetone–isopropanol–water	45.5	45.5	46.6	10.1
		Acetone–MEK–water	42.2	42.2	45.7	17.7
Schlünder [11]	(A.3)	Acetone–isopropanol–water	45.4	45.4	47.2	13.1
		Acetone–MEK–water	41.4	41.4	42.6	10.0
Thome and Shakir [13]	(A.4)	Acetone–isopropanol–water	–24.8	24.8	27.9	12.8
		Acetone–MEK–water	–32.4	32.4	34.2	11.0
Bajorek and Lloyd [14]	(A.6)	Acetone–isopropanol–water	–24.3	24.3	27.2	12.4
		Acetone–MEK–water	–33.2	33.2	34.3	8.8

with mixture properties to calculate the ideal heat transfer coefficient.

The thermophysical and transport properties required to compare the correlations were calculated by selecting appropriate correlations and mixing rules from Reid et al. [23] and Holland et al. [24]. Non-random two liquid (NRTL) equation [23] which is known to estimate the activity coefficients of non-ideal multicomponent mixtures with good accuracy was used to generate the vapor–liquid equilibrium data. The selected correlations and mixing rules were then used in Aspen Plus to estimate the properties. The performance of the literature correlations is given in Table 3.

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