Condensation and vaporization heat transfer of low-GWP mixtures

S Bortolin, M Azzolin, A Berto, C Guzzardi and D Del Col

University of Padova, Department of Industrial Engineering
Via Venezia 1, 35131 Padova, Italy
Corresponding author e-mail: stefano.bortolin@unipd.it

Abstract. The present work investigates the heat transfer performance of non-azeotropic ternary low-GWP (Global Warming Potential) mixtures during condensation and vaporization inside a 8.0 mm inner diameter channel. The tested mixtures are R455A, which is a ternary blend of R32, R1234yf and R744 (21.5/75.5/3.0% by mass composition), and R452B, which is a ternary blend of R32, R1234yf and R125 (67.0/26.0/7.0% by mass composition). R455A presents a GWP100-yr value equal to 146 and a temperature glide of 10 K at pressure $p=17.6$ bar, while R452B is characterized by higher GWP, equal to 676, but lower temperature glide, about 1 K at $p=22.9$ bar. Condensation tests have been run with the two mixtures R455A and R452B at a mean saturation temperature (average value between dew point and bubble point temperatures at constant pressure and given composition) equal to 40 °C and mass velocity ranging between 100 and 600 kg m$^{-2}$ s$^{-1}$. Flow boiling tests have been performed at around 18 °C mean saturation temperature and mass velocity ranging between 200 and 400 kg m$^{-2}$ s$^{-1}$. The effects of vapor quality, mass velocity and heat flux on the heat transfer coefficient are investigated. Moreover, the adoption of zeotropic mixtures poses the problem of how to extend the correlations developed for pure fluids. In the present paper, the contribution of the mass transfer resistance is assessed and the corrections needed in the models are discussed.

1. Introduction
European and international organizations have recently taken actions for a gradual phase-out of high Global Warming Potential (GWP) refrigerants. The F-gas Regulation 517/2014 (European Union, [1]) set a 79% cut in the hydrofluorocarbons (HFCs) supply by 2030, with respect to 2015 data of HFC sales. Furthermore, the Kigali Amendment to the Montreal Protocol on Substances that Deplete the Ozone Layer (United Nations, [2]) is focused on a gradual reduction in the HFCs employment by 80-85% by 2040. This implies that the refrigeration and air-conditioning industry has to replace the traditionally employed refrigerants maintaining the efficiency of industrial devices: the employment of low-GWP fluids represents a necessary step to limit the carbon dioxide emissions of the installations. From the merely economic point of view, the unit price of the currently employed high-GWP refrigerants (e.g. R410A) has recently increased and in some countries taxes associated with the GWP value of the operating fluid are foreseen.

The search for alternative refrigerants is focused primarily on the use of natural fluids and on the employment of new synthetic refrigerants having low-GWP. Natural refrigerants (e.g. hydrocarbons and ammonia) are suggested as good alternatives to HFCs due to their thermodynamic properties and very low GWP; however, they are often flammable or toxic. Pure hydrofluolefins (HFOs) R1234yf and R1234ze(E) can be considered as substitutes for R134a because of their low GWP value. McLinden et al. [3] estimated GWP, flammability, critical temperature and other thermodynamic parameters of about...
30 HFOs, recognizing the potential of R1234yf and R1234ze(E). With regard to the heat transfer performance at the condenser, Del Col et al. [4, 5] experimentally studied heat transfer and pressure drop of R1234yf and R1234ze(E) during condensation at 40 °C inside a 0.96 mm diameter circular minichannel. Unfortunately, single-component low-GWP refrigerants cannot cover all the applications (e.g. drop-in refrigerants for R410A and R404A) and some zeotropic blends of HFOs and HFCs have been proposed. However, zeotropic mixtures introduce an additional mass transfer resistance that leads to a degradation of the heat transfer performance and thus models developed for pure fluids cannot be directly applied (Del Col et al. [6], Azzolin et al. [7]). Therefore it is important to collect new data with these mixtures that can be used for the assessment of available two-phase heat transfer correlations (for both condensation and flow boiling) or to develop new models.

1.1. Condensation heat transfer coefficient

Although several experimental and numerical investigations have been conducted on ternary non-azeotropic HFCs mixtures (Sami and Grell [8], Cavallini et al. [9]), relatively poor studies have been published on condensation performance of ternary HFOs/HFCs blends. Kondou et al. [10] measured the heat transfer coefficients and pressure drop of R744/R32/R1234ze(E) mixtures at different mass compositions during condensation and evaporation inside a horizontal microfin tube with equivalent inner diameter of 5.35 mm. In both condensation and vaporization processes, the heat transfer coefficients of the mixtures were strongly penalized by the additional mass transfer resistance. Azzolin et al. [11] measured the condensation heat transfer coefficients and two-phase frictional pressure drops of ternary low-GWP zeotropic mixtures R455A and R452B inside a minichannel (0.96 mm inner diameter) and a conventional tube (8.0 mm inner diameter). They found that mixture R452B presents higher heat transfer coefficients with respect to R455A and, at the same operating conditions, the heat transfer coefficient inside the 0.96 mm diameter minichannel is higher than the one measured inside the 8.0 mm diameter channel.

1.2. Flow boiling heat transfer coefficient

Regarding flow boiling heat transfer coefficient measurements, experimental works carried out with the aim to investigate the flow boiling characteristics of non-azeotropic ternary mixtures of HFCs and HFOs are limited in the literature. Lillo et al. [12] recently investigated the flow boiling heat transfer performance of R452A, which is a ternary non-azeotropic blend of R32, R125 and R1234yf at 11.0/59.0/39.0% by mass composition with a temperature glide of about 3 K, inside a 6.0 mm inner diameter channel. The heat transfer coefficients of the mixture were found to be dependent on heat flux and mass velocity. Local heat transfer measurements during flow boiling of three low GWP HFOs/HFCs mixtures inside a horizontal microfin tube of hydraulic diameter equal to 5.45 mm were performed by Kedzierski and Kang [13]. They tested the mixtures R452B, R448A and R449A. The mixture R452B exhibited the highest heat transfer coefficient, due to the smaller temperature glide.

1.3. Present study

The present study is aimed at investigating the condensation and flow boiling heat transfer performance of two ternary low-GWP non-azeotropic mixtures, named R455A and R452B, inside a 8.0 mm inner diameter channel. R455A is a blend of R1234yf, R32 and R744 at 75.5/21.5/3.0% by mass composition and can be employed for commercial refrigeration in low temperature applications. R452B is composed of R1234yf, R32 and R125 at 26.0/67.0/7.0% by mass composition and it is suitable for air-conditioning and heat pumps applications. Both these blends belong to the ANSI/ASHRAE A2L class. In Table 1, some thermodynamic and transport properties of the tested refrigerant blends are reported, compared to those of the main pure fluid components, R32, R1234yf and R125. Properties are computed using NIST Refprop 10 (Lemmon et al. [14]).
### Table 1. Properties of mixtures and pure components at 40 °C and 18.2 °C mean saturation temperature computed using NIST Refprop 10 (Lemmon et al. [14]).

|     | R32  | R1234yf | R125  | R455A | R452B |
|-----|------|---------|-------|-------|-------|
| GWP | 677  | <1      | 3170  | 146   | 676   |

|     | Mean saturation temperature equal to 40 °C |
|-----|-----------------------------------------|
| $p_{\text{sat}}$ [bar] | 24.78 | 10.18 | 20.08 | 17.62 | 22.87 |
| $\rho_l$ [kg m$^{-3}$] | 893.0 | 1033.8 | 1088.4 | 988.9 | 924.7 |
| $\rho_g$ [kg m$^{-3}$] | 73.3 | 57.8 | 142.5 | 61.0 | 79.8 |
| $T_{\text{dew}}$ [°C] | 44.9 | | 40.6 | | |
| $T_{\text{bubble}}$ [°C] | 35.1 | | 39.4 | | |

|     | Mean saturation temperature equal to 18.2 °C |
|-----|-----------------------------------------|
| $p_{\text{sat}}$ [bar] | 14.02 | 5.61 | 11.47 | 10.07 | 12.97 |
| $\rho_l$ [kg m$^{-3}$] | 988.5 | 1116.2 | 1228.3 | 1082.7 | 1024.7 |
| $\rho_g$ [kg m$^{-3}$] | 38.7 | 31.1 | 73.9 | 31.5 | 42.1 |
| $T_{\text{dew}}$ [°C] | 23.8 | | 18.8 | | |
| $T_{\text{bubble}}$ [°C] | 12.6 | | 17.6 | | |

#### 2. Experimental apparatus and data reduction

The test section consists of a counterflow tube-in-tube heat exchanger, with inner diameter equal to 8.0 mm. During condensation tests, the subcooled refrigerant is sent by a magnetic-driven pump to a tube-in-tube evaporator where it is vaporized and superheated using hot water as secondary fluid. The superheated refrigerant then passes through a pre-condenser where it partially condenses reaching the desired vapor quality conditions at the inlet of the measuring sector. After the measuring sector, the refrigerant is collected in a post-condenser. The secondary fluid used to remove the condensation heat flow rate in the measuring sector is water and it is provided at different temperatures by means of thermal baths and electrical heaters. Differently, when flow boiling tests are performed, the subcooled refrigerant is sent by the gear pump directly to the test section where it is heated and vaporized using hot water. After the test section, the refrigerant enters in the post-condenser, where it is fully condensed and subcooled.

The quasi-local heat transfer coefficient is obtained by measuring the heat flow rate $q$ exchanged on the water side over the surface area $A$:

$$\text{HTC} = \frac{q}{A \Delta T_{\text{LMTD}}} \quad (1)$$

where the heat flow rate is determined on the water side from the measured water mass flow rate ($\dot{m}_w$) and water temperatures at the inlet ($T_{w,\text{in}}$) and outlet ($T_{w,\text{out}}$) of the test section:

$$q = \dot{m}_w c_w |T_{w,\text{out}} - T_{w,\text{in}}| \quad (2)$$

while the logarithmic mean temperature difference is determined from the saturation and wall temperatures measured at the inlet and outlet of the measuring sector.

$$\Delta T_{\text{LMTD}} = \frac{(T_{\text{sat, in}} - T_{\text{wall, in}}) - (T_{\text{sat, out}} - T_{\text{wall, out}})}{\ln \left( \frac{T_{\text{sat, in}} - T_{\text{wall, in}}}{T_{\text{sat, out}} - T_{\text{wall, out}}} \right)} \quad (3)$$

The saturation and wall temperatures are measured by means of T-type thermocouples. In particular, the wall temperatures at the inlet or outlet of the test section are obtained as the mean value of the readings of four thermocouples, inserted into four equidistant axial grooves around the perimeter of the test tube.
(Cavallini et al. [15]). Detailed description of the experimental apparatus and data reduction procedure is reported in Del Col [16]. The uncertainty analysis is carried out applying the law of propagation of uncertainty and the complete procedure is described thoroughly by Del Col et al. [5]. Table 2 lists type B uncertainty of the measured parameters. Considering the whole experimental database, the mean expanded uncertainty (coverage factor $k = 2$) on the heat transfer coefficient and vapour quality during convective condensation and flow boiling is reported in Table 3 for both the mixtures.

**Table 2.** Type B experimental uncertainty of measured parameters.

| Parameter                  | Uncertainty |
|---------------------------|-------------|
| Temperature               | ±0.05 °C    |
| Temperature difference    | ±0.03 °C    |
| Absolute pressure         | ±0.1% full scale |
| Water flow rate           | ±1% of the reading |
| Refrigerant flow rate     | ±0.4% of the reading |
| Differential pressure     | ±0.06 kPa   |

**Table 3.** Experimental uncertainties for heat transfer coefficient and vapor quality during condensation and flow boiling tests.

|          | Condensation | Flow boiling |
|----------|--------------|--------------|
|          | $u_{HTC}$ [%] | $u_x$ [1]    | $u_{HTC}$ [%] | $u_x$ [1]    |
| R455A    | min | max | min | max | min | max | min | max |
| R455A    | 2.3 | 6.5 | 0.004 | 0.013 | 2.9 | 7.6 | 0.004 | 0.012 |
| R452B    | 2.5 | 7.8 | 0.004 | 0.015 | 3.1 | 8.4 | 0.004 | 0.013 |

3. Condensation heat transfer results

Condensation tests have been run with the mixtures R455A and R452B at mean saturation temperature (between dew and bubble) equal to 40 °C and mass velocity ranging between 100 and 600 kg m$^{-2}$ s$^{-1}$. The vapor quality change in the test section is always lower than 0.19 to determine a quasi-local heat transfer coefficient. In Figures 1 and 2 the experimental heat transfer coefficients of R455A and R452B are reported against vapor quality. For both the blends, the heat transfer coefficients are found to increase with mass velocity and vapor quality. At the same operative conditions, the heat transfer coefficients of R455A are lower than those of R452B, due to the reduced fraction of R32 and higher temperature glide. Indeed, the heat transfer coefficient of R455A is lower by 30% and 25% compared to the one of R452B at $G = 200$ kg m$^{-2}$ s$^{-1}$ and $G = 600$ kg m$^{-2}$ s$^{-1}$, respectively, considering the whole range of vapour qualities. Several works in the literature (Fronk and Garimella [17], Macdonald [18]) have pointed out that typically larger temperature glide blends undergo larger degradation in heat transfer rates compared to mixtures with smaller temperature glide. Indeed, the temperature glide is related to the composition of the mixture and it is responsible for the decrease of the driving temperature difference during condensation between saturation and wall temperature as the vapor quality diminishes, due to the fact that the phase change process is not isothermal. Additionally, the preferential condensation of the less volatile component of the mixture leads to a concentration gradient in the vapor phase. The condensation of large fractions of the less volatile component at the interface causes its depletion in the vapor phase and a corresponding increase in the liquid phase at the interface (related to the locally augmented concentration of the more volatile component vapor at the liquid-vapor interface). The results are a lower interfacial temperature, due to the greater concentration of the more volatile component at the interface, which presents the lower saturation temperature, and an additional mass transfer resistance, due to the fact that the less volatile component has to diffuse through the vapor phase to proceed condensation at the interface.
3.1. Correlations assessment

Different predicting approaches have been proposed for modelling condensation heat transfer of zeotropic mixtures. Due to the different volatility of the blend components, which causes the condensation process to be non-isothermal, continuous changes in the composition of liquid and vapor phases lead to the build-up of mass transfer resistances interfering with the heat transfer mechanisms. The method proposed in the present work to account for this additional resistance is the equilibrium method which was first proposed by Silver [19] and Bell and Ghaly [20]. This method assumes complete mixing and overall equilibrium in the liquid and vapor phases. In the equilibrium method, the sensible heat is removed from the vapor and transferred to the interface by convection, where the heat transfer coefficient is evaluated under the hypothesis that only vapor is present using the mass flux and properties of the vapor.

By adopting the Silver [19] and Bell and Ghaly [20] approach (named SBG correction in the figures), the heat transfer coefficients of a mixture can be evaluated by correcting the heat transfer coefficient calculated using a model for pure fluid with an additional heat transfer resistance in the vapor. A detailed description of the model can be found in Azzolin et al. [11].

The pure vapour condensation models that were chosen for the evaluation of the condensate heat transfer coefficient are the ones by Shah [21] and Cavallini et al. [22]. As can be seen from Figure 3 and Figure 4, the condensation heat transfer coefficients of the mixture R455A inside the 8.0 mm channel is generally underpredicted when the models are corrected for mixtures, and in particular deviations between experimental data and models predictions are larger at low mass velocities.

The best predictions for both the mixtures are provided by the Cavallini et al. [22] model coupled with the SBG correction (mean absolute deviation $e_{AB} = 5.9\%$ and standard deviation $\sigma_N = 5.9\%$ for R452B; $e_{AB} = 16.1\%$ and $\sigma_N = 8.9\%$ for R455A). The Shah [21] model with SBG correction is able to predict the whole experimental database of R455A with $e_{AB} = 17.2\%$ and $\sigma_N = 7.2\%$, while the R452B data are predicted with $e_{AB} = 10.7\%$ and $\sigma_N = 11.4\%$. 

![Figure 1. R455A experimental heat transfer coefficients versus vapor quality at different mass velocity $G$ [kg m$^{-2}$ s$^{-1}$].](image1)

![Figure 2. R452B experimental heat transfer coefficients versus vapor quality at different mass velocity $G$ [kg m$^{-2}$ s$^{-1}$].](image2)
4. Flow boiling heat transfer results

In Figure 5 the heat transfer coefficients of the mixtures R455A and R452B measured inside the 8.0 mm channel are reported versus vapour quality at mass velocities $G$ equal to 200 and 400 kg m$^{-2}$ s$^{-1}$, mean saturation temperature equal to 18.2 $^\circ$C and heat flux $q'$ between 14 and 17 kW m$^{-2}$. The heat transfer coefficients of both R455A and R452B become higher when increasing the mass velocity. Comparing the performance of the two blends, R452B displays higher heat transfer coefficients due to the higher fraction of R32 and the lower temperature glide, if compared to R455A.

Figure 6 reports the heat transfer coefficients of the mixtures R455A and R452B measured inside the 8.0 mm channel at two different heat fluxes ($q'\,=\,15.5\pm10\%\,\text{and}\,q'\,=\,25\pm10\%\,\text{kW m}^{-2}$), mass velocity $G$ equal to 400 kg m$^{-2}$ s$^{-1}$ and mean saturation temperature equal to 18.2 $^\circ$C. The effect of the heat flux on the heat transfer coefficient is higher for the mixture R452B compared to R455A. This can be explained considering the physical phenomena which characterize the flow boiling heat transfer with non-azeotropic mixtures. As the blend is heated, the more volatile component (for both the mixtures, R32) evaporates faster than the less volatile one (R1234yf). This leads to a change in the composition of the liquid and vapor phases and introduces a concentration gradient at the interface that rises the bubble point. Thus, in addition to the heat for evaporation some additional heat is required for the sensible heating of the liquid and vapor phases. The high-temperature glide mixture, due to the mass diffusion resistance and to the greater sensible heat, shows a reduced increase of the heat transfer coefficient with heat flux, if compared to the low-temperature glide mixture.

4.1. Assessment of correlations

Flow boiling heat transfer correlations developed for pure fluids are expected to provide inaccurate results when applied to non-azeotropic mixtures due to the additional mass transfer resistance. In the present paper, the experimental data have been compared against predictions from the Gungor and Winterton [23] and the Sun and Mishima [24] correlations modified to account for the additional mass transfer resistance. The Gungor and Winterton [23] model has been modified as proposed by Shah [25], applying the Thome and Shakir [26] correction factor to the nucleate boiling contribution and the Bell and Ghaly [20] correction factor to the convective boiling contribution. For the Sun and Mishima [24] model, which was originally developed for channels with inner diameter $\leq 6.5$ mm considering only the nucleate boiling contribution, the heat transfer coefficient can be calculated applying the Thome and Shakir [26] correction factor to the boiling number.
The heat transfer coefficients evaluated with the two correlations corrected for blends are reported in Figures 7 and 8. An improvement in the prediction accuracy has been found for both the models: the Sun and Mishima [24] correlation corrected for zeotropic mixtures is able to predict the whole presented experimental database with a mean absolute deviation \( e_{AB} = 20.0\% \) and a standard deviation \( \sigma_N = 11.8\% \) (for R455A \( e_{AB} = 12.2\% \) and \( \sigma_N = 9.6\% \), for R452B \( e_{AB} = 27.8\% \) and \( \sigma_N = 8.1\% \)). The Gungor and Winterton [23] model corrected for zeotropic mixtures gives a mean absolute deviation equal to 12.1% and a standard deviation equal to 9.6% (for R455A \( e_{AB} = 16.4\% \) and \( \sigma_N = 10.3\% \), for R452B \( e_{AB} = 7.7\% \) and \( \sigma_N = 6.7\% \)).
5. Conclusions
Condensation and flow boiling heat transfer coefficients of two ternary zeotropic mixtures in a 8.0 mm diameter channel are reported in the present paper. The tested fluids are R455A (GWP equal to 146), ternary blend of R32, R1234yf and R744 at 21.5/75.5/3.0% by mass composition, and R452B (GWP equal to 676), mixture of R32, R1234yf and R125 at 67.0/26.0/7.0% by mass composition.

For both condensation and vaporization tests, the mixture R452B presents higher heat transfer coefficients by about 25-30% with respect to R455A due to the lower temperature glide. Since the phase change process of zeotropic mixtures is not isothermal and continuous changes in the composition of liquid and vapor phases lead to the build-up of a mass transfer resistance, therefore models developed for pure fluids need to be corrected. Good results have been obtained with the correction proposed by Silver [19] and Bell and Ghaly [20] which introduces an additional mass transfer resistance proportional to the temperature glide. When condensation tests are considered, the best prediction for both the mixtures is provided by the Cavallini et al. [22] model coupled with the SBG correction ($e_{AB} = 5.9\%$ and $\sigma_N = 5.9\%$ for R452B; $e_{AB} = 16.1\%$ and $\sigma_N = 8.9\%$ for R455A). When flow boiling tests are considered, good accuracy is obtained using the Gungor and Winterton [23] model modified as proposed by Shah [25] ($e_{AB} = 7.7\%$ and $\sigma_N = 6.6\%$ for R455A and $e_{AB} = 16.4\%$ and $\sigma_N = 10.2\%$ for R455A).

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Nomenclature

| Symbol | Definition                | Unit         |
|--------|---------------------------|--------------|
| $A$    | Heat transfer area        | m$^2$        |
| $c$    | Specific heat             | [J kg$^{-1}$ K$^{-1}$] |
| $e_{AB}$ | Mean absolute deviation | [%]         |
| $G$    | Mass velocity             | [kg m$^{-2}$ s$^{-1}$] |
| $HTC$  | Heat transfer coefficient | [W m$^{-2}$ K$^{-1}$] |
| $\dot{m}$ | Mass flow rate          | [kg s$^{-1}$] |
| $\rho$ | Density                   | [kg m$^{-3}$] |
| $\Delta T$ | Temperature difference  | [K]         |
| $\sigma_N$ | Standard deviation       | [-]         |
| $p$    | Pressure                  | [bar]        |
| $q$    | Heat flow rate            | [W]         |
| $q'$   | Heat flux                 | [W m$^{-2}$] |
| $T$    | Temperature               | [°C]         |
| $u$    | Uncertainty               | [%]         |
| $x$    | Vapour quality            | [-]         |

Greek symbols

| Symbol | Definition                |
|--------|---------------------------|
| $\Delta T$ | Temperature difference  |
| $\sigma_N$ | Standard deviation       |
| $\rho$ | Density                   |

Subscripts

| Symbol | Definition    |
|--------|---------------|
| $g$    | Vapour        |
| $in$   | Inlet         |
| $l$    | Liquid        |
| $LMTD$ | Logarithmic mean temperature difference |
| $out$  | Outlet        |
| $sat$  | Saturation    |
| $w$    | Water         |
| $wall$ | Wall          |

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