Heat transfer degradation during condensation of non-azeotropic mixtures

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Abstract. International organizations call for a reduction of the HFCs production and utilizations in the next years. Binary or ternary blends of hydrofluorocarbons (HFCs) and hydrofluoroolefins (HFOs) are emerging as possible substitutes for high Global Warming Potential (GWP) fluids currently employed in some refrigeration and air-conditioning applications. In some cases, these mixtures are non-azeotropic and thus, during phase-change at constant pressure, they present a temperature glide that, for some blends, can be higher than 10 K. Such temperature variation during phase change could lead to a better matching between the refrigerant and the water temperature profiles in a condenser, thus reducing the exergy losses associated with the heat transfer process. Nevertheless, the additional mass transfer resistance which occurs during the phase change of zeotropic mixtures leads to a heat transfer degradation. Therefore, the design of a condenser working with a zeotropic mixture poses the problem of how to extend the correlations developed for pure fluids to the case of condensation of mixtures. Experimental data taken are very helpful in the assessment of design procedures. In the present paper, heat transfer coefficients have been measured during condensation of zeotropic mixtures of HFC and HFO fluids. Tests have been carried out in the test rig available at the Two Phase Heat Transfer Lab of University of Padova. During the condensation tests, the heat is subtracted from the mixture by using cold water and the heat transfer coefficient is obtained from the measurement of the heat flux on the water side, the direct measurements of the wall temperature and saturation temperature. Tests have been performed at 40°C mean saturation temperature. The present experimental database is used to assess predictive correlations for condensation of mixtures, providing valuable information on the applicability of available models.

1. Introduction

In the last three decades, growing attention to global warming and environmental issue has led to a reconsideration of the fluids traditionally employed in refrigeration and air-conditioning applications. Starting from 1987, the international environmental agreement called Montreal Protocol, signed by all UN and EU members, established the phase-out of ozone depleting CFCs (chlorofluorocarbons), such as R12 and R502. With the Copenhagen Amendment to the Montreal Protocol (1992) also HCFCs (hydrochlorofluorocarbons), such as R22, have been affected by the regulation. Then the Kyoto Protocol (1997) fixed emission reduction targets for the six main greenhouse gases, including HFCs...
(hydrofluorocarbons) such as R134a and R32. The EU Regulation No. 517/2014 disposed the phase-out of most refrigerants commonly used in refrigeration and air conditioning systems, in particular R404A, R410A and R134a, due to their high GWP (Global Warming Potential). The aim is to reach a two-thirds reduction of EU F-gas emissions by 2030 compared to 2014 levels. In 2016 the Kigali Amendment to the Montreal Protocol was adopted to significantly reduce the consumption and production of HFCs on a global scale.

As HFCs need to be replaced to meet European and international requirements, considerable research has been fostered on possible substitutes, both natural and synthetic. Recently, low GWP hydrofluoroolefins (HFOs) such as R1234yf (GWP<4) and R1234ze(E) (GWP<1) have been studied as possible candidates for the replacement of R134a. Currently, no pure fluids competitive in terms of thermodynamic performance, low GWP and non-flammability are available as alternatives to R410A in chiller and air-conditioning applications and to R404A in commercial and transport refrigeration. For these applications a possible solution should be represented by refrigerant mixtures of HFOs and other HFCs (i.e. R32, R125). These mixtures present a temperature variation during phase change at constant pressure, due to the temperature glide between the dew and the bubble point, which can lead to a better matching between the refrigerant and the water temperature profiles. On the other hand, the additional mass transfer resistance which occurs during the phase change of zeotropic mixtures causes a heat transfer degradation.

In the literature, few studies are available on condensation and evaporation of HFOs/HFCs mixtures. Hossain et al. [1] investigated the flow boiling heat transfer characteristics of the mixture R1234yf/E/R32 (mass fraction 55/45%) in a horizontal tube with inner diameter of 4.35 mm, comparing them with those of R410A and R32 at the same operating conditions. The local heat transfer coefficient of the mixture was found to be lower than that of R32 and R410A in the whole quality region. With regard to R1234yf(E), the heat transfer coefficient of the zeotropic blend is lower only in the high quality region. Kondou et al. [2] investigated the condensation and evaporation heat transfer coefficients of the mixtures R744/R32/R1234ze(E) and R32/R1234ze(E) at different composition in a horizontal microfin tube (5.35 mm equivalent inner diameter). The authors found that both condensation and in particular evaporation heat transfer coefficients of the mixtures are penalized with respect to those of single components and they tried to quantify the contribution of the additional mass transfer resistance. Wang et al. [3] studied the condensation heat transfer characteristics of R32/R1234yf non-azeotropic mixtures (mass fraction 52/48% and 77/23%) inside a horizontal smooth tube with inner diameter equal to 4 mm. They developed also a prediction model for the forced convective condensation of zeotropic mixtures in reasonable agreement with experimental data.

In this paper, two mixtures of R1234yf(E) and R32 at different mass compositions (23/77% and 46/54% by mass) have been investigated during condensation inside a single circular cross section microchannel with inner diameter equal to 0.96 mm. Tests have been carried out on the experimental apparatus available at the Two-Phase Heat Transfer Lab of the University of Padova. The new experimental data are compared to those of pure components R1234yf(E) and R32 in order to analyze the heat transfer penalization due to the mass transfer resistance of these zeotropic mixtures and to assess predictive models.

2. Test facility
The heat transfer test section is located in the experimental apparatus schematized in Figure 1. In the primary loop, the refrigerant is subcooled in a post-condenser and then dried up before entering an independently controlled gear pump, magnetically coupled to a variable-speed electric motor. The mass flow rate is measured by a Coriolis effect mass flow meter. Hence, the fluid passes through a mechanical filter and a tube-in-tube heat exchanger where the primary fluid can be vaporized and superheated using hot water flowing in a secondary loop with PID-controlled electrical heaters. The refrigerant is finally sent through the test section for heat transfer measurements. The test section is characterized by a pre-conditioning sector, used to reach the desired thermodynamic conditions of the refrigerant at the inlet of the measuring sector. The refrigerant pressure at the inlet of each sector is
measured by means of relative pressure transducers, whereas a differential pressure transducer is employed to measure pressure drop along the measuring sector. All the temperatures are detected using T-type thermocouples which have been calibrated following the technique in Del Col et al. [4]. Two thermal baths are employed on the secondary fluid side: the first serves brine at 5 °C to the post-condenser while the second sets the temperature of the distilled water entering the measuring and pre-conditioning sectors. In every test run, when the apparatus is working in steady state conditions, measurements of thermo-fluid-dynamic parameters are recorded for 50 s with a time step of 1 s and then averaged.

![Figure 1](image-url)

**Figure 1.** Experimental test rig: PS pre-conditioning sector; MF mechanical filter; FD filter dryer; PV pressure vessel; CFM Coriolis-effect mass flow meter; TV valve; P pressure transducer; DP differential pressure transducer; T thermocouple.

### 2.1. Heat transfer coefficient test section

The test section is composed by two parts: a 50 mm long pre-conditioning sector and a 230 mm long measuring sector. Two independent distilled water loops serve the the pre-conditioning sector and the measuring sector and each of them is provided by a flow regulating valve to set the flow rate measured by a dedicated Coriolis effect mass flow meter. When necessary, the water entering the pre-conditioning sector is additionally heated up.

Both the pre-conditioning and measuring sectors work as counter-current heat exchangers and have been obtained from a 8 mm external diameter copper rod having a 0.96 mm internal bore and an inner surface roughness $Ra$ equal to 1.3 μm. The external surface of the copper rod is machined to embed the secondary fluid channel (Figure 2), which is externally closed by plastic sheath. The tortuous path geometry on the water side enables good water mixing for precise local coolant temperature and meanwhile it reduces the external heat transfer resistance in favour to low uncertainty measurements. The two sectors are connected between them and with the experimental apparatus using stainless steel adiabatic tubes to allow thermal separation and the insertion of two pressure ports.

On the coolant side, the inlet and outlet water temperatures are measured by T-type thermocouples at the ends of each sector, while the water temperature differences across both sectors are measured by multi-junction copper-constantan thermopiles. In the measuring sector, the local water temperature profile is determined from the measurements of 15 thermocouples placed in the water channel while the local wall temperatures are gauged by 13 thermocouples embedded in the wall. More details on the test section are given in Matkovic et al. [5].
3. Mixture composition

In this work, condensation heat transfer characteristics of two R32/R1234ze(E) mixtures with different compositions (0.23/0.77 and 0.46/0.54 by mass) have been investigated. In Table 1, some thermophysical and transport properties of the refrigerant blends at 40 °C saturation temperature are reported, compared with those of pure fluid components, R32 and R1234ze(E). Properties are computed using NIST Refprop 9.1 (Lemmon et al., [6]) with the Akasaka model [7], specifically developed for the evaluation of thermodynamic properties of R32/R1234ze(E) mixtures.

From Table 1 it can be noticed that for R32/R1234ze(E) mixture at 23/77% composition the temperature glide is equal to 11 K, while at 46/54% it is equal to 8.2 K. These mixtures present a GWP\textsubscript{100-year} value that is significantly lower than those of traditionally used refrigerants, such as R404A (GWP\textsubscript{100-year}=3922), R410A (GWP\textsubscript{100-year}=2088) and R32 (GWP\textsubscript{100-year}=675).

Table 1. Properties of pure R32 and R1234ze(E) and R32/R1234ze(E) mixtures at 40 °C saturation temperature (data from NIST Refprop 9.1, Lemmon et al. [6]).

| Fluid          | p\textsubscript{sat} [bar] | \(\rho\)_l [kg m\textsuperscript{-3}] | \(\rho\)_g [kg m\textsuperscript{-3}] | \(\mu\)_l [μPa s] | \(\lambda\)_l [W m\textsuperscript{-1} K\textsuperscript{-1}] | \(t\)\textsubscript{dew} [°C] | \(t\)\textsubscript{bub} [°C] | Glide [K] | GWP [-] |
|----------------|---------------------------|-----------------------------------|-----------------------------------|------------------|----------------------|-----------------|-----------------|----------|-------|
| R32            | 24.8                      | 893.0                             | 73.3                              | 95.0             | 0.115                 | /               | /               | /        | 675   |
| R1234ze(E)     | 7.7                       | 1111.3                            | 40.7                              | 167.0            | 0.069                 | /               | /               | /        | 0.97  |
| R32/R1234ze(E) | 13.1                      | 1075.3                            | 56.3                              | 143.8            | 0.083                 | 45.6            | 34.6            | 11       | 155   |
| 23/77%         |                           |                                   |                                   |                  |                      |                 |                 |          |       |
| R32/R1234ze(E) | 17.4                      | 1017.9                            | 64.8                              | 122.0            | 0.094                 | 44.4            | 36.2            | 8.2      | 309   |
| 46/54%         |                           |                                   |                                   |                  |                      |                 |                 |          |       |

4. Data reduction

The local heat flux is determined from the slope of the water temperature profile along the test section (obtained from the thermocouples inserted along the coolant path):

\[
q' = \frac{m\textsubscript{water}c\textsubscript{water}}{\pi d} \frac{dT\textsubscript{water}(z)}{dz}
\]

where \(z\) is the axial coordinate along the tube and \(dT\textsubscript{water}(z)/dz\) is the derivative of the polynomial equation interpolating the measured water temperature. Thus a proper interpolating function of the measured water temperatures is needed (complete description of the whole procedure is reported in Del Col et al. [8]).

The heat flow rate transferred to the secondary fluid up to a certain position \(z\) is obtained integrating the local heat flux from the refrigerant inlet to the position \(z\):
Finally, the local heat transfer coefficient inside the microchannel is obtained as:

\[ \alpha(z) = \frac{q'(z)}{T_{SAT}(z) - T_w(z)} \]  

where \( T_w \) is the wall temperature, measured with the thermocouples embedded in the wall while \( T_{SAT} \) is the saturation temperature calculated from the pressure, enthalpy and mixture composition \( X_{mix} \), assuming an equilibrium state:

\[ T_{SAT}(z) = f(p(z), h(z), X_{mix}) \]  

Along the measuring sector, the refrigerant enthalpy at the position \( z \) can be calculated as follows:

\[ h(z) = h_{in,MS} - \frac{q(z)}{m_{ref}} \]  

During a condensation test, the refrigerant enters the pre-conditioning sector as superheated vapour and is cooled down by the secondary fluid. The enthalpy of the entering vapour in the pre-conditioning sector is determined from the measured temperature and pressure, while the refrigerant enthalpy at the inlet of the measuring sector \( h_{in,MS} \) is determined from a thermal balance on the water side.

The pressure of the mixture is measured at the inlet and outlet of the measuring sector. The local pressure could be obtained from a linear interpolation of inlet and outlet measured values but in the present case a more precise iterative procedure is needed. In fact, the pressure profile is not linear along the microchannel and the interpolation between inlet and outlet pressure should account for the frictional pressure drop in the stainless steel tubes and in the copper microchannel, the pressure variations due to changes in the geometry and the pressure recovery due to momentum variation during condensation. A detailed description of the models applied to determine these local and distributed contributions is available in Del Col et al. [9].

As for the refrigerant temperature, the vapor quality along the test section is evaluated using the local pressure, the local enthalpy and the mixture composition:

\[ x(z) = f(p(z), h(z), X_{mix}) \]  

Experimental uncertainties of the measured parameters (Type B) are reported in Table 2.

| Parameter                                                                 | Uncertainty          |
|---------------------------------------------------------------------------|----------------------|
| Temperature                                                               | ± 0.05°C             |
| Temperature difference (with thermopile)                                  | ± 0.03°C             |
| Water flow rate in pre-conditioning sector                                | ± 0.2 % at 10 kg h\(^{-1}\) |
| Water flow rate in measuring sector                                       | ± 0.14 % at 10 kg h\(^{-1}\) |
| Refrigerant flow rate                                                    | ± 0.2 % at 2 kg h\(^{-1}\) |
| Absolute pressure                                                        | ± 5 kPa              |
| Pressure difference (higher than 1 kPa)                                  | ± 0.12 kPa           |
| Pressure difference (lower than 1 kPa)                                   | ± 0.1 %              |
5. Experimental results and comparison to pure components

Experimental results obtained during condensation of R32/R1234ze(E) mixtures at 23/77% and 46/54% composition in a 0.96 mm diameter microchannel are reported in Figure 3. Condensation tests have been performed at a pressure of 13.7 bar and 17.7 bar respectively for the 23/77% and the 46/54% mixture. The local heat transfer coefficients for the two mixtures are plotted against local vapour qualities for mass velocities ranging from 200 to 800 kg m\(^{-2}\)s\(^{-1}\).

![Figure 3. Experimental local heat transfer coefficients versus vapor quality for R32/R1234ze(E) mixtures at 23/77% and 46/54% mass compositions.](image)

For both tested mixtures, the heat transfer coefficient was found to increase with vapor quality and mass velocity although at low mass velocity and vapor quality it does not vary significantly. From Figure 3, it can be noticed the influence of R32 mass fraction on heat transfer coefficient values of the mixture. Indeed, as the mass fraction of R32 is increased from 23% to 46%, heat transfer coefficients are enhanced on average by about 7% at \(G = 800\) kg m\(^{-2}\)s\(^{-1}\), by 7.4% at \(G = 600\) kg m\(^{-2}\)s\(^{-1}\), by 8% at \(G = 400\) kg m\(^{-2}\)s\(^{-1}\) and by 13% at \(G = 200\) kg m\(^{-2}\)s\(^{-1}\). Heat transfer coefficients of 46/54% are higher than those of 23/77% mixture for all mass velocities due to the greater liquid thermal conductivity and smaller temperature glide.

In Figs. 4 and 5, heat transfer coefficients of the two R32/R1234ze(E) mixtures inside 0.96 mm inner diameter microchannel are compared to those of pure components R32 and R1234ze(E) at the same working conditions. At \(x = 0.3\) (Fig. 4) the heat transfer coefficients of the 46/54% mixture are roughly the same of those of R1234ze(E) for all the mass velocities. At higher vapor qualities (\(x = 0.6\), as shown in Figure 5, the condensation heat transfer coefficients of pure R1234ze(E) are higher than those of the 46/54% mixture by about 4% at \(G = 800\) kg m\(^{-2}\)s\(^{-1}\), by 2% at \(G = 600\) kg m\(^{-2}\)s\(^{-1}\), by 6.5% at \(G = 400\) kg m\(^{-2}\)s\(^{-1}\) and by 22% at \(G = 200\) kg m\(^{-2}\)s\(^{-1}\).

With regard to Figs 4 and 5, the heat transfer coefficients of R32 are always higher than those of R32/R1234ze(E) mixtures and pure R1234ze(E). The increase of the heat transfer coefficient of R32 compared to that of R32/R1234ze(E) mixture at 46/54% composition ranges from 20 to 28% at \(x = 0.3\) and from 35 to 46% at \(x = 0.6\).
6. Prediction of the heat transfer coefficient

The experimental heat transfer coefficient data taken in the 0.96 mm diameter microchannel have been compared to predictions of Cavallini et al. [10] model. This model was originally proposed to predict condensation heat transfer coefficients of pure fluids inside horizontal smooth tubes with internal diameter greater than 3 mm. As underlined in Matkovic et al. [5], this model can be reasonably applied in the case of R32 and R1234ze(E) during condensation inside a 0.96 mm microchannel at mass velocities equal or higher than 200 kg m$^{-2}$ s$^{-1}$. In Figure 6 predicted heat transfer coefficients are plotted versus experimental ones for mass velocities ranging from 200 to 800 kg m$^{-2}$ s$^{-1}$. It is evident that Cavallini et al. [10] model overestimates the experimental data with an absolute deviation $\epsilon_{AB}$ equal to 10.9% and a standard deviation of about 6.1%.

As reported in Del Col et al. [11], during condensation of zeotropic mixtures, the effect of the additional mass transfer resistance cannot be neglected. The heat transfer mechanisms occurring during condensation of zeotropic mixtures are the same as for pure fluids, convective condensation and film condensation during stratified flow, with the difference that the condensation process is non-isothermal and there is sensible heat to be removed both in the liquid and vapor phase. The sensible heat removed from the vapour is more significant due to the low heat transfer coefficients associated with this process. Diffusion resistances in both the vapour and the liquid phases are present due to the continuous change in the compositions of both phases, and between them the mass transfer resistance in the vapour phase is the key point for calculations. When considering mixtures, models developed for pure vapor condensation can be applied if complete mixing in both liquid and vapor phase is reached and overall equilibrium is maintained, with a correction term through which the additional resistance of the mixture due to the mass transfer is properly accounted for.
Figure 6. Experimental data of R32/R1234ze(E) mixtures at 23/77% and 46/54% mass compositions compared to values calculated with Cavallini et al. (2006) model.

The Cavallini et al. (2006) model can be applied to zeotropic mixtures using the Bell and Ghaly [12] and the Silver correction [13]. In this procedure the heat transfer coefficient of the mixture is calculated as a function of the condensate heat transfer coefficient $\alpha_c$ (calculated with the model for pure fluids but using thermophysical properties of the mixture), of the heat transfer coefficient of the vapor phase flowing alone in the duct $\alpha_g$ (calculated with the Dittus-Boelter equation), and of the ratio between the sensible flow rate heating the vapor and the total heat flow rate $\partial q_s / \partial q_t$, as reported in Equation (7):

$$\alpha_{mix} = \left( \frac{1}{\alpha_f} + \frac{\partial q_{sg}}{\partial q_t} \right)^{-1} \tag{7}$$

The ratio of the sensible to total heat duty can be expressed with good approximation as follows:

$$\frac{\partial q_{sg}}{\partial q_t} \approx x c_p g \left( \frac{dT}{dh} \right) \tag{8}$$

If the ratio $dT/dh$ remains approximately constant during condensation the ratio can be rewritten as a function of the temperature glide $\Delta T_{GL}$ and the enthalpy change of isobaric condensation $\Delta h_{mix}$:

$$\frac{dT}{dh} \approx \frac{\Delta T_{GL}}{\Delta h_{mix}} \tag{9}$$

The comparison between the experimental data and heat transfer coefficients predicted by Cavallini et al. (2006) corrected with Silver-Bell-Ghaly approach is shown in Figure 7. Predicted data are in good agreement with experimental ones, with an absolute deviation $e_{AB}$ of 7.4 % and a standard deviation of 5.4%.
7. Conclusions
This work reports experimental heat transfer coefficients during condensation of two R32/R1234ze(E) zeotropic blends at different mass compositions (23/77% and 46/54%), inside a 0.96 mm diameter horizontal smooth microchannel. These zeotropic mixtures display a temperature glide of about 11 K and 8 K, respectively. Condensation tests have been carried out with mass velocity ranging between 200 kg m$^{-2}$s$^{-1}$ and 800 kg m$^{-2}$s$^{-1}$ at a pressure of 13.7 bar and 17.7 bar, respectively, for the 23/77% and the 46/54% mixture. The heat transfer coefficient of both mixtures increases with vapor quality and mass velocity. When the mass fraction of R32 is increased from 23% to 46%, the heat transfer coefficient increases by 7% at $G = 800$ kg m$^{-2}$s$^{-1}$, by 8% at $G = 400$ kg m$^{-2}$s$^{-1}$ and 13% at $G = 200$ kg m$^{-2}$s$^{-1}$ due to better properties (higher thermal conductivity) and smaller temperature glide. With comparison to pure components, at mass velocities ranging from 200 kg m$^{-2}$s$^{-1}$ and 800 kg m$^{-2}$s$^{-1}$ the R32/R1234ze(E) mixture at 46/54% composition shows heat transfer coefficients which are lower than those of R1234ze(E) at high vapor qualities but comparable at low vapor qualities.

The experimental heat transfer coefficients have been compared with those predicted by the Cavallini et al. (2006) model coupled with the Silver-Bell-Ghaly correction to account for the additional mass transfer resistance. This model shows a good accuracy in predicting experimental data, displaying an absolute deviation of 7.4% and a standard deviation of 5.4%.

Nomenclature

| Symbol | Unit          | Description                     |
|--------|---------------|---------------------------------|
| $c_p$  | [J/(kg·K)]   | Specific heat                   |
| $d_i$  | [m]           | Hydraulic diameter              |
| $e_{AB}$ | [-]          | Absolute deviation              |
| $G$    | [kg/(m²·s)]  | Mass velocity                   |
| $h$    | [J/kg]        | Specific enthalpy                |
| $m$    | [kg/s]        | Mass flow rate                  |
| $p$    | [Pa]          | Pressure                         |
| $q$    | [W]           | Heat flow rate                   |
| $q'$   | [W/m²]        | Heat flux                        |
| $t$    | [°C]          | Temperature                      |
| $T$    | [K]           | Temperature                      |
| $x$    | [-]           | Thermodynamic vapour quality     |
| $X$    | [-]           | Mass fraction                    |
| $z$    | [m]           | Position                         |
Greek symbols
\( \alpha \) [W/(m² K)] Heat transfer coefficient \( \lambda \) [W/(m K)] Thermal conductivity
\( \Delta h \) [J/kg] Specific enthalpy difference \( \mu \) [μPa s] Dynamic viscosity
\( \Delta T \) [K] Temperature difference \( \rho \) [kg/m³] Density

Subscripts
bub Bubble in Inlet s Sensible
dew Dew l Liquid SAT Saturation
f Film mix Mixture t Total
g Vapor MS Measuring section water Water
GL Glide ref Refrigerant w Wall

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