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Forced convective boiling in binary mixtures

GIAN PIERO CELATA, MAURIZIO CUMO and TOMMASO SETARO
ENEA ENE-IMPE, Thermal Process Engineering Division, C.R.E. Casaccia-Via Anguillarese, 301-00060, Rome, Italy

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Abstract—ENEA started in 1990 a new research programme on 'Thermal Hydraulics Of Mixtures' (THOM) focused on the heat transfer in forced convective boiling of refrigerant mixtures. The main aim of the research is to accomplish new experimental data on binary mixtures in forced convective boiling and particularly on three main topics: convective boiling heat transfer, hysteresis in nucleate boiling incipience and critical heat flux. A first preliminary step of the research, started in the middle of 1990, consisted in the thermal hydraulic characterization of the pure components of the binary mixture chosen, namely R 12 (CCl_2F_2) and R 114 ($C_2Cl_2F_4$). At the end of 1991 (and still in progress) the main investigation on the heat transfer performance of different compositions of an R 12/R 114 mixture started. In the present paper, a review of previous works, together with a description of the first experimental results on heat transfer coefficients is given. Finally a comparison of experimental data with the available correlations, is accomplished.

1. INTRODUCTION

MANY INDUSTRIALIZED nations are concentrating on the R & D of the advanced technology in the efficient use of energy including waste heat recovery from various industrial processes with moderate and/or lower temperature levels. Increasing attention to heat pump systems exactly reflects this demand, since they are able to take heat energy from a lower temperature source, to use mechanical work so as to upgrade the temperature and then to reject both heat from the lower temperature source and the mechanical work to the higher temperature sink. In this peculiar advanced technology, the particular importance of selecting the optimum working fluid of the system has to be stressed. Thus, nonazeotropic binary mixtures become attractive to the R & D of advanced vapour-compression heat pump systems. Nevertheless, in spite of the interest in using nonazeotropic refrigerant mixtures for increasing the coefficient of performance of heat pumps to achieve a better energy conservation, little attention has been paid to the boiling of liquid mixtures in the presence of a forced flow. In the following sections, the present state of the art in this field is reviewed.

2. AN OVERVIEW ON BINARY MIXTURES FLOW BOILING RESEARCH

2.1. *Experimental research*

Research on forced convective boiling of binary mixtures has not yet been conducted in an exhaustive way. Only over the last ten years has a more refined evaluation of thermophysical and thermodynamic properties of binary mixtures been performed, and in particular an evaluation of heat transfer characteristics. Sivagnanam and Varma [1] also verified the presence of hysteresis at the onset of subcooled boiling using aqueous mixtures in forced convection outside a vertical heating wire provided along the centre line of a glass tube, and particularly revealed an increase of the wall superheating at the onset of subcooled boiling with the increase of the mole fraction of the more volatile component. Mishra *et al.* [2] made an experimental research using a binary mixture of R 12 and R 22 with various mole fractions to evaluate the local heat transfer coefficients in forced convective boiling in a horizontal channel. The experimental results showed a lower heat transfer coefficient of the binary mixture with respect to the simple linear interpolation between the values corresponding to the two pure components. This result was confirmed also by Fink *et al.* [3] with a different mixture, obtained using R 11 and R 113, in the nucleate boiling region, as shown in Fig. 1. In the same figure, the negligible influence of the velocity on the heat transfer coefficient is confirmed. Fink *et al.* [3] also verified the non-dependence from the fluid subcooling of the heat transfer coefficient of the mixtures, as known for the pure components. A further study was made by Ross *et al.* [4], devoted to heat pump applications, using an R 152a and R 13B1 mixture in horizontal forced convective boiling inside a channel. Ross *et al.* assessed that at very low pressures (0.1 MPa) the heat transfer regime is mainly nucleate boiling dominated. Recently, Jung *et al.* [5] have performed tests with a nonazeotropic binary mixture of R 22 and R 114, using a horizontal test section; the results obtained by Jung *et al.* [5] verify the heat transfer coefficient trends already known for nonazeotropic binary mix-

NOMENCLATURE

A parameter, equation (4)
Bo boiling number, q''/Gh_{fg}
C specific heat [kJ kg^{-1} K^{-1}]
D_i inner diameter [m]
D liquid mass diffusivity [m^2 s^{-1}]
F nucleate boiling factor, equation (1)
G specific mass flow rate [kg m^{-2} s^{-1}]
h heat transfer coefficient [kW m^{-2} K^{-1}]
h_{fg} latent heat of vaporization [kJ kg^{-1}]
h_m mass transfer coefficient [m s^{-1}]
k thermal conductivity [W m^{-1} K^{-1}]
M molecular weight
m parameter defined in equation (4)
$\dot{m}$ mass flow rate [kg s^{-1}]
n parameter defined in equation (4)
Pr Prandtl number, $\mu C_p/k$
p pressure [MPa]
q'' heat flux [kW m^{-2}]
R radius [m]
Re Reynolds number, $uD\rho/\mu$
S suppression factor, equation (1)
Sc Schmidt number, $\mu/\rho D$
T temperature [K]
u velocity [m s^{-1}]
X quality
X_{tt} Lockhart–Martinelli parameter, $(\mu_L/\mu_V)^{0.1}(\rho_V/\rho_L)^{0.5}(1-X/X)^{0.9}$
x liquid mole fraction of the more volatile component
x_M liquid composition based on mass of the more volatile component
y vapour mole fraction of the more volatile component
y_M vapour composition based on mass of the more volatile component.

Greek symbols
α thermal diffusivity [m^2 s^{-1}]
μ dynamic viscosity [kg m^{-1} s^{-1}]
ρ density [kg m^{-3}]
σ surface tension [N m^{-1}].

Subscripts
1 pertains to the more volatile component
2 pertains to the less volatile component
bulk pertains to the bulk
i ideal
in inlet
L pertains to the liquid phase
mac macroscopic
mic microscopic
mix pertains to the mixture
s,sat pertains to saturation conditions
sub subcooled
tp two-phase
V pertains to the vapour phase
w wall.

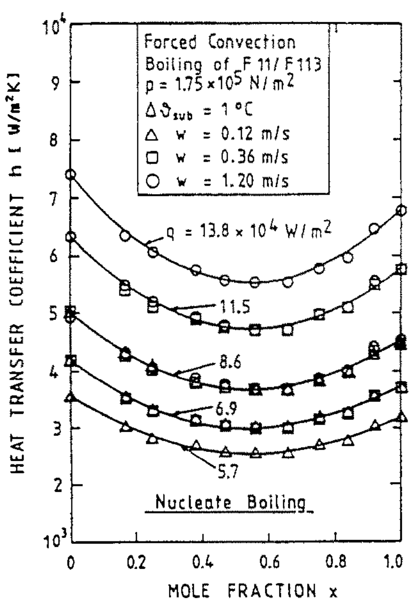


FIG. 1. Boiling heat transfer coefficient as a function of composition at different liquid velocity and heat flux, for R 11/R 113 mixtures (Fink *et al.* [3]).

tures as shown in Fig. 2, i.e. the heat transfer coefficient has a reduction for low values of mole fraction, tends to a constant value for intermediate values of the mole fraction (0.2 < mole fraction < 0.6) and then increases sharply for mole fractions of R 22 higher than 0.8. Another interesting investigation has been performed by Kedzierski and Didion [6] who visualized the nucleate flow boiling of an R 22/R 114 mixture in a horizontal channel. They verified a comparable amount of nucleation between the more volatile component, R 22, and the mixture, even though the mixture was mostly R 114 by mole, while R 114 exhibited much more nucleation than either R 22 or the R 22/R 114 mixture. Hihara and Saito [7] performed experiments with an R 22/R 114 mixture in a horizontal tube and studied the variation of the circumferentially averaged heat transfer coefficient with overall composition. As shown in Fig. 3, heat transfer coefficients for mixtures are much lower than those for pure R 22 and R 114. Takamatsu *et al.* [8], using R 22/R 114 mixtures, presented experimental results for heat transfer and pressure drop during forced convective boiling inside a horizontal

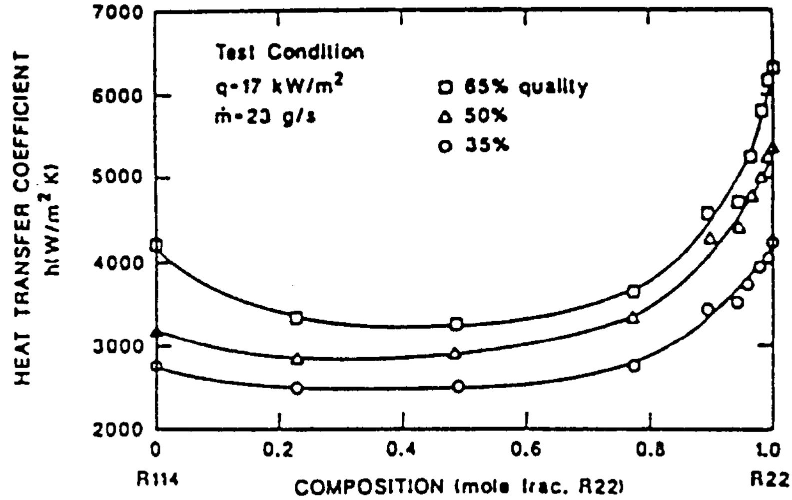


FIG. 2. Boiling heat transfer coefficient as a function of composition at different steam quality, for R 22/R 114 mixtures (Jung *et al.* [5]).

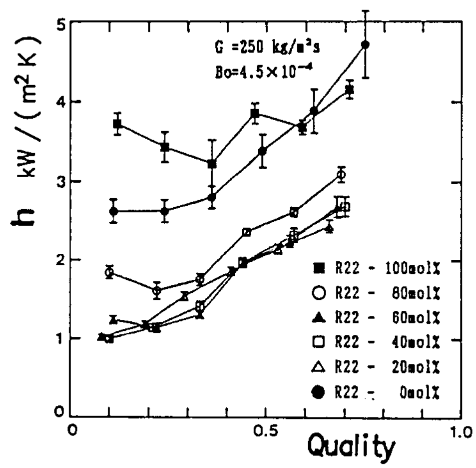


FIG. 3. Boiling heat transfer coefficient as a function of steam quality at different compositions, for R 22/R 114 mixtures (Hihara and Saito [7]).

tube with internal spiral grooves, heated by means of water flowing countercurrently in an outer annulus. They noted that the temperature distribution of the mixture is convex downward for a low mole fraction of the less volatile component, R 114, while is convex upward for a high mole fraction of the less volatile component, R 114. Bennett and Chen [9] have made one of the few studies with vertical channels on binary mixtures in forced convective boiling. They used an aqueous mixture of ethylene/glycol and evaluated the local heat transfer coefficient with different mixture compositions and at different qualities (0 < quality < 30%). Kawano [10] made another study with binary mixtures in forced convective boiling along tubes on the shell side of a vertical shell-and-tube heat exchanger, using R 114/R 152a mixtures, and verified the lower heat transfer coefficients of mixtures with respect to those of single-component fluids.

The above analysis of the experimental research devoted to binary mixtures shows a lack of experimental data in this field and particularly in the vertical forced convective flow, where very few have been performed in the last two decades, as previously reported by Stephan [11] and Shock [12] and recently reviewed by Steiner [13].

2.2. *Heat transfer correlations*

The empirical evaluation of the heat transfer coefficient in forced convective boiling has been performed by different authors using modified heat transfer correlations, derived from the original ones developed for pure components. Most of the common derivations are obtained from two typical relationships for the heat transfer coefficient in forced convective boiling of pure fluids. The first relation is derived from the well known correlation by Chen [14] for pure liquids:

$$h = h_{\mathrm{mac}}F + h_{\mathrm{mic}}S \qquad (1)$$

where

$$h_{\mathrm{mac}} = 0.023\frac{k_{\mathrm{L}}}{D_{\mathrm{i}}}Re_{\mathrm{L}}^{0.8}\,Pr_{\mathrm{L}}^{0.4},$$

$$Re_{\mathrm{L}} = \frac{GD_{\mathrm{i}}(1-X)}{\mu_{\mathrm{L}}},$$

$$F = \begin{cases} 1 & \text{if } \dfrac{1}{X_{\mathrm{tt}}} \leqslant 0.1 \\ 2.35\left[\dfrac{1}{X_{\mathrm{tt}}}+0.213\right]^{0.736} & \text{if } \dfrac{1}{X_{\mathrm{tt}}} > 0.1 \end{cases},$$

$$\frac{1}{X_{\mathrm{tt}}} = \left(\frac{\rho_{\mathrm{L}}}{\rho_{\mathrm{V}}}\right)^{0.5}\left(\frac{\mu_{\mathrm{V}}}{\mu_{\mathrm{L}}}\right)^{0.1}\left(\frac{X}{1-X}\right)^{0.9},$$

$$h_{mic} = 0.00122 \frac{k_L^{0.79} Cp_L^{0.45} \rho_L^{0.49}}{\sigma^{0.5} \mu_L^{0.29} \rho_V^{0.24} h_{fg}^{0.24}} \Delta T_{sat}^{0.24} \Delta p_{sat}^{0.75},$$

$$\Delta T_{sat} = T_w - T_{sat},$$

$$\Delta p_{sat} = p_{sat}(T_w) - p_{sat}(T_{sat}),$$

$$S = \frac{1}{1 + 2.53 \times 10^{-6} Re_{tp}^{1.17}},$$

$$Re_{tp} = Re_L F^{1.25}.$$

In equation (1), h_{mic} is the microscopic heat transfer coefficient, i.e. the nucleate boiling contribution to the heat transfer, and h_{mac} is the macroscopic heat transfer coefficient, i.e. the convective boiling contribution to the heat transfer.

The second relation for the heat transfer coefficient is based on the relationship:

$$\frac{h}{h_L} = f(X_{tt}, Bo), \tag{2}$$

where h_L is the heat transfer coefficient if the pure liquid was flowing through the tube, X_{tt} is the Lockhart–Martinelli parameter and Bo is the boiling number.

Bennett and Chen [9] have proposed an extension of equation (1) to binary mixtures. They derived the h_{mic} coefficient using an expression proposed by Forster and Zuber [15] for pool boiling, modified, taking into account the greater thermal gradient in the vapour generating zone near the wall due to the forced convection, defining a suppression factor which is a function of the two-phase Reynolds number. This suppression factor, obtained from a solution given by Florschuetz and Khan [16], tends to unity when the flow rate tends to zero and is equal to zero for flow rates tending to infinity. The h_{mac} coefficient is obtained from the Dittus–Boelter–Kraussold equation (Stephan [11]). Bennett and Chen [9] also introduced a corrective factor to take into account the effective mass transfer on the thermal driving force. The final expression of their correlation is given by:

$$h_{mix} = h_{mac} F_{mix} + h_{mic} S_{mix}, \tag{3}$$

where

$$F_{mix} = F f(Pr_L) \left[\frac{\Delta T}{\Delta T_s}\right]_{mac},$$

$$f(Pr_L) = \left[\frac{Pr_L + 1}{2}\right]^{0.444},$$

$$\left[\frac{\Delta T}{\Delta T_s}\right]_{mac} = 1 - \frac{(1 - y_M) q''}{\rho_L h_{fg} h_m \Delta T_s} \left.\frac{dT_{sat}}{dx_M}\right|_{p_{bulk}},$$

$$h_m = 0.023 \frac{D}{D_i} Re_{tp}^{0.8} Sc^{0.4},$$

$$S_{mix} = \frac{S}{1 - \frac{C_{pL}(y_M - x_M)}{h_{fg}} \frac{dT_{sat}}{dx_M} \left(\frac{\alpha}{D}\right)^{1/2}},$$

$$S = \frac{1}{1 + 2.53 \times 10^{-6} Re_{tp}^{1.17}},$$

$$Re_{tp} = Re_L [f(Pr_L) F]^{1.25}.$$

Bennett and Chen [9] validated their correlation using the experimental data obtained with aqueous mixtures of ethylene glycol; the correlation is able to predict experimental data within an error of $\pm 15\%$.

Mishra *et al.* [2] correlated their experimental data, obtained using R 12/R 22 mixtures, with the expression:

$$\frac{h}{h_L} = A \left(\frac{1}{X_{tt}}\right)^m (Bo)^n. \tag{4}$$

Mishra *et al.* [2] gave the coefficient values for two different mixture composition, as:

(a) R 12 23–27%, R 22 77–73%: $A = 5.64$, $m = 0.23$, $n = 0.05$,

(b) R 12 41–48%, R 22 59–52%: $A = 21.75$, $m = 0.29$, $n = 0.23$.

The correlation of Mishra *et al.* [2] shows a mean deviation of $\pm 30\%$ on the data of composition type (a) while a better performance is obtained if used on the data of composition type (b).

3. PRESENT RESEARCH

The programme on Thermal Hydraulics Of Mixtures (THOM) focused on the heat transfer in forced convective boiling of refrigerant mixtures up to the dryout condition. In the following sections a description of the experimental loop, together with the experimental results, is given. A comparison of the experimental data with the existing correlation is also accomplished.

3.1. *Experimental apparatus*

The experimental loop, schematically represented in Fig. 4, consists mainly of a piston pump, an electric heater, a condenser and a storage tank. The maximum operating pressure of the loop is 3.5 MPa, while the maximum specific flow rate is 1800 kg m^{-2} s^{-1}; the available electrical power is 10 kW for the electric heater and 15 kW for the test section. The test section is an industrial stainless steel (AISI 316), circular duct uniformly heated (Joule effect) over a length of 2300 mm, with an inner diameter of 7.57 mm and a wall thickness of 0.975 mm. The test section instrumentation consists of 0.5 mm, K-type insulated thermocouples distributed according to the scheme of Fig. 5 for the wall (eleven) and the fluid (fifteen) temperature measurements. Thermocouples located at the same section of the channel have been connected to give directly the temperature difference between the outer wall and the bulk fluid, $\Delta T = T_{w,o} - T_{bulk}$. Each couple of sensors has been chosen with the same calibration curve to avoid any offset error. The temperature difference, ΔT, is reduced by the temperature

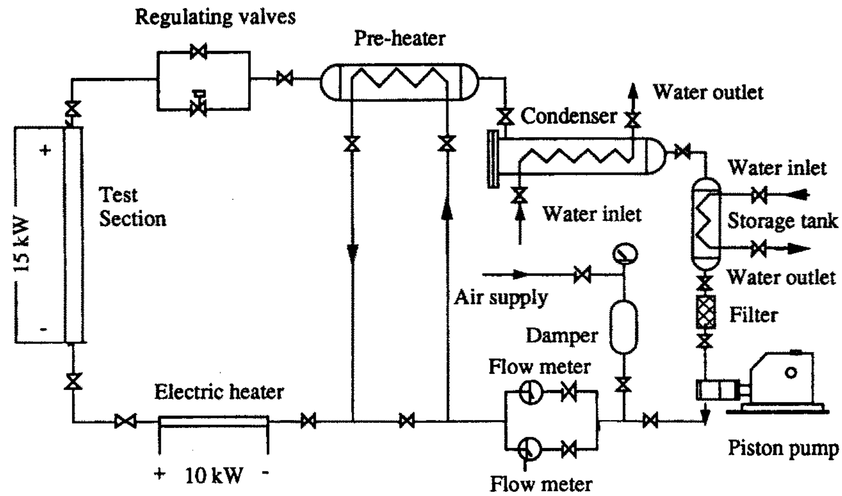


FIG. 4. Schematic of the THOM loop (ENEA).

drop across the wall, $T_{w,o}-T_{w,i}$, calculated using the Fourier equation. From this point on, when speaking of wall temperature, T_w, we intend the inner tube wall temperature, i.e. $T_w = T_{w,i}$. The remaining four thermocouples for fluid temperature measurements provide the absolute fluid temperature trend. Two pressure transducers measure the pressure at the inlet and outlet of the test section. A turbine flowmeter measures the volumetric flow rate at the inlet of the test section. The heating power has been gauged with

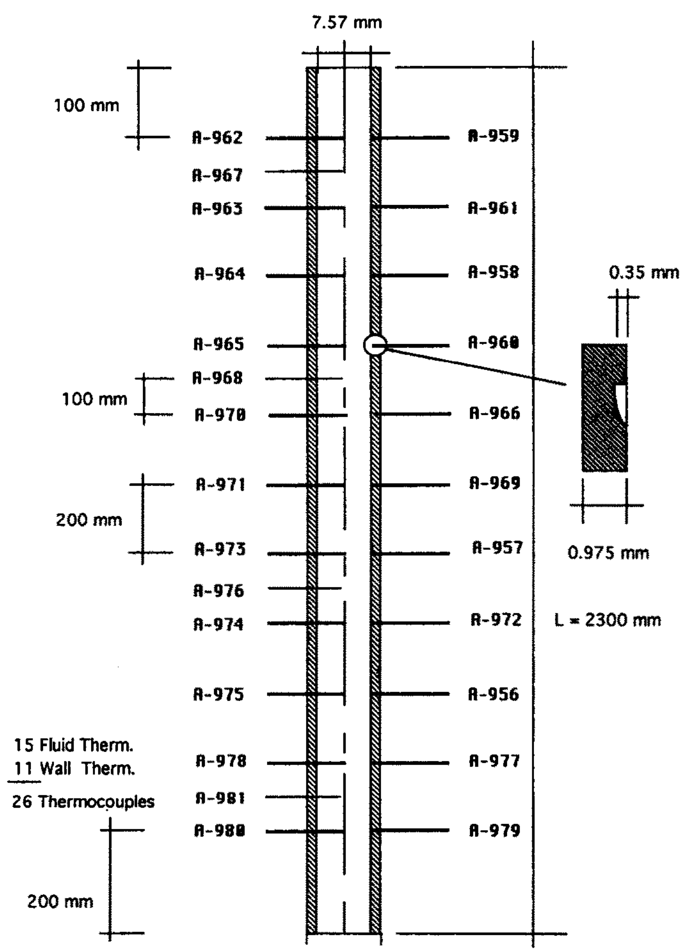


FIG. 5. Schematic of the THOM test section (ENEA).

a wattmeter. The fluid flow is upwards with subcooled inlet conditions. The system was charged with the mixture of R 12 and R 114 of the required composition by introducing known masses of each refrigerant into the storage tank.

3.2. *Physical properties evaluation*

For the analysis of the data and comparison with the predictive methods, all thermodynamics properties, such as density, composition, enthalpy, and saturation temperature, for both pure and mixed refrigerants were calculated by the Carhahan–Starling–DeSantis (CSD) equation of state, as proposed by Morrison and McLinden [17].

Specific heat of liquid is calculated using a linear mass fraction weighting method, as:

$$Cp_{L,mix} = x_{M1}Cp_{L,1} + x_{M2}Cp_{L,2}. \quad (5)$$

Liquid thermal conductivity, $k_{L,mix}$, is evaluated using the correlation proposed by Filippov [18]:

$$k_{L,mix} = x_{M1}k_{L,1} + x_{M2}k_{L,2} - 0.72x_{M1}x_{M2}(k_{L,2} - k_{L,1}). \quad (6)$$

It should be noted that the components were chosen so that $k_{L,2} \geqslant k_{L,1}$.

Liquid viscosity, $\mu_{L,mix}$, is derived with the method proposed by Kandlikar *et al.* [19], as:

$$\ln(\mu_{L,mix}) = x\ln(\mu_{L,1}) + (1-x)\ln(\mu_{L,2}). \quad (7)$$

Actually, Kandlikar *et al.* [19] used a mass fraction in equation (7). However, it was found that the mole fraction better fitted the data. Thus, the mole fraction was used throughout this study.

Vapour viscosity, $\mu_{V,mix}$, was obtained from Wilke's correlation [20], given by:

$$\mu_{V,mix} = \frac{y_1\mu_{V,1}}{y_1 + y_2P_{12}} + \frac{y_2\mu_{V,2}}{y_2 + y_1P_{21}}, \quad (8)$$

where P_{ij} is defined as

$$P_{ij} = \frac{[1 + (\mu_{V,i}/\mu_{V,j})^{0.5}(M_iM_j)^{0.25}]^2}{[8(1 + M_i/M_j)]^{0.5}}, \quad (9)$$

and M is the molecular weight.

Surface tension of the mixture, σ_{mix}, is calculated using a linear mole fraction weighting method, as:

$$\sigma_{mix} = x\sigma_1 + (1-x)\sigma_2. \quad (10)$$

3.3. *Experimental results*

A test matrix was defined to investigate the different parameters affecting the heat transfer coefficients of binary mixtures. One value of inlet subcooling and three fluid inlet flow rates were chosen while, because of the significant dependence of heat transfer on the pressure, a wide range of pressures was investigated.

Table 1.

	Heat transfer experiments
Fluid	R 12 (CCl_2F_2), R 114 ($C_2Cl_2F_4$)
x (R 12)	0, 0.136, 0.377, 0.586, 0.767, 1
p [MPa]	1.0, 1.2, 1.5, 1.8, 2.0, 2.2
$\dot{m}$ [kg s^{-1}]	0.025, 0.047, 0.07
$\Delta T_{sub,in}$ [K]	12

Globally, the range of variation of the parameters in the tests performed is given in Table 1.

The heat transfer coefficient obtained in the boiling regime is generally lower than the ideal heat transfer coefficient, h_i, defined using the linear mole fraction method, as:

$$h_i = xh_1 + (1-x)h_2. \quad (11)$$

In Fig. 6, a typical trend of the ratio between actual and ideal heat transfer coefficient, h/h_i, vs the more volatile component mole fraction, x (R 12) is shown for three different mass fluxes, with the pressure as a parameter; the inlet subcooling is equal to 12 K. The degradation of heat transfer coefficient as a function of the mixture composition appears to be dependent on saturation pressure, increasing with the saturation pressure. Looking at the top graph of Fig. 6, in which the vapour/liquid mole fraction difference for the more volatile component ($y-x$) is plotted for the two examined pressures, we can also verify the correspondence of the maximum degradation of heat transfer coefficient with the maximum value of the ($y-x$) difference, confirming a typical behaviour already observed and studied in pool boiling of mixtures [21]. Figure 7 shows the variation of the mixture heat transfer coefficient with the mixture composition. The deviation from the ideal behaviour of the mixture (equation (11)) is evident, i.e. the non-linear fashion of the h–x curve. Lines in Figs. 6 and 7 represent the best fit of the data. The experimental uncertainty of the heat transfer coefficient has been evaluated within $\pm 15\%$.

3.4. *Data analysis*

The data analysis is concerned with the evaluation of the performance of existing correlations for the heat transfer coefficient in forced boiling of binary mixtures, as detailed in the previous paragraphs. Figure 8 shows the comparison of the theoretical heat transfer coefficients in the convective boiling region, obtained using the correlations by Chen (graph a), by Bennett and Chen (graph b) and by Mishra *et al.* (graph c), with the experimental heat transfer coefficients. The correlation recommended by Chen [14], even if developed for pure fluids, shows a good performance and seems to equate the Bennett and Chen [9] correlation. The correlation proposed by Mishra *et al.* [2] gives a less accurate prediction of the experimental heat transfer coefficients. However, if we plot the data points percent of the three correlations

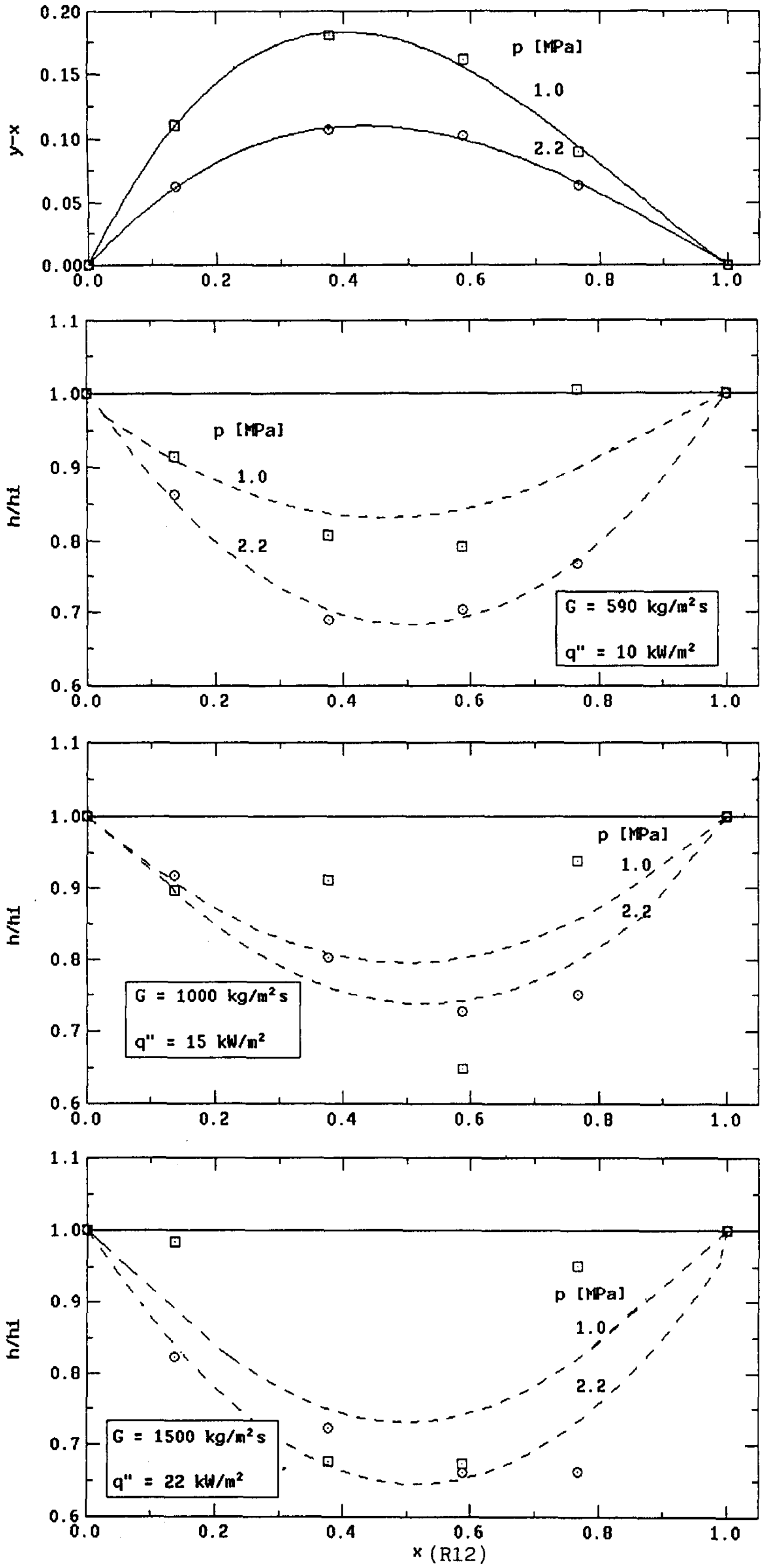


FIG. 6. Vapour/liquid mole fraction difference as a function of composition (top graph) and relative decrease of the heat transfer coefficient as a function of composition at various mass fluxes and pressures, for R 12/R 114 mixtures (ENEA).

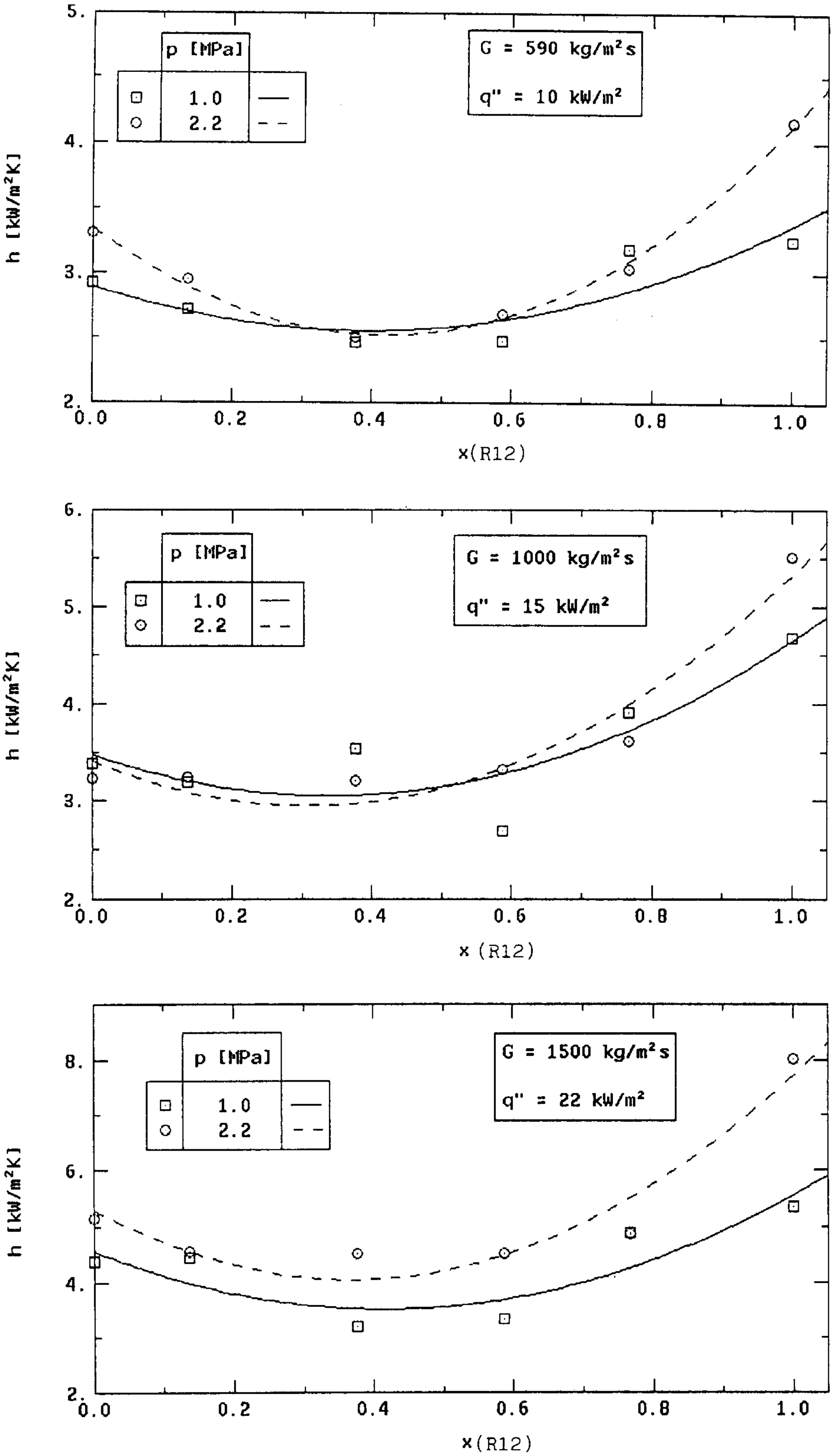
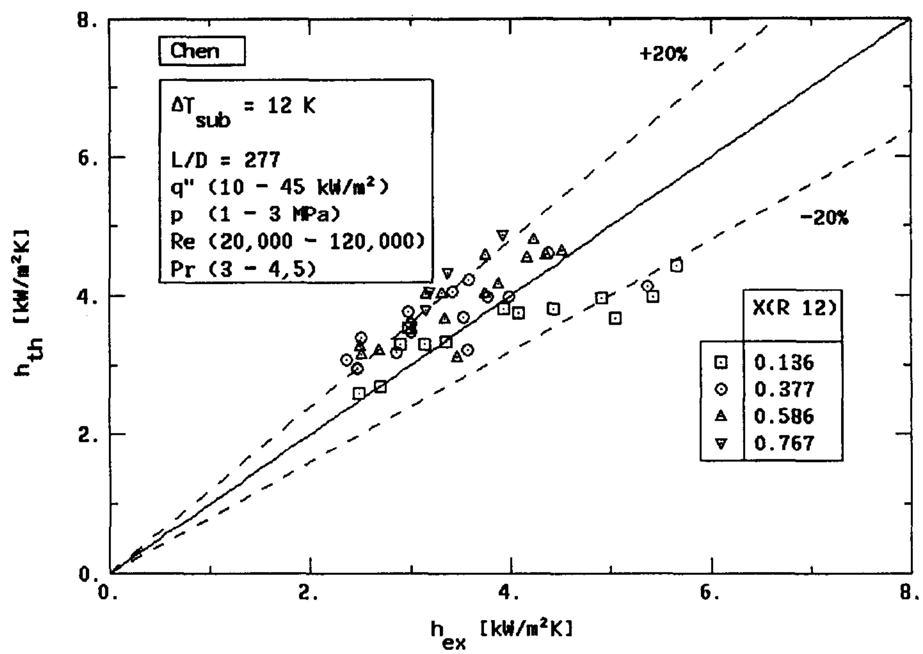
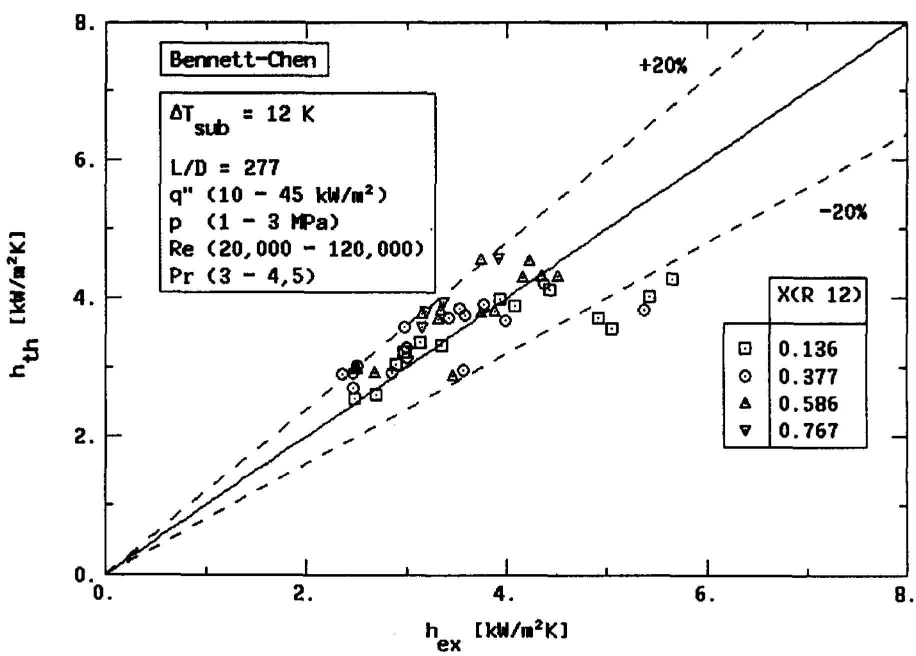


FIG. 7. Heat transfer coefficient as a function of composition at various mass fluxes and pressures, for R 12/R 114 mixtures (ENEA).

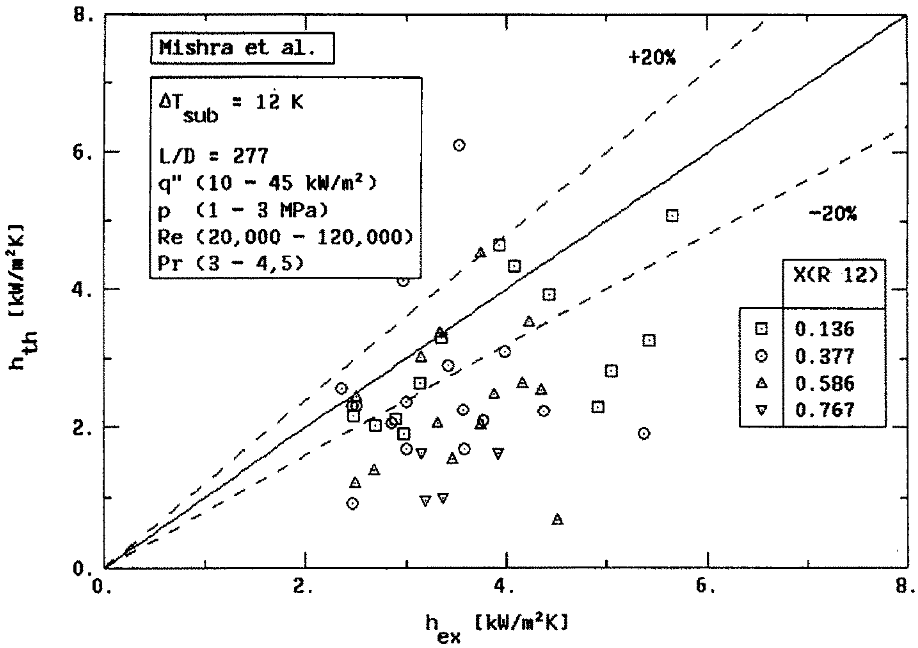


(a)



(b)

FIG. 8. Comparison of available correlations by Chen [14] (graph a), by Bennett and Chen [9] (graph b) and by Mishra *et al.* [2] (graph c), with the experimental heat transfer coefficients (ENEA).



(c)

FIG. 8.—*Continued.*

vs the different error bands ranging from ± 10 to $\pm 40\%$, it is possible to verify that the Bennett–Chen correlation has the best performance through the whole error band range, as clearly shown in Fig. 9.

Thus, the Chen correlation upgrade to binary mixtures, made by Bennett and Chen, is justified. Finally, the Bennett–Chen correlation shows an error greater than the claimed one in the original recommendation, around ± 25–30%, due, probably, to the use of a Refrigerants mixture instead of an aqueous solution.

4. CONCLUSIONS

The first analysis of the experimental data on binary mixtures R 12/R 114 in upflow forced convective boiling shows that the degradation of heat transfer coefficient as a function of the mixture composition appears to be depending on both saturation pressure and mass flux. The comparison of theoretical heat transfer coefficients in the convective boiling region, obtained using the correlations by Chen [14], Bennett

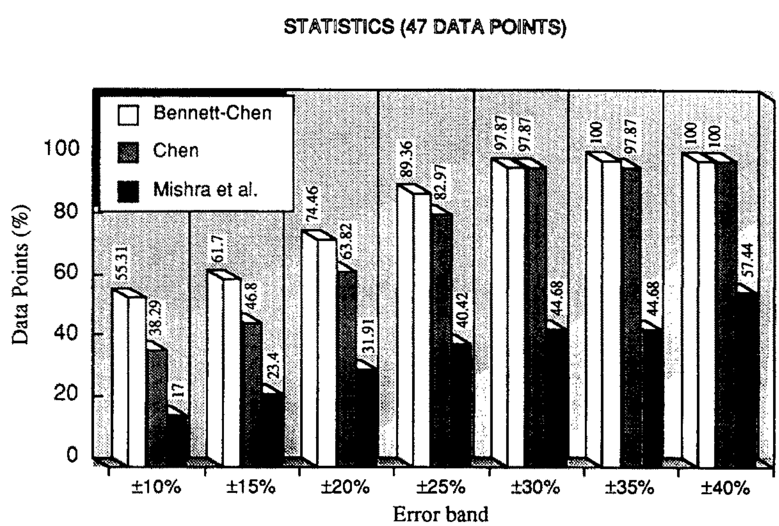


FIG. 9. Comparison of different heat transfer correlation performances using different mass fluxes, pressures and composition of R 12/R 114 mixtures, as detailed in the test matrix (ENEA).

and Chen [9] and by Mishra *et al.* [2], with experimental heat transfer coefficients confirm the good performance of the Bennett–Chen correlation in predicting heat transfer coefficients of binary mixtures also in the case of Refrigerants mixtures.

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