

Heat transfer to mixtures of acetone, isopropanol and water under subcooled flow boiling conditions—I. Experimental results

U. WENZEL and B. HARTMUTH

Department of Chemical and Materials Engineering, University of Auckland, Auckland, New Zealand

and

H. MÜLLER-STEINHAGEN†

Department of Chemical and Process Engineering, University of Surrey, Guildford GU2 5XH, U.K.

(Received 24 July 1992 and in final form 27 August 1993)

Abstract—Flow boiling heat transfer coefficients for ternary mixtures of acetone, isopropanol and water were measured over a wide range of process parameters such as heat flux, subcooling, flow velocity and concentration. The measurements cover the regimes of convective heat transfer, transition region and fully developed subcooled boiling. The effect of process parameters on the heat transfer coefficients in the various regimes is discussed.

INTRODUCTION

SUBCOOLED boiling is characterized by the generation of vapour bubbles at the heater surface while the bulk temperature of the liquid is still below the saturation temperature. Bubbles detaching from the heat transfer surface collapse and condense in the subcooled liquid bulk. While this situation basically occurs in almost every evaporator, it is particularly significant in reboilers of low pressure distillation columns. The hydrostatic head from the supply lines to the evaporator can be a multiple of the saturation pressure in the first separation stage (especially for the common vertical long evaporator types), causing the liquid in the entrance region of the evaporator to be considerably subcooled. Depending on the total operating pressure, subcooled boiling will, therefore, occur over a considerable length of the evaporator, representing up to 50% of the total heat duty. Despite its significant role, subcooled boiling of mixtures has, so far, received little attention.

In this paper (Part I) heat transfer measurements for subcooled flow boiling of acetone, isopropanol and water mixtures flowing upward in a vertical annulus are presented. The measured data are listed in ref. [1]. In a companion paper (Part II) a calculation procedure for the heat transfer coefficient is described, which is applicable for convective, transition and boiling heat transfer.

TEST EQUIPMENT

Figure 1 shows a flow diagram of the test rig used for the present investigation. The fluid was pumped through a closed circuit consisting of temperature controlled storage tank, pump, flow meter and annular test section. The flow velocity of the liquid was usually measured with a magnetic flow meter or with an orifice plate, if the electrical conductivity of the liquid was too low. The bulk temperature of the fluid was measured with thermocouples located in two mixing chambers, before and after the test section. The gage pressure inside the annulus was measured locally with a differential pressure transmitter. The complete test rig was made of stainless steel 316.

The test section consists of an electrically heated cylindrical heating rod made of stainless steel, which is shown in Fig. 2. The heater is surrounded by a cylindrical pipe to form a concentric annulus. The dimensions of the test section are:

diameter of the heating rod	10.67 mm
outer diameter of annulus	25.40 mm
length of heated section	100.0 mm
heated length to thermocouple location	82.60 mm
entrance length up to thermocouple location	298.6 mm.

All measurements were performed with a single heater manufactured by Drew Chemicals according to specifications by Heat Transfer Research Inc. (HTRI). The surface roughness of the heater was measured using a surtronic 3 instrument manufactured by Rank Taylor Hobson, Leicester, England. The mean R_a value was $0.328 \mu\text{m}$; the mean R_p value was $1.018 \mu\text{m}$.

† Author to whom correspondence should be addressed.

NOMENCLATURE

j	j -factor [-]
$l_{h,s}$	length of heated section [m]
l_{th}	heated length to thermocouple location [m]
n	exponent [-]
Nu	Nusselt number [-]
Pr	Prandtl number [-]
\dot{q}	heat flux [W m^{-2}]
s	distance between thermocouple and heater surface [m]
T	temperature [K]
ΔT_{sub}	subcooling [K].

Greek symbols

α	heat transfer coefficient defined by equation (7) [$\text{W m}^{-2} \text{K}^{-1}$]
α_0	heat transfer coefficient defined by equation (2) [$\text{W m}^{-2} \text{K}^{-1}$]
λ	thermal conductivity [$\text{W m}^{-1} \text{K}^{-1}$].

Indices

b	liquid bulk
i	index
mix1	mixing chamber one
mix2	mixing chamber two
sat	saturation
th	thermocouple
w	wall.

The local wall temperature of the heater is measured with three thermocouples located in one cross-section just below the surface of the heated section. The temperature drop between the thermocouple location and the heater surface is a function of the heat flux and the ratio between the distance of each thermocouple from the surface and the thermal conductivity of the heating rod material (s/λ). This ratio was determined in numerous calibration measurements using a Wilson plot technique. The surface temperature of the heater is then calculated as

$$T_w = \frac{1}{3} \sum_{i=1}^3 \left(T_{th,i} - \dot{q} \frac{s_i}{\lambda_i} \right) \quad (1)$$

using the reading of all three thermocouples. A fourth

thermocouple was connected to a temperature controller to avoid overheating of the test heater.

EXPERIMENTAL PROCEDURE

All measurements were taken after the system had reached steady-state for the given conditions. The flow velocity and the heat flux were varied, starting with the highest heat flux and the lowest fluid velocity to eliminate hysteresis effects due to inactivated bubble nucleation sites. This effect is, for instance, mentioned by Müller-Steinhagen *et al.* [2].

The local heat transfer coefficient is defined as:

$$\alpha_0 = \frac{\dot{q}}{T_w - T_b} \quad (2)$$

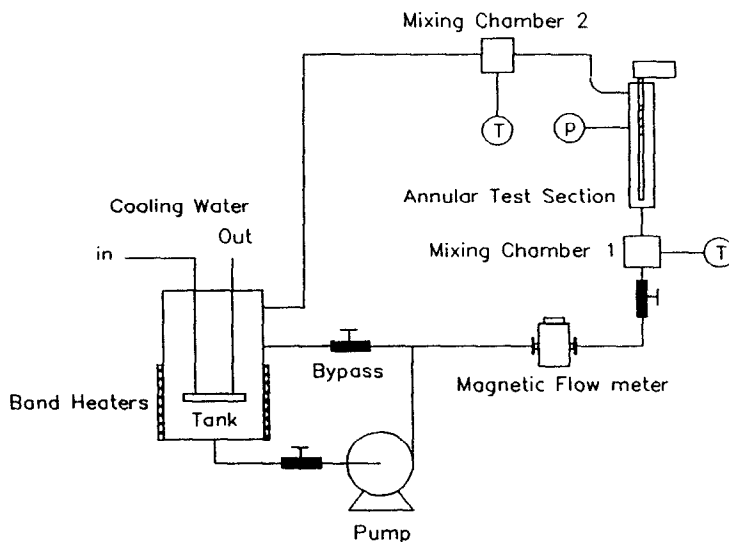


FIG. 1. Test loop.

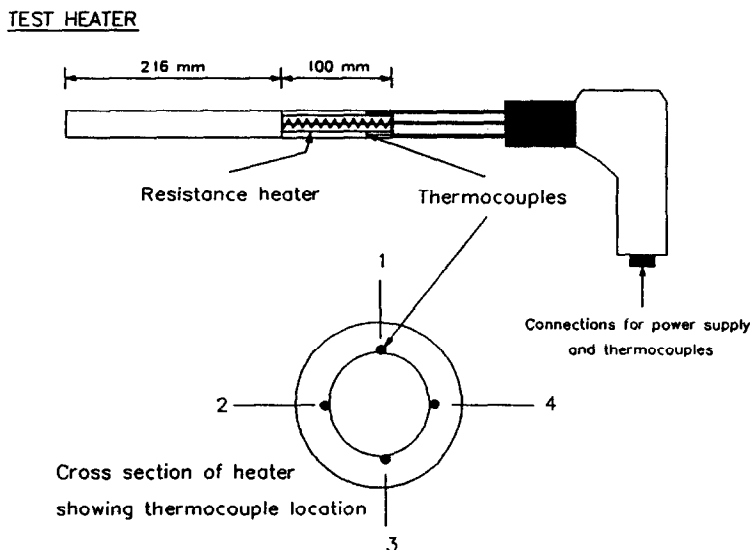


FIG. 2. Test heater.

where the local bulk temperature T_b of the liquid is calculated with

$$T_b = T_{\text{mix}1} + \frac{l_{\text{th}}}{l_{\text{h.s.}}} (T_{\text{mix}2} - T_{\text{mix}1}) \quad (3)$$

assuming perfect insulation and a linear bulk temperature distribution over the length of the heated section. The fluid composition was determined with a gas chromatograph using two liquid samples before and after each run.

Several tests of the reproducibility of the experimental results showed a variation of the heat transfer coefficients by less than 7%. A complete data bank containing all experimental results can be found in ref. [3].

EXPERIMENTAL PARAMETERS

The liquids used for the present measurements were acetone/isopropanol/water mixtures. This ternary system contains one azeotropic point at a concentration of 67.5 mol-% isopropanol and 32.5 mol-% water for a pressure of 120 kPa. Figure 3 shows an equilateral triangular graph indicating the compositions of the 45 different fluid mixtures investigated. Each corner of the ternary mixture triangle represents a single component fluid, the sides of the triangle are the three binary mixtures, while all the points inside the triangle stand for ternary mixtures. A total number of 2430 measurements were taken covering the regimes of convective heat transfer, transition boiling and fully developed subcooled boiling. The standard range of experimental parameters is shown in Table I.

For certain concentrations and heat fluxes, flow velocity and subcooling have been varied in smaller

increments to demonstrate the effect of these parameters more clearly.

RESULTS

Figure 4 shows the typical shape of the boiling curve for a mixture consisting of 12.5 mol-% acetone, 12.5 mol-% isopropanol and 75 mol-% water. The heat transfer coefficient is almost independent of the heat flux for high flow velocity and low heat fluxes, while low flow velocities and high heat fluxes result in a strong dependence of the heat transfer coefficient on the heat flux. These two areas of the boiling curve are the convective heat transfer regime and the subcooled boiling regime. The influence of several parameters on the heat transfer coefficients in the convective and boiling regimes will be discussed in the following.

Convective heat transfer

Effect of heat flux. The heat transfer coefficients are almost independent of the heat flux. The slight increase of the convective heat transfer coefficients with increasing heat flux is caused by the increasing wall temperature resulting in increased natural convection and reduced fluid viscosity close to the heat transfer surface.

Effect of subcooling. Heat transfer is affected by a change in subcooling, i.e. liquid temperature, due to the changing physical properties of the fluid. The viscosity is the physical property which is the most sensitive to changes of the fluid temperature resulting in higher heat transfer coefficients for higher fluid temperatures (lower subcooling).

The viscosity was calculated using the generalized corresponding states method by Teja, described in ref. [4].

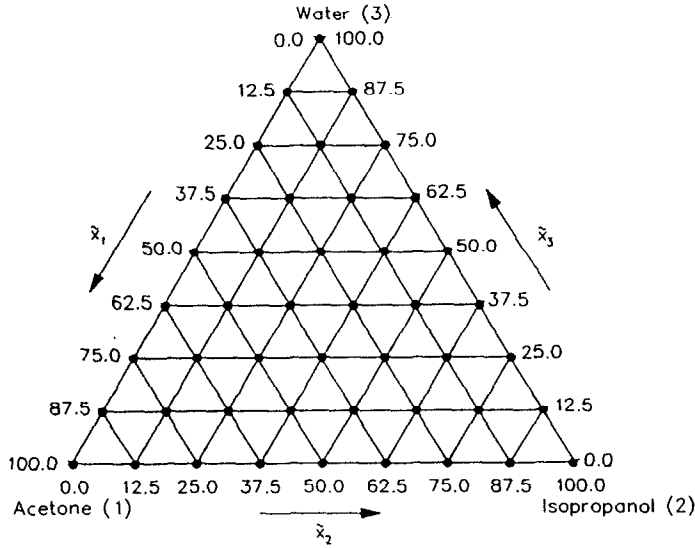


FIG. 3. Investigated acetone/isopropanol/water mixture.

Table 1. Range of parameters

Heat flux q [kW m^{-2}]	40, 80, 120, 190, 260, 400
Flow velocity v [m s^{-1}]	0.1, 0.3, 0.9
Subcooling ΔT_{sub} [K]	10, 18, 25

Effect of flow velocity. The flow velocity has a strong influence on the heat transfer coefficient. Figure 4 shows that the heat transfer increases with increasing fluid velocity. This relationship is demonstrated more

clearly in Fig. 5, showing the j -factor as a function of the Reynolds number. The data shown in this figure have been randomly selected from the large number of convective heat transfer measurements to provide an equal distribution over the full range of Reynolds numbers investigated. The j -factor is a dimensionless number defined as,

$$j = \frac{Nu}{Pr^{0.33}} \quad (4)$$

which describes heat transfer as a function of the

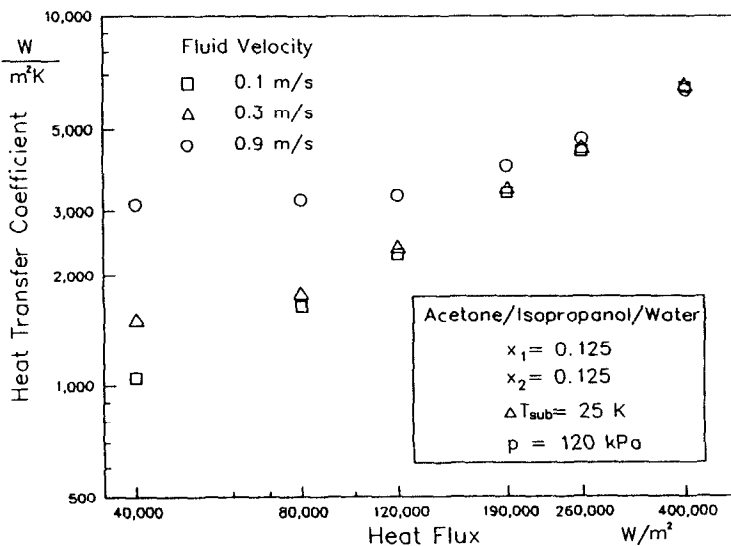
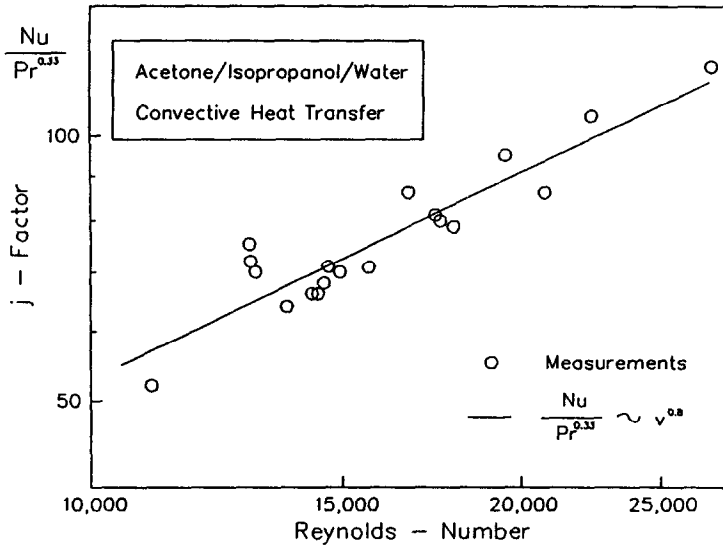


FIG. 4. Heat transfer coefficients vs heat flux.

FIG. 5. *j*-factor vs Reynolds number.

Reynolds number, independent of the physical properties of the fluid. The data presented in Fig. 5 can be curve-fitted by a line with the slope 0.8. This indicates that the heat transfer coefficients are proportional to the fluid velocity to the power of 0.8, which is the relationship used, for instance, in the Dittus/Boelter correlation [5], for convective heat transfer under turbulent conditions. The data points shown in this graph represent a variety of fluid compositions and subcoolings to obtain the widest possible range of Reynolds numbers.

Effect of fluid composition. To show the influence of the fluid composition on the heat transfer coefficient a three-dimensional illustration is used, see Fig. 6. This illustration uses the ternary mixture triangle of Fig. 3 as a base area. Each point on this triangle represents one fluid mixture. The outer corners of this surface stand for the heat transfer coefficients of the three pure fluids, the outer border lines represent the heat transfer coefficients of binary mixtures and all other points illustrate the heat transfer coefficients of ternary mixtures. The heat transfer coefficients shown in Fig. 6 were measured for a heat flux of 40 kW m^{-2} , a fluid velocity of 0.9 m s^{-1} and a subcooling of 25°C .

The highest heat transfer coefficient under these conditions was measured for water. A small addition of acetone or isopropanol to pure water causes the heat transfer coefficient to drop drastically to almost 50% of the water value. Larger additions of isopropanol to water give approximately 35% lower heat transfer coefficients than the coefficients measured for the same addition of acetone. The heat transfer coefficients of ternary mixtures containing a constant percentage of water, which are represented by lines connecting the left and right borders of the surface in Fig. 6, decrease almost linearly with increasing isopropanol content of the mixture.

The shape of the surface shown in Fig. 6 is solely a function of the physical properties of the fluid. It is the thermal conductivity of the fluid which has the biggest influence, because of its large change with fluid composition

$$\lambda_{\text{H}_2\text{O}} > 4 \cdot \lambda_{\text{acetone, isopropanol}}$$

and its importance for convective heat transfer.

Boiling heat transfer

Effect of heat flux. The correlation between the heat transfer coefficient and the heat flux can be expressed as:

$$\alpha_0 = C \dot{q}^n. \quad (5)$$

The exponent n has a value of 0.8 for a subcooling of 10°C , while a subcooling of 25°C results in a value of 0.85. This is due to the definition of the heat transfer coefficient

$$\begin{aligned} \alpha_0 &= \frac{\dot{q}}{(T_w - T_{\text{sat}}) + (T_{\text{sat}} - T_b)} \\ &= \frac{\dot{q}}{(T_w - T_{\text{sat}}) + \Delta T_{\text{sub}}}. \end{aligned} \quad (6)$$

The large difference between saturation and bulk temperature for high subcoolings obscures the effect of the heat flux on the wall temperature T_w , resulting in a higher value of the exponent n . The value of 0.8 of the exponent agrees quite well with the result of a correlation given by Gorenflo in ref. [6] for saturated liquids.

Effect of subcooling. The influence of subcooling on the heat transfer coefficient defined in equation (2) cannot be discussed directly, because this definition of the heat transfer coefficient already contains a dependence on the subcooling, as shown in equation

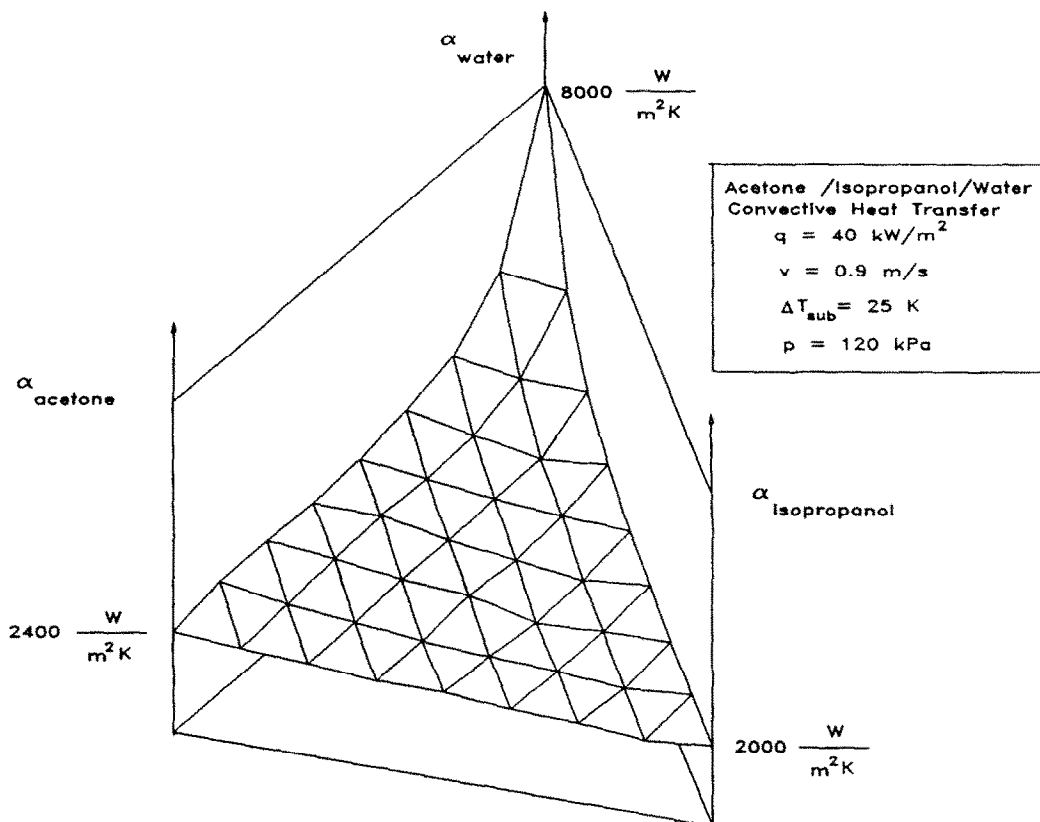


FIG. 6. Convective heat transfer coefficients of acetone/isopropanol/water mixtures.

(6). The effect of subcooling is, therefore, partly due to the definition of the heat transfer coefficient. To discuss the influence of the subcooling on the heat transfer coefficient a different definition of the heat transfer coefficient was chosen

$$\alpha = \frac{\dot{q}}{T_w - T_{sat}} \quad (7)$$

It was assumed that there was no concentration gradient in the liquid mixture over the length of the heated section due to the collapse of vapour bubbles under subcooled boiling conditions. The saturation temperature of the mixture at the thermocouple location was, therefore, calculated using the mixture composition at the inlet of the heated section.

Figures 7(a)–(c) show the heat transfer coefficient defined by equation (7) and the wall temperature as a function of subcooling. Figure 7(a) illustrates that subcooling has almost no effect on the boiling heat transfer coefficients for pure liquids. Contrariwise, a strong effect was found for binary mixtures of acetone/water and isopropanol/water containing small solvent concentrations, see Fig. 7(b). An increase in subcooling from 0 to 12°C causes a drop of the heat transfer coefficient by approximately 20% and a corresponding rise of the wall temperature. The addition of a third component to the binary mixtures

results in a less significant influence of the subcooling as shown in Fig. 7(c).

Effect of fluid composition. Figure 8 illustrates the influence of the fluid composition on the fully developed subcooled boiling heat transfer coefficient. The same type of three-dimensional graph is used as in Fig. 6. All experimental data as well as 'binary' diagrams for most mixtures are given in ref. [1]. The heat transfer coefficients shown in Fig. 8 were measured for a heat flux of 260 kW m⁻², a fluid velocity of 0.1 m s⁻¹ and a subcooling of 18°C. The maximum heat transfer coefficients were measured for the pure components, with the highest value found for water. The heat transfer coefficients of the binary and ternary mixtures are considerably lower than the values of the pure components or values gained by linear interpolation between these values. Schlünder [7] explains this trend by the preferential evaporation of the more volatile component at the liquid vapour interface, causing a local rise of the saturation temperature. For a constant heat flux, the rise of the saturation temperature results in a higher wall temperature and, therefore, a lower apparent heat transfer coefficient. The extent of the reduction of the heat transfer coefficient depends on the evaporation rate, the liquid-side mass transfer resistance and the shape of the vapour-liquid phase equilibrium diagram. The

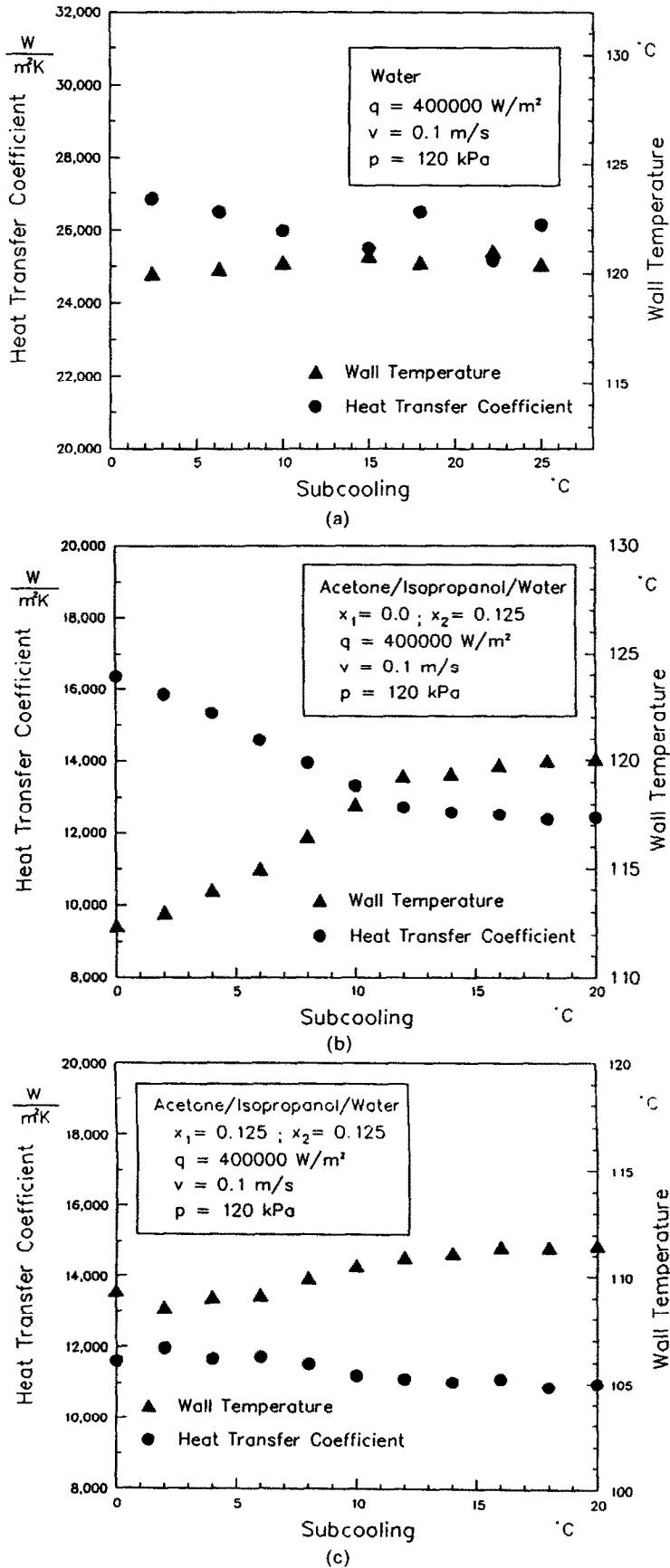


FIG. 7. (a)–(c) Heat transfer coefficient (equation (7)) and wall temperature vs subcooling.

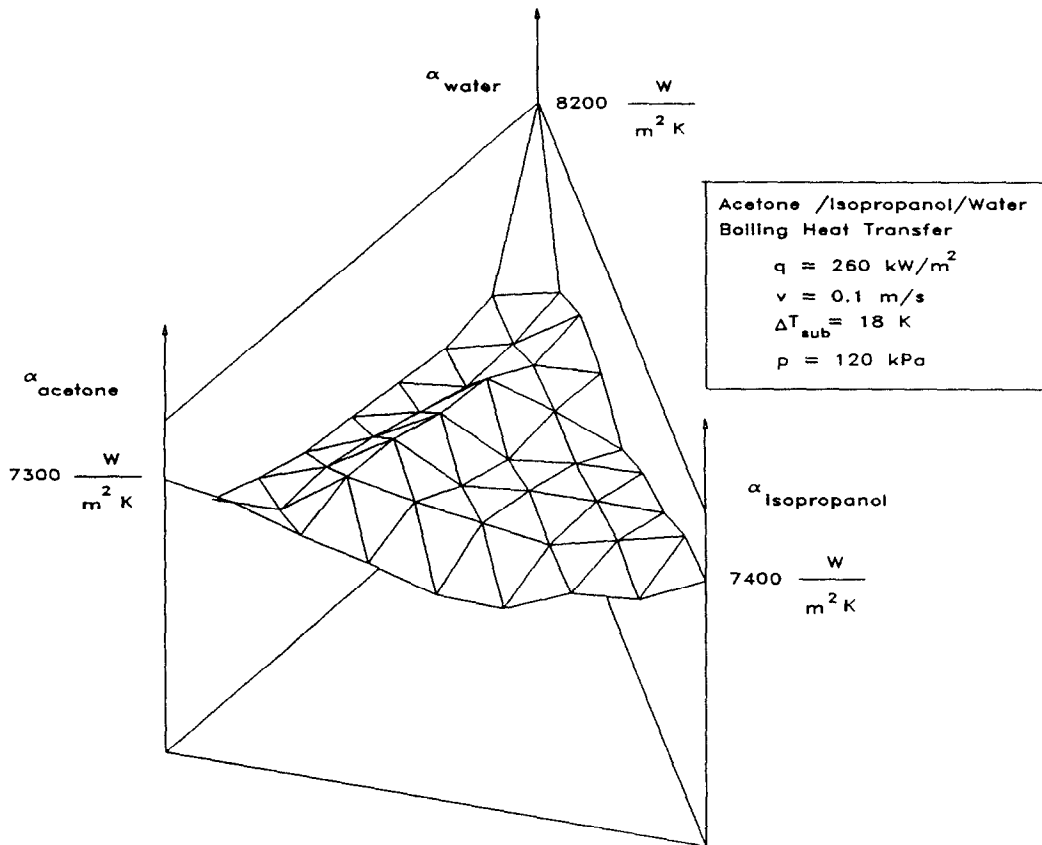


Fig. 8. Nucleate boiling heat transfer coefficients of acetone/isopropanol/water mixtures.

strongest effect is found if the difference between the equilibrium vapour and liquid concentrations is large and the slope of the bubble point line is steep. For the solutions used in the present investigation, this occurs for low solvent concentrations of acetone/water and isopropanol/water mixtures. As a result, we observe a drop of the heat transfer coefficient of almost 50% due to only a small addition of acetone or isopropanol to pure water.

Fluid velocity. Figure 4 shows that the heat transfer coefficient for fully developed subcooled boiling is independent of the flow velocity. This result was found for all mixtures and subcoolings. It indicates that the heat transfer is not controlled by macro or bulk convection but by the micro convection induced by the bubble formation and detachment mechanisms.

More interesting with respect to the present investigation is the observation that the reduction of heat transfer coefficient for the mixtures is not affected by changes in the flow velocity. This means that the higher level of turbulence for higher fluid velocities does not decrease the liquid-side mass transfer resistance.

CONCLUSIONS

Flow boiling heat transfer coefficients for ternary mixtures of acetone, isopropanol and water have been measured for different heat fluxes, subcoolings and

flow velocities covering the regimes of convective heat transfer, transition area and fully developed nucleate boiling.

The convective heat transfer coefficients of the mixtures show the same dependency on heat flux, subcooling and fluid velocity composition as the pure components. Heat transfer depends on fluid composition and temperature because of the changing physical properties of the fluid. The heat flux has almost no influence, while the fluid velocity has the dominant effect on the heat transfer.

Heat transfer in the fully developed subcooled boiling regime depends on the heat flux, the subcooling and the fluid composition, while it is independent of the fluid velocity. This indicates that higher fluid velocities do not decrease the liquid-side mass transfer resistance. The degree of subcooling does not influence heat transfer to single component fluids, while it has a noticeable effect on binary and to a lesser extent ternary mixtures. The subcooled boiling heat transfer coefficients for binary and ternary mixtures are considerably lower than the values gained by a linear interpolation between the heat transfer coefficients for the single component fluids.

REFERENCES

1. B. Hartmuth, Untersuchung des Wärmeübergangs bei unterkühltem Sieden in ternären Mischungen, Diplo-

- marbeit am Lehrstuhl A für Thermodynamik der Technischen Universität München, München, Germany (1992).
2. H. M. Müller-Steinhagen, A. P. Watkinson and N. Epstein, Subcooled-boiling and convective heat transfer to heptane flowing inside an annulus and past a coiled wire, *J. Heat Transfer* **108**, 922–933 (1986).
 3. U. Wenzel, Saturated pool boiling and subcooled flow boiling of mixtures at atmospheric pressure, Ph.D. Thesis, University of Auckland, Auckland, New Zealand (1992).
 4. A. S. Teja and P. Rice, Generalized corresponding states method for the viscosity of liquid mixtures, *Ind. Engng Chem. Fundam.* **20**, 77–81 (1981).
 5. F. W. Dittus and L. M. K. Boelter, *Heat Transfer in Automobile Radiators of the Tubular Type*, Vol. 2, p. 13. University of California Press, California (1930).
 6. D. Gorenflo, Behältersieden, *VDI-Wärmeatlas*, Sect. Ha (4th. edn.). VDI, Düsseldorf (1984).
 7. E. U. Schlünder, Über den Wärmeübergang bei der Blasenverdampfung von Gemischen, *Verfahrenstechnik* **16**, 692–698 (1982).