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International Journal of Heat and Mass Transfer 50 (2007) 3774–3779

International Journal of HEAT and MASS TRANSFER

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Marangoni condensation of steam–ethanol mixtures on a horizontal tube

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Received 1 August 2006; received in revised form 13 February 2007
Available online 12 April 2007

Abstract

For condensation of binary mixtures where the less volatile constituent has the higher surface tension the condensate film is potentially unstable and various ‘‘modes’’ of condensation are seen depending primarily on composition and vapour-to-surface temperature difference. Of particular interest is the so-called ‘‘pseudo-dropwise’’ mode of condensation where the appearance of the condensate closely resembles that of dropwise condensation of a pure fluid on a hydrophobic surface. In recent years the so-called Marangoni condensation problem has been studied experimentally in great detail by Utaka and co-workers [4–8,11] for condensation of steam–ethanol mixtures on small vertical plane surfaces. It has been found that very small concentrations of ethanol in the liquid phase can give rise to significantly larger heat-transfer coefficients than found with pure steam.

In the present investigation the problem has been studied using a horizontal condenser tube. Heat flux and vapour-to-surface temperature difference have been measured for steam–ethanol mixtures over a wide range of composition at atmospheric pressure. The results show the same trends as those found by Utaka for vertical surfaces. Differences in detail can be explained by geometry considerations and strong dependence of heat-transfer coefficient on vapour-to-surface temperature difference and vapour velocity, both of which vary around the perimeter of the horizontal tube.

1. Introduction

Condensation of vapour mixtures has been widely studied for many years. In most cases the focus has been on the diffusion process in the vapour phase which results in the so-called mass-transfer resistance and diminution of the heat transfer. The vapour phase convection-with-diffusion process in forced and free convection of binary mixtures is now well understood (see Fujii [1]). For certain binary mixtures a mode of condensation, whose appearance resembles that of dropwise condensation of a pure vapour on a hydrophobic surface, has been observed (Mirkovich and Missen [2], Fujii et al. [3], Utaka and co-workers [4–8,11]. Most notably it has been found that, notwithstanding the vapour-phase diffusion resistance and with quite small velocity (0.4 m/s), vapour-side heat-transfer coefficient enhancement up to around 8 times can be obtained by adding very small amounts (0.5% or less) of ethanol to the boiler feed water (Utaka and Wang [8]).

So-called Marangoni or pseudo-dropwise condensation may occur when the more volatile constituent has the smaller surface tension e.g. an ethanol–water mixture. An explanation for this behavior has been given by Hijikata et al. [9] who pointed out that in these circumstances the condensate film is potentially unstable. Any deformation in the film would be expected to give a lower film surface temperature (nearer that of the wall) where the film is thinner and a higher temperature (nearer that of the vapour) at a crest. For phase equilibrium at the interface the less volatile constituent is then more concentrated in the liquid at a crest than in a valley. The surface tension gradient in this

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0017-9310/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ijheatmasstransfer.2007.02.011

Nomenclature

C_{iL}	ethanol mass fraction in water at room temperature
C_L	ethanol liquid mass fraction during condensation
C_V	ethanol vapour mass fraction during condensation
q	heat flux
T_v	vapour temperature
T_w	wall temperature
$T_{w,ave}$	mean wall temperature
$T_{w,top}$	wall temperature at top of tube
α	heat-transfer coefficient, $q/\Delta T$
α_{top}	$q/\Delta T_{top}$
ΔT	vapour-to-surface temperature difference $T_v - T_{w,ave}$
ΔT_{top}	vapour-to-surface temperature difference at top of tube $T_v - T_{w,top}$
ϕ	angle measured form top of tube

case causes liquid to be drawn to crest and the irregularity in the film to grow. When pseudo dropwise condensation occurs the effect of composition on surface tension presumably outweighs the effect of temperature (generally lower surface tension at higher temperature for fixed composition) and the pressure gradient resulting from change of interface curvature.

For methanol–water and ethanol–water mixtures Fujii et al. [3] found that the condensate film took different appearances according to the prevailing conditions (composition, heat flux, pressure). The dropwise appearance was generally found at higher water concentrations and lower condensation rates. At higher ethanol concentrations, where the mixture tends to azeotropic behavior the appearance became that of a smooth film. Between these extremes an irregular streaky appearance and a more regular "ring" appearance (vertical condensate ridges around the tube) was seen. The different modes presumably are governed by the balance of the forces mentioned above. Reported heat-transfer coefficients for the condensate film (i.e. excluding gas-phase diffusion resistance) were highest (around 5 or 6 times higher than Nusselt [10] theory values) for the drop regime.

More recently Utaka and co-workers have studied this phenomenon. In a series of experiments using small vertical plane surfaces the dependence of the vapour-to-surface heat-transfer coefficient on vapour composition (Utaka and Wang [6]), surface subcooling (Utaka and Terachi [4]) and cross flow velocity (Utaka and Kobayashi [7]) have been systematically investigated. Utaka and Nishikawa [11] used a laser light absorption technique to measure the film thickness over time at a given location on a surface during condensation of an ethanol–water mixture. It was confirmed that a film of thickness at least 1 μm was always present. With fixed vapour composition and velocity, the heat-transfer coefficient was found to be relatively low at small surface subcooling and subsequently to increase steeply before decreasing again as the mode of condensation progressed to a smooth film with increased subcooling. The effect of velocity was to raise the heat-transfer coefficient, presumably due to reduction in the gas-phase diffusion resistance as well as to the shear stress effect on the condensate surface. Under optimum conditions (low ethanol concentration and high vapour velocity) heat-transfer coefficients (including diffusion resistance in the vapour) up to 8 times those found for pure steam were reported.

2. Apparatus and procedure

The stainless-steel test apparatus, shown schematically in Fig. 1, consisted of a closed loop, with vapour generated in an electrically-heated boiler (maximum power 12 kW). The vapour was directed vertically downward through a calming section before flowing over the horizontal, water-cooled, test condenser tube. Excess vapour passed to an auxiliary condenser from which the condensate returned to the boiler by gravity. The copper test condenser tube had an outside diameter of 12.2 mm, active heat-transfer length 90 mm and was fitted with four embedded wall thermocouples located as shown in Fig. 2. PTFE sleeves were

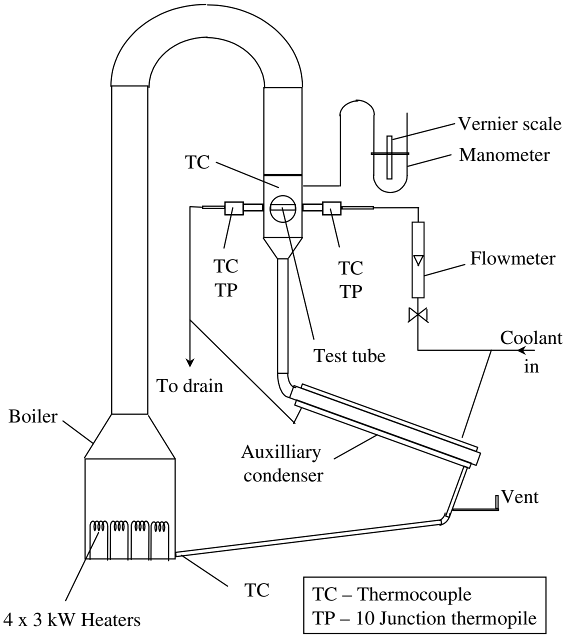

Fig. 1. Apparatus.

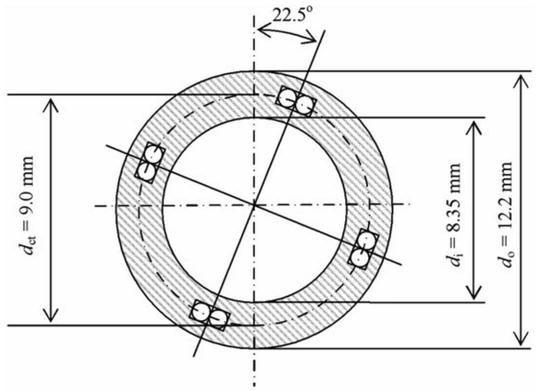

Fig. 2. Location of thermocouples in test tube wall.

inserted at both ends of the tube so that the cooled length of tube was the same as that exposed to vapour. Before installation the tube was thoroughly cleaned with a mixture (distilled water (2000 ml), sulphuric acid (100 ml) and sodium dichromate (200 g)) before rinsing with distilled water. Film condensation was always obtained when using pure steam. All tests were done at a little above atmospheric pressure.

The cooling water temperature rise was measured using a ten-junction thermopile. Care was taken to ensure adequate coolant mixing prior to temperature measurement and sufficient isothermal immersion of the thermopile leads in the vicinity of the junctions. The heat flux was found from the coolant flow rate and temperature rise. A small predetermined correction (dependent on coolant flow rate) for the dissipative temperature rise of the cooling water in the tube and mixing boxes was incorporated in the calculation of the heat-transfer rate. The surface temperatures were those indicated by the embedded thermocouples with a small correction for the depth of the thermocouples below the condensing surface. The vapour velocity at approach to the test section was found from the measured power input to the boiler (together with the condensate return temperature) with a small, predetermined correction for the heat loss from the well-insulated apparatus (see Lee and Rose [12]). All thermocouples were calibrated in a high-precision constant temperature bath against a platinum resistance thermometer, accurate to 0.005 K. The accuracy of the thermo e.m.f. measurement was 2 μV, equivalent to 0.005 K for the ten-junction thermopile. The coolant flow rate was measured with a float-type flow meter accurate to 2%.

Tests were conducted using mass fractions of ethanol (in the water at room temperature prior to testing) of 0.05%, 0.1%, 0.5% and 1.0%. Vapour velocities at approach to the condenser tube were 0.15, 0.24, 0.35, 0.56 and 0.75 m/s. For each composition and velocity, measurements were taken for a range of coolant flow rates. The coolant inlet temperature was near to 20 °C in all cases.

Full details of the apparatus and procedure are given by Murase [13].

3. Results

3.1. Visual observations

With increasing coolant flow rate the appearance of the condensate was generally observed to progress from film condensation with small ripples at the lowest vapour-to-surface temperature differences, through a range of flow rates where pseudo-dropwise condensation prevailed and subsequently, at the highest coolant flow rates and temperature differences, back to film condensation. Complete film condensation was only seen at the highest vapour-to-surface temperature differences and with low vapour velocity. Under certain conditions the mode of condensation was pseudo dropwise on the upper part of the tube (where the temperature difference was least) and wavy film condensation on the lower part.

3.2. Heat transfer results

The results are shown in Fig. 3 in the form of graphs of heat flux versus vapour-to-surface temperature difference based on the arithmetic mean surface temperature. Separate figures are shown for each vapour approach velocity and points showing the different compositions are distinguished on each graph. Also shown are the Nusselt [10] equation for pure quiescent steam and an equation given by Rose [14] based on the model of Shekriladze and Gomelauri [15] which takes account of vapour velocity. The heat fluxes are generally significantly higher, particularly at higher vapour velocities, than the pure vapour values. It is also seen that, at higher ethanol concentration and low ΔT, the heat flux is lower than the calculated values for pure steam. In all cases the rate of increase of heat flux decreases at the higher ΔTs (denoting a fall in heat-transfer coefficient). In the intermediate range of ΔT the heat flux is sensitive to composition, with higher heat fluxes at lower ethanol concentrations. The sensitivity to composition becomes less at higher values of ΔT.

In Fig. 4 the same data are plotted with separate figures for each composition and different symbols representing different vapour velocities. The trends mentioned above are again apparent. It is seen that the heat flux increases strongly with velocity except at the lower values of ΔT.

In Fig. 5 heat-transfer coefficients are plotted where it may be seen that at the lowest ethanol concentration (mass fraction in the liquid at room temperature 0.05%) and higher vapour velocities, the maximum heat-transfer coefficient, occurring at ΔT around 7 K, was almost four times that for pure steam.

4. Discussion

The dependence of heat transfer on temperature difference and composition is evidently related to change in mode of condensation. Vapour velocity enhances heat

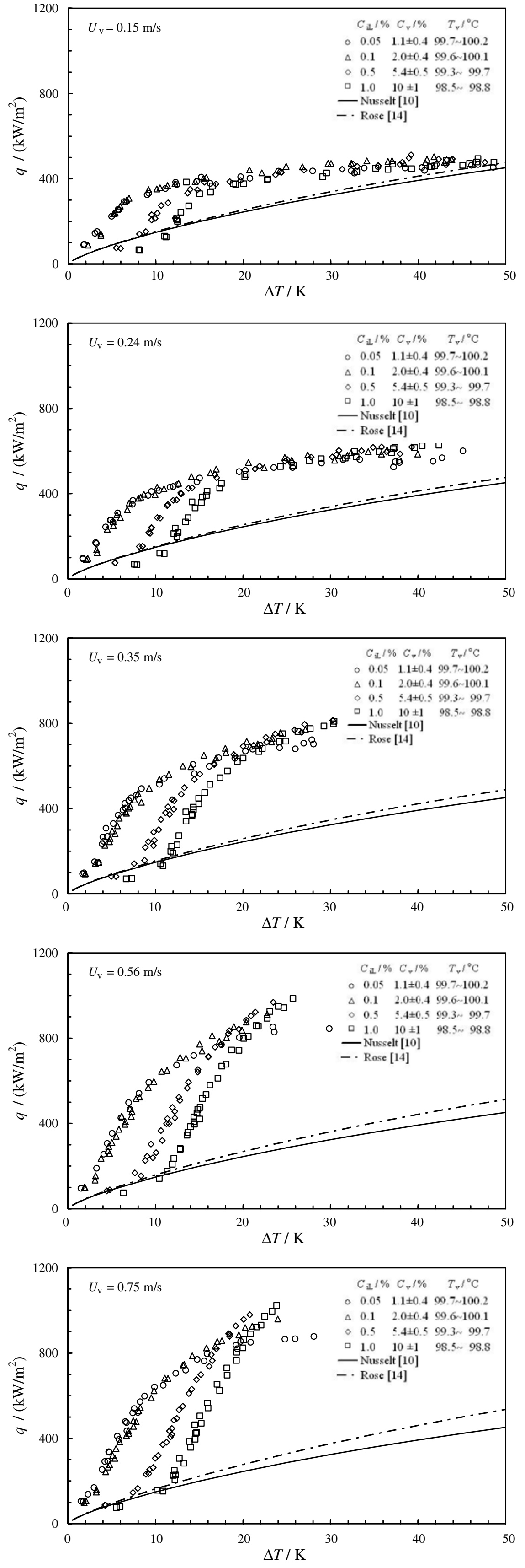

Fig. 3. Heat flux versus vapour-to-surface temperature difference for different vapour approach velocities.

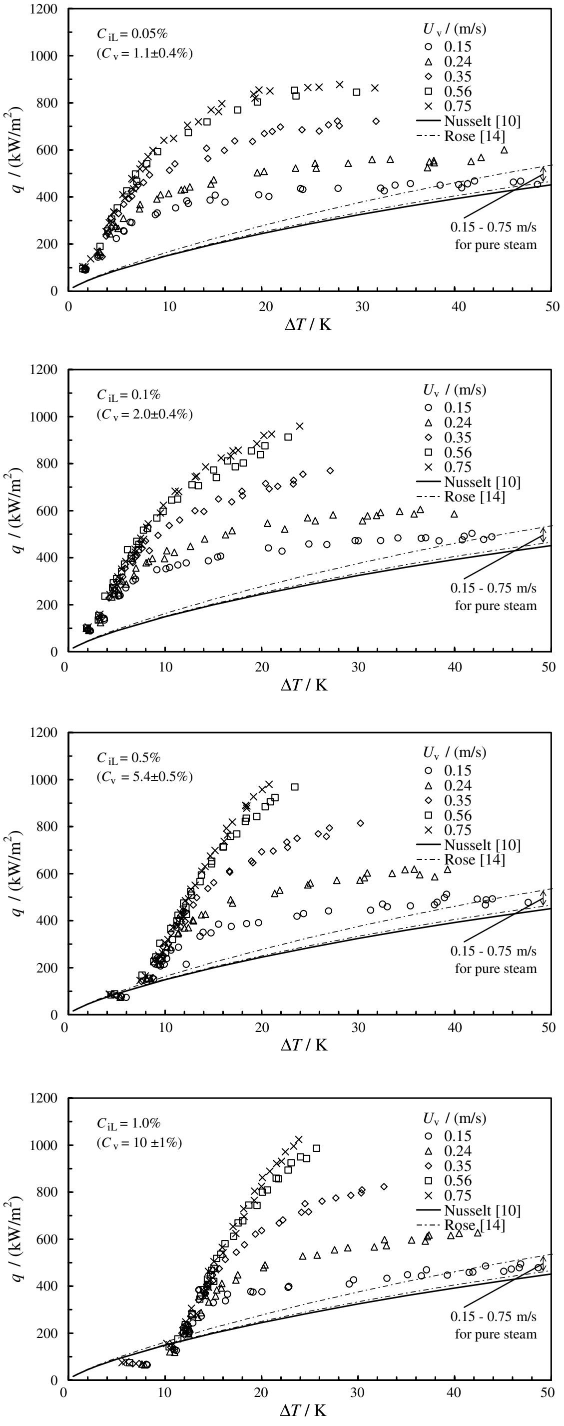

Fig. 4. Heat flux versus vapour-to-surface temperature difference for different vapour compositions.

transfer by reducing the diffusion resistance in the vapour phase as well as by affecting the motion of the condensate. Similar trends to those indicated above have been found for small vertical plates by Utaka and co-workers. Fig. 6 show the present results and those of Utaka and Wang [6] results for nominally similar conditions (C_v and U_v). Differences in detail and higher heat-transfer coefficients in the earlier work may be in part due to difference in

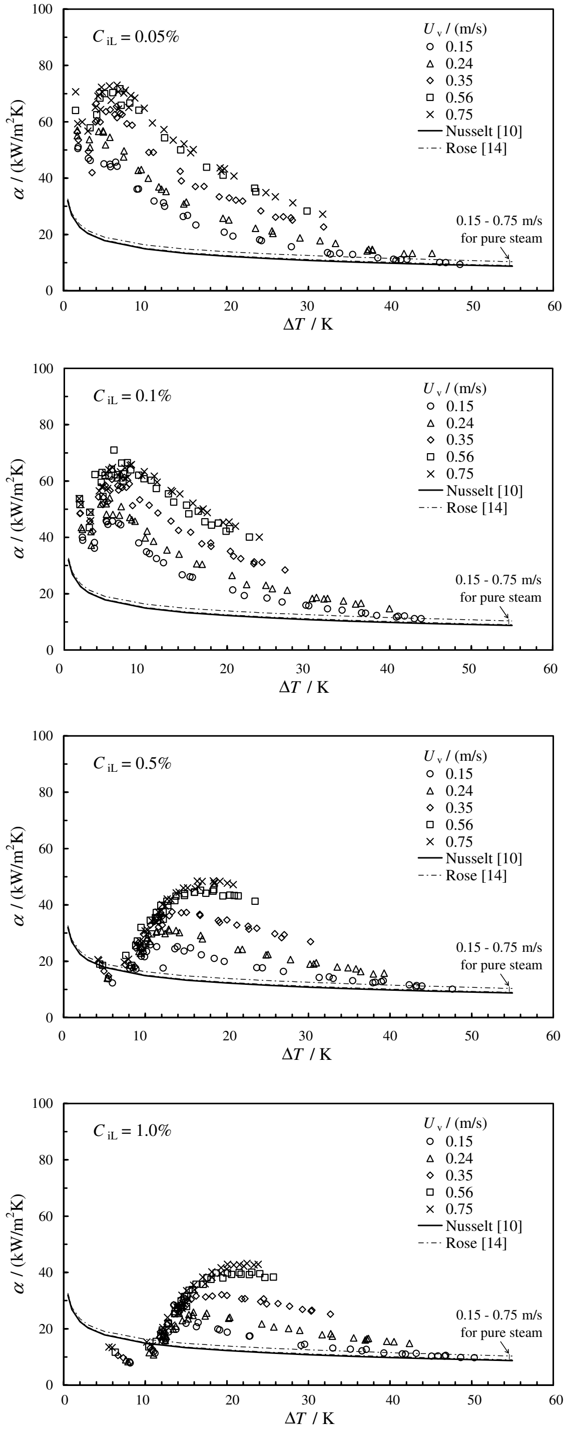

Fig. 5. Heat-transfer coefficient versus temperature difference for different compositions.

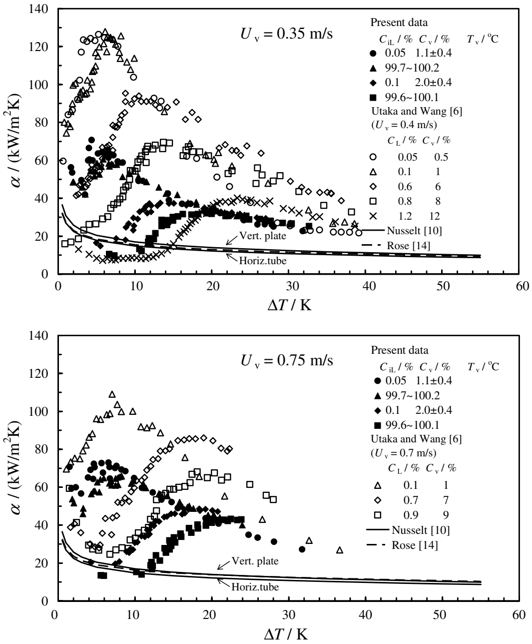

Fig. 6. Comparison of present data with Wang and Utaka [6].

geometry (small vertical plate versus horizontal tube) and consequent effect on condensate drainage (the ratio of heat-transfer coefficients, according to Nusselt theory, for the vertical plate used by Utaka and Wang [6]) to the horizontal tube used in the present work is around 1.9). Also significant is the fact that the heat-transfer coefficient depends strongly on vapour velocity and vapour-to-surface temperature difference, both of which are essentially uniform over the small plane surface and both vary strongly around the horizontal tube. For the tube the velocity varies from zero at the top stagnation point to around twice the approach velocity at an angle from the top of the tube of approximately $\pi/2$, and further around the tube is zero following separation of the vapour boundary layer. The surface temperature was found to vary in a sinusoidal fashion from a maximum at the top to a minimum at the bottom of the tube as illustrated in Fig. 7, giving a corresponding variation of ΔT around the tube. To demonstrate the sensitivity of heat-transfer coefficient to local surface temperature the data of Utaka and Wang [6] are compared in Fig. 8 with the present data for the same conditions as in Fig. 6. Here the present heat-transfer coefficients have been calculated using the temperature difference at the top of the tube. The two sets of data are more closely in accord. (Note that α_{top} is not strictly the heat-transfer coefficient at the

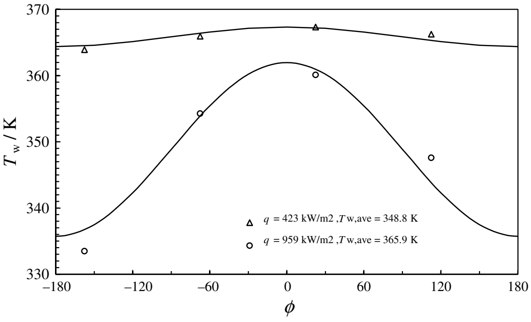

Fig. 7. Specimen surface temperature distributions $C_{iL} = 0.1\%$, $U_v = 0.75$ m/s.

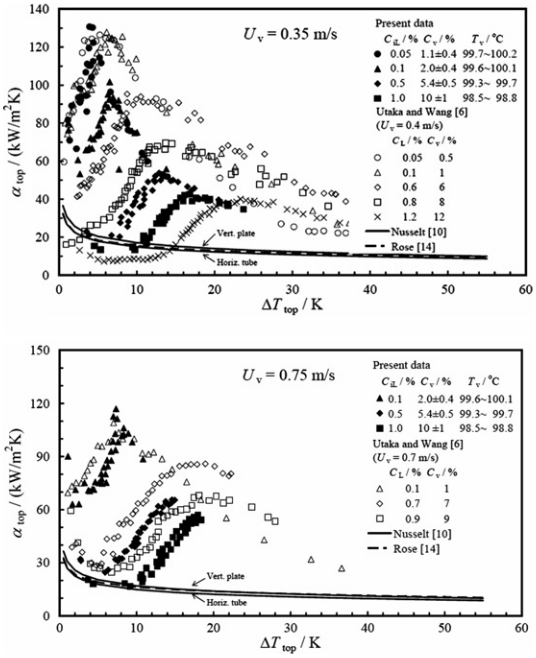

Fig. 8. Comparison with vertical plate data of Wang and Utaka [6] using vapour-to-surface temperature difference at the top of the tube.

top of the tube since it is based on the mean heat flux for the whole tube).

5. Conclusions

Data for condensation of steam–ethanol mixtures on vertical plane surfaces and horizontal tubes show that significant heat-transfer enhancement may be obtained through addition of very small amounts of ethanol to the boiler feed water. The trends found for the horizontal tube are the same as those found earlier for a small plane surface. Differences in detail are due geometry effect on condensate flow as well as variation in vapour velocity and temperature around the tube. The mechanism of Marangoni or pseudo-dropwise condensation is understood in principle but no methods are available for prediction of heat transfer.

A preliminary report on the work described here was presented [16] at the 17th International Symposium on Transport Phenomena, Toyama, Japan (2006).

References

[1] T. Fujii, Theory of Laminar Film Condensation, Springer-Verlag, 1991.
[2] V.V. Mirkovich, R.W. Missen, Study of condensation of binary vapors of miscible liquids, Canadian J. Chem. Eng. 39 (1961) 86–87.
[3] T. Fujii, N. Osa, S. Koyama, Free convection condensation of binary mixtures on a smooth tube: condensing mode and heat transfer coefficient of condensate. in: Proc. Engng. Foundation Conference on Condensation and Condenser Design. St. Augustine, Florida, ASME, 171–182 (1993).
[4] Y. Utaka, N. Terachi, Measurements of condensation characteristic curves for binary mixture of steam and ethanol vapor, Heat Transfer Jpn. Res. 24 (1995) 57–67.
[5] Y. Utaka, T. Kenmotsu, S. Yokoyama, A study on Marangoni condensation (measurement and observation for steam and ethanol vapor mixture), Proc. 11th Int. Heat Transfer Conf. 6 (1998) 397–402.
[6] Y. Utaka, S. Wang, Promotion of steam condensation heat transfer using solutal Marangoni condensation, Trans. JSME (in Japanese) 68 (2002) 148–156.
[7] Y. Utaka, H. Kobayashi, Effect of velocity on condensation heat transfer for steam-ethanol binary vapor mixture. in: Proc. 6th ASME-JSME Thermal Engng. Joint Conf., CD paper TED-AJ03-604 (2003).
[8] Y. Utaka, S. Wang, Characteristic curves and promotion effect of ethanol addition on steam condensation heat transfer, Int. J. Heat Mass Transfer 47 (2004) 4507–4516.
[9] K. Hijakata, Y. Fukasaku, O. Nakabeppu, Theoretical and experimental studies on the pseudo-dropwise condensation of a binary vapor mixture, Trans. ASME J. Heat Transfer 118 (1996) 140–147.
[10] W. Nusselt, Die Oberflachenkodensation des Wasserdampfes, Z. Vereines Deutsch. Ing. 60 (1916) 569–575.
[11] Y. Utaka, T. Nishikawa, Unsteady measurement of condensate film thickness for solutal Marangoni condensation, Proc. 12th Int. Heat Transfer Conf. (2002) 893–898.
[12] W.C. Lee, J.W. Rose, Forced convection film condensation on a horizontal tube with and without non-condensing gases, Int. J. Heat Mass Transfer 27 (1984) 519–528.
[13] T. Murase, Surface tension effects in condensation heat transfer: condensation on wire-wrapped tubes and Marangoni condensation of mixtures. PhD Thesis, Univ. London, UK (2006).
[14] J.W. Rose, Effect of pressure gradient in forced convection film condensation on a horizontal tube, Int. J. Heat Mass Transfer 27 (1984) 39–47.
[15] I.G. Shekriladze, V.I. Gomelauri, Theoretical study of laminar film condensation of a flowing, Int. J. Heat Mass Transfer 9 (1966) 581–591.
[16] T. Murase, H.S. Wang, J.W. Rose, Marangoni condensation of steam–ethanol mixtures on a horizontal tube, in: Proc. 17th International Symposium on Transport Phenomena, Toyama, Japan (2006).