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Analysis and experimental study of the heterogeneous nucleation process in the boiling of mixed refrigerants

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ABSTRACT

The heterogeneous nucleation process is important for the study of the nucleate boiling of the mixed refrigerants. In this paper, the onset of heterogeneous bubble nucleation of non-azeotropic mixtures was analysed based on the change in the Gibbs free energy. From the calculated results, the critical radius, change in availability, onset of boiling (ONB) superheat and heat flux were determined. The results showed that the critical radius and the maximum change in the availability of the mixtures was higher than that of the corresponding pure fluids. The ONB superheat first increased and then decreased when the mole fraction of the high boiling point component was increased. In addition, experimental tests were conducted to measure the ONB temperature and heat flux of R124, R22 and R124/R22 mixtures on a copper surface at the pressure of 0.7–0.85 MPa. The ONB superheat and heat flux first increased and then decreased when the concentration of R124 was increased. Additionally, the experimental ONB superheat and heat flux were used to validate the correlations of other models as well as the model presented here. Our model shows much better agreement with the experimental data. Most of the experimental results had an error of +20% to −40%.

1. Introduction

Boiling of mixtures occurs in many industrial processes [1–4]. Because of the mass transfer resistance and the rise of local boiling point at the liquid-vapor interface, the boiling performance of mixtures is much worse than the pure refrigerants [5]. Efforts have been made to study the deterioration of boiling heat transfer in mixtures [5–7]. And the boiling nucleation site density, bubble departure diameter and frequency, bubble growth rate have been studied and heat transfer correlations have been developed [8,9].

However, in the actual chemical process, the temperature differences of hot and cold flows were found to be relatively small [1–3,10–12]. Additionally, it is unclear if the boiling of mixtures occurs in these processes. Cao et al. [10] experimentally studied the ‘mixed refrigerants’ heat transfer performance of a plate-fin heat exchanger in a single-stage cryogenic cycle. The mixed refrigerants evaporated in the cold channel and condensed in the hot channel. In the study, the temperature differences of the hot and cold flow varied from 8 to 25 °C when the refrigeration temperature was lower than −160 °C, and the wall superheat for the liquid mixtures was approximately in the range of 4–13 °C. Ardhapurkar et al. [11] studied the temperature profile of a helical heat exchanger in a mixed refrigerant Joule-Thomson cryocooler. The apparent log mean temperature differences (ALMTD) of the hot and cold flow varied from 2 to 21 °C when the system was stable. Pang et al. [1] experimentally studied the thermal performance of brazed plate heat exchangers (BPHE) in a mixed-refrigerants Joule-Thomson cooler. Three BPHEs were used as recuperative heat exchangers in the Linde-Hampson refrigeration cycle, and the temperature differences were in the range of 4.4–29.5 °C. As known, the heat transfer performance of boiling is much better than natural convection; thus, it is important to study the heterogeneous nucleation process and determine the onset of boiling (ONB) temperature of mixed refrigerants.

Heterogeneous nucleation of pure fluid has been studied extensively [13]. Models for the nucleation process and correlations for the ONB superheat and heat flux have been proposed [14]. Hsu [15] was the first to study the nucleation process in a cavity. He assumed that a linear temperature gradient exists on the superheat wall and the bubble nucleus will not grow if the minimum temperature of the surrounding liquid is lower than the bubble temperature. In his model, the size of a bubble was proportional to the

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Nomenclature

T	temperature (K)
q	heat flux (w m^{-2})
x	distance (m)/mole fraction of the liquid phase
y	concentration of the vapor phase
k	temperature gradient (K m^{-1})
P	pressure (Pa)
r	bubble radius (m)
V	volume (m^3)
h	specific enthalpy (J mol^{-1})
s	specific entropy (J mol^{-1} K^{-1})
c_{pl}	liquid specific heat (J mol^{-1} K^{-1})
c_{pv}	vapor specific heat (J mol^{-1} K^{-1})
R	gas constant (J mol^{-1} K^{-1})
h_{fg}	latent heat (J mol^{-1})
k_l	thermal conductivity (W m^{-1} K^{-1})
Q'	dimensionless heat flux
Delta T'	dimensionless superheat

Greek symbols

σ	surface tension (N m^{-1})
θ	contact angle (°)
μ	chemical potential (J mol^{-1})
Ψ	availability (J mol^{-1})
λ	thermal conductivity (W m^{-1} K^{-1})

Subscripts

bub	bubble point
dew	dew point
ONB	onset of boiling
b	bubble
l	liquid phase
v	vapor phase
in	initial state
sat	saturation
w	wall
pure	pure fluid
A	part A
B	part B
1	component 1
2	component 2
c	critical
sur	surface
v	volume

radius of the cavity mouth; thus, the contact angle in the model was a constant. What's more, this model was improved by many researchers [16–22].The effect of the wettability was considered by Basu et al. [20]. The thermal layer thickness was modified by Sato et al. [16], and in the study of Davis et al. [17], a more precise expression was obtained. Finally, the thermal hydraulic field was considered, and the linear temperature gradient in the model was modified by Kandliker et al. [18,19].

More recently, researchers have attempted to account for the heterogeneous nucleation process from the perspective of the change in chemical potential. Wu et al. [23] developed a heterogeneous nucleation model based on the second law of thermodynamics, and the growth and collapse of a bubble nucleus was dependent on the change of a function. Cheng et al. [24,25] developed a thermodynamic model for the heterogeneous nucleation process based on the change in the Gibbs free energy and the availability function. In their model, the temperature gradient and the wettability were considered. Additionally, the critical radius of a bubble nucleus, the wall temperature gradient and the heat flux were obtained. Yuan et al. [26] developed a heterogeneous bubble nucleation model. The effect of the insoluble gas and the line tension were considered.

Compared with the large number of studies on the heterogeneous nucleation process of pure fluids, similar studies are rare for mixtures. No theoretical model was found and few experimental studies have been performed. Han et al. [27] studied the bubble behavior and heat transfer characteristics during saturated pool boiling of R11/R113 mixtures near the ONB temperature, and the mixtures usually showed a greater ONB temperature difference than pure fluid. Inoue et al. [28] studied the pool boiling performance of ethanol/water mixtures with a surface-active agent. They found that the ONB heat flux for pure liquids were lower than for mixtures, and the maximum ONB heat flux occurred when the concentration of water was 0.5. They believed that the higher ONB heat flux for the mixtures was due to the mass transfer effect and the preferential evaporation of the more volatile liquid in the vicinity of the heated surface. But before the boiling occur, the small bubble placed on the heated surface is in "local thermal equilibrium" state [24]. The quantity of the liquefied molecules are equal to the vaporized molecules. Thus, it seems that 'the preferential evaporation of the more volatile liquid' is not the reason of the higher ONB heat flux of the mixtures.

In our perspective, because of the effect of mixtures, the entropy of the liquid and vapor phase of the mixed refrigerants are different from the pure substances. And the chemical potential of the mixtures are changed. Thus, it is harder for a bubble embryo to nucleate in liquid mixtures. To disclose the nucleation mechanism of a mixed fluid, the heterogeneous nucleation process in the boiling of binary mixtures was analysed based on Cheng's model [24,25]. In this paper, the critical radius under different wall superheats and different liquid concentrations was calculated, the ONB superheat and heat flux for different liquid concentration were obtained, and the change in availability was analysed. Additionally, a visualization study was performed to investigate the ONB temperature and heat flux of pure fluids and mixtures.

2. Analysis of the heterogeneous nucleation process in the boiling of binary mixtures

Based on Cheng's model [24,25], a model for the heterogeneous nucleation process of binary mixtures was developed. The schematic diagram is shown in Fig. 1, and the following assumptions were made:

(1) The temperature at the top of the bubble was at least equal to the bubble point temperature (the saturation temperature of the liquid mixtures under the operating pressure) under the operating pressure.

$$T(x_b) \geqslant T_{bub} \tag{1}$$

(2) The temperature profile of the superheated liquid layer must be determined. And based on the Cheng's model [24,25] and Hsu's model [15], the temperature was assumed to be linear.

$$T_l = T_w - kx \tag{2}$$