



# Analysis and experimental study of nucleation site densities in the boiling of mixed refrigerants



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## ABSTRACT

The reduction of the boiling nucleation site density in mixed liquid refrigerants is one of the main reasons for the reduction of the boiling heat transfer coefficient. Instead of the mass diffusion effect, this work proposes the concentration fluctuation effect to account for the reduction in the boiling site density. Evidence is provided to support this hypothesis, and a model adopting the fluctuation theory is developed. In addition, experimental tests were conducted to investigate the boiling site densities of i-butane, propane, R134a/R32 and i-butane/propane under pressures in the range of 0.25–1 MPa and wall superheats in the range of 3–25 °C. For the pure refrigerants, the boiling nucleation site density showed a linear relation with wall superheat. Additionally, a correlation is proposed to predict the boiling site density based on the present data. In the case of binary mixtures, the boiling site density first decreased and then increased as the concentration of the high boiling point fraction was increased. The experimental attenuation factors of boiling site densities were calculated for binary mixtures. Qualitative calculations from the proposed model showed good agreement with the experimental data.

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## 1. Introduction

Boiling of mixtures occurs in many chemical processes [1–7]. As a result, the heat transfer coefficient (HTC) is crucial for the design of heat exchangers. However, there can be large variations in the boiling processes of pure and mixed refrigerants, and it is impossible to guarantee thermal performance when mixed refrigerants are employed in heat exchangers which use pure refrigerants.

Boiling of mixtures has been investigated extensively in the past decades [8–12]. Many efforts have been made to study the deterioration of boiling heat transfer in mixtures [13]. Various explanations have been proposed to account for the differences in boiling heat transfer performance between pure refrigerants and their mixtures. A summary of the studied causes was provided by Celata et al. (1994) [13]:

- (1). Increase in the local boiling point and mass transfer resistance.
- (2). Changes in physical properties of the mixtures.
- (3). The effect of mixtures on nucleation.

- (4). Retardation of the main mechanisms of heat transfer.

In this paper, efforts have been made to study the reduction of nucleation sites during the boiling of binary mixtures.

During the boiling process, bubbles inside the liquid grow from active nucleation sites, whose density depends on the surface micro-roughness (size distribution of cavities, distribution of the half cone angles of the cavities, minimum cavity mouth radius, etc. [14]), the surface tension of the liquid, the physical properties of the heating surface and the liquid, and the wall superheat [15]. Due to the large uncertainty for finding the proper cavities and the pressure fluctuation by the bubble generated, the statistical method does not appear to predict the nucleation site density accurately [16]. As a result, based on the mechanisms and experimental studies of pure fluids (mostly water), many nucleation site models and correlations have been proposed [14,15,17–20] and continually corrected [16,21–25].

Compared to the large number of studies on the boiling nucleation site densities of pure refrigerants, similar studies are relatively rare for binary mixtures. Hui et al. (1984) [26] measured the boiling site density and the HTC of ethanol/water and ethanol/benzene mixtures at 0.101 MPa for a heated vertical brass disk. The mass diffusion effect was considered as the main reason for the strong effect of composition on the boiling site density. Benjamin

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**Nomenclature**

$T$	temperature (K)
$x$	molar fraction in the liquid phase
$n$	the number of molecule
$S$	entropy ( $\text{J}\cdot\text{K}^{-1}$ )
$N$	molar mass (mole)
$V$	volume ( $\text{m}^3$ )
$W$	probability
$k$	Boltzmann constant ( $\text{J}\cdot\text{K}^{-1}$ )
$P$	pressure (MPa)
$R$	gas constant ( $\text{J}\cdot\text{kg}^{-1}$ )
$R_c$	critical radius (m)
$M$	molar mass ( $\text{g}\cdot\text{mol}$ )
$i$	latent heat ( $\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$ )
$v$	specific volume ( $\text{m}^3\cdot\text{kg}^{-1}$ )
$D_d$	departure diameter (m)
$A$	area of the heated wall ( $\text{m}^2$ )
$g$	acceleration of gravity ( $\text{m}\cdot\text{s}^{-2}$ )
$Na$	boiling site density ( $\text{sites}\cdot\text{m}^{-2}$ )
$Pr$	Prandtl number
$R_a$	center line average (m)
$C_p$	specific heat capacity ( $\text{J}\cdot\text{kg}\cdot\text{K}$ )
$y$	molar fraction in the vapor phase

*Greek symbols*

$\Delta T$	temperature difference ( $^{\circ}\text{C}$ )
$\Delta S$	entropy difference ( $\text{J}\cdot\text{K}^{-1}$ )
$\varphi$	attenuation factor

$\mu$	specific chemical potential ( $\text{J}\cdot\text{kg}^{-1}$ )
$\sigma$	surface tension ( $\text{N}\cdot\text{m}^{-1}$ )
$\theta$	contact angle ( $^{\circ}$ )
$\rho$	density ( $\text{kg}\cdot\text{m}^{-3}$ )
$\gamma$	surface-liquid interaction parameter
$\eta$	dimensionless surface micro roughness parameter
$\rho$	molar density (mole/ $\text{m}^3$ )

*Subscripts*

<i>dew</i>	dew point
<i>bub</i>	bubble point
ONB	on set of boiling
<i>lsh</i>	limits of superheat
1	the high boiling point component
2	the low boiling point component
<i>l</i>	liquid phase
<i>v</i>	vapor phase
<i>sat</i>	saturation
<i>w</i>	wall
<i>exp</i>	experiment
<i>cal</i>	calculation
<i>pure</i>	pure fluid
<i>mix</i>	mixture
<i>eff</i>	effective

et al. (1997) [27] investigated the nucleation site density of acetone/carbon tetrachloride and n-hexane/carbon tetrachloride under atmospheric pressure. They considered the effects of surface micro-roughness and the physical properties of the liquid, and proposed a correlation based on their previous study and experimental data for pure liquids. In their opinion, a part of the driving force provided by temperature was utilized to overcome the resistance caused by mass diffusion. Thus, an attenuation factor related to the diffusion coefficient was applied to the temperature difference term in their correlation. In these studies, the mass diffusion effect was regarded as the main reason for the reduction in the boiling nucleation site density of mixtures.

In the classical theory of homogeneous nucleation, nucleation sites are generated as a result of density fluctuations in the liquid (Katz, 1975 [28]). For the case of mixtures, this paper proposed that the concentration of the low-boiling point component should be aggregated in a localized region before the nucleation sites are generated. This concentration fluctuation effect is the main reason for the reduction in the number of boiling nucleation sites generated inside liquid mixtures.

This paper, firstly, provides evidence for the concentration fluctuation effect by analysing the experimental data for onset of boiling (ONB) and limits of superheat. Furthermore, by using the classical fluctuation theory [29], a model is developed to account for the reduction of boiling nucleation site density in binary mixtures. Secondly, a visualization study was carried out to investigate the boiling nucleation site densities of i-butane, propane, R134a/R32 and i-butane/propane mixtures under various pressures. A correlation was developed for pure refrigerants, based on the experimental data. Additionally, experiments on mixed refrigerants show that the boiling site densities first decrease and then increase as the fraction of the high-boiling point component increases. The attenuation factors for the reduction of boiling site densities were calculated for mixed refrigerants. Lastly, by using

the proposed model, the qualitative results of calculations were compared with the experimental data.

**2. Analysis of the nucleation process in binary mixtures***2.1. Evidence of the concentration fluctuation effect*

It is generally understood that under a static steady state, liquids do not vaporize until their temperature reaches the limit of superheat. To prove the attenuation effect of concentration fluctuation on the formation process of nucleation sites, the experimental data for ONB and the limits of superheat are studied here for binary mixtures.

Han et al. (2004) [30] studied the bubble behavior and heat transfer characteristics during saturated pool boiling of R11/R113 mixtures near the ONB temperature under atmospheric pressure. Table 1 shows the experimental results of the ONB temperature in their study. And the mixtures usually showed a greater ONB temperature difference ( $\Delta T_{\text{ONB}}$ ) than pure fluid. But in their study, the reason of higher ONB temperature difference for mixtures was not presented. We propose that the higher ONB temperature difference for mixtures is due to the concentration fluctuation effect.

In addition, extensive experimental studies were conducted to investigate the limits of superheat for pure refrigerants and mixed

**Table 1**  
ONB temperatures of R11/R113 mixtures from the study of Han et al. (2004) [30].

Mass fraction of R11 (%)	$T_{\text{bub}}(^{\circ}\text{C})$	$T_{\text{exp}}(^{\circ}\text{C})$	$\Delta T_{\text{ONB}}(^{\circ}\text{C})$
100	23.71	35.0	11.29
81.5	25.79	40.9	15.11
53.6	29.91	43.1	13.19
28.0	35.63	51.0	15.37
13.9	40.43	54.9	14.47
0	47.58	61.0	13.42

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