SPECIAL ISSUE

Nucleate boiling of FC-87/FC-72 zeotropic mixtures on a horizontal copper disc

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Abstract This paper presents nucleate boiling experimental results, at atmospheric pressure, for heat fluxes $q \leq 40$ kW/m^2, for FC-87/FC-72 binary mixtures in molar fractions of 0/100, 25/75, 50/50, 75/25, 85/15 and 100/0, at saturation temperatures for pure fluids and bubble points for mixtures. The test section was an upward facing copper disc of 12 mm diameter and 1 mm thickness. The experimental heat transfer coefficient was compared with the correlations of Rohsenow (1952), as reported by Rohsenow et al. (Handbook of heat transfer, McGraw-Hill, New York, 1998), Stephan and Abdelsalam (Int J Heat Mass Transfer 23;73–78, 1978) and Cooper (Int Chem Eng Symp Ser 86:785–792, 1984) for pure fluids and the semi-empirical models of Stephan and Körner (Chem Ing Tech Jahrg 7:409–484, 1969), Thome (J Heat Transfer 104:474–478, 1982), Fujita et al. (1996), as reported by Rohsenow et al. (Handbook of heat transfer, McGraw-Hill, New York, 1998), Fujita and Tsutsui (Int J Heat Mass Transfer 37(1):291–302, 1994) and Calus and Leonidopoulos (Int J Heat Mass Transfer 17:249–256, 1973) for mixtures.

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List of symbols

c_{pl}	liquid specific heat (J/kgK)
C_{sf}	constant in Rohsenow correlation
D	mass diffusivity (m^2/s)
d_b	departure bubble diameter (m)
g	acceleration due to gravity (m/s^2)
h	heat transfer coefficient (kW/m^2 K)
h_{lv}	latent heat of vaporization (J/kg)
K	correction factor
k_l	liquid thermal conductivity (W/mK)
M	molecular weight (kg/kmol)
p	pressure (N/m^2)
p_r	reduced pressure
Pr	Prandtl number
q	heat flux (kW/m^2)
R_a	surface roughness (μm)
T	temperature (K, °C)
x	liquid molar fraction of the more volatile component
y	vapor molar fraction of the more volatile component

Greek symbols

ΔT_{si}	$i = 1, 2$, wall superheating for pure fluids (K)
ΔT	temperature difference (K)
ΔT_{db}	temperature difference between the dew point and the bubble point (K)
θ	contact angle (°)
μ	dynamic viscosity (Pa s)
ρ	density (kg/m^3)
σ	surface tension (J/m^2)

Subscripts

bp	bubble point
C	Cooper
db	the dew point minus the bubble point
id	ideal
l	liquid

R   Rohsenow
SA   Stephan–Abdelsalam
sat   saturation
th   theoretical
v   vapor
w   wall

1 Introduction

Zeotropic mixtures have been employed as a fast and rel-atively cheap industrial alternative to replace phased-out chlorofluorocarbons (CFCs). They can be used as a refrigerant in evaporators under a vast number of opera-tional conditions, for a constant pressure and different temperatures, depending on the molar fraction, in different applications.

Several experimental results for the nucleate boiling of mixtures show a decrease in the heat transfer coefficients in comparison with those for a single component substance with the same physical properties, Fujita and Tsutsui [1], and also with the linearly interpolated values of the heat transfer coefficients of the pure components [1, 2].

The objective of this study is to present the main results, following an experimental analysis, for the heat transfer coefficient, h, in nucleate boiling regime, at atmospheric pressure, for FC-87(C_5F_{12})/FC-72(C_6F_{14}) binary mixtures in molar fractions of 0/100, 25/75, 50/50, 75/25, 85/15 and 100/0, at saturation temperatures for pure fluids and vapor bubble points for mixtures. This analysis includes the comparison of h with correlations for pure fluids and models for mixtures.

2 Literature review

The heat transfer coefficient, h, for nucleate boiling of a binary mixture is defined as the ratio of the heat flux q to the difference between the wall temperature, T_w, and the bubble-point temperature, T_{bp}, Carey [3]:

$$h = \frac{q}{[T_w - T_{bp}(p,x)]} \tag{1}$$

where p and x represent the pressure and the liquid molar fraction corresponding to the most volatile fluid. In Eq. (1), when $x = 0$ or 1, $T_{bp} = T_{sat,1}$ or $T_{sat,2}$.

Van Wijk et al. [4] explained that the reduction in the h value for the mixtures, as considered above, compared with the h value for pure fluids with the same physical proper-ties, is related to the more volatile molar fraction of the vapor created on the wall being greater than that of the bulk mixture. Thus, to maintain the boiling and the equilibrium between the vapor and the liquid phases, the more volatile

component should be vaporized in order to allow the bubble growth. The consequence of this local decrease in the more volatile molar fraction of the mixture, causing an increase in the wall superheating for a heat transfer rate, is a decrease in the heat transfer coefficient, as defined by Eq. (1). This effect is equivalent to a reduction in the effective superheating of the liquid mixture. The description for the nucleate boiling mechanism of mixtures proposed by Van Wijk et al. [4] is based on the dependence of h on the difference between the more volatile molar fraction of the vapor, y, and that of the liquid, x, $|y - x|$.

Benjamim and Balakrishnan [5] considered that the extent of the decrease in h for mixtures is dependent on the thermophysical properties, the apparent reduction in the bubble departure rate and the decrease in the departure bubble diameter, along with the non-linear dependence of the properties on the molar fraction. Sterling and Tichacek (as reported by Thome [6]) explained that the lower value of h for mixtures is due to the additional resistance of the more volatile component mass diffusion inside the vapor bubble. Tolubinskiy and Ostrovskiy [7] considered that the nucleate boiling of mixtures is accompanied not only by phase-changes but also by the distillation of the mixtures with mass transfer of the mixture components in the liquid phase as well as between the liquid and vapor phases.

Thome [8] postulated that the h is influenced by parameters that depend on the mixture concentration, such as the evaporation rate, the diffusion rate and the density of active nucleation sites, the evaluation of which is a com-plex problem. The molar fraction inside the vapor bubble is a function of the molar fraction of the mixture and this has an influence on the superheating degree of the liquid phase for the nucleation. In order to simplify, Thome [8] pro-posed that the reduction in the heat transfer coefficient for mixtures is a function of ΔT_{db} (temperature difference between the dew point and the bubble point) for each mixture concentration. The lowest h value is found for the highest value of ΔT_{db}.

Other models, proposed by Calus and Rice [9], Calus and Leonidopoulos [10] and Kandlikar [11], consider the effect of the mass diffusivity on the decrease in nucleate boiling heat transfer for mixtures.

In the following, a synthesis of five models and three empirical correlations used to analyze the experimental results for the mixtures and pure fluids, respectively, will be presented.

2.1 Semi-empirical models for mixtures

Five models for the calculation of heat transfer coefficient for mixtures will be considered: Stephan and Körner [12], Thome [8], Fujita et al. (in Ref. [13]), Fujita and Tsutsui [14] and Calus and Leonidopoulos [10], whose the procedure is

summarized in the following. The ideal heat transfer coefficient for a mixture, h_{id}, as defined by Stephan [15], is:

$$h_{id} = \frac{q}{\Delta T_{id}} \tag{2}$$

where ΔT_{id} represents the corresponding wall superheating, and is calculated, for a binary mixture, as in the following equation, [14]:

$$\Delta T_{id} = \sum_{i=1}^{2} (x_i \Delta T_{si}) \tag{3}$$

where x_1, x_2, and ΔT_{s1}, ΔT_{s2} represent the molar fraction for each component $(x_1 + x_2 = 1)$ and the wall superheating for pure components of the mixture $(\Delta T_{s1} = T_w - T_{sat,1}, \Delta T_{s2} = T_w - T_{sat,2})$, respectively. The linear dependence of ΔT_{id} with ΔT_{s1} and ΔT_{s2} with the molar fraction over estimates the ideal nucleate boiling heat transfer coefficient, for a mixture, because the effect of ΔT_{db} was not considered in the Eqs. (2, 3). These models consider a reduction factor $(1 + K)^{-1}$ that multiplies h_{id}:

$$\frac{h}{h_{id}} = \frac{1}{1 + K} \tag{4}$$

The relations for the correction factor K, for the five models considered in this review, are summarized in Table 1.

In Eq. (5), Table 1, the values of A_0, lying between 0.42 and 3.56, in the model of [12], are dependent on the mixture components and for no tested fluids these authors recommend $A_0 = 1.53$. Equations (5) and (9) depend on the mass transfer driving force $|y - x|$ but in Eq. (9) the role of the empirical constant A_0 is played by the expression:

$$\left[\left(\frac{\alpha}{D} \right)^{0.5} \left(\frac{c_{pl}}{h_{lv}} \right) \left(\frac{dT}{dx} \right) \right]$$

where α, D, c_{pl}, h_{lv} and dT/dx represent the thermal diffusivity, the mass diffusivity, the liquid specific heat, the latent heat of vaporization and the slope of the bubble point line, respectively. The other models are dependent on the temperature differences ΔT_{db} and ΔT_{id} but Eq. (8) includes, also, the heat flux.

2.2 Correlations for pure fluids

The empirical correlations proposed by Rohsenow [13], Stephan and Abdelsalam [16] and Cooper [17] will be considered to calculate the heat transfer coefficient for pure fluids. The correlation of Rohsenow, gives $h = h_R$, by the following equation:

$$h_R = \mu_l h_{lv} \left(\frac{g(\rho_l - \rho_v)}{\sigma} \right)^{0.5} \left(\frac{c_{pl}}{C_{sf} h_{lv} Pr^s} \right)^3 (T_w - T_{sat})^2 \tag{10}$$

where $s = 1$ for water and 1.7 for the other fluids. The coefficient C_{sf} is dependent on the fluid, wall material and the surface conditions, [3].

The correlation of [16], with $h = h_{SA}$, is:

$$h_{SA} = 207 \left(\frac{k_l}{d_b} \right) \left(\frac{q d_b}{k_l T_{sat}} \right)^{0.745} \left(\frac{\rho_l}{\rho_v} \right)^{0.585} Pr^{0.533} R_a^{0.133} \tag{11}$$

where d_b:

$$d_b = 0.0149 \theta \left(\frac{2\sigma}{g(\rho_l - \rho_v)} \right)^{0.5} \tag{12}$$

is the bubble diameter and θ the contact angle, in degrees, [3, 16]. R_a is the roughness of the surface and Pr_l the liquid Prandtl number.

The third and simplest correlation considered here is that of [17], which allows the calculation of $h = h_C$ considering only the reduced pressure (p_r), the molecular weight (M) and the heat flux (q):

$$h_C = 55 p_r^b (-0.4343 \ln p_r)^{-0.55} M^{-0.5} q^{0.67}, \tag{13}$$

with,

$$p_r = \frac{p}{p_c} \tag{14}$$

where p_c represents the critical pressure and

$$b = 0.12 - \log R_a \tag{15}$$

In the Eq. (15), the surface roughness (R_a), as reported by Passos and Reinaldo [18] does not seem to be very well interpreted by Eqs. (13) and (15) and $b = 0.12$.

All the properties, in Eqs. (10–15) are considered at saturation temperature (T_{sat}).

Table 1 Correction factor K for different models

Reference	K	Equation
Stephan and Körner [12]	$A_0\|y - x\|(0.88 - 0.12p);$ p in bar	(5)
Thome [8]	$\frac{\Delta T_{db}}{\Delta T_{id}}$	(6)
Fujita et al. (1998), in Rohsenow et al. [13]	$\frac{\Delta T_{db}}{\Delta T_{id}} \left[1 - \exp\left(-2.8 \frac{\Delta T_{id}}{\Delta T_s} \right) \right]$	(7)
Fujita and Tsutsui [14]	$\frac{\Delta T_{db}}{\Delta T_{id}} \left[1 - 0.8 \exp\left(-q/10^5 \right) \right];$ q in W/m^2	(8)
Calus and Leonidopoulos [10]	$\left[\|y - x\| \left(\frac{\alpha}{D} \right)^{0.5} \left(\frac{c_{pl}}{h_{lv}} \right) \left\| \frac{dT}{dx} \right\| \right]$	(9)

Where $\Delta T_s = T_{sat,1} - T_{sat2}$

3 Experiment

3.1 Experimental set up

The nucleate boiling tests were performed in FC-72, FC-87 and FC-87/FC-72 mixture pools, at atmospheric pressure, at LEPTEN of the Federal University of Santa Catarina, in Brazil, during the Master of Science research of [19], in the Mechanical Engineering Graduate Program.

Figure 1 shows the experimental set up, consisting of a boiling chamber (8) installed in the center of an external chamber (9), both assembled between two horizontal stainless steel plates of 200 × 200 × 10 mm (12 and 13). The boiling chamber is a vertical glass tube with a 90 mm inner diameter and 175 mm height, the test section and the working fluid being inside. The external chamber has a square cross-section of 170 × 170 mm and transparent lateral walls of plexy-glass (9). The test condition temperature of the working fluid, the saturation temperatures for the pure fluids or the bubble point as a function of the molar fraction for the mixture, is imposed by a forced flow of water in the space created between the glass tube of the boiling chamber and the plexy-glass wall of the external chamber. The water temperature is controlled by a cryostat. Inside the boiling chamber, in the upper part, there is a serpentine condenser (7) cooled by water whose temperature is controlled by a second cryostat. The boiling chamber is equipped with a pressure transducer (3) and valves (2 and 6). Four E thermocouples (4), three of them in the liquid and the other in the vapor, allow the monitoring of the test condition temperature, which is controlled by cold water flowing inside the serpentine (7).

The test section, Fig. 1, (10), and Fig. 2, (1), consisting of a copper disc of 12 mm diameter and 1 mm thickness, is heated by a skin heater of 11.6 ± 0.05 Ω, Fig. 2, (2), bounded on the bottom side of the disc with epoxy resin, and fixed also with epoxy resin in a PVC support, Fig. 1 (10) and Fig. 2 (5).

The copper surface of the disc in contact with the working fluid was polished using emery paper #600, corresponding to a roughness R_a of 1.1 μm, and is horizontally mounted, upward facing. Three E thermocouples are fitted, close to the center, on the downward facing side of the copper disc.

A DC power supply, HP6030A, is connected to the skin heater and controlled by a PC using LABVIEW and the data acquisition and initial treatment are carried out by a HP34970A. The heating of the disc was controlled by increasing the heat flux.

Figure 3 shows a still photograph of the FC-87 nucleate boiling process, for a heat flux of 30 kW/m². The average copper disc temperature, T_w, was 42.5°C.
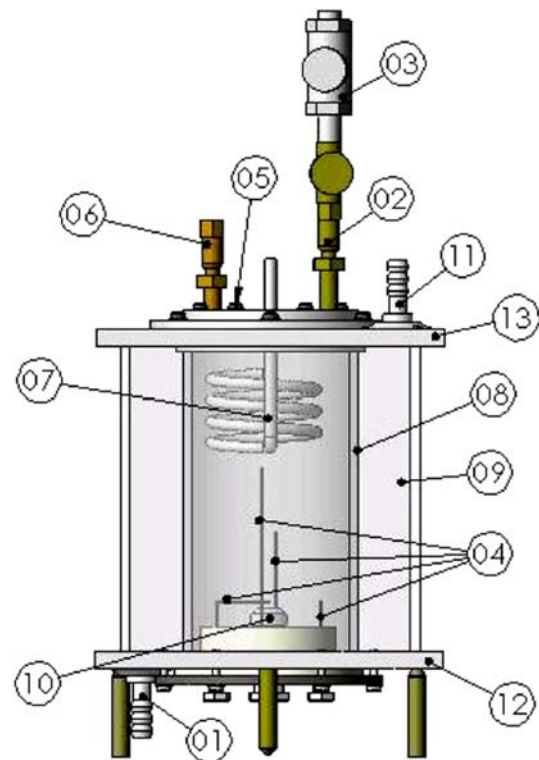
Fig. 1 Experimental set-up: *1* Bath water entrance, *2* valve, *3* pressure transducer, *4* thermocouples, *5* plexy-glass window, *6* valve, *7* condenser, *8* boiling chamber, *9* plexyglass chamber, *10* test section support, *11* bath water exit, *12*, *13* stainless steel plates
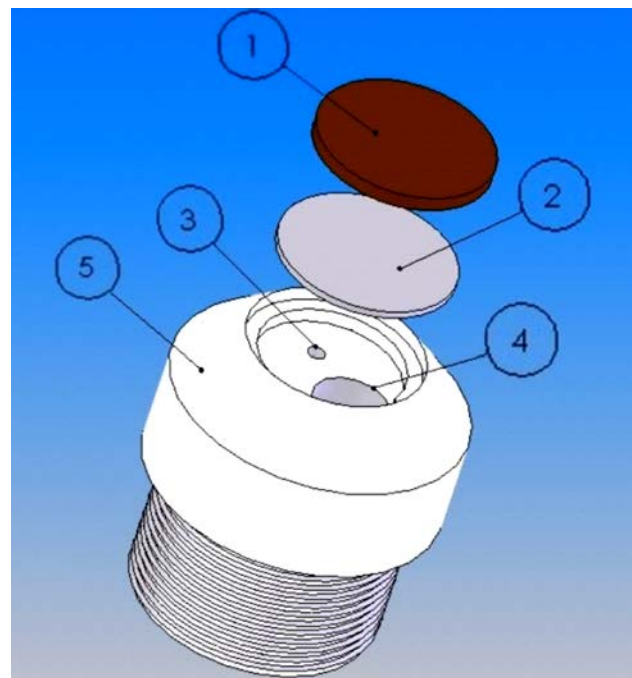
Fig. 2 View of test section assembly. *1* Copper disc (test section), *2* skin-heater, *3*, *4* holes to cable feed-throughs, *5* PVC support of the test section

Fig. 3 FC-87 nucleate boiling, $q = 30$ kW/m^2, $T_w = 42.5$°C

Table 2 Bubble points for the FC-87/FC-72 mixtures

(%FC-87/%FC-72)	Temperatures (°C)		
	Exp.	Theor.	ΔT
0/100	29.1	29.1	–
25/75	45.1	47.1	2.0
50/50	38.5	40.1	1.6
75/25	32.1	34.4	2.3
85/15	31.0	32.5	1.5
100/0	56.6	56.6	–

Fig. 4 Liquid and vapor temperatures during a test, $q = 40.01$ kW/m^2

3.2 Experimental procedure

Before each series of measurements, a vacuum was created in the boiling chamber which was then fed with the working fluid. By setting the temperature of the water bath, the test conditions represented by atmospheric pressure and the saturation temperature (for pure fluids) or bubble point for each molar fraction of the mixture were imposed. This condition was regulated by monitoring the pressure and the temperature inside the boiling chamber. Once the test conditions had stabilized, the heat flux was imposed in the range of 0–40 kW/m^2. At least 292 experimental points were obtained: 96 for pure fluids and 196 for mixtures.
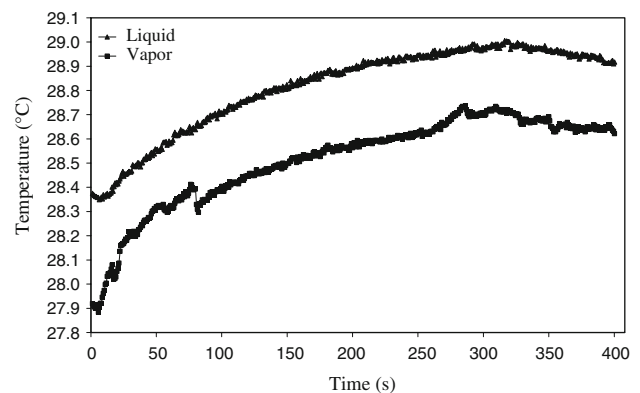
The test section was polished using #600 emery paper and the surface was then cleaned using acetone and dried with an air jet. This procedure was repeated before each series of measurements.

The boiling point, for each molar fraction, was experimentally determined and compared with the physical properties of FC-72, from EES [20] and FC-87, from REFPROP [21], and the difference between the measured and the theoretical values (EES and REFPROP data) varied between 1.5 and 2.3°C, as shown in Table 2.

Figure 4 shows the liquid and vapor temperatures during a test. On average, the difference between the liquid and the vapor temperatures was on average equal to 0.3°C. This difference is related to the influence of the serpentine condenser, in the vapor region where the thermocouple is located. For each test, 90 data points were acquired after a time period of 300 s, during a period of 65 s, and the

average values for the copper disc temperature and heat flux were calculated.

The uncertainty of the difference between the nominal and actual molar fraction values varied between (+0.8%) and (−1.4%). The maximum experimental uncertainty for the heat flux was 2.2%. The experimental uncertainty of the heat transfer coefficients, for heat fluxes between 3 and 10 kW/m^2, corresponding to natural convection, varied between 9.5 and 5.8, respectively, and, on average, was 6.1%. For a heat flux between 12 and 40 kW/m^2, corresponding to the nucleate boiling regime, the experimental uncertainty varied, on average, between 4.4 and 3.8%, respectively, with maxima of 5.8 and 4.8%, respectively.

4 Results

4.1 Experimental results

Figure 5 shows the partial boiling curves fitting the experimental points during single phase natural convection and nucleate boiling, for a heat flux up to 40 kW/m^2, for FC-87/FC-72 binary mixtures in molar fractions of 0/100, 25/75, 50/50, 75/25, 85/15 and 100/0, at saturation temperatures for pure fluids and vapor bubble points for
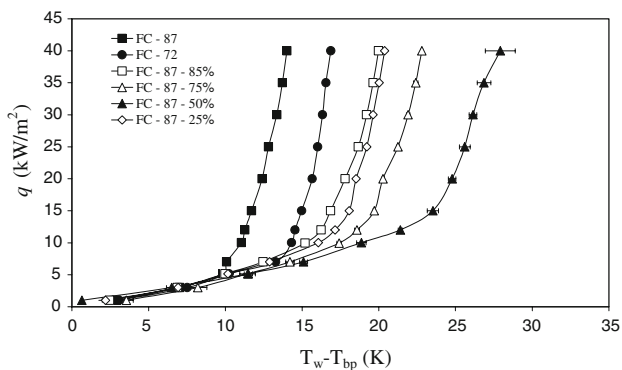
Fig. 5 Partial boiling curves

mixtures. Each experimental point represents an average of at least three experimental points, for the same test conditions, which were obtained following the experimental procedure explained in the previous section. A good repeatability of the results can be observed. In the region characterizing the nucleate boiling regime, for a heat flux between 12 and 40 kW/m^2, the curves for the mixtures are shifted to the right indicating lower values for the heat transfer coefficient for mixtures compared with those for pure fluids. The lowest values of h were obtained for the mixture with a molar fraction of 50%.

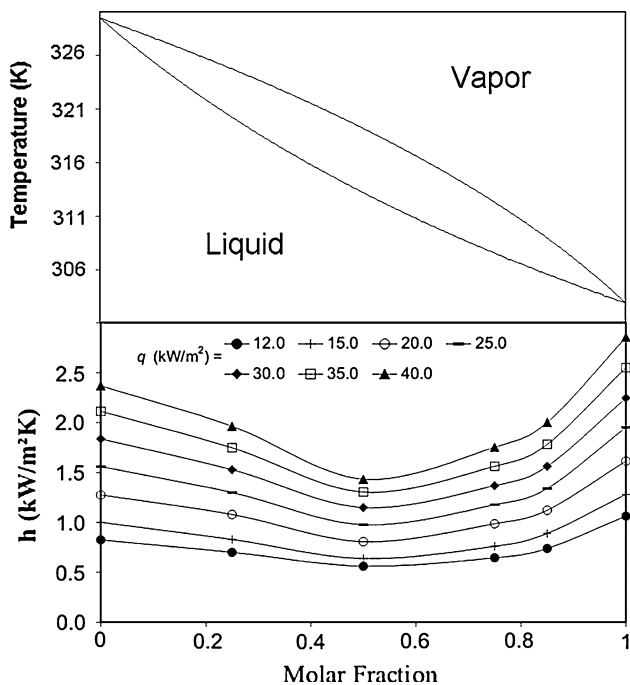Figure 6 shows, in the lower part, the curves fitting the experimental points for the heat transfer coefficient h,

Fig. 6 *Lower part* heat transfer coefficient for the FC-87/FC-72 mixtures against the liquid molar fraction x of the most volatile component (FC-87); *upper part* phase-diagram for the FC-87/FC-72 mixture
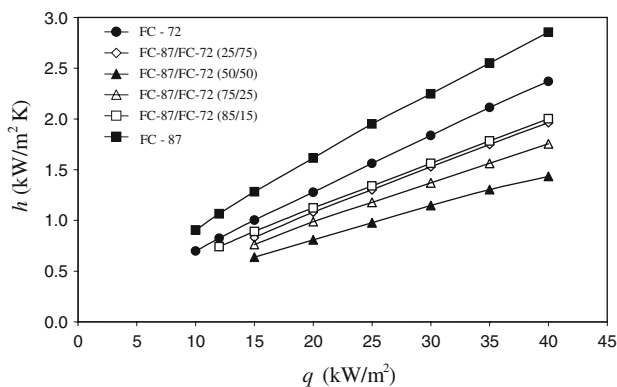

Fig. 7 Experimental results as a function of heat flux, for different concentrations

computed using Eq. (1), as a function of the molar fraction, for heat fluxes between 12 and 40 kW/m^2, and, in the upper part, the corresponding theoretical phase diagram for the FC-87/FC/72 binary mixtures. For all the cases, the minimum loci of the curves for h occurred for a mixture with a molar fraction of 50/50, that corresponds to the maximum difference between the dew point and the bubble point, ΔT_{db}, as shown in the phase diagram.

Figure 7 shows the experimental heat transfer coefficients for the four tested concentrations of FC-87/FC-72 mixture and also for the pure fluids. For all heat fluxes the h values for the 50/50 mixture are the lowest. In fact, the maximum theoretical temperature difference ΔT_{db}, approximately 6 K, occurs for this concentration as we can observe in the phase diagrams, in the upper part of Fig. 6. For the nucleate boiling regime, the relation between the experimental heat transfer coefficients h and the heat flux q, $h = Cq^n$, is given as $n = 0.86$, and $n = 0.83$ considering only the four mixtures. This is close to the upper limit of published results, where n is a value between 0.6 and 0.8, as reported by Stephan [2].

4.2 Comparison of h with models and correlations

Figures 8 and 9 show the comparison of the experimental heat transfer coefficients with those calculated using the correlations of Rohsenow, Eq. (10), with $C_{sf} = 0.0065$ and 0.0079 for FC-72 and FC-87, respectively, Stephan and Abdelsalam, Eqs. (11, 12) and Cooper, Eqs. (13–15), for FC-87 and FC-72, respectively. For FC-87 the average absolute deviations of 9, 76.7 and 79.4% were found comparing with Cooper, Stephan–Abdelsalam and Rohsenow's correlations, respectively. A similar trend was observed for FC-72, with average absolute deviations of 11.8, 36 and 58.8% compared with Cooper, Stephan–Abdelsalam and Rohsenow's correlations, respectively.

It is important to observe that the maximum value of the heat flux, 40 kW/m^2, tested in this study is equivalent to 26
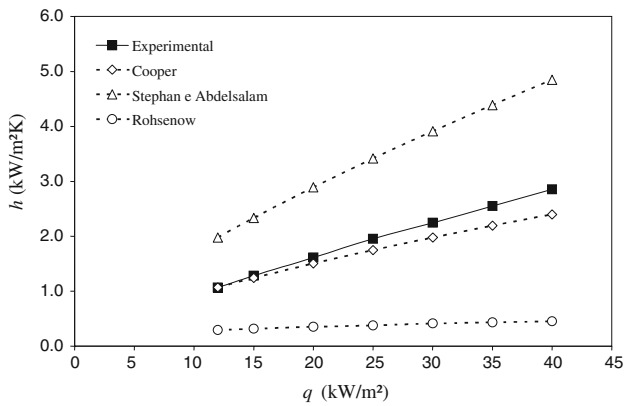
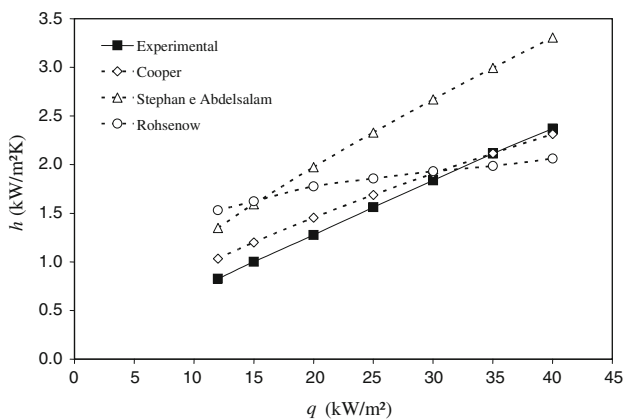**Fig. 8** Comparison of h with the correlations for FC-87

Fig. 9 Comparison of h with the correlations for FC-72

Fig. 10 Comparison of h with the models for FC-87/FC-72 mixture for heat flux of 30 kW/m^2
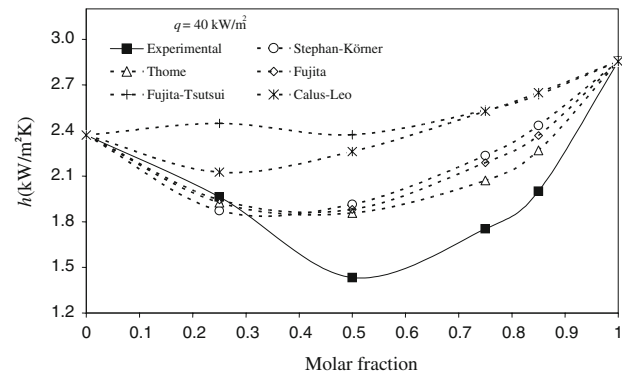
Fig. 11 Comparison of h with the models for FC-87/FC-72 mixture for heat flux of 40 kW/m^2

Table 3 Deviations between h_{exp} and h_{th}

Model	Average deviation (AD)
Thome	13.6
Fujita et al.	19.4
Stephan and Körner ($A_0 = 1.53$)	19.6
Stephan and Körner ($A_0 = 3.36$)	9.7
Calus and Leonidopoulos	35.2
Fujita and Tsutsui	39.3

$$AD = \sum \frac{\left| h_{th} - h_{exp} \right|}{h_{exp}}$$

Calus–Leonidopoulos and Fujita and Tsutsui gave deviations higher than 35%.

and 22.5% of the critical heat fluxes, calculated using Zuber's equation, Carey [3]: 153 and 178 kW/m^2, for FC-72 and FC-87, respectively. The maximum value of the heat flux is also lower than the heat flux which characterizes the transition from the isolated to the coalesced region of the nucleate boiling regime, as calculated using the Moissis and Berenson correlation, as presented by Carey [3] and verified by Cardoso and Passos [22]. This region of the boiling curve, for low and moderate heat fluxes, is characterized by particular difficulties in correlating experimental results with the above correlations for pure fluids.

Figures 10 and 11 show the comparison of experimental results for h with those calculated by the models listed in Table 1, for heat fluxes of 30 and 40 kW/m^2. Except for the cases with a molar fraction of 25/75, all the calculated values are higher than the experimental results.

Table 3 summarizes the average deviations AD of the experimental results for the heat transfer coefficients from those calculated using the semi-empirical models listed in Table 1. The lowest average deviations, <20%, were obtained by comparison with the models of Stephan–Korner, Thome and Fujita et al., whereas comparisons with
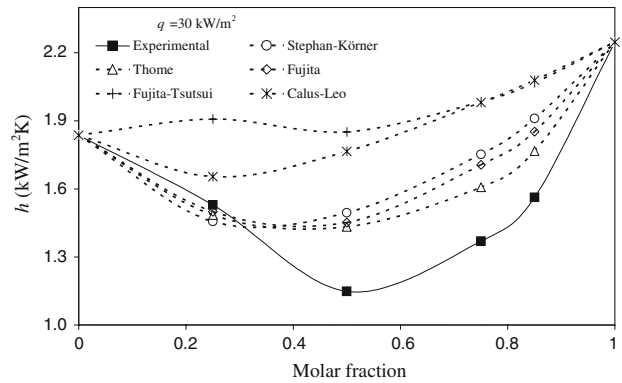
5 Conclusions

The following conclusions can be drawn from this study:

1. The linearly interpolated heat transfer coefficients, h, for the nucleate boiling regime, for FC-72 and FC-87, as a function of the molar fraction, were always higher

than those for FC-87/FC-72 zeotropic mixtures, and for FC-87 this value was 20% higher than that for FC-72;

2. For FC-87/FC-72 mixtures, with molar fractions of 25/75, 50/50, 75/25 and 85/15, the highest h was obtained with a molar fraction of 85/15, this being 30% less than the h value for FC-87. The lowest h value was found for a molar fraction of 50/50, which is 50 and 36% less than those for FC-87 and FC-72, respectively. For 25/75 and 75/25 molar fractions, h was 33 and 39%, respectively, less than the values for FC-87;

3. For FC-72 and FC-87, the experimental heat transfer coefficients for the nucleate boiling regime were compared with those calculated by Cooper [17], Stephan and Abdelsalam [16] and Rohsenow correlations. The lowest average absolute deviations were 9 and 11.8% compared with Cooper's previous correlation;

4. The experimental mixture heat transfer coefficients, for heat fluxes between 15 and 40 kW/m^2, were compared with those calculated by the models proposed by Stephan and Körner [12], with $A_0 = 1.53$, Thome [8], Fujita et al. see [13], Fujita and Tsutsui [14] and Calus and Leonidopoulos [10] and the average linear deviations were 19.6, 13.6, 19.4, 39.3, and 35.2, respectively. Using $A_0 = 3.36$ the comparison with Stephan–Körner's model gives an average deviation of 9.7%.

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