

A Review on Nucleate Pool Boiling Heat Transfer of Binary Mixtures

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Abstract: NPB of many binary mixtures has been studied earlier like methanol-water, ethanol-water, methanol-benzene, acetone-water and isopropanol-water etc. This area of research has been on the rise in the last few years due to its various applications like separation of mixture, chemical engineering and refrigeration etc. This process is used in various industries to increase the energy efficiency which leads to reduction in environmental pollution. Pool boiling of binary mixtures is more complex than boiling of an individual liquid component since it involves mass transfer along with heat transfer. The boiling of binary mixture is immanently complex due to non-uniqueness of boiling point, limited mass transfer and strong variation of mixture properties. The HTC of a binary mixture is known to deteriorate as compared to its individual liquid components. Many co-relations have been developed to determine the HTC but they are only applicable for their own experiment and fail to predict the experimental data of other investigations. This paper aims to provide a brief review of boiling heat transfer of binary mixtures of fluids.

Key words: NPB – nucleate pool boiling, binary mixtures, HTC – heat transfer coefficient, CHF – critical heat flux, VLE – vapour liquid equilibrium, environmental pollution.

Introduction

NPB is characterized by boiling of a liquid or a mixture of liquids by a heated surface with the liquid or mixture being static. There are various applications of NPB which includes separation of mixture in industries (such as separation of hydrogen by boiling ethanol-water), chemical engineering, refrigeration, and many other industries. Nucleate pool boiling is an effective mode of heat transfer which helps to reduce the use of fossil fuels and thus reduce pollution. The boiling of binary mixture is quite different than that of a pure liquid because of addition of mass transfer along with the heat transfer. The boiling HTC of a binary mixture is generally lower

than the ideal HTC of an ideal fluid. There are two main reasons for this. The mass transfer process causes a dip in the effective wall superheat and thus the calculated HTC using the observed wall superheat is lower than the ideal HTC. The other reasons are that the physical properties of a binary mixture changes non-linearly, so the calculated HTC is lower than the ideal HTC. Many co-relations have been developed in literature to predict the HTC of a binary mixture during a nucleate pool boiling. But the predicted values are found to differ than the observed values. The relations are only limited for low heat flux. Additionally, some co-relations require some adjusting parameters for predicting the boiling HTC which may not be possible for some cases.

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Critical Heat Flux (CHF)

CHF is defined as the maximum heat flux which is attainable by a given system at a given pressure. The accurate prediction and enhancement of CHF is necessary to design a boiling arrangement with high efficiency and safety features. The CHF is dependent on various parameters such as surface roughness, bubble initiation time, bubble growth rate etc.

The heat flux of a nucleate pool boiling system is dependent on three heat transfer mechanisms:

1. Heat flux of evaporation to produce and develop the bubbles (q''_e).
2. Heat flux of quenching produced after bubble departure due to the thermal boundary layer surrounding it (q''_q).
3. Heat flux of convection transferred to the liquid phase outside the zone of influence of the bubbles (q''_c).

$$q''_{NB} = q''_e + q''_q + q''_c \quad (1)$$

Heat Transfer Coefficient (HTC)

Convection is a heat transfer process, which is characterized by the motion of fluid molecules with respect to a heated surface. The rate of heat transfer in a convection is directly dependent on the heat transfer coefficient of the fluid. The HTC of a system can be determined from the following expression:

$$Q = hA\Delta T \quad (2)$$

where Q = Total heat transfer, h = Heat transfer coefficient, A = Surface area of the heating surface and $\Delta T = (\Delta T_s - \Delta T_f)$, difference between surface temperature and fluid temperature.

Many investigators have proposed various correlations to calculate the HTC of a binary system. They have related the HTC with many thermo-physical properties. Some of them are discussed in Figure 2.

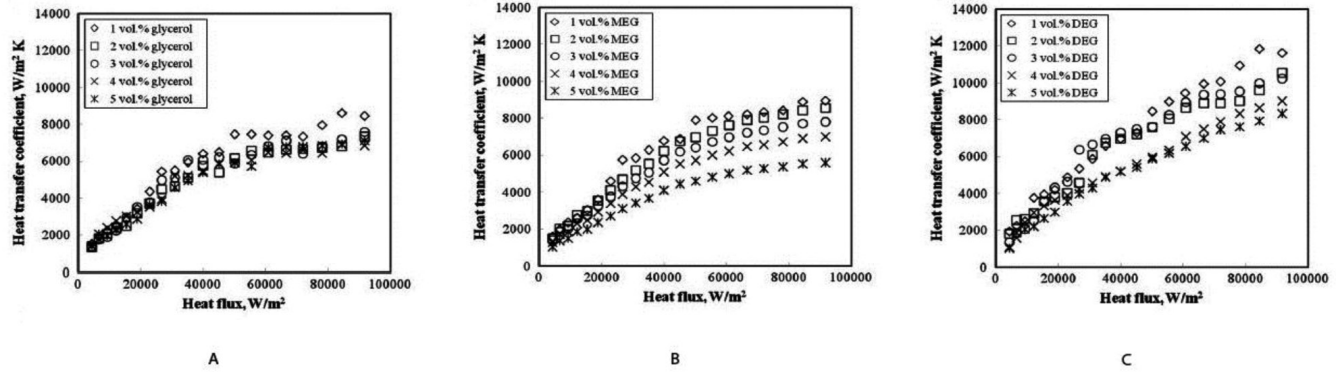


Figure 1: Heat transfer coefficient varying with the heat flux. (A) Glycerol/water, (B) ethylene glycol/water and (C) di-ethylene glycol/water.

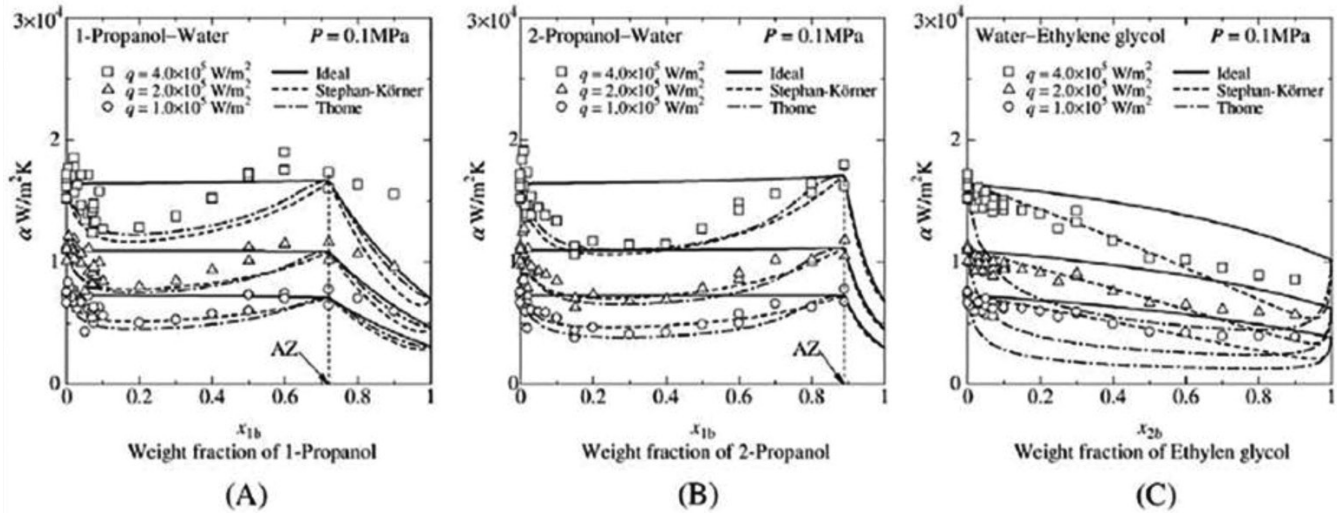


Figure 2: Heat transfer coefficient as a function of weight fraction: (A) 1-propanol/water (B) 2-propanol/water and (C) ethylene glycol/water.

Nomenclature

A	area, m^2	Subscript	
B_0	boiling number	0	reference
D	diffusivity constant, $m^2 s^{-1}$	1 or 2	component number
f	fugacity	bp	boiling point range
g	gravity acceleration, $m s^{-2}$	id	ideal
H_{fg}	heat of vapourization, $J kg^{-1}$	l	liquid
k	thermal conductivity, $W m^{-1} s^{-1}$	s	liquid-vapour interface
K, K_0, K_p	constants	sat	saturation
N	nucleation site	v	vapour
P_r	Prandtl number	Greek Symbols	
p	pressure, atm	α	boiling heat transfer coefficient, $W m^2 K^{-1}$
q	heat, W		
ΔT	temperature difference	$\hat{\alpha}$	thermal diffusivity, $m^2 s^{-1}$
x	liquid mole fraction	β	mass transfer coefficient, $m^2 s^{-1}$ or contact angle, rad
y	vapour mole fraction		
		σ	surface tension, $N m^{-1}$
		ρ	mass density, $kg m^{-3}$
		ϕ	association parameter

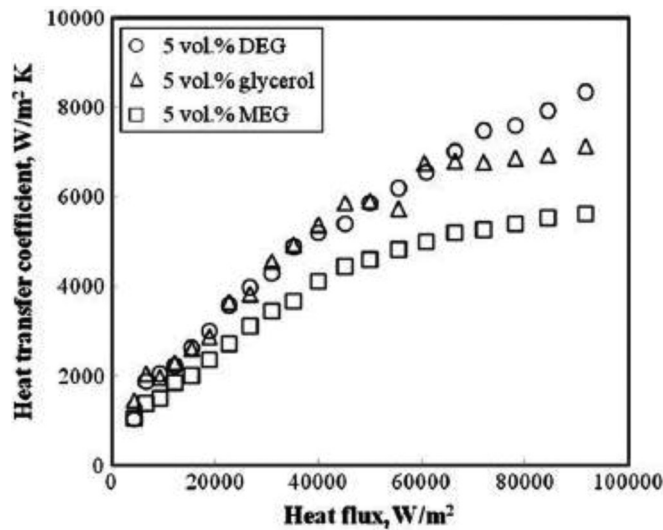


Figure 3: Comparison of heat transfer coefficients of three binary mixtures (diethylene glycol, glycerol, and ethylene glycol) operating at similar conditions.

Literature Review

Heat Transfer in Pure Liquids due to Boiling

Many co-relations have been created to predict the heat transfer due to boiling in pure liquids. Many people have done a detailed research on boiling of pure liquids

like Vinayak and Balakrishnan (2004), Alavi Fazel et al. (2008) along with Stephan and Abdelsalm (1980), Gorenflo (1993) and McNelly (1953). It is necessary to calculate the HTC of a pure liquid to predict the transfer rate of the binary mixture.

Many experiments have been conducted to predict the heat transfer coefficient of pure liquids in sub-atmospheric pressures. The characteristics of pool boiling in subatomic pressures is quite different than at normal atmospheric pressures. Raben et al. (1965), Cole and Shulman (1967), Van Stralen (1956) and Stralen et al. (1975) studied nucleate pool boiling of water at sub-atmospheric pressures. Boiling water at sub-atmospheric pressures which resulted in low-frequency bubble departure, creates surface temperature gradients and surface temperature oscillations. Gangto et al. (2019) observed that macroscale enhancement like porous mesh and porous foam enhances the HTC as well as CHF. The microstructure enhancement like micro-fins, micro-channels increases the HTC but enhancement of CHF varies.

Boiling Heat Transfer in Binary Mixtures

Many co-relations have been made to find the correlation between the heat transfer coefficients of binary mixture

to that of the pure liquid components. Stephan and Körner (1969) proposed an equation which showed the importance of a factor in the reduction of heat transfer in binary mixture. They suggested that the wall superheat for any binary mixture can be calculated by the addition of wall superheat and excess superheat. Many investigators later provided the equation and values of the empirical constant after altering it for specific binary arrangements. Investigator like Jungnickel et al. (1979) modified the Stephan and Körner equation by adding the heat flux multiplier. They predicted the HTC for refrigerant mixtures by experimenting on a copper plate.

Calus and Rice (1972) developed an empirical system which was based on the theories proposed by Scriven (1959) and Van Stralen (1967) on bubble growth. They presented the data for acetone-water and isopropanol-water binary mixture and the individual components on boiling under free convection. Their correlation was unable to apply the accurate pure component correlations or the data available at that time because their methods yield the pool boiling coefficients directly. Their model couldn't predict the suppression which was seen in their own experiment.

Calus and Leonidopoulos (1974) proposed an equation entirely based on literature. Their equation could not represent the composition of heat transfer precisely, it provided a mean error which was lower than the Calus and Rice (1972) and Stephan and Körner (1969) equations. Schlünder (1982) introduced a correlation of the pure component in which the parameter was the difference between the saturation temperatures. His correlation presented the dependency of HTC on heat flux density and pressure. Thome (1983) took into account that the saturation temperature rises at the interface of liquid-vapour of a bubble. He introduced a new parameter to reduce the temperature difference which was the boiling range (it is the difference between the dew point and the boiling point temperatures at a given condition).

Later on, Thome and Shakir (1987) calculated a new factor which was proposed by Schlünder (1982) i.e. the mass transfer correction factor. They made an assumption to form their correlation which was that near the heating surface the bubble point temperature is not constant. Wenzel et al. (1995) followed a method, which was quite similar to that of Schlünder (1982), but they had to use the mass transfer equation to determine the actual value of interface concentration at the bubble boundary. The interface temperature was then determined by using the interface concentration. Fujita and Tsutsui (1994) altered the correlation proposed by Thome and Shakir (1987) by adding a term dependent

on heat flux and replacing the mass transfer term. Fujita et al. (1996) later altered the Fujita and Tsutsui (1994) equation by altering the term dependent on heat with an ideal wall superheat dependent term.

Fujita and Tsutsui (1994) utilized a vertical tube to measure the HTC of binary mixtures. Two different correlations were proposed by them which was based on the fact that the decrease of heat transfer drop is mainly due to the drop in effective temperature difference. Another assumption was made by them that the bubble point temperature is function of heat flux. This function was determined by observing the heat transfer data of different types of binary mixtures which includes organic, azeotropic and non-azeotropic, aqueous mixtures. The modified equation provided by Fujita et al. (1996) made it applicable for a wider use. They found that the decrease in heat transfer is large for high heat flux, high mole fraction difference and large boiling range. Inoue et al. (2002) introduced a correlation with a tuning parameter. They measured the HTC of ammonia-water and the individual components. Unal (1986) reported a new correlation which was based on experimental technique of dimensional analysis. Vinayak and Balakrishnan (2004) conducted an experiment using acetone-isopropanol-water and acetone-butanone-water mixtures to calculate the HTC. They developed a correlation which involved the use of mass and thermal diffusivity coefficients.

The major drawback of this correlation was that the association factor of both the components is required to calculate the mass diffusivity coefficient which may not be available for all systems. Chao et al. (2018) worked with zeotropic mixtures (R134a/R245fa) and observed an increase in CHF as compared to the individual liquids but the onset of nucleate boiling was delayed. Investigators like Cai et al. (2018) used superheated ethanol-water for the nucleate pool boiling experiment. Their experimental data was in sync with the literature data. Jiaji et al. (2018) conducted an experiment to study the mass diffusion in NPB of binary mixtures and calculated the bubble growth rate. The average bubble growth rate constant, β of R134a, R142b and isobutene was equal to 3.7932 which is in agreement with the literature.

Some major co-relations for NPB heat transfer are shown in Table 1.

Vapour-liquid Equilibrium in a Binary Mixture

The vapour-liquid data must be known to predict the boiling characteristics. Many factors like HTC, CHF and incipience are greatly dependent on the properties

Table 1: Major co-relations for NPB heat transfer

<i>Author and year</i>	<i>Correlations</i>
Palen and Small (1964)	$\frac{\alpha}{\alpha_{id}} = \exp(-0.027 \Delta T_{bp})$
Stephan and Köröner (1969)	$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + K_0 y - x (0.88 + 0.13 p)}$
Calus and Rice (1972)	$\frac{\alpha}{\alpha_{id}} = \left[\frac{1}{1 + y - x (\hat{\alpha}/D_{AB})^{0.5}} \right]^{0.7}$
Calus and Leonidopoulos (1974)	$\Delta T = (\Delta T_1 x_1 + \Delta T_2 x_2) \left[1 + (y - x) \left(\frac{k}{D_{12}} \right)^{0.5} \left(\frac{c_l}{\Delta h_p} \right) \left(\frac{dT}{dx} \right) \right]$
Jungnickel et al. (1980)	$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + K_0 y - x (\rho_v/\rho_l)(q/A)^{0.48+0.1x_1}}; \alpha_{id} = x_1 \alpha_1 + x_2 \alpha_2$
Schlünder (1982)	$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + \frac{\Delta T_{bp}}{\Delta T_{id}} y_1 - x_1 \left[1 - \exp \left(-\frac{B_0 q}{\beta_L \rho_L H_{fg}} \right) \right]}$
Schlünder (1983)	$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + y_1 - x_1 \left\{ 1 - \exp(-B_0 q / \beta_L \rho_L H_{fg}) \right\} (T_{s1} - T_{s2}) \Delta T_{id}}$
Unal (1986)	$\frac{\alpha}{\alpha_{id}} = \frac{1}{[1 + (b_2 + b_3)(1 + b_4)][1 + b_5]}; b_4 = 152 P_r^{3.9}; b_5 = 0.92 y - x ^{0.001} P_r^{0.66}$ $b_2 = (1 - x) \ln \left(\frac{1.01 - x}{1.01 - y} \right) + x \ln \left(\frac{x}{y} \right) + y - x ^{1.5}; \quad b_3 = 0 \text{ for } x \geq 0.01 \quad \text{or}$ $b_3 = (y/x)^{0.1} - 1 \text{ for } x < 0.01$
Thome and Shakir (1987)	$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + \frac{\Delta T_{bp}}{\Delta T_{id}} \left[1 - \exp \left(-\frac{B_0 q}{\beta_L \rho_L H_{fg}} \right) \right]}$
Wenzel et al. (1995)	$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + (\alpha_{id}/\phi)(T_s - T_{sat})}; \frac{y_2 - x_1}{y_2 - x_2} = \exp \left(-\frac{\phi}{B_0 \beta_L \rho_L \Delta h_p} \right)$
Fujita and Tsutsui (1994)	$\alpha_{id} = \frac{1}{1 + K \Delta T_{bp} / \Delta T_{id}}; K = 1 - 0.8 \exp(-1 \times 10^{-5} (q/A))$

(Contd.)

Table 1 (Contd.)

Author and year	Correlations
Fujita and Tsutsui (1996)	$\alpha_{id} = \frac{1}{1 + K \Delta T_{bp} / \Delta T_{id}}; K = 1 - \exp \left[\frac{-60q}{\rho_v H_{fg}} \left(\frac{\rho_v^2}{\sigma g (\rho_l - \rho_v)} \right)^{1/4} \right]$
Inoue et al. (2002)	$\alpha_{id} = \frac{1}{1 + K \Delta T_{bp} / \Delta T_{id}}; K = 1 - 0.75 \exp(-0.75 \times 10^{-5} (q/A))$
Vinayak and Balakrishnan (2004)	$\frac{\alpha}{\alpha_{id}} = \left[1 - \left(y - x \sqrt{D_{AB} / \hat{\alpha}} \right) \right]$

of fluid. The interactions between solute-solute, solute-solvent and solvent-solvent particles can be understood by the study of thermo-physical properties of mixtures such as density, refractive index etc. Many investigators have predicted the VLE data. Some of them are discussed in table below.

Thermo-physical properties like pressure can be calculated using the ideal and non-ideal mixture relationships. The chemical potential of any substance 'a' in the liquid is equal to the chemical potential of

the substance in the vapour. Fugacity of a liquid is dependent on the chemical potential, and the fugacity of the substance in the liquid is also equal to the fugacity of the substance in the vapour,

$$f_{a,l} = f_{a,v} \quad (3)$$

The fugacity of the substance 'a' in the vapour is dependent on the mixture pressure,

$$f_{a,v} = \Phi_a y_a P_{\text{mix}} \quad (4)$$

Author	Mixture used	Pressure range
Mokbel et al. (2012)	Glycerol-water, glycerol + 1,3-propanediol	32 kPa to 163 kPa
Rane et al. (2016)	1,2-propanediol + 1,3-propanediol	6 kPa to 50 kPa
Soujanya et al. (2010)	Methanol-water, glycerol-water Methanol-glycerol	15 kPa to 95 kPa 32.02 kPa and 45.3 kPa
Song et al. (2017)	1-(ethoxymethoxy)-propane + ethanol 1-(ethoxymethoxy)-propane + 1-propanol	101.3 kPa 101.3 kPa
Peng et al. (2017)	Ethanol-water	101.32 kPa
Kochenburger et al. (2017)	R14 + R1234yf, R23 + R1234yf, R218 + R1234yf, R728 + R1234yf, R740 + R1234yf, R170 + R23	5 MPa

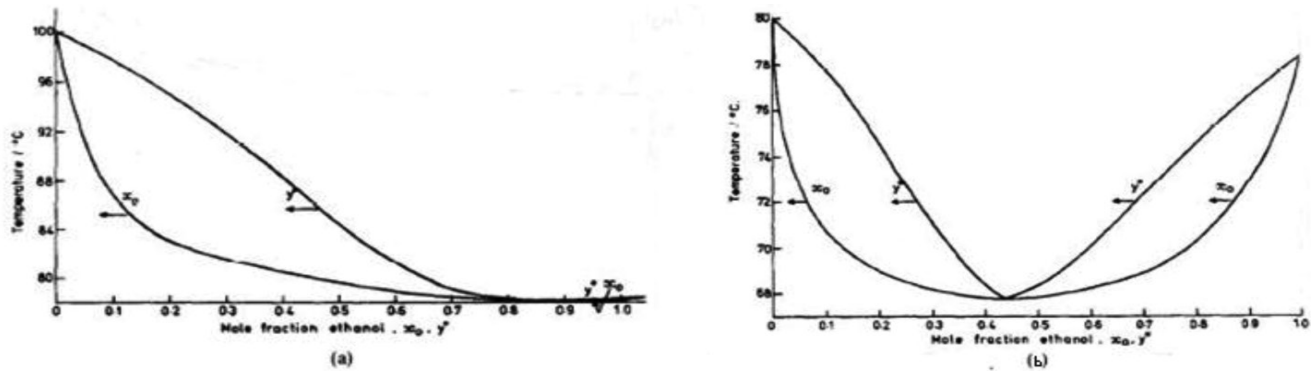


Figure 4: Phase equilibrium of binary mixtures: (a) ethanol-water and (b) ethanol-benzene.

The fugacity of the substance 'a' in the liquid is dependent on the saturation pressure of the substance 'a',

$$f_{a,l} = \gamma_a x_a P_{\text{pure},a} \quad (5)$$

where Φ_a = fugacity coefficient, γ_a = activity coefficient, y_a = vapour mole fraction of substance 'a' and x_a = liquid mole fraction of substance 'a'.

Comparing the equations, we get:

$$\Phi_a y_a P_{\text{mix}} = \gamma_a x_a P_{\text{pure},a} \quad (6)$$

The above equation can be rewritten as

$$P_{\text{mix}} = \frac{\gamma_a x_a P_{\text{pure},a}}{\Phi_a y_a} \quad (7)$$

Using $y_a + y_b = 1$ and $x_a + x_b = 1$ and assuming $\Phi_a = 1$ (for low pressures), equation (8) can be written as

$$P_{\text{mix}} = x_a \gamma_a P_{\text{pure},a} + (1 - x_a) \gamma_b P_{\text{pure},b} \quad (8)$$

For known values of x_a and y_b , the vapour-liquid equilibrium data can be obtained.

Conclusion

Fossil fuels is the most used form of fuel used in any major industry. The use of fossil fuels have numerous disadvantages, the major one being increase in pollution levels. Nucleate pool boiling provides an effective way to transfer heat because of high HTC and CHF. This provides multiple advantages:

1. The energy efficiency of the applicant increases i.e. less amount of heat or energy is required. This helps in less use of fossil fuels and thus helps to reduce the environmental pollution levels.
2. It saves a lot of time as nucleate pool boiling increase HTC which results in obtaining the desired heating effect in less time.
3. All the above factors combined helps to save a lot of money for the industries.

When the NPB data of binary mixtures is compared with the data of pure liquid, it is observed that there is reduction in nucleation site density, bubble departure diameter and heat flux. The reasons are as follows:

1. There is reduction in the temperature driving force due to increase in the boiling point of the microlayer that is because the lighter component evaporates preferentially during the bubble growth.
2. Mass diffusion of the lighter components that is caused by preferential evaporation of the lighter components is much slower than heat transfer.

3. There is a significant non-linearity in the mixture properties with the change in composition.
4. The composition of the mixture also influences bubble dynamics consequently; it affects the HTC.

Many correlations have been determined to calculate the heat transfer of a binary system. Fujita and Tsutsui model predicted the HTC within a range of -25% to +25%. The equation proposed by Fazel and Jamialahmadi (2013) was within a range of -15% to +15%. The major drawbacks with these models are they are accurate for only specific conditions. For example: the Fujita and Tsutsui model is applicable only for low concentrations ($0 < X < 0.3$) and high heat fluxes ($q > 20 \text{ W/m}^2$). Thus, there is a need to propose a model that can predict the HTC for any binary system. It has also been observed that very little work has been done to calculate the HTC for binary mixtures at sub-atmospheric pressure. Further research on this area to increase the CHF and HTC of a Nucleate Pool Boiling Heat Transfer system of binary mixtures with the use of novel engineered surfaces is required.

Acknowledgements

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