An experimental investigation of heat transfer in natural convection boiling of multicomponent mixtures

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The present investigation deals with experimental study related to natural convection boiling of acetone-isopropanol-water ternary mixtures on a horizontal plain tube at atmospheric pressure. The heat transfer coefficients of ternary mixtures depend on the concentrations of the light components and heat flux. The wall superheat required for the boiling of any composition of ternary mixture is greater than that either of pure components. The correlation proposed by Thome and Shakir for multicomponent mixtures has been found to correlate data within absolute mean error of ± 5.16%.

Boiling is a physical process of wide industrial application and thus has been the subject of intensive research for the past several decades. However, major research efforts have been confined to the boiling of single component liquids on horizontal plain tubes. It is appropriate to emphasize that boiling of binary and ternary mixtures has engineering relevance in the design of reboilers and evaporators, which have wide application in the chemical, petrochemical, refrigeration, pharmaceutical, and other allied process industries. The mechanism of liquid mixtures boiling is significantly different from that of single component liquids. This has been explained by Stralen and Cole. In fact, the rate of vapour bubble growth in single component liquid is governed by the heat flow from the heating surface towards the vapour bubble boundary to satisfy the heat requirements of the growing bubbles. However, during boiling of liquid mixtures the heat diffusion is linked with the mass diffusion of the more volatile component of the mixture. Thus, this component gets exhausted rapidly in the liquid phase adjacent to the vapour bubble. Consequently, a low concentration of the more volatile component results thereby slowing down the bubble growth.

Nearly all the studies of mixture boiling deal with nucleate pool boiling of binary liquid mixture as the study of multicomponent mixture boiling is much more tedious because of the large number of experiments required to cover the composition range of all the components. Boiling of multicomponent liquid mixtures is of special practical interest for extractive distillation system where liquid mixtures of widely different volatilities are partly separated in a reboiler. It seems there are three studies in the published literature for boiling of ternary liquid mixtures on plain tube. It is, therefore, necessary that more number of experiments on ternary liquid mixtures should be performed to collect data and thus to support for the development a comprehensive correlation for the prediction of heat transfer coefficient.

The present investigation is an effort in this direction to study experimentally heat transfer during natural convection boiling of acetone-isopropanol-water ternary mixtures at atmospheric pressure.

Experimental Set-up

The experimental set-up used in the present investigation is shown schematically in Fig. 1. More details of test apparatus and experimental procedure are described in Kadhum et al. The plain heating tube used for boiling of liquids was fabricated from a solid brass rod (70% Cu, and 30% Zn). The tube has an axial hole of 18 mm diameter drilled from one end of the rod up to a length of 145 mm, leaving 22 mm undrilled to avoid the longitudinal heat flow. The outer diameter of the tube is 31.2 mm and the effective heated length is 108 mm (the length in which the cartridge heater is housed). The outer surface of the tube has been finished by a 400 grade emery paper to ensure almost identical finish. Four thermocouple holes in the wall thickness of the heating tube have been drilled in a pitch circle diameter of 25 mm. Each hole is of 2 mm diameter having a length of 91
mm. Calibrated copper-constantan thermocouples, \( T_w \) and \( T_l \) are placed inside the wall and in the pool around heating tube corresponding to wall thermocouples positions to measure wall and bulk liquid temperatures, respectively. The tube has been heated using a cartridge heater capable of providing a maximum heat flux of 57,624 W/m². The power supplied to the heater was stabilized by a servo-voltage stabilizer, modulated by an auto transformer and measured by a calibrated precision grade wattmeter. A calibrated sensitive pressure/vacuum gauge, installed on the top of the vessel, measures pressure/vacuum inside it.

Procedure—Double distilled water, reagent grade acetone and isopropanol are used to prepare acetone-isopropanol-water liquid mixtures. The vessel is filled with a given test liquid mixture to a height of 100 mm above the top of the heating tube surface. The heater is set to a heat flux of 57,624 W/m². Deaeration of the liquid pool is carried out by heating the liquid and removing the soluble gases through a bubbler. The stabilization of the heating surface has been done by boiling the liquid for about fifty hours. The procedure for stabilization has always been carried out before experiments with new heating tube-mixture combination. Once the heating tube is stabilized for a mixture the process of data recording is initiated. At steady state, the readings of wall and liquid thermocouples, and wattmeter are recorded. Experiments have been conducted for decreasing the values of heat flux. This procedure is used to avoid the possibility of hysteresis in the boiling curve, as reported by Kartsounes, Jensen and Hsu, and Hahne et al. Experiments have been carried out for various values of heat flux, viz., 52901, 44399, 35897, 27395, 18893, and 10391 W/m². To ensure that the surface of a given heating tube is not fouled, some experiments are repeated to verify it.
While conducting the experiments, samples of liquid mixtures were drawn before and after each experiment to determine their densities to check if any changes in the composition of the liquid mixture had taken place during the boiling. The results showed a negligible variation of within ±0.01 mole fraction.

Data reduction—The heat transfer coefficient for the heating tube having outer surface temperature \( T_{w0} \) and boiling liquid temperature, \( T_i \), is calculated as:

\[
h = \frac{Q}{A (T_{w0} - T_i)} \quad \ldots (1)
\]

where \( T_i \) is the arithmetic average of the bulk liquid temperatures measured at three points around the heating tube by thermocouples and \( A(\pi D_o L) \) is the surface area of the plain tube. The outer surface temperature \( T_{w0} \) is calculated from the value of \( T_{wb} \), which is the arithmetic average of the four wall thermocouple readings, using Fourier’s conduction equation as reported by Hahne et al.\textsuperscript{10} and Webb and Pais\textsuperscript{11}:

\[
T_{w0} = T_{wb} - \left( \frac{qD_o}{2k_1} \right) \ln \left( D_o / D_{in} \right) \quad \ldots (2)
\]

where \( D_{in} \) is the thermocouple pitch circle diameter.

Uncertainty analysis is carried out following the procedure recommended by Schultz and Cole\textsuperscript{12} and the maximum uncertainty in the values of heat transfer coefficients has been estimated to be ±4.6%.

Results and Discussion

To check, whether the experimental set-up is capable of generating reliable and acceptable data, experiments on single component liquids (acetone, isopropanol, and distill water) and binary azeotropic liquid mixture have been conducted. The data thus obtained are compared with well known and widely accepted correlations for single component boiling. For this purpose two correlations have been chosen after a review of the literature, regarding the composition of the intermediate volatile component in vapour phase, it can be either higher or lower than the bulk liquid composition depending on its boiling point, i.e., volatility.

\[
(hD/o_{k}) = 0.225 [qD/o_{k}\mu_{0.69} [p/D/o]^{0.31} \times \left( (\rho - \rho_{o})/\rho_{o} \right)^{0.33} [\mu C_{p}/o_{k}]^{0.69} \quad \ldots (3)
\]

The correlation proposed by Cooper is claimed to be tested against a large number of experimental data and has been reported to give a good fit. Hence, average heat transfer coefficients are computed for the boiling of acetone, isopropanol, and water on plain tube at atmospheric pressure using Cooper’s correlation as presented below:

\[
h = C[((p/p_{o})^{0.12} - 0.2 \log (R_{p})) - \log (p/p_{c})]^{0.55} \quad \ldots (4)
\]

\[
C = 95 \text{ for water, and } C = 55 \text{ for other liquids.}
\]

Recently, Webb and Pais\textsuperscript{14} have recommended the Cooper correlation for the prediction of boiling heat transfer coefficients of pure liquids with the values of \( R_{p} \) varying from 0.3 to 0.6 \( \mu \). The best prediction has been obtained for the present data with \( R_{p} \) equal 0.5 \( \mu \). It is important to note that the Cooper correlation is not applicable for the boiling of azeotropic mixtures and hence the data for azeotropic mixture are not compared with this correlation.

The standard deviation (S.D.) and mean absolute error (M.A.E.) values for both correlations Eqs (3) and (4) are 10.21 and 9.94%, and 10.01 and 8.72% respectively.

From the above data, it is noted that both correlations correlate the present data excellently and the Cooper correlation [Eq. (4)] predicts better than the McNelly correlation [Eq. (3)]. An indirect implication of these results is that present experimental set-up provides reliable data and hence experimental data for mixtures boiling are logically expected to be reliable. The subsequent part of the paper deals with ternary mixture boiling on plain tube.

In a ternary liquid mixture, one of the components is the more volatile having the less boiling point, the other intermediate volatile having the intermediate boiling point and still another component is the least volatile having the highest boiling point. As a result of it, the vapour-liquid compositions differ from each other and is dictated by the phase equilibrium thermodynamics. The vapour phase will have a higher composition of the more volatile component than in the bulk liquid phase, and the corresponding liquid phase will have less of the more volatile component. As regards the composition of the intermediate volatile component in vapour phase, it can be either higher or lower than the bulk liquid composition depending on its boiling point, i.e., volatility.

Nucleate pool boiling curve—Nucleate pool boiling curve has a unique significance as regards the boiling heat transfer. This represents heat flux, \( q \) versus wall superheat \( \Delta T \) plot on log-log scale covering free convection, nucleate pool boiling, transition, and film boiling regions. Since experimental data of present investigation pertain to nucleate pool boiling region a plot made between \( q \) and the boiling heat transfer. This represents heat flux, \( q \) versus wall superheat \( \Delta T \) plot on log-log scale covering free convection, nucleate pool boiling, transition, and film boiling regions. Since experimental data of present investigation pertain to nucleate pool boiling region a plot made between \( q \).
and $\Delta T$ has been made to represent the boiling curve for this region.

Fig. 2 represents nucleate pool boiling curves for the boiling of acetone-isopropanol-water mixtures on plain tube at atmospheric pressure. An inspection of the plot reveals that 'boiling curves' of different compositions of same mixture possess the same characteristic features. It can be further observed from Fig. 2 that the wall superheats required for the boiling of ternary mixtures on plain tube are greater than those required for the boiling of their pure components. All the data points of the ternary liquid mixtures lie below those of pure components, i.e., water, isopropanol and acetone. This established that there is a degradation in boiling heat transfer coefficients of ternary liquid mixtures due to mass diffusion and the variation of thermophysical properties.

In the case of ternary liquid mixture, the degree of degradation in boiling heat transfer coefficient depends on the requirements for the different species (in the mixture) to diffuse to the growing bubble interface from the bulk liquid in order to maintain equilibrium. Thus, it is possible to expect that the degradation in boiling heat transfer coefficients of ternary mixture, $(h_i/h_{id})$ should be a function of $|Y-X|$, which is the summation of $|Y-X|$ of the more and the intermediate volatile components in the mixture as follows:

$$|Y-X| = |Y_1-X_1| + |Y_2-X_2| \quad \ldots (5)$$

The indices 1 and 2 represent the more (acetone) and the intermediate (isopropanol) volatile components, respectively. This degradation in heat transfer coefficient is primarily due to the mass diffusion which increases the liquid temperature near the vicinity of a bubble and impedes the heat from the heating surface to the bubble and also to some extent due to thermophysical properties.

Fig. 3 has been drawn to represent the variation of dimensionless heat transfer coefficient $(h_i/h_{id})$ against $|Y-X|$. The linear mixing law was used to calculate the ideal heat transfer coefficient, $h_{id}$. From the plot it can be noticed that on plain tube the value of $(h_i/h_{id})$ decreases with the increase in the value of $|Y-X|$.

The experimental heat transfer coefficients of the ternary mixtures have been compared with predictions using the correlation for multicomponent mixtures due to Thome and Shakir\textsuperscript{(15)}. The Thome and Shakir correlation has the advantage that it is applicable to binary as well as multicomponent mixtures. In that correlation the boiling range factor, $\Delta T_{BR}$ takes into account the effects of phase equilibrium and in other words the deteriorative effect of mass diffusion on heat transfer coefficient. Thome and Shakir correlation is reproduced below:

$$(h_i/h_{id}) = \left[1 + \frac{h_{id}}{q} \Delta T_{BR} \right]^{-1} \left[1 - \exp \left(-\frac{q}{\rho} \beta_1 h_{id} \right) \right]^{-1} \quad \ldots (6)$$

They recommended the Stephan and Abdelsalam\textsuperscript{(16)} correlation for calculating the value of $h_{id}$ of Eq.(6). Stephan-Abdelsalam have used regression analysis of more than 5000 data points collected from 72 papers. The boiling range, $\Delta T_{BR}$ is the difference between the dew point and bubble point temperatures at a given bulk liquid composition. The mass transfer coefficient, $\beta_1$ in the liquid phase has been kept equal to 0.0002 m/s and $B$, an empirical scaling parameter, which accounts for the fraction of heat consumed for the bubble formation, has been set equal to 1.0 as used by Schlunder\textsuperscript{(17)} in his correlation.

It is worth-mentioning that the value of liquid mass transfer coefficient in the above correlation
is very difficult to predict for liquid mixtures as it requires a priori knowledge of the mass diffusivity. Since Schlunder has observed that its value typically ranges from about 0.0001 to 0.0005 m/s for physical and chemical gas absorption, it seems reasonable to choose the midpoint value of 0.0003 m/s as a fixed value instead of 0.0002 as recommended by Thome. Since there is a very limited amount of data available for nucleate pool boiling of multicomponent mixtures, the accuracy and range of applicability of correlations are not well defined.

The predictions of nucleate pool boiling heat transfer coefficient values, $h_{\text{pred}}$ from Eq. (6) have been compared with the experimental ones, $h_{\text{expl}}$ of the present investigation in Fig. 4 for the boiling of acetone-isopropanol-water ternary mixtures on plain tube at atmospheric pressure. Good agreement between predicted and experimental values with standard deviation, (S.D.) and mean absolute error, (M.A.E.) of 6.03 and 5.16, respectively were observed.

The procedure for evaluation of the thermophysical properties of acetone-isopropanol-water ternary mixtures and data have been reported elsewhere in Kadhum.

Conclusions

Data for nucleate pool boiling of acetone-isopropanol-water ternary mixtures of various compositions at atmospheric pressure were obtained for plain tube. Based upon the results obtained, the following conclusions may be made:

1. The wall superheat required for the boiling of any composition of acetone-isopropanol-water ternary mixture is greater than that either of pure components.

2. The degradation in the boiling heat transfer coefficients for the present ternary mixtures increases with increase in the value of the summation of the vapour-liquid mole fraction difference of the more volatile component $|Y - X_I|$, i.e., acetone and that of the intermediate volatile component $|Y_2 - X_2|$, i.e., isopropanol.

3. The correlation due to Thome and Shakir for multicomponent mixtures predicts the experimental heat transfer coefficients of ternary mixtures for plain tube at atmospheric pressure with a maximum deviation of ±15%.

Nomenclature

- $A$ = surface area, m$^2$
- $B_0$ = empirical scaling factor
- $C_p$ = liquid specific heat, J/kg K
- $D$ = diameter of heating tube, m
- $d$ = bubble departure diameter, m
- $h$ = average heat transfer coefficient, W/m$^2$K
- $h_0$ = ideal heat transfer coefficient from Eq. (6), W/m$^2$K
- $h_b$ = differential latent heat of vaporization, J/kg
- $k$ = thermal conductivity, W/mK
- $L_e$ = effective heated length, m
- $M_w$ = molecular weight, g/mol
- $p$ = pressure, kPa
- $Q$ = input power, W
- $q$ = heat flux, W/m$^2$
- $R_s$ = average surface roughness, μm
- $T$ = average temperature, K
- $\Delta T$ = temperature difference, K
- $X$ = mole fraction in liquid phase
- $Y$ = mole fraction in vapour phase
- $\rho$ = density, kg/m$^3$
- $\sigma$ = surface tension, N/m
- $\mu$ = dynamic viscosity, N.s/m$^2$
- $\alpha$ = thermal diffusivity, m$^2$/s

Subscripts

- b = bubble point
- BR = boiling range, ($= T_d - T_b$), K
- d = dew point
- expl = experimental
- i = inner
- l = liquid
- o = outer
- pred = predicted
- s = bulk liquid
- t = tube
- th = pitch circle diameter of wall thermocouples
- v = vapour
- wo = outer wall
- 1 = more volatile component
- 2 = intermediate volatile component
- 3 = less volatile component

References