Heat transfer in boiling of mixtures in a thermosiphon reboiler

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Boiling heat transfer data for water, methanol and their seven binary mixtures are presented. The heat transfer section consisted of an electrically heated stainless steel tube of 25.56 mm i.d. and 1900 mm length. The heat flux values ranged from 4.1 to 4.30 kW/m². The liquid submergence levels were maintained at 100, 75, 50 and 30 per cent. All the data were generated at atmospheric pressure. A significant reduction in the heat transfer coefficient is observed for binary mixtures attributable to the effect of mass diffusion on heat transfer. Possible reasons for the low heat transfer rates in mixtures are suggested in relation to bubble growth and nucleation.

Vertical thermosiphon reboilers with boiling of mixtures on the tube side are common process equipment in the petroleum and chemical industries. The process fluid entering the tubes of heat exchanger gets heated and moves upwards due to density difference between the liquid and boiling two phase fluid. The boiling of liquids in vertical tubes of closed loop circulation systems as above, finds its application also extending to refrigeration, power plants and nuclear reactors.

The nucleate boiling heat transfer coefficient in binary or multicomponent liquid mixtures is strongly influenced by the composition of the mixture. The heat transfer coefficient were found to be lower than that from the pure component liquids. Similar observations were also made in flow boiling of liquid mixtures. With natural circulation boiling of liquid mixtures, as encountered in thermosiphon reboiler, therefore, the composition becomes an additional controlling parameter.

The available data on boiling of liquid mixtures give insufficient and inaccurate information about the local heat transfer coefficient and circulation rate under the influence of governing parameters. In spite of the industrial importance of the design of reboilers for distillation columns, the studies on thermosiphon boiling of liquid mixtures in vertical tubes have been reported only by a few workers.

Therefore, the present experimental study, was undertaken to obtain such data for natural circulation boiling of binary mixtures and to study the effect of heat flux and submergence on heat transfer to boiling binary mixtures in a single vertical tube thermosiphon reboiler.

Experimental Procedure

The main unit was a U-shaped circulation loop made of two long vertical tubes and one short horizontal tube. The experimental reboiler and cooling system used in this study was described in detail elsewhere. In order to monitor the heat transfer surface temperatures along the tube length, twenty one copper-constantan thermocouples were spot welded on the outer surface of the tube at intervals of 50 mm upto a length of 200 mm from the bottom end and 100 mm over the remaining length. A copper-constantan thermocouple probe was placed in the viewport to measure the inlet liquid temperature. The temperature of the boiling liquid before entry to the vapour-liquid separator was measured by another traversing thermocouple probe. The entire set up was thoroughly lagged with asbestos rope and glass wool and finally covered with a thin aluminium sheet to reduce the heat losses, which were less than ± 2.5 per cent.

For conducting a series of runs, the test liquid was boiled off for about 8-10 h to remove the last traces of dissolved air which was indicated by the disappearance of the air bubbles in the bubbler. Then a desired heat flux was impressed and cooling water flow rate to the condenser was regulated to give a maximum temperature rise consistent with no loss of vapour due to inadequate condensation. The liquid level in the down-flow pipe was adjusted by adding/dRAINing the requisite amount of test liquid. When steady state conditions were established, the readings of the ammeter, voltmeter, thermocouples and rotameters were recorded. The liquid level in the downflow pipe was ob-
Table 1—Range of experimental parameters

<table>
<thead>
<tr>
<th>System</th>
<th>$q \times 10^{-4}$, W/m²</th>
<th>$S$, %</th>
<th>$\Delta T_{sub}$ °C</th>
<th>Concentration of more volatile component, wt, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distilled water</td>
<td>0.57-4.3</td>
<td>30, 50</td>
<td>0.5-4.6</td>
<td>--</td>
</tr>
<tr>
<td>Methanol</td>
<td>0.41-2.1</td>
<td>30, 50</td>
<td>1.0-3.7</td>
<td>--</td>
</tr>
<tr>
<td>Methanol-Water</td>
<td>0.57-2.9</td>
<td>30, 50</td>
<td>0.5-4.6</td>
<td>5, 10, 18, 26, 30, 38, 58</td>
</tr>
<tr>
<td></td>
<td></td>
<td>75, 100</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

served. The maximum liquid head used in the present study corresponded to the liquid level equal to the top end of the reboiler tube. This condition has been termed as 100% submergence. The experimental data were generated for four different levels of liquid submergence and various heat fluxes. The liquid samples from the inlet of the reboiler tube, the vapour-liquid separator and the condensed vapour were withdrawn with the help of a specially designed double walled sampling device made of glass through drain cocks $C_1$, $C_2$, $C_3$, and $C_4$, respectively. The temperature between the two walls of sampling device was maintained at 0°C by placing melting ice in it. This arrangement proved quite effective for instant and complete cooling of the sample and thus avoiding the losses of vapour. After cooling the samples, the inner tube was taken out and kept in a refrigerator. The refractive index of the samples were determined and the composition read from the refractive index composition chart of the system under study. A similar procedure was followed for various compositions of binary liquid mixture.

Distilled water and methanol were used to prepare binary mixtures of methanol-water of required compositions. The composition of binary mixtures were judiciously selected based on the behaviour of $(y-x)$ versus $x$ plot of the system. The operating parameters with each binary liquid mixture were heat flux, submergence and inlet liquid subcooling. The mixture concentrations and range of other experimental parameters studied are tabulated in Table 1.

Based on thermal equilibrium mode the circulation rates and liquid bulk temperature distribution in the thermosiphon reboiler have been determined by making a heat balance on the test section. In order to determine the liquid circulation rate, it was required to know the length of effective non-boiling or sensible heating region over which the liquid temperature varied linearly. The effective boiling and non-boiling zones over the entire heated length were determined from the amount of net vapour generation. This could be

![Fig. 1—Variation of wall temperature along the tube length with heat flux as parameter for methanol-water system](image1)

![Fig. 2—Variation of wall temperature along the tube length with submergence as parameter for methanol-water system](image2)
obtained by the vapour condensed in the condenser. The details have been discussed earlier. The vapour fraction of two-phase mixture at the exit of test section is:

\[ x^o = \frac{M_v}{m} \]  \hspace{1cm} \ldots (1)

The local heat transfer coefficients in boiling as well as non-boiling sections were calculated by dividing wall heat flux with local values of temperature difference between the wall and liquid as expressed by equation:

\[ h = \frac{q}{(T_w - T_L - \Delta T_w)} \]  \hspace{1cm} \ldots (2)

where the temperature drop, \( \Delta T_w \), between the thermocouple bead and the inside surface was estimated using the equation of conductive heat transfer with internal heat generation for cylindrical wall.

The length-mean values of heat transfer coefficient in the boiling region were calculated as given below:

\[ \bar{h}_B = \frac{q}{(\Delta T_w)_{lm}} \]  \hspace{1cm} \ldots (3)

Results and Discussion

Figs 1 and 2 shows the typical variation of wall temperature with heat flux and liquid submergence as parameters for methanol-water system. The wall temperature, \( T_w \), rises at a fast rate with \( Z \) from its inlet (lower) end up to a point beyond which a steep fall sets in followed by a gradual decrease, over the remaining portion of the heated tube. However, the curves at higher heat flux values get shifted to higher wall temperatures. The typical behaviour, as observed above, remaining the same at other concentrations of liquid mixtures, the values of wall temperatures, location of peak values and the lengths of various zones are different. The location of wall temperature peaks get shifted towards the tube inlet and the curves move to lower values of \( T_w \) as the liquid submergence is reduced from 100 to 30 per cent. At the submergence of 100 and 75 per cent, all the regimes of heat transfer are present while at lower submergences of 50 and 30 per cent the initial portions of curves before peak are almost absent. The liquid temperature increases linearly along the tube length as observed for pure liquids, up to a point where it attains the saturation value (bubble point). The liquid temperature remains saturated with its temperature increasing at a very low rate. This may be attributed to the vapourization of liquid accompanied with the decrease in the concentration of low boiling component as the liquid moves upwards. The bubble point thus increases along the tube length inspite of the reduction in hydrostatic head.

The typical variation of wall and liquid temperatures as observed indicates that there exist different regimes of heat transfer in a reboiler tube. The linear rise in the temperature of liquid as it moves upwards through the tube results from sensible heating under uniform heat flow. Since the degree of subcooling in the present study is small, the wall temperatures attain values above saturation right in the close vicinity of tube inlet. When the minimum wall superheat required is attained, the bubbles start nucleating at the surface but collapse there due to the presence of subcooled liquid core. The onset of subcooled boiling thus creates additional turbulence at the surface. This explains why the linearly increasing wall temperature corresponding to convective heat transfer, starts varying at decreasing rate eventually becoming zero at peak values. Once the bulk liquid temperature attains saturation value, the bubbles generated at the surface grow to their maximum size and get detached resulting in the existence of vapour phase in the tube. All the heat supplied
The typical variation of heat transfer coefficient along the test section length with heat flux as parameter has been shown in Fig. 4. At approximately the same value of inlet liquid subcooling and submergence, the characteristic behaviour of curves for higher values of heat flux get shifted to higher heat transfer coefficients than those for lower heat flux values. As the heat flux is raised, the initial portions of the curves (decreasing and slowly rising) get shortened shifting the point of steep rise towards tube inlet. Fig. 5 shows an effect of submergence on the variation of $h$ with $Z$ at a constant heat flux. The curves, at the same value of heat flux, generally shift to higher values of heat transfer coefficient as the submergence is lowered from 100 to 30 per cent.

The characteristic behaviour of $h$ versus $Z$ curves are indicative of heat transfer mechanism governed by various parameters. The initial portions of curves with $h$ decreasing may be assigned to the entrance effect. This is observed to be pronounced at low heat flux values and / or high submergence where the heat transfer is dominantly convective. The increase in $h$ with $Z$ at a slow rate, after the entrance effect may also be due to the convective heat transfer followed by progress-
ively increasing subcooled boiling. The steep rise in the values of heat transfer coefficient corresponds to the onset of saturated boiling. The enhancement of transfer coefficient at varying rates along the tube length may be accounted for by the additional turbulence due to nucleation, growth and detachment of vapour bubbles resulting in two phase with different flow patterns. With increase in the value of heat flux larger number of nuclei for bubble generation become active enhancing the heat transfer coefficient. The changes in the characteristic behaviour of transfer coefficient profiles with submergence seems mainly due to the interaction between circulation rate and bubble dynamics in the tube. The circulation rate increases with $S$, resulting in the dominant convective heat transfer with appearance of entrance effect and slowly rising $h$ regions. At low submergences, the circulation rate falls and the nucleate boiling dominates showing its effect as observed and discussed. The point, where saturated boiling sets in, gets shifted to a lower level in the tube as the submergence is reduced from 100 per cent probably due to the change in circulation rates. The decrease in the value of liquid submergence reduces the driving force for liquid circulation and hence its rate through reboiler tube. At a lower rate of liquid circulation the rate of change in its temperature along the tube length becomes higher and the saturation temperature is attained at much smaller length from the inlet. When the submergence is low, the net flow of liquid is small and local circulation between the tube wall and liquid bulk dominates resulting in a condition similar to pool boiling.

Fig. 6 shows the effect of mixture concentration on variation of $h$ with $Z$, at same heat flux and liquid submergence. While the main feature making the shape of the curves are same at all concentrations as observed earlier, the relative magnitudes, positions of boiling incipience and nature of variations change with composition. The change in the behaviour of heat transfer coefficient profiles for binary mixtures may be attributed mainly to the physical properties of mixtures being different from those of their constituents and presumably the effect of mixture composition on the mechanism of the boiling process. Further, with the progress of vaporization, the larger quantities of more volatile component go into the vapour phase reducing its concentration into the liquid phase, the saturation temperature increases and the above effect is intensified. But the higher concentration of vapour phase than that of the coexisting liquid phase in the tube presents the necessary concentration gradient for mass diffusion which might affect adversely the bubble growth rate. Thus the
variations of \( h \) with \( Z \) for binary systems are governed by all the parameters involved for pure components and the additional effect of mass diffusion on bubble growth rate in a very complex manner.

The average values of heat transfer coefficient for saturated boiling of binary mixtures have been plotted against their concentrations in Figs 7 and 8, respectively. These figures clearly demonstrate that \( h_B \) decreases with increase in concentration of more volatile component in the mixture up to a certain value beyond which it increases showing a minima. The general nature of variation for a system remains essentially the same at all heat fluxes and liquid submergences. The shifting of curves with \( q \) and \( S \) follows the same trend as observed earlier for pure components and mixtures. The mixture concentration at which the heat transfer coefficient becomes minimum is nearly the same at all heat fluxes and submergences for a system. The experimentally obtained values of \( h_B \) for mixtures are invariably lower than their weighted average at any given concentration. The difference between the two values is maximum at the concentration corresponding to the minimum value of \( h_B \) of a system.

The reduction in the values of heat transfer coefficient for mixtures as compared to the weighted average of the constituent components may be attributed to the change in properties on mixing and the effect of mass diffusion on boiling process. When a vapour bubble forms and grows in a liquid mixture, the concentration of more volatile component in the vapour bubble, which is in equilibrium with the surrounding liquid, becomes higher than that in the surrounding liquid. The difference between the vapour and liquid phase compositions \((y-x)\) provides the necessary concentration gradient for the diffusion of more volatile component from the bubble to the liquid. This process being just opposite of the bubble growth and hence the rate of bubble growth gets retarded. The micro-convective contribution to heat transfer, thus gets reduced resulting in a lower heat transfer coefficient than the expected value of weighted average. The rate of diffusion and hence the retardation of bubble growth is maximum at a concentration at which \((y-x)\) is maximum. This is the reason why the values of \( h_B \) are minimum at the concentrations which are almost same as those corresponding to the maximum value of \((y-x)\).

**Conclusion**

The values of heat transfer coefficient during boiling of binary mixtures are generally lower than the weighted average of the pure component values. This reduction may be due to the expected retardation of bubble growth rate by the diffusion of more volatile component from the vapour bubble to the saturated liquid.

**Nomenclature**

- \( h \): heat transfer coefficient, W/m²·°C
- \( \bar{h} \): average heat transfer coefficient, W/m²·°C
- \( h_B \): average boiling heat transfer coefficient, W/m²·°C
- \( m \): circulation rate, kg/s
- \( M_L \): liquid flow rate from condenser, kg/s
- \( q \): heat flux, W/m
- \( S \): submergence, %
- \( T \): temperature, °C (K)
- \( \Delta T \): temperature difference, \((T_a - T_L)\), °C
- \( \Delta T_{ob} \): degree of subcooling, \((T_s - T_L)\), °C
- \( x^* \): vapour fraction.
- \( x \): mass fraction of component in liquid phase
- \( y \): mass fraction of more volatile component in vapour phase
- \( Z \): distance along the test section, m

**Subscripts**

- \( B \): boiling
- \( i \): inlet
- \( L \): liquid
- \( im \): logarithmic average
- \( o \): outlet
- \( OB \): onset of boiling
- \( w \): wall
References