

NUCLEATE POOL BOILING HEAT TRANSFER OF MULTICOMPONENT MIXTURES

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Steady state pool boiling heat flux data has been obtained for acetone-isopropanol-water and acetone-MEK(methyl ethyl ketone)-water ternary mixtures. The data shows that to obtain a given heat flux, the wall superheat required is greater for mixtures than for the pure components constituting the mixture. The measured heat transfer coefficients were compared with predictions from literature correlations for multicomponent mixtures. In all the cases, overestimation or underestimation of the data was observed. Therefore, a new correlation has been proposed for the heat flux in terms of Archimedes number, Prandtl number, surface-liquid interaction parameter, modified Jakob number, dimensionless surface roughness group, properties-profile parameter and an effective temperature driving force. In general, the effective temperature driving force in binary mixtures is less than that encountered in pure components and is obtained by incorporating the binary diffusivity of the mixture. In multicomponent systems, the multicomponent diffusion coefficients have to be incorporated into the expression for the effective temperature driving force. The heat flux correlation predicts the present experimental data as well as literature data, satisfactorily. The heat flux was found to be a function of the difference between the equilibrium vapour and liquid concentration, $(y - x)$ of the light component(s) and the minimum heat flux occurs at the maximum of $(y - x)$ of the light component(s).

Keywords: pool boiling; multicomponent mixture; diffusion; vapour-liquid equilibrium.

INTRODUCTION

Boiling is a physical process of considerable practical significance. Nucleate pool boiling of liquid mixtures finds many applications in chemical, petrochemical, pharmaceutical, air-separation and refrigeration and air-conditioning industries. Nucleate pool boiling of multicomponent mixtures, when compared to pure liquids, is characterized by a reduction in the nucleation site density, the bubble departure diameter and the heat flux. This is due to (a) reduction in temperature driving force because of increase in the boiling point of the microlayer (the liquid layer trapped under a growing bubble) which is due to the preferential evaporation of the light components during the bubble growth; (b) the mass diffusion of the light components to the microlayer (caused by 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 multicomponent mixtures. However, most boiling research has been limited to the behaviour of pure components or binary mixtures. One of the earliest studies on ternary mixture

boiling on plain surfaces was by Grigor'ev *et al.* (1968) who performed an experimental investigation on pool boiling of acetone-methanol-water and acetone-ethanol-water ternary systems. Stephan and Preusser (1979) measured pool boiling heat transfer coefficients in acetone-methanol-water and methanol-ethanol-water ternary systems. Sardesai *et al.* (1986) measured pool boiling heat transfer coefficients in hydrocarbon mixtures with up to five components. Bajorek *et al.* (1989) reported experimental measurements of pool boiling heat transfer coefficients in six aqueous ternary systems. Kadhum *et al.* (1994) carried out an investigation on an acetone-isopropanol-water ternary system. Krupiczka *et al.* (1999) conducted an experimental study on methanol-isopropanol-water ternary system.

Unlike pool boiling of pure components and binary mixtures, for which many correlations are available in the literature, there are only a few correlations available in the literature to estimate the pool boiling heat transfer rates of multicomponent mixtures. These are from Palen and Small (1964), Stephan and Preusser (1979), Schlünder (1983), Thome and Shakir (1987), and Bajorek and Lloyd (1997). However, both the data and the correlations are limited and covered very few mixtures so that the correlations for multicomponent liquid mixtures have not been validated conclusively as yet. This formed the motivation for the present study. Many mixture boiling hypotheses, experimental studies and correlations have been summarized by Thome and Shock (1984), and Bajorek and Lloyd (1997).

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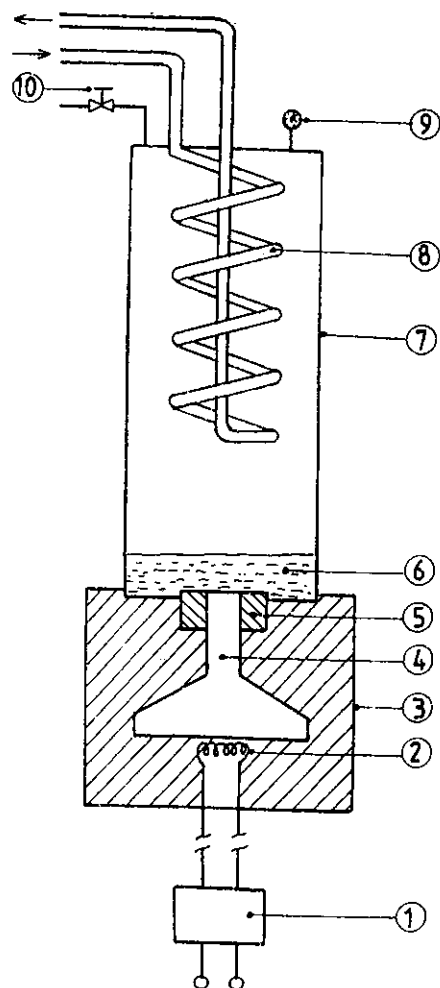
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EXPERIMENTAL

Experimental-Set up

The experimental set up (Figure 1) 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 26 cm and its inner diameter is 9.3 cm. 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 vapour 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



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. Pressure gauge
10. Relief valve

Figure 1. Schematic of the experimental set-up.

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. A provision was made to measure temperature at four points along the axis of the heating block by 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.

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 mole-fraction, and ternary mixtures covering the concentration range of acetone from 0.1–0.9 keeping the concentrations of the other two components equal. About 150 ml of liquid mixture was boiled in each run. The variables measured were the heat flux, the heating surface (wall) temperature and the liquid temperature. The heat transfer coefficient was calculated using an appropriate form of Newton's law of cooling. After a particular voltage was applied to 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 0.1K for about two minutes. Then the temperatures along the axis of the heating block were noted and using these, the surface temperature was evaluated by extrapolation. The heat flux was calculated using Fourier's law. Before taking the first reading of each run the liquid and the heating surface are heated for nearly two hours to ensure degassing and aging. For a given composition, boiling experiments were conducted at different excess temperatures (wall superheats). The boiled liquid was condensed back into the test section using the cooling coils. Therefore, the bulk liquid composition does not change during the course of the experiments. Prior to the start of each boiling experiment, the heating surface was polished with 4/0 grade emery paper and the centreline 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.

Experimental Uncertainty

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

RESULTS

The variation of heat flux with wall superheat for different compositions of the two ternary systems studied is shown in Figures 2 and 3. From the figures, it was observed that for a given heat flux the wall superheat required is greater for mixtures than for the constituent pure liquids or, for a given

wall superheat, the heat fluxes of mixtures are significantly less than the pure component values. It was also noticed from Figures 2 and 3 that the heat flux for a given wall superheat decreases and then increases with increase in the concentration of acetone. As can be seen from the figures, the heat flux at a given wall superheat decreases up to $x_1 = 0.3$, $x_2 = 0.35$ and then increases as the concentration of acetone increases in the acetone(1)–isopropanol(2)–water(3) system, whereas in the acetone(1)–MEK(2)–water(3) system the heat flux 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 flux or heat transfer coefficient with increase in the concentration of the more volatile component has been observed previously in the case of binary mixtures. Hui and Thome (1985) found that the heat transfer coefficient decreases and then increases with increase in ethanol concentration (for all sub coolings) for ethanol–water systems. Benjamin and Balakrishnan (1997) 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.

COMPARISON OF PRESENT EXPERIMENTAL DATA WITH EXISTING MULTICOMPONENT CORRELATIONS

One of the objectives of this investigation is to compare the present experimental data with the existing multicomponent pool boiling correlations in the literature. The correlations incorporate an ‘ideal’ heat transfer coefficient and a correction term for the presence of other liquids. The ideal heat transfer coefficient h_{id} , is the heat transfer coefficient the mixture would be expected to have without any mass transfer effects. The correlations chosen for comparison are those of Palen and Small (1964), Schlünder (1983), Thome and Shakir (1987), and Bajorek and Lloyd (1997).

Palen and Small (1964) Correlation

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

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

Palen and Small (1964) determined the ideal heat transfer coefficient using the McNelly (1953) correlation proposed for pure liquids using average physical properties

$$h_{id} = 0.225 \left(\frac{k_l}{d_b} \right) \left(\frac{d_b q}{\lambda_l \mu_l} \right)^{0.69} \left(\frac{P d_b}{\sigma} \right)^{0.31} \times \left(\frac{\rho_l - \rho_v}{\rho_v} \right)^{0.33} \left(\frac{C_{p,l} \mu_l}{k_l} \right)^{0.69} \quad (2)$$

The term $(T_{bo} - T_{bi})$ is the temperature difference between the vapour 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.

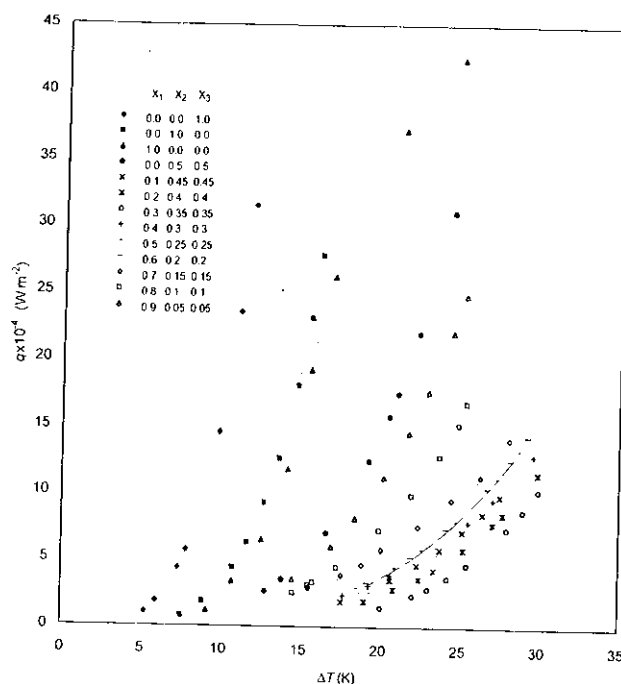


Figure 2. Nucleate boiling curves for acetone(1)–isopropanol(2)–water(3) system (x_1 = composition of acetone; x_2 = composition of isopropanol; x_3 = composition of water).

The Palen and Small (1964) correlation has been compared extensively with boiling data of binary mixtures on plain tubes. However, it has been claimed that the correlation can be extended to mixtures of three or more components. In the present study the thermophysical and transport properties required to compare the correlation were calculated by selecting appropriate correlations and mixing rules for the present ternary systems from Reid *et al.* (1988). The selected correlations and mixing rules were then used in Aspen Plus Steady State Simulation Package, Version 10.1-0, to estimate the properties. The physical properties of the mixtures are given in Tables 1 and 2. Figure 4 shows the comparison of the present experimental data with the predictions of the Palen and Small (1964) correlation. As can be seen from the figure, the agreement of the data with the correlation is poor with the predictions being higher by about 45% for the two systems.

Schlünder (1983) Correlation

Based on the film theory of mass transfer, Schlünder (1983) developed a semi-empirical correlation for nucleate boiling in multicomponent mixtures

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

Schlünder (1983) 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 vapour side mass transfer resistance

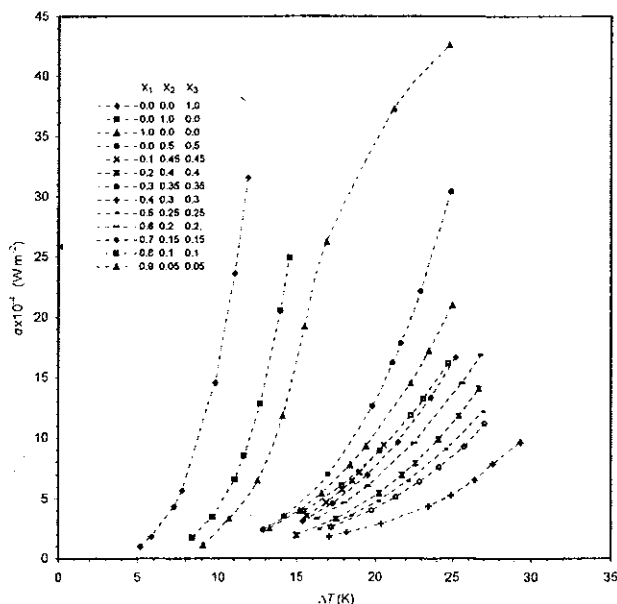


Figure 3. Nucleate boiling curves for acetone(1)-MEK(2)-water(3) system (x_1 = composition of acetone; x_2 = composition of MEK; x_3 = composition of water).

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^{-1} . 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 vapour and liquid are in thermodynamic equilibrium at the interface and that the concentration difference of more volatile components between the liquid and the vapour at the interface is the same as that in the bulk.

The physical properties required for comparison of the Schlünder (1983) correlation with the present data were estimated as mentioned earlier. The Non-Random Two Liquid (NRTL) equation (Reid *et al.*, 1988) which is known to estimate the activity coefficients of non-ideal multicomponent mixtures with good accuracy was used to generate the vapour-liquid equilibrium data. The comparison of the present experimental data with the predictions of the Schlünder (1983) correlation is shown in Figure 5. Here again, the agreement is poor with the predicted values being higher, by about 40%, for the two systems studied.

Thome and Shakir (1987) Correlation

Thome and Shakir (1987) analysed the expression for the slope of the curve used by Schlünder (1983) and concluded that the actual slope is predicted by the expression from Schlünder (1983) 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:

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

Thome and Shakir (1987) used the Stephan and Abdelsalam (1980) correlation for pure components with mixture properties to calculate the ideal heat transfer coefficient. The Stephan and Abdelsalam (1980) correlation used by them is,

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

Figure 6 shows the comparison of the present experimental data with those predicted by the Thome and Shakir (1987) correlation. The physical properties required for comparison of the Thome and Shakir (1987) correlation with the present data were estimated as discussed earlier. Once again, poor agreement was observed with the predicted values being lower this time; by 25% for the acetone(1)-isopropanol(2)-water(3) system and 32% for the acetone(1)-MEK(2)-water(3) system.

Bajorek and Lloyd (1997) Correlation

Bajorek and Lloyd (1997) 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 (1983) approach arrived at the following expression for a ternary mixture:

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

where,

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

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

the multicomponent transport terms are,

$$\psi_1 = \frac{k_l q \zeta_1}{\rho_l \lambda \det[D] h_{id} \sqrt{Le}} \quad (9)$$

$$\psi_2 = \frac{k_l q \zeta_2}{\rho_l \lambda \det[D] h_{id} \sqrt{Le}} \quad (10)$$

where ζ_1 and ζ_2 are the eigen values 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 (1980) correlation for pure components (equation 5) with mixture properties to calculate the ideal heat transfer coefficient.

The comparison of the present experimental results with the predictions of the Bajorek and Lloyd (1997) correlation is shown in Figure 7. The multicomponent diffusion coefficients were estimated using a method from Taylor and Krishna (1993) discussed in more detail later. The physical properties were estimated as before. However, the predic-

Table 1. Thermophysical and transport properties of acetone(1)-isopropanol(2)-water(3) ternary system.

Mole fraction	T_s, K	$\rho_l, kg/m^3$	$\rho_v, kg/m^3$	$\lambda \times 10^{-3}, J/kg$	$C_{pl}, J/(kg K)$	$\mu_l \times 10^3, Ns/m^2$	$k_l, W/(m K)$	$\sigma \times 10^3, N/m$	$\alpha_l \times 10^6, m^2/s$	$\bar{D} \times 10^9, m^2/s^2$	$\sum (v-x) > 0$
$x_1 = 0.1, x_2 = 0.45$	349.7	766.6	1.43	962.6	3776	0.374	0.1616	37.5	5.58	3.15	0.2403
$x_1 = 0.2, x_2 = 0.4$	343.1	767.7	1.52	903	3665	0.369	0.1592	36.1	5.66	3.05	0.3116
$x_1 = 0.3, x_2 = 0.35$	340.8	763.9	1.60	842.5	3537	0.354	0.1564	34.1	5.79	3.14	0.3125
$x_1 = 0.4, x_2 = 0.3$	338.8	760.6	1.68	787.8	3378	0.338	0.1541	32.0	6.00	3.32	0.2815
$x_1 = 0.5, x_2 = 0.25$	335.6	759.4	1.76	739.5	3191	0.324	0.1528	30.1	6.31	3.59	0.2351
$x_1 = 0.6, x_2 = 0.2$	333.8	756.9	1.84	691.8	2996	0.307	0.1513	28.0	6.67	4.07	0.1818
$x_1 = 0.7, x_2 = 0.15$	332.2	754.7	1.92	645.8	2799	0.289	0.1501	25.9	7.11	4.75	0.1270
$x_1 = 0.8, x_2 = 0.1$	330.8	752.6	2.00	600.6	2613	0.271	0.1491	23.7	7.58	5.69	0.0749
$x_1 = 0.9, x_2 = 0.05$	329.9	750.3	2.07	555.2	2455	0.252	0.1481	21.5	8.04	6.98	0.0304

Table 2. Thermophysical and transport properties of acetone(1)-MEK(2)-water(3) ternary system.

Mole fraction	T_s, K	$\rho_l, kg/m^3$	$\rho_v, kg/m^3$	$\lambda \times 10^{-3}, J/kg$	$C_{pl}, J/(kg K)$	$\mu_l \times 10^3, Ns/m^2$	$k_l, W/(m K)$	$\sigma \times 10^3, N/m$	$\alpha_l \times 10^6, m^2/s$	$\bar{D} \times 10^9, m^2/s^2$	$\sum (v-x) > 0$
$x_1 = 0.1, x_2 = 0.45$	344.5	776.2	1.64	727.7	3265	0.362	0.1661	38.9	6.55	2.39	0.1212
$x_1 = 0.2, x_2 = 0.4$	342.4	771.8	1.69	678.9	3351	0.349	0.1625	36.8	6.28	3.05	0.1422
$x_1 = 0.3, x_2 = 0.35$	339.3	769.2	1.76	647.1	3293	0.337	0.1598	34.8	6.48	2.75	0.2186
$x_1 = 0.4, x_2 = 0.3$	337.3	765.8	1.81	622.2	3169	0.324	0.1572	32.7	6.74	2.75	0.2421
$x_1 = 0.5, x_2 = 0.25$	335.8	762.1	1.87	601.5	3014	0.309	0.1549	30.5	6.74	4.00	0.1822
$x_1 = 0.6, x_2 = 0.2$	334.5	758.7	1.92	583.1	2849	0.294	0.1528	28.3	7.07	4.52	0.1505
$x_1 = 0.7, x_2 = 0.15$	332.7	756.2	1.98	566.2	2686	0.279	0.1513	26.1	7.45	5.11	0.1111
$x_1 = 0.8, x_2 = 0.1$	331.5	753.1	2.04	548.3	2540	0.264	0.1497	23.8	7.82	5.94	0.0696
$x_1 = 0.9, x_2 = 0.05$	329.9	751.0	2.09	530.0	2419	0.249	0.1485	21.6	8.18	6.97	0.0308

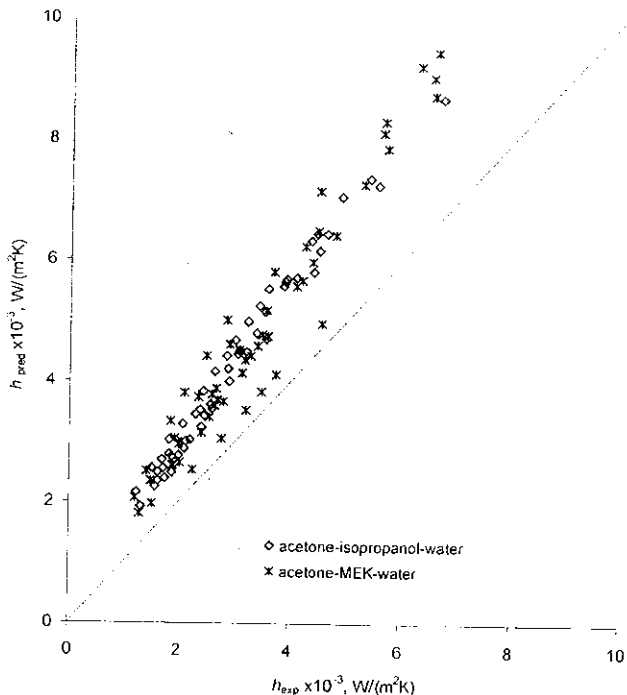


Figure 4. Comparison of present experimental data with Palen and Small (1964) correlation (h_{ii} based on McNelly (1953) correlation).

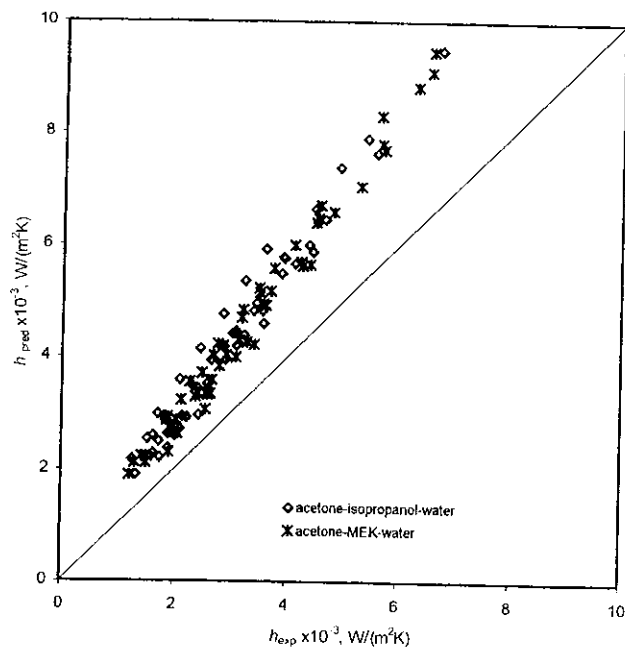


Figure 5. Comparison of present experimental data with Schlünder (1983) correlation (h_{ii} based on linear mixing law).

tions are lower than the experimental data by 24% for the acetone(1)–isopropanol(2)–water(3) ternary system and 33% for the acetone(1)–MEK(2)–water(3) ternary system.

CORRELATION FOR HEAT FLUX

The models in the previous section compared with the present experimental data show systematic overprediction or underprediction. Most of these models and the correlations to calculate ideal heat transfer coefficient 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. The models do not take into account the effect of surface micro-roughness in mixture boiling and the surface–liquid interactions. Furthermore, as Bajorek and Lloyd (1997) have said, the methods for calculating the ideal heat transfer coefficient implicitly assume that the effect of the surface on mixture boiling is the same as it is in the boiling of pure components. That is, if there is a significant effect of the mixture composition on activation and/or deactivation of nucleation sites, it is accounted for only as well as can be predicted in computing the ideal heat transfer coefficient. The models, except for that of Bajorek and Lloyd (1997) do not consider the multicomponent diffusion coefficients. These facts indicate that additional parameters are needed to correlate data of multicomponent mixture boiling phenomena.

Benjamin and Balakrishnan (1999) developed a model for heat flux for binary mixtures. They related the effective and superficial temperature driving forces as,

$$\frac{\Delta T_{eff}}{\Delta T} = \left[1 - \left(|y - x| \sqrt{\frac{D_{AB}}{z_{mix}}} \right)^{0.5} \right] \quad (11)$$

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 vapour and liquid concentration, $(y-x)$, is greater than zero and for the heavy component is less than zero. In the above expression, 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 $(D_{AB}/\alpha_{mix})^{0.5}$ 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 a ternary mixture, it is possible for there to be two light components and one heavy component or one light and two heavy components. Therefore, the reduction in the temperature driving force depends on the diffusion of the light components to the bubble interface in order to maintain equilibrium. Unlike binary mixtures where a single diffusivity $D_{AB} = D_{BA}$ 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 term, 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 (1993) as follows:

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

$$[D] = [B]^{-1}[\Gamma] \quad (12)$$

The elements of the thermodynamic correction factor matrix $[\Gamma]$ were calculated using the NRTL model (Taylor and Krishna, 1993) for multicomponent mixtures.

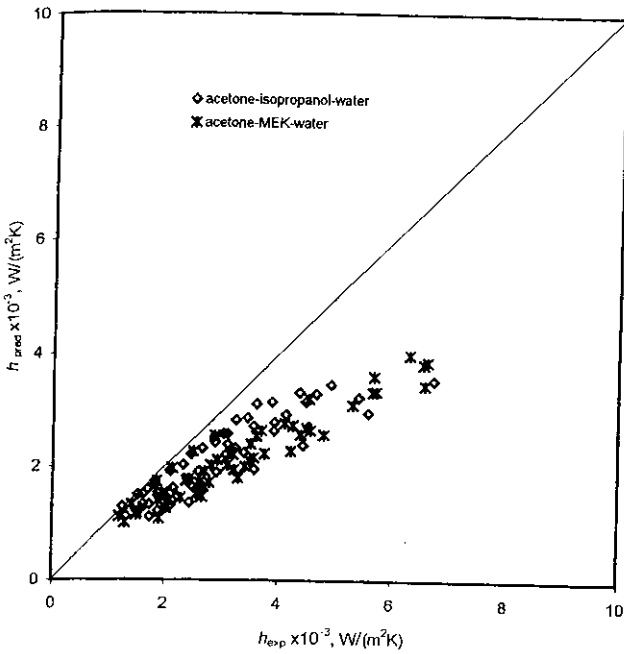


Figure 6. Comparison of present experimental data with Thome and Shakir (1987) correlation (h_{id} based on Stephan and Abdelsalam (1980) correlation).

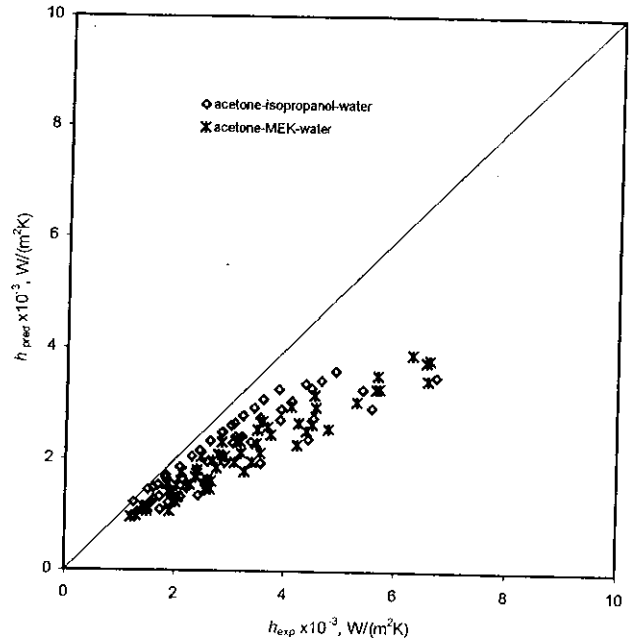


Figure 7. Comparison of present experimental data with Bajorek and Lloyd (1997) correlation (h_{id} based on Stephan and Abdelsalam (1980) correlation).

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

$$B_{ii} = \frac{x_i}{D_m} + \sum_{\substack{k=1 \\ k \neq i}}^n \frac{x_k}{D_{ik}} \quad (13)$$

$$B_{ij} = -x_i \left(\frac{1}{D_{ij}} - \frac{1}{D_m} \right) \quad (14)$$

The Maxwell-Stefan diffusivities (Taylor and Krishna, 1993) for each binary pair in a multicomponent mixture were estimated using,

$$D_{ij} = (D_{ij}^0)^{(1+x_i-x_j)/2} (D_{ji}^0)^{(1+x_j-x_i)/2} \quad (15)$$

where the infinite dilution diffusivities D_{ij}^0 were calculated using the correlation given by Wilke and Chang (1955) (Taylor and Krishna, 1993).

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

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 (equation 11) was modified and extended to ternary mixtures. Thus, the modified temperature driving force due to diffusion of light components in a ternary mixture may be written as,

$$\frac{\Delta T_{eff}}{\Delta T} = \left[1 - \sum_{i=1}^2 \left((y_i - x_i) \sqrt{\frac{\bar{D}}{\alpha_{mix}}} \right)^{0.5} \right] \quad (17)$$

where \bar{D} is the determinant of Fick matrix of diffusion coefficients.

Using the effective temperature driving force and other parameters involved in multicomponent mixture boiling, the heat flux data obtained at various wall superheats at different compositions of the two ternary mixtures, acetone(1)-isopropanol(2)-water(3) and acetone(1)-MEK(2)-water(3) were correlated

$$q = 1.34 \times 10^{-5} (Ar_{mix})^{-0.95} (Pr_{mix})^{-0.9} (\gamma_{mix})^{1.3} \times (Ja_m)^{0.85} \left(\frac{R_a P}{\sigma_{mix}} \right)^{0.73} (Bo_{mix}^*)^{-1.36} (\Delta T_{eff})^2 \quad (18)$$

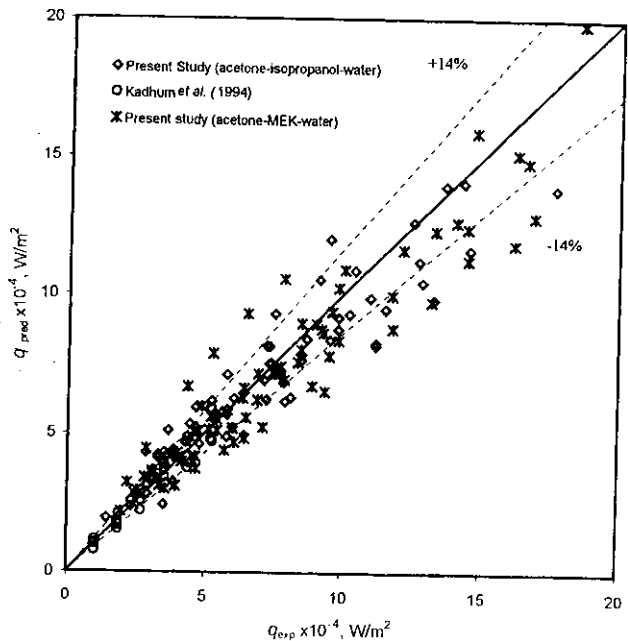


Figure 8. Performance of the heat flux correlation proposed for ternary mixtures.

The dimensionless groups used and their physical significance have been summarized earlier by Benjamin (1996). The mixture physical properties (Tables 1 and 2) were used to estimate the dimensionless groups. The performance of the correlation is shown in Figure 8. The correlation was further validated with the boiling data of Kadhum *et al.* (1994) obtained 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 of average was measured using a Perth-o-meter. The R_a value obtained was $0.256 \mu\text{m}$ which was then used in the boiling heat flux correlation (equation 18). This is also shown in Figure 8 and the data (both present and Kadhum *et al.* (1994)) fit the correlation with a mean absolute error of 14% and an average error of +0.18%.

The range of parameters used in the correlation are:

$$386597 < Ar_{mix} < 590292$$

$$4.05 < Pr_{mix} < 8.74$$

$$17.1 < \gamma_{mix} < 46.3$$

$$11.5 < Ja_m < 48.0$$

$$0.44 < \left(\frac{R_a P}{\sigma_{mix}} \right) < 2.45$$

$$9.6 \times 10^{-9} < Bo_{mix}^* < 9.25 \times 10^{-8}$$

$$6.0 < \Delta T_{eff} < 24.2$$

Uncertainty Analysis

The relative uncertainties in using the proposed correlation can be estimated using the method due to Moffat (1988). 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 heatflux is $\pm 2\%$ and the uncertainty in heat transfer coefficient is $\pm 2.4\%$. The uncertainty in estimating Ja_m is $\pm 1.4\%$. The uncertainty in the measurement of R_a is $\pm 1\%$. Therefore, the uncertainty in estimating Bo_{mix}^* is $\pm 2\%$ and the uncertainty in $(R_a P / \sigma_{mix})$ is $\pm 1\%$. These errors were obtained by applying propagation of errors rule to the respective expressions. Therefore, the relative uncertainty in the proposed heat flux correlation is 4.14%.

DISCUSSION

Mixture Boiling Characteristics

Figures 2 and 3 show the boiling curves for various compositions of the two ternary systems studied. The physical process of boiling of multicomponent mixtures is significantly different from that of boiling a pure liquid. During the boiling of a liquid mixture, due to the vapour-liquid phase equilibrium characteristics, the vapour phase has a composition different from that of the liquid phase. Hence, as a liquid mixture evaporates on the heating surface, the vapour contains more of the more volatile components. Therefore, the microlayer is depleted of the more volatile components and this causes diffusion of

the more volatile components from the bulk liquid towards the microlayer. Since mass diffusion is much slower than heat diffusion, bubble growth in mixtures is mass transfer controlled and a part of the driving force is utilized in overcoming this mass transfer resistance and there is a reduction in temperature driving force. Therefore, in order to achieve a given heat flux in a multicomponent mixture, the wall superheat required is greater than that for the constituent pure liquids or for a given wall superheat the heat fluxes for mixtures are less than for the pure component liquids constituting the mixture.

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 vapour pressure of the boiling mixture. When the concentration difference between the vapour and the liquid ($y-x$) is a maximum, the relative volatility is a minimum.

Stephan and Körner (1969) 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 (1981) 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 (1989) 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 flux or heat transfer coefficient is a function of ($y-x$) of the more volatile component and the minimum heat transfer coefficient or heat flux occurs near the composition at which ($y-x$) is a maximum in binary mixtures.

Heat flux of ternary mixtures is a function of composition and wall superheat for a given pressure and heating surface.

$$q = f(x_1, x_2, x_3, \Delta T) \quad (19)$$

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 Figures 9 and 10.

Figure 9 shows the variation of heat flux at different wall superheats and the variation of difference between the equilibrium vapour and liquid concentrations for all the components with the concentration of acetone for the acetone(1)-isopropanol(2)-water(3) ternary system. Figure 10 shows similar results for the acetone(1)-MEK(2)-water(3) ternary system. The solid curves are the predictions from equation 18 and are found to reproduce the experimental heat fluxes well over the whole composition range of acetone. It is seen from the figures that except for $x_1 = 0.1$, $x_2 = 0.45$ of the acetone(1)-MEK(2)-water(3) ternary system wherein 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 flux predominantly to be a function of ($y-x$) of acetone. From Figure 9, it can be seen that the ($y-x$) of acetone increases, reaches a maximum at $x_1 = 0.3$, $x_2 = 0.35$ and then decreases as the concentration of acetone increases in the acetone(1)-isopropanol(2)-

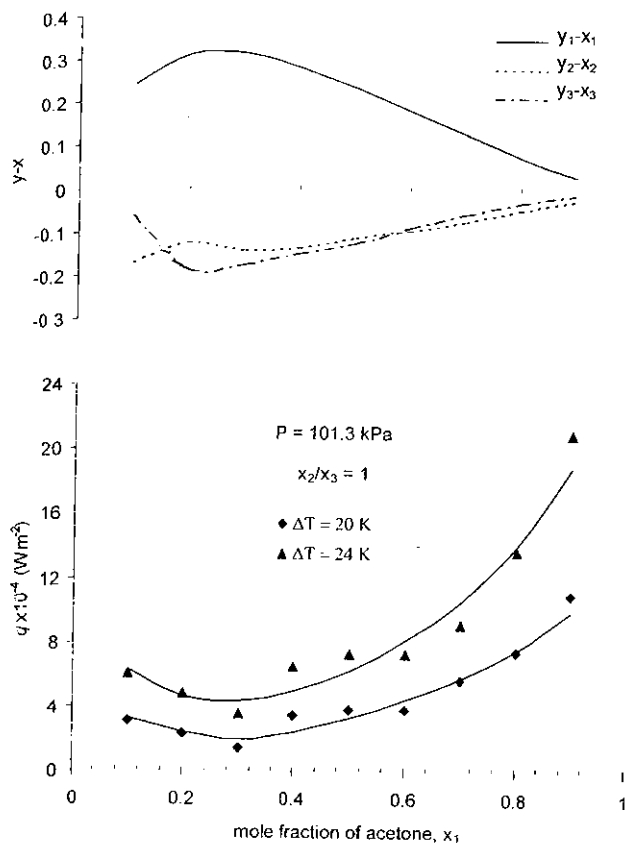


Figure 9. Dependence of heat flux on $(y-x)$; system: acetone(1)–isopropanol(2)–water(3).

water(3) ternary system. Correspondingly it can be observed that heat flux decreases, reaches a minimum at $x_1 = 0.3$, $x_2 = 0.35$ and then increases. A similar trend is observed for the acetone(1)–MEK(2)–water(3) ternary system and is shown in Figure 10. 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 flux correspondingly decreases, reaches a minimum at $x_1 = 0.4$, $x_2 = 0.3$ and then increases. From Figures 9 and 10, it can be seen that in the present study, heat flux is a function of $(y-x)$ of acetone and the minimum in heat flux occurs at the maximum $(y-x)$ of acetone. Therefore the decreasing–increasing trend of heat flux with composition shown in Figures 2 and 3 can be attributed to the variation in $(y-x)$ of acetone.

CONCLUSIONS

Heat flux 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) The wall superheat required to achieve a given heat flux is greater for mixtures than for the constituent pure components. 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)

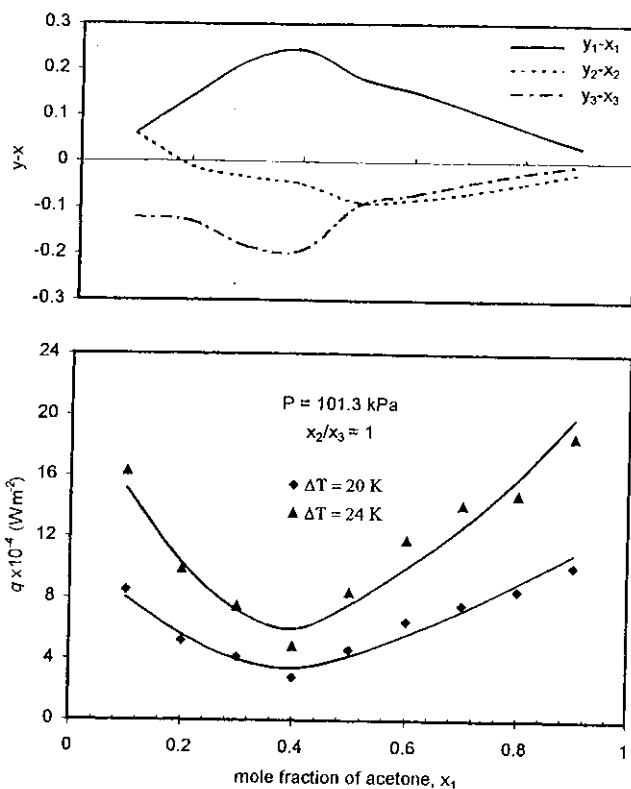


Figure 10. Dependence of heat flux on $(y-x)$; system: acetone(1)–MEK(2)–water(3).

to the bubble interface. Therefore, to obtain a given heat flux, additional temperature driving force is required for multicomponent mixtures.

- (2) The experimental data were compared with the existing correlations for pool boiling of multicomponent mixtures in the literature. The correlations either over-predicted or under-predicted the present experimental data. This indicates that additional parameters are needed to correlate multicomponent boiling data. Therefore, the heat flux data obtained in the present study were correlated in terms of the Archimedes number, Prandtl number, surface–liquid interaction parameter, modified Jakob number, dimensionless surface roughness group, properties–profile parameter and an effective temperature driving force. The heat flux correlation proposed predicts the present experimental data and the data from the literature satisfactorily.
- (3) 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 vapour–liquid equilibrium data of the light component(s).
- (4) It was found from the present study that the heat flux is a function of the difference between the equilibrium

vapour and liquid concentration, $(y-x)$ of the light component (acetone) and the minimum heat flux 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 flux on $(y-x)$.

NOMENCLATURE

Ar	Archimedes number, $(g/v_l^2)(\sigma/\rho_l g)^{1.5}$
Bo_{mix}^*	properties-profile parameter, $(\rho_l R_g^2 g/\sigma)$
B	inverse of binary diffusion coefficient, $s\ m^{-2}$
B_0	coefficient in Schlünder correlation
C_p	specific heat, $J/(kg\ K)$
d	diameter, m
dT/dx	slope of the boiling curve
D	mass diffusivity, $m^2\ s^{-1}$
\bar{D}	determinant of matrix of Fickian diffusion coefficients, $m^4\ s^{-2}$
ΔT	wall superheat, $T_w - T_s$, K
g	gravitational constant, $m\ s^{-2}$
h	heat transfer coefficient, $W/(m^2\ K)$
Ja_m	modified Jakob number, $(\rho_l C_p \Delta T_{eff}/\rho_l \lambda)$
k	thermal conductivity, $W/(m\ K)$
Le	Lewis number, α/D
M	molecular weight, $kg\ mol^{-1}$
P	pressure, $N\ m^{-2}$
Pr	Prandtl number, $(C_p \mu/k)$
q	heat flux, $W\ m^{-2}$
R_w	centre line average, μm
T	temperature, K
V	molar volume at normal boiling point, $m^3\ mol^{-1}$
x	liquid mole fraction
y	vapour mole fraction

Greek symbols

α	thermal diffusivity, $m^2\ s^{-1}$
β	mass transfer coefficient, $m\ s^{-1}$
γ	surface-liquid interaction parameter, $\sqrt{(k_w \rho_w C_{p,w})/(k_l \rho_l C_{p,l})}$
λ	latent heat of vapourisation, $J\ kg^{-1}$
μ	viscosity, $N\ s\ m^{-2}$
ν	kinematic viscosity, $m^2\ s^{-1}$
ρ	density, $kg\ m^{-3}$
σ	surface tension, $N\ m^{-1}$

Subscripts and superscripts

b	bubble
bp	boiling range
eff	effective
i, j	components
id	ideal
l	liquid
mix	mixture
n	number of components
s	saturation
v	vapour
w	wall
1, 2, 3	more, intermediate and less volatile components

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