Boiling Heat Transfer With Binary Mixtures: Part II—Flow Boiling in Plain Tubes

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Flow boiling heat transfer with pure fluids comprises convective and nucleate boiling components. In flow boiling of binary mixtures, in addition to the suppression effects present in pool boiling, the presence of flow further modifies the nucleate boiling characteristics. In the present work, the flow boiling correlation by Kandlikar (1990, 1991b) for pure fluids is used as the starting point, and the mixture effects derived in Part I (Kandlikar, 1998) of this paper are incorporated. Three regions are defined on the basis of a volatility parameter, \( v_i = (c_i/\Delta h_{li})(\kappa/D_i)^{1/2} \) \((y_i - x_i)\Delta T/\Delta x_i \). They are: region I—near azotropic, region II—moderate diffusion-induced suppression, and region III—severe diffusion-induced suppression. The resulting correlation is able to correlate over 2500 data points within 8.3 to 13.3 percent mean deviation for each data set. Furthermore, the \( \alpha - x \) trend is represented well for R-12/R-22, R-22/R-114, R-22/R-152a, R-500, and R-132a/R-123 systems. Electrically heated stainless steel test sections as well as fluid-heated copper test sections are both covered under this correlation.

1 Introduction

Heat transfer in flow boiling of binary mixtures is receiving increasing attention in the refrigeration industry as refrigerant mixtures are being evaluated to replace the conventional pure refrigerants. The advantages of using refrigerant mixtures over pure refrigerants include the improvement of coefficient of performance, better match with the product thermal load, and safer, environmentally friendly refrigerants. The pure refrigerant systems in reality contain binary mixtures with the presence of dissolved oil. While ternary refrigerants (four components with oil) are currently being tested as potential replacements, a fundamental understanding of binary systems is essential before attempting multicomponent heat transfer modeling. Among other applications, chemical, petrochemical and process industry applications of binary mixtures are noteworthy.

2 Review of Literature

There are relatively few studies available in the open literature on modeling the flow boiling heat transfer compared to those on pool boiling of binary mixtures. Table 1 provides a summary of some of the available correlations. One of the early developments was proposed by Calus et al. (1973) who extended the pool boiling suppression factor derived by Calus and Rice (1972). Calus et al. modified an existing flow boiling correlation, which included only the convective term, and introduced an additional correction factor to account for the rise in the saturation temperature at the liquid–vapor interface of a bubble. As their correlation did not include a nucleate boiling term, it underpredicted the results considerably at higher heat flux.

Bennett and Chen (1980) presented a correlation scheme based on the Chen (1966) correlation. The convective component was modified to incorporate the bubble interface temperature. The suppression factor suggested by Calus and Leonidopoulos (1974) for pool boiling was introduced in the nucleate boiling term with some modifications. The mass transfer coefficient in the liquid near a bubble interface was calculated from a correlation with the same form as the Dittus–Boelter correlation for heat transfer, but employing the Sherwood and Schmidt numbers and the same values of constants, although these constants were obtained independently from their own data sets. Jung (1988) found that the Chen correlation overpredicted the heat transfer coefficient for pure refrigerants, and the Bennett and Chen correlation was unable to correlate the heat transfer data for refrigerant mixtures.

Jung (1988) conducted an extensive study on the flow boiling of refrigerant mixtures of R-12/R-152a, including azo trope R-500, and R-22/R-114. He developed a correlation shown in Table 1, using an ideal heat transfer coefficient for mixtures in flow boiling, similar to that employed for pool boiling by earlier investigators. This approach implicitly incorporates the convective component of flow boiling in the averaging scheme. Jung’s correlation utilizes the phase equilibrium data, and critical pressure and temperature, along with 25 constants, which were determined empirically from their own experimental data sets.

Kandlikar (1991a) extended his earlier pure component flow boiling correlation (Kandlikar, 1990) to binary mixtures as shown in Table 1. It was postulated that only the nucleate boiling component would be affected in binary systems due to diffusion effects, which were modeled after a semi-empirical approach proposed by Calus and Rice (1972) for pool boiling. The results were satisfactory when compared to Jung’s (1988) data. However, with the availability of new data sets, it was seen that the Kandlikar (1991a) correlation for binary mixtures yielded larger errors, usually underpredicting the mixture effects. One of the main reasons was the inability of the underlying Calus and Rice’s suppression model to predict the severe suppression seen for mixtures with large volatility differences as shown in Part I.

3 Objectives of the Present Work

From this discussion it can be seen that there is a need for developing a flow boiling correlation applicable to binary systems and azo tropes, particularly with the current interest in refrigerant mixtures. The present work is aimed toward incorporating the pool boiling model developed in Part I of this paper in a flow boiling model for binary mixtures. The new model is
The subscripts NBD and CBD in Eq. (1) refer to the nucleate boiling dominant and the convective boiling dominant regions, for which the respective \( \alpha_{TP} \) are given by:

\[
\alpha_{TP,NBD} = 0.6683C_0^{-0.2}(1-x)^{0.8}a_{LO} + 1058.0B_0^{0.7}(1-x)^{0.8}F_{PI}a_{LO}
\]

and

\[
\alpha_{TP,CBD} = 1.136C_0^{-0.9}(1-x)^{0.8}a_{LO} + 667.2B_0^{0.7}(1-x)^{0.8}F_{PI}a_{LO}
\]

Additionally, for horizontal tubes with Froude number, \( F_{RILO} \), less than 0.4, a multiplier (25F_{RILO})^{0.324} is applied to the first terms in Eqs. (2) and (3). For \( F_{RILO} > 0.04 \), and for vertical tubes no correction is needed. This correction is usually not needed for the range of mass fluxes employed in the refrigerant evaporators.

\( F_{PI} \) in Eqs. (2) and (3) is a fluid-surface parameter related to the nucleation characteristics. Table 2 lists its value for several refrigerants. The single-phase heat transfer coefficient, \( \alpha_{LO} \), is obtained from the Petukhov and Popov (1963), and Gnielinski (1976) correlations, as suggested later by Kandlikar (1991b).

Kandlikar and Popov (1963) for \( 0.5 < F_{RILO} \leq 2000 \) and \( 10^4 \leq Re_{LO} < 5 \times 10^6 \):

\[
Nu_{LO} = \alpha_{LO}D/\lambda_{LO} = Re_{LO} Pr_{L}/(f/2)/[1.07 + 12.7(Pr_{L}^{2/3} - 1)(f/2)^{0.5}]
\]

Gnielinski (1976) for \( 0.5 < F_{RILO} \leq 2000 \) and \( 2300 < Re_{LO} < 10^4 \):

\[
Nu_{LO} = \alpha_{LO}D/\lambda_{LO} = (Re_{LO} - 1000) Pr_{L}(f/2)/[1.0 + 12.7(Pr_{L}^{2/3} - 1)(f/2)^{0.5}]
\]
Table 2  Fluid-surface parameter $F_{fl}$ for refrigerants in copper or brass tubes

<table>
<thead>
<tr>
<th>Fluid</th>
<th>$F_{fl}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>1.00</td>
</tr>
<tr>
<td>R-11</td>
<td>1.30</td>
</tr>
<tr>
<td>R-12</td>
<td>1.50</td>
</tr>
<tr>
<td>R-13B1</td>
<td>1.31</td>
</tr>
<tr>
<td>R-22</td>
<td>2.20</td>
</tr>
<tr>
<td>R-113</td>
<td>1.30</td>
</tr>
<tr>
<td>R-114</td>
<td>1.24</td>
</tr>
<tr>
<td>R-124</td>
<td>1.9</td>
</tr>
<tr>
<td>R-134a</td>
<td>1.63</td>
</tr>
<tr>
<td>R-152a</td>
<td>1.10</td>
</tr>
</tbody>
</table>

For all fluids in stainless steel tubes, $F_{fl}=1.0$

The diffusion coefficient $D_{12}$ was calculated as follows:

$$D_{12} = (D_{12}^0)^{2/3}(D_{12}^S)^{1/3}$$  \(13\)

$D_{12}^0$ and $D_{12}^S$ in this equation are self-diffusion coefficients given by

$$D_{12}^0 = 1.1782 \times 10^{-16} \frac{(\phi M_e)^{1/2} T}{\eta e_{fl} V_{m,1}}$$  \(14\)

$\phi$ is the association factor for the solvent (2.26 for water, 1.9 for methanol, 1.5 for ethanol, 1.9 for ethylene glycol, and 1.0 for unassociated solvents, including benzene, methane, and refrigerants).

4.3 Flow Boiling Model for Binary Mixtures. The diffusion-induced suppression factor $F_D$ given by Eq. (7) is derived for pool boiling. Before this factor can be applied to the nucleate boiling component in flow boiling, a valuable insight is obtained from a plot of wall superheat versus heat transfer coefficient obtained by Kandlikar and Raykoff (1977), shown in Fig. 1. The experiments were conducted with subcooled flow of water/ethylene glycol solutions over a flat 9.5 mm circular heater placed flush on the lower wall of a 3 mm x 40 mm horizontal flow channel. Figure 1 shows the variation of heat transfer coefficient with wall superheat for pure water, and for 20–80 percent mass concentration of water/ethylene glycol solution. The heat transfer coefficient with pure water is almost constant at lower values of wall superheat. Nucleation begins at ONB (onset of nucleate boiling), and $\alpha$ increases slowly with wall superheat. This region is similar to the CBD region. Beyond a certain value of wall superheat, $\alpha$ begins to rise rapidly due to increased nucleation activity, and this region corresponds to the NBD region. The results for the aqueous ethylene glycol solution, also shown in Fig. 1, indicate the same trend of slowly increasing $\alpha$ in the CBD region, but the sharp increase in $\alpha$ corresponding to the NBD region is not seen even for a superheat of 33°C. This shows that the nucleate boiling dominant region is not present, or pushed significantly toward the higher wall superheats for binary mixtures. Therefore, it can be surmised that the convective mechanism is the dominant mechanism in flow boiling of binary mixtures.

From this discussion, it can be concluded that the CBD region extends into considerably higher values of wall superheats for flow boiling of binary mixtures. For conditions near azetotropic compositions, the pure component correlation should, however, hold good. For the conditions where the mixture effects are significant, the diffusion-induced suppression factor could be
applied to the nucleate boiling component in the CBD region correlation. The volatility parameter, $V_i$, given by Eq. (6), derived for pool boiling of binary mixtures, is again utilized here to define three regions with different levels of diffusion-induced suppression. The criteria identifying these regions are obtained by analyzing over 2500 data points for five different binary systems reported by five investigators. Table 3 gives the details of the experimental conditions and ranges of parameters for these data sources. The data contains the entire range of concentrations, including the azoetropes of R-12/R-22 and R-12/R-152a.

The following correlation is presented to predict the flow boiling of binary mixtures. The properties of the mixtures at saturation are employed in the following equations.

**Flow Boiling Correlation for Binary Mixtures**

**Region I:** Near-azeotropic region; $V_i < 0.03$,

$$\alpha_{TP,B} = \text{larger of } \left\{ \begin{array}{l} \alpha_{TP,B,NBD} \\ \alpha_{TP,B,CBD} \end{array} \right\}$$  \hspace{1cm} (15)

In this region, the general correlation for pure fluids is applicable. $\alpha_{TP,B,NBD}$ and $\alpha_{TP,B,CBD}$ are obtained from Eqs. (2) and (3), respectively, using the mixture properties. The fluid-surface parameter is obtained as the mass fraction-averaged value given by the following equation:

$$F_{FI} = x_1 F_{FI,1} + x_2 F_{FI,2}$$  \hspace{1cm} (16)

Table 2 lists $F_{FI}$ for different refrigerants flowing in copper or brass tubes. $F_{FI}$ for all liquids in stainless steel tubes is 1.0.

The near-azeotrope region covers azoetropes and low-volatility difference mixtures. Although the nucleate boiling may be slightly affected for these mixtures, its effect on the flow boiling heat transfer coefficient is insignificant.

**Region II:** Moderate diffusion-induced suppression region, $0.03 < V_i < 0.2$, and $Bo > 1E - 4$,

$$\alpha_{TP,B} = \alpha_{CBD,LO} = 1.136Co^{-0.9}(1 - x)^{0.8}\alpha_{LO}$$

$$+ 667.2Bo^{0.7}(1 - x)^{0.8}F_{FI}\alpha_{LO}$$  \hspace{1cm} (17)

$F_{FI}$ for mixtures is obtained from Eq. (16). In the moderate diffusion-induced suppression region, the nucleation effects are suppressed due to the mass diffusion resistance, and the convective heat transfer becomes dominant. In the CBD region, the bubble growth is primarily limited to the early stages in the growth cycle. The correlation for the CBD region without any suppression factor is therefore able to predict this region well.

**Region III:** Severe diffusion-induced suppression region, $(a)$ for $0.03 < V_i < 0.2$ and $Bo \leq 1E - 4$, and $(b)$ $V_i \geq 0.2$,

$$\alpha_{TP,B} = 1.136Co^{-0.9}(1 - x)^{0.8}\alpha_{LO}$$

$$+ 667.2Bo^{0.7}(1 - x)^{0.8}F_{FI}\alpha_{LO}F_D$$  \hspace{1cm} (18)

This region covers the two ranges as indicated in $(a)$ and $(b)$ above. $F_D$ is obtained from

$$F_D = 0.678[1 + (c_{p,L}/\Delta h_{LO})] \times (\kappa/D_{12})^{1/2} \times (y_1 - x_1)(dT/dx_1)]^{-1}$$  \hspace{1cm} (19)

where $dT/dx_1$ is the slope of the bubble point temperature versus $x_1$ curve. Equation (19) is simplified from Eq. (7) to eliminate the iteration for the equilibrium concentrations at the interface. Instead, bulk liquid and vapor concentrations are used along with the slope of the bubble point curve. The error in $F_D$ due to this simplification is small, usually less than 5 percent for the refrigerant mixtures, and $F_D$ is applied only to the nucleate boiling term. The fluid-surface parameter $F_{FI}$ in these equations is obtained from Eq. (16).

The severe diffusion-induced suppressed region is dominated by the convective effects. The nucleate boiling contribution in this region is further reduced due to the large difference in composition between the two phases, and the resulting mass diffusion resistance at the liquid–vapor interface of a growing bubble.

### 4 Results and Discussion

Table 3 shows the details of the experimental data sets used to compare the results from the correlation. The data covers a broad range of the volatility parameter, $V_i$. Table 4 shows the mean deviation from each data set listed in Table 3. The ranges of $Bo$, $Co$, and $V_i$ for each data set are also listed in Table 4. The correlation is applicable for qualities from near zero to about 0.8. The dryout and near-dryout regions are not covered by the correlation. The properties of the mixtures are evaluated using REFPROP by NIST (1995).

It can be seen from Table 4 that the mean absolute error for each data set is between 8.3 and 13.3 percent. Additional discussion on the comparison for specific ranges of $V_i$ covering different suppression regions is presented in the following paragraphs.

**Region I—Near-Azeotropic Region.** In this region, the compositions of the two phases are nearly equal. The data by Jung et al. (1988) on azo trope R-500 are representative of this region. R-500, which is an azo trope of R-12 and R-152a, shows a small volatility difference, with $V_i$ in the range of 0.0006–0.002. Figure 2 shows a comparison between the predicted and the experimental variation of $\alpha$ with quality $x$. It can be seen that the nucleate boiling dominant region at lower $x$ and the convective boiling dominant region at higher $x$ are well represented by the correlation. The mean absolute error for this data set is 11.4 percent.

Some of the experimental data of Hihara et al. (1989) also fall in this region for the R-22/R-12 system. Figure 3 shows a comparison between the predicted and the experimental values. It can be seen that the trend in $\alpha$ versus $x$ is well represented by the correlation showing the transition between NBD and CBD regions. No diffusion-induced suppression is observed in the entire region and the pure fluid correlation correctly represents $\alpha$ and its trends with $x$.

**Region II—Moderate Diffusion-Induced Suppression Region.** This region covers the range $0.03 < V_i < 0.2$ with $Bo > 1E - 4$. In this region, the nucleate boiling dominant region is not present, and the heat transfer is mainly in the CBD region. However, the diffusion-induced suppression is quite moderate, and does not affect the nucleate boiling term in the CBD region.

The data of Celata et al. (1993) for R-12/R-114 falls in this
Table 4 Parameter ranges of data sources and comparison with correlation

<table>
<thead>
<tr>
<th>Data source</th>
<th>Binary System</th>
<th>Bo x 10^-5</th>
<th>Co</th>
<th>V</th>
<th>Mass Abs. Deviation, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jung et al. (1988)</td>
<td>R-12/R-152a</td>
<td>5.1-5.3</td>
<td>0.52-1.8</td>
<td>0.023-0.044</td>
<td>8.3%</td>
</tr>
<tr>
<td>R-12/R-153a</td>
<td>5.1-5.3</td>
<td>0.01-1.45</td>
<td>0.013-0.022</td>
<td>10.6%</td>
<td></td>
</tr>
<tr>
<td>R-22/R-114</td>
<td>7.5-77</td>
<td>0.1-1.83</td>
<td>0.0006-0.002</td>
<td>11.4%</td>
<td></td>
</tr>
<tr>
<td>R-500</td>
<td>7.5-77</td>
<td>0.1-1.83</td>
<td>0.0006-0.002</td>
<td>11.4%</td>
<td></td>
</tr>
<tr>
<td>Hihara et al. (1989)</td>
<td>R-12/R-32</td>
<td>47-61</td>
<td>0.01-1.91</td>
<td>0.013-0.064</td>
<td>13.3%</td>
</tr>
<tr>
<td>R-22/R-114</td>
<td>37-60</td>
<td>0.025-1.64</td>
<td>0.07-0.07</td>
<td>9.0%</td>
<td></td>
</tr>
<tr>
<td>Takamatsu et al. (1993)</td>
<td>R-22/R-114</td>
<td>0.018-3.2</td>
<td>0.28-0.34</td>
<td>9.2%</td>
<td></td>
</tr>
<tr>
<td>Celata et al. (1993)</td>
<td>R-12/R-114</td>
<td>9.4-88</td>
<td>0.52-1.83</td>
<td>0.06-0.15</td>
<td>8.9%</td>
</tr>
<tr>
<td>Murata and Hashizume (1993)</td>
<td>R-134a/R-123</td>
<td>20-185</td>
<td>0.004-0.05</td>
<td>0.28-0.34</td>
<td>12.1%</td>
</tr>
</tbody>
</table>

Figure 4 shows the data from Celata et al. (1993) for R-22/R-114 obtained in an electrically heated stainless steel test section. $F_{HI}$ for this case is 1.0 in the entire range of concentrations. As seen from Fig. 4, the correlation is able to predict the data well. The mean absolute error with this data set is 8.9 percent. Similar observations are made with Jung's (1988) data sets for R-22/R-114 and R-22/R-152a mixtures employing an electrically heated stainless-steel test section. Figure 5 shows the comparison with Jung's R-12/R-152a data falling in the moderate diffusion-induced suppression region, with a good agreement between the observed and predicted trends.

Region III—Severe Diffusion-Induced Suppression. In this region, the nucleate boiling mechanism is strongly affected by the mass diffusion effects, and the nucleate boiling component in the CBD region is suppressed considerably. A large number of data investigated in the present work falls in this region.

Figure 6 shows Murata and Hashizume's (1989) data for R-134a/R-123 system. This data set is obtained using a copper tube with an electrical heater element wrapped around it. The fluid-surface parameter $F_{HI}$ given by equation (16) is applied for this case. Again the correlation does a good job in predicting the heat transfer coefficient in this region as well, with a mean error of 12.1 percent for the entire data set.

Figure 7 shows the data of Takamatsu et al. (1993) obtained using a copper test section exchanging heat with hot water. This corresponds to a fluid-heated test section. The fluid-dependent parameter $F_{HI}$ is applicable to copper and brass tubes. The correlation is able to represent this data set as well and the trends in $\alpha$ versus $x$ are also accurately represented. The mean absolute error is seen from Table 4 to be 9.2 percent for the entire data set.

Figure 8 shows the data set Hihara et al. (1989) for R-22/R-114 under the severe diffusion-induced suppression region. These data were obtained in an electrically heated stainless-steel test section for which $F_{HI} = 1.0$ applies. The agreement between the correlation and the data is very good, with the mean absolute error seen from Table 4 as 9.0 percent.

Additional Comments. The experimental error in a binary system is higher than that with pure components due to additional uncertainties associated with equilibrium phase compositions. The saturation temperature along the length of the evaporator tube varies due to changing liquid and vapor compositions as well as changing pressure. Additional factors are introduced.
5 Conclusions

A flow boiling correlation is developed for binary systems. It incorporates the diffusion-induced suppression factor developed in Part I of this paper. The basic correlation by Kandlikar (1990) for pure liquids is extended to cover the binary systems under three regions depending on the volatility parameter: region I—near-azeotropic region, region II—moderate diffusion-induced suppression region, and region III—severe diffusion-induced suppression region.

2 The correlation is compared with the experimental data for five binary systems of refrigerant mixtures reported in the literature. The overall absolute mean deviation for over 2500 data points is around 10 percent.

3 The trend in the heat transfer coefficient $\alpha$ versus quality $x$ is well represented by the correlation in all three regions.

4 The correlation performs equally well for fluid-heated as well as electrically heated test sections. Also, the tube material effect, stainless steel or copper/brass, is well accounted by the fluid dependent parameter, which reduces to 1.0 for stainless-steel test sections.

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References


NIST, 1995, REFPROP, National Institute for Science and Technology, Washington, DC.
