

Review

Climate Change and Refrigerants: Thermodynamic Properties of Low-GWP Fluids for Domestic Applications and Binary Systems for Low-Temperature Options

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Abstract: The most commonly used refrigerants are potent greenhouse gasses that can contribute to climate change. Hydro-Fluoro-Olefins are low Global Warming Potential fluids. A summary of our experimental research activity on the thermodynamic properties of two environmentally friendly Hydro-Fluoro-Olefins, namely R1234yf and R1234ze(E), is reported. In particular, the measurements were performed with an isochoric apparatus and the apparatus specifically built to reach temperatures down to about 100 K. The data elaboration confirms the validity of the choice and that R1234yf and R1234ze(E) can be adopted in many domestic applications. Moreover, considering the reduction of the flammability issues of R1234yf and R1234ze(E), the properties of binary systems containing these fluids and carbon dioxide were analyzed. The presented mixtures could be very interesting for low-temperature applications such as cascade cycles.

Keywords: GWP; environmentally friendly refrigerants; HFO

1. Introduction

In recent years, the scientific community has been focused its effort on the investigation on the so-called fourth-generation refrigerants referred to as low Global Warming Potential (GWP) refrigerants, to decrease environmental issues and climate change caused by the emissions of traditional refrigerants.

For example, while, in Europe, the HydroChloroFluoroCarbons (HCFCs) emissions are reaching zero, on the other hand, the consumption of HydroFluoroCarbon (HFCs) is growing very fast. However, since the HFCs are high-GWP fluids, there is a need to reduce the impact to climate change due to their emissions into the atmosphere. Among the family of HFCs, there are several fluids used for air conditioning, foams and other field, but a lot of them have a very high GWP. One of the most famous is the 1,1,1,2-tetrafluoroethane (R134a in the ASHRAE designation) that is widespread in use in domestic and industrial plants that have a GWP of 1300 and is one of the substances controlled by the Kigali Amendment to the Montreal Protocol [1,2].

Governmental regulations and taxes influence the increasing motivation behind low-GWP refrigerants. The European Union (EU) F-Gas regulations [3] impose that the GWP of household air conditioner refrigerants must remain less than 150 by 2020. In addition, they proposed to decrease at least two-thirds of the 2010 emissions of fluorinated greenhouse gases by 2030 in the EU.

More recently (February 2017), the European Commission [4] adopted a proposal to ratify the Kigali amendment to the Montreal Protocol to constantly restrict the production and application of HFCs [5].

The challenge to find refrigerants that are at the same time low GWP, secure, and have a high performance is ongoing.

McLinden et al. [6,7] presented a well organised and comprehensive review of new environmentally friendly refrigerants, implementing a selection based on safety, environmental, and thermodynamic properties' criteria.

They took into account several and important factors for the selection, such as low flammability, low GWP, efficiency, and proper capacity. The authors found a very limited group of compounds, most of them HydroFluoroOlefins (HFOs), that could be possible substitutes for old refrigerants.

The HFOs have a carbon-carbon double-bond that causes them to have a very short lifetime in the atmosphere. Nevertheless, their disadvantages are that often, they are flammable or even toxic. Moreover, frequently, the thermophysical properties of these alternative refrigerants are not suitable for engineering applications. To fix these shortcomings, researchers are studying many blends of the former and the latter refrigerants in order to take into account the benefits of many compounds and balance their disadvantages [7].

Thanks to its thermodynamic properties, one of the most used refrigerants for domestic supplies is R134a [2].

As stated before, R134a has a very high level of GWP; therefore, the restriction of GWP 150 obliges the replacement in domestic refrigerator and freezers. Despite many of the pure HFOs and hydrocarbons being flammable, these alternatives to HFCs are being used in 90% of domestic use [8].

Hydrocarbons could be considered a very good alternative to R134a because of their high energy performance and could have several environmental advantages. However, in many countries, the safety regulations forbid the use of such compounds. For particular usages, such as domestic refrigeration, isobutane (R600a in the ASHRAE designation) could be considered as a good replacement for R134a [9].

For example, in Europe and Japan, the domestic regulations allow hydrocarbons such as R600a, despite many technical issues. But in many countries with more restrict safety regulations, this fluid is considered dangerous due to its flammability. For instance, in the United States, the amount of hydrocarbons for domestic use is restricted to 57 g for each charge, while in Europe, it is limited to 150 g [5].

For all these reasons, the fourth generation of synthetic refrigerants with a low GWP and low level of flammability has been arising a lot of interest by researchers and markets.

In this study, a review of the thermodynamic properties of two of the most important HFOs, namely 2,3,3,3-tetrafluoroprop-1-ene (R1234yf in the ASHRAE designation) and *trans*-1,3,3,3-Tetrafluoropropene (R1234ze(E) in the ASHRAE designation) is presented. The interest in these fluids characterized by GWP values near to 1 was confirmed by the Bobbo et al. [10] review in which it was shown that the thermophysical properties of R1234yf and R1234ze(E) are the most investigated, also in terms of thermal conductivity, viscosity and surface tension.

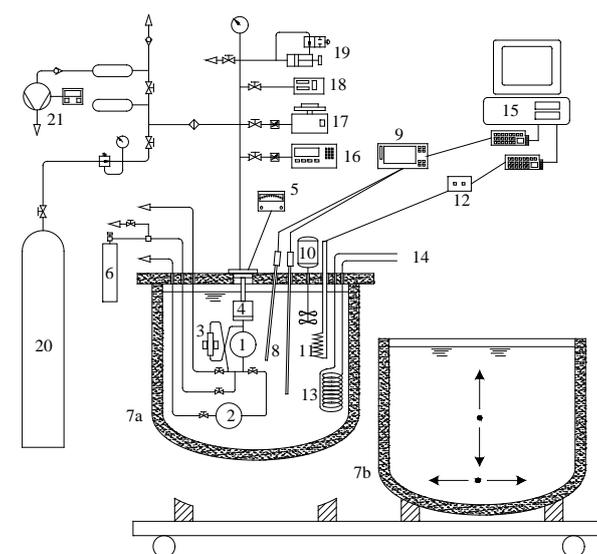
Regarding domestic refrigeration, in the last decade, a huge number of papers have focussed on low-GWP replacements for R134a [9,11–15]. Among the proposed options, R152a presents a relatively low GWP but it is flammable, such as R32 and R600a. For these reasons, the most suitable candidates should be selected between HFOs and their mixtures with conventional and natural refrigerants.

In addition, considering the flammability issues of the two HFOs, an analysis of thermodynamic properties of different binary systems containing these fluids and carbon dioxide is presented.

2. Materials and Methods

The Pressure-Specific Volume-Temperature, PvT and Pressure-Specific Volume-Temperature-Composition, $PvTz$ measurements for the low GWP refrigerants and their binary systems were performed using an isochoric apparatus. The triple points of pure fluids and the Solid-Liquid Equilibrium (SLE) of their blends were measured with an apparatus specifically built to reach temperatures down to about 100 K.

The isochoric apparatus is formed by an AISI 304 stainless-steel isochoric sphere and two thermostatic baths that work at low temperatures (from 210 to 298 K) and high temperatures (from 303 to 390 K), respectively. A schematic view of the experimental setup is shown in Figure 1. For details of the setup, the measurement procedure and the uncertainties are given in previous works [16,17] and only a summary of the information is reported in this section. The spherical cell (1) containing the refrigerant sample is connected to a differential diaphragm pressure transducer (4) coupled to an electronic null indicator (5). In the new setup, the transducer and sphere were placed vertically and a magnetic pump (3) for mixing the sample was connected to the sphere. Temperature is controlled by a PID device and measured with a calibrated resistance thermometer (8). The measured binary systems were prepared by a gravimetric method.



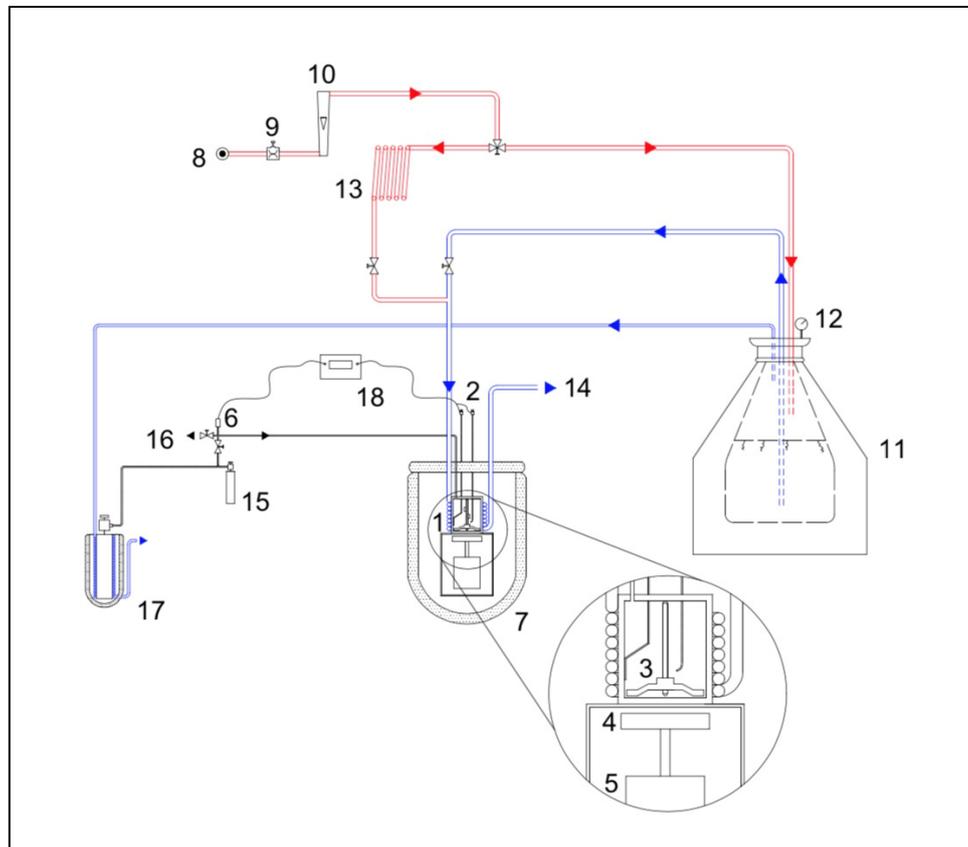
1.	Constant volume spherical cell	12.	Power system
2.	Auxiliary cell	13.	Cooling coil
3.	Magnetic pump	14.	Connections to auxiliary thermostatic bath
4.	Differential pressure transducer	15.	Acquisition system
5.	Electronic null indicator	16.	Bourdon gage
6.	Charging system	17.	Dead weight gage
7.	Thermostatic baths	18.	Vibrating cylinder pressure gage
8.	Platinum thermo-resistances	19.	Precision pressure controller
9.	Thermometric bridge	20.	Nitrogen reservoir
10.	Stirrer	21.	Vacuum pump system
11.	Heater		

Figure 1. Schematic view of the isochoric apparatus.

To perform the experimental runs, the sample charged in the isochoric sphere was allowed to stabilize at the temperature set-point for an hour before the pressure measurements were taken. After recording the measured values, the set-point of the thermostatic bath was changed to the next set temperature. The measurement procedure was then repeated.

The isochoric measurements have the following expanded uncertainties at the 95% confidence level (coverage factor of 2): 1.2 mg for the masses, 0.03 K for the temperature, 1 kPa for the pressure, 0.3 cm³ for volumes, 0.001 for the mole fractions, and 0.005 m³ kg⁻¹ for specific volumes.

The experimental setup for the triple point temperatures and SLE measurements is based on the cooling curve method [18,19] which is a dynamic technique of measurement and does not require the visual observation of phase behavior. Figure 2 shows the experimental setup. The detailed descriptions of the experimental setup and the testing procedure were reported in previous works [20–22]. Only summary information is included below.



1.	Measuring cell	10.	Rotameter
2.	Type T thermocouples	11.	Liquid nitrogen Dewar tank
3.	Stirrer	12.	Liquid nitrogen Dewar manometer
4.	Magnet	13.	External heating coil
5.	Electric engine	14.	Nitrogen outlet
6.	Pressure transducer	15.	Charging bottle
7.	Dewar flask	16.	Vacuum pump system
8.	Dry air supplier	17.	Recovering bottle
9.	Mass flow controller	18.	Acquisition system

Figure 2. Schematic view of the Solid-Liquid Equilibrium (SLE) apparatus.

The apparatus includes a measuring cell (1) consisting of a stainless-steel cylinder. The cover of the cylinder has two holes that house two T-type thermocouples (2). To avoid any premature stratification of the sample and to ensure homogeneity during the sample liquefaction and crystallization, a stirrer (3) was placed inside the cylinder. The measuring cell is surrounded by a copper coil that exchanges heat with the cell through its contact surface and by means of a working fluid (air or liquid nitrogen). The cell and the copper coil are placed inside a thermally insulated Dewar flask (7). The apparatus has two separate circuits: a compressed air circuit and a liquid nitrogen circuit. One end of the compressed air circuit is connected to the dry air supplier (8). A mass flow control (9) was installed downstream

the dry air supplier to adjust the air flow rate, while a rotameter (10) measured the air flow rate. On the other end, the compressed air circuit is connected both to the thermally insulated liquid nitrogen Dewar tank (11) and to an external heating coil (13) that is connected to the copper coil surrounding the measuring cell. The two circuits allowed to carry out measurements in two operating modes: cooling and heating modes. During the cooling mode, a cooling curve is drawn in real time and the sample solidification results in a slope modification of the temperature trend in correspondence to the freezing point. During the heating mode, the sample melting point was identified on the time-temperature curve.

The combined uncertainties associated with the measurements of mass, temperature, and mole fraction are ± 0.6 , ± 1 K, and ± 0.0005 , respectively.

3. Results

Table 1 presents the Global Warming Potential (GWP) of some of the most widespread refrigerants, both natural and synthetic, together with their main physical properties. The reported GWP values were collected from WMO 2018 [23]. The physical properties (molar mass (M), normal boiling point temperature (T_b), critical temperature (T_c), and critical pressure (P_c)) were collected from REFPROP 10.0 [24].

Table 1. Global Warming Potential (GWP) and physical properties of the studied refrigerants.

Refrigerant	GWP	M (kg kmol ⁻¹)	T _b (K)	T _c (K)	P _c (kPa)
R744	1	44.010	194.69	304.13	7377.3
R32	705	52.024	221.50	351.26	5782.0
R600a	<1	58.122	261.40	407.81	3629.0
R134a	1360	102.030	247.08	374.21	4059.3
R1234yf	<1	114.040	243.67	367.85	3382.2
R1234ze(E)	<1	114.040	254.18	382.51	3634.9

The GWP of a compound compares its global warming impact to the impact due to the emission of a similar amount of a reference compound, usually taken as carbon dioxide (R744 in the ASHRAE designation) [25]. This value is estimated during a time horizon that is usually assumed to be equal to 100 years. In particular, the GWP results from the combination of the cumulative radiative forcing, both direct and indirect effects, and atmospheric lifetime of a compound (trace gas), together with the time horizon for its evaluation [6]. The radiative forcing is the change in net irradiance at the tropopause due to the change in atmospheric concentration of a trace gas resulting from a pulse release of that gas.

Although the concept of GWP has several weaknesses, such as that it neglects the effect of reaction products resulting from breakdown and the assumption of a global mean concentration [6], this index is easy and the most commonly used environmental metric.

The GWPs of blends are estimated as the mass-weighted averages of components' GWPs.

The United Nations Environment Programme (UNEP) [26] proposed a classification scheme, distinguishing between very low (or ultra-low) (<~30), very low (<~100), low (<~300), moderate (<~1000), high (<~3000), very high (<~10,000) and ultra-high (>~10,000) GWP fluids.

In recent years, many experimental vapor pressure and PvT data in the superheated vapor phase were measured by our research group with the isochoric apparatus for R1234yf and R1234ze(E) [27–30], as summarized in Figure 3.

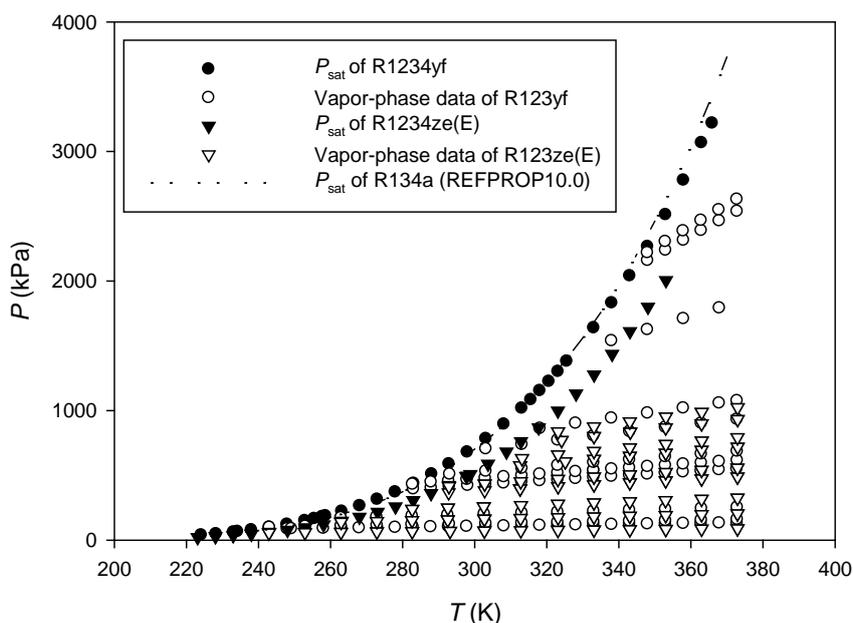


Figure 3. Vapor pressure (P_{sat}) and PvT data in the superheated vapor phase measured with the isochoric apparatus for R1234yf and R1234ze(E). The vapor pressure of R134a calculated from REFPROP 10.0 is reported.

3.1. R1234yf

As it is possible to see from Figure 3, the vapor pressure data for R134a, here reported as regressed values, overlap with the measurements of R1234yf, suggesting that it can be substituted by the proposed HFO. In addition, the critical temperatures of the two fluids are also very similar, as reported in Table 1. In addition, the normal boiling point and the critical pressure are also quite similar to the ones of the other fluids.

By means of the SLE set-up, we also measured its triple point, which was found to be 122.88 K on average [20]. This temperature also allows R1234yf to be considered for low-temperature applications, combining it with other fluids.

The main issue regarding R1234yf is its flammability, thus, this study aimed to find non-flammable binary systems.

3.2. R1234ze(E)

The critical parameters and vapor pressure (reported in Table 1) of R1234ze(E) are also very interesting, and are similar to many HFCs, suggesting that it can also be an interesting alternative for domestic applications. In particular, R1234ze(E) was shown to have higher critical temperature than R1234yf and R134a, which is a critical parameter thermodynamic parameter for a good performance.

By means of the SLE set-up, we also measured its triple point, which was found to be 167.9 K on average [22]. The measurements were obtained both in the heating and in the cooling mode, and the final value was determined by averaging the values.

Moreover, for R1234ze(E), the measuring binary system is a valuable perspective to increase the number of applications of this fluid.

3.3. Binary Systems

We measured many binary systems with both the isochoric and the SLE set up. Regarding low GWP mixtures, our research group focused the research activity on blends with R744, as this fluid is non-flammable and has a GWP = 1. R744 + R1234yf and R744 + R1234ze(E) were measured both in

the two-phase and in the superheated vapor region [31,32]. The results obtained are summarized in Tables 2 and 3 for R744 + R1234yf and R744 + R1234ze(E), respectively.

Table 2. Measurements at bulk compositions z_1 and average specific volumes v for R744 (1) + R1234yf (2) binary system over the temperature range ΔT and pressure range ΔP .

Series	N. Points	z_1	v ($\text{m}^3 \text{kg}^{-1}$)	ΔT (K)	ΔP (kPa)
1	17	0.051	0.026	225–373	62–976
2	17	0.160	0.029	223–373	96–973
3	16	0.266	0.027	224–373	148–1119
4	15	0.365	0.026	223–373	198–1273
5	16	0.480	0.026	225–373	271–1381
6	16	0.574	0.029	224–373	312–1354
7	16	0.662	0.018	223–373	420–2292
8	15	0.754	0.029	224–373	460–1667

Table 3. Measurements at bulk compositions z_1 and average specific volumes v for R744 (1) + R1234ze(E) (2) binary system over the temperature range ΔT and pressure range ΔP .

Series	N. Points	z_1	v ($\text{m}^3 \text{kg}^{-1}$)	ΔT (K)	ΔP (kPa)
1	14	0.155	0.028	233–363	104–943
2	14	0.254	0.029	233–363	149–1001
3	14	0.331	0.029	233–363	171–1063
4	14	0.459	0.029	233–363	255–1178
5	14	0.563	0.028	233–363	340–1366
6	14	0.642	0.030	233–363	397–1380
7	14	0.733	0.028	233–363	504–1601
8	14	0.859	0.028	233–363	634–1909

The P - T diagram of R744 + R1234ze(E) is also reported in Figure 4 as an example.

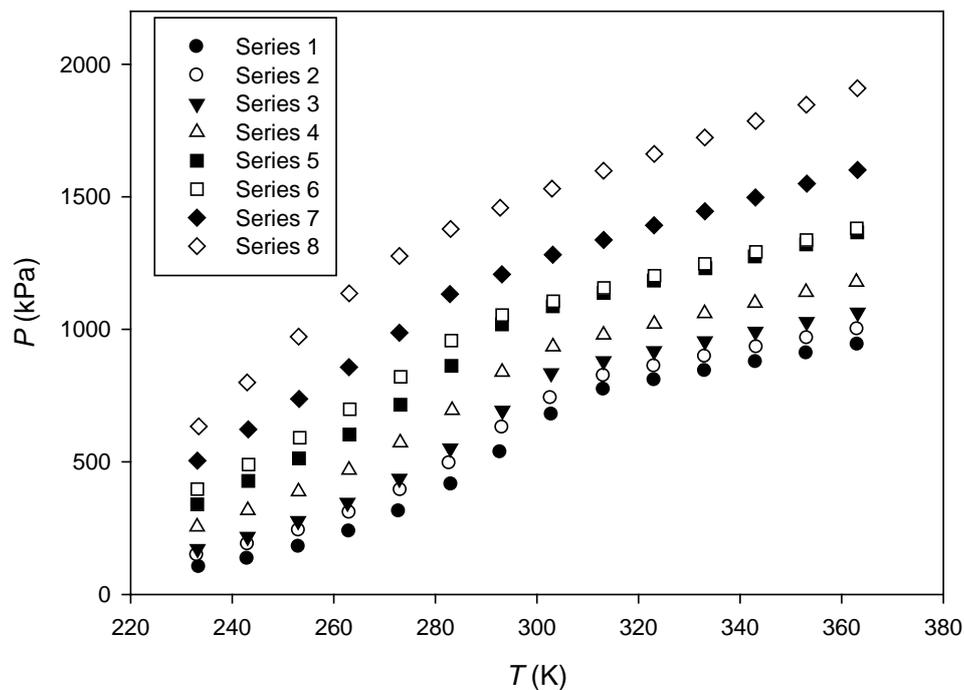


Figure 4. Two-phase and vapor phase $PvTz$ data measured with the isochoric apparatus for the R744 + R1234ze(E) binary system.

Finally, Figures 5 and 6 report the SLE obtained for R744 + R1234yf and R744 + R1234ze(E), respectively [33].

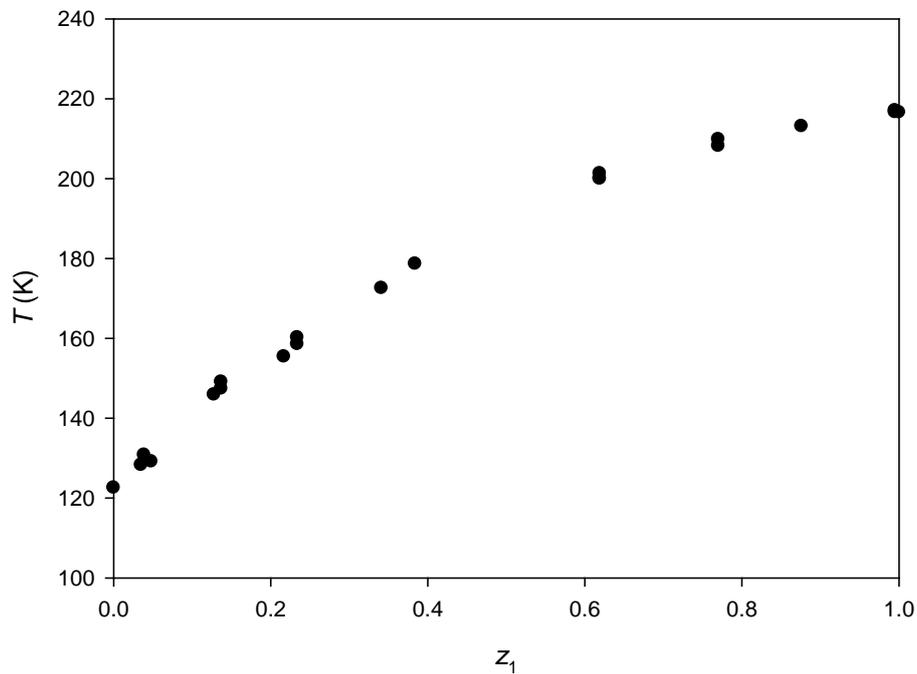


Figure 5. SLE for the R744 (1) + R1234yf (2) binary pair.

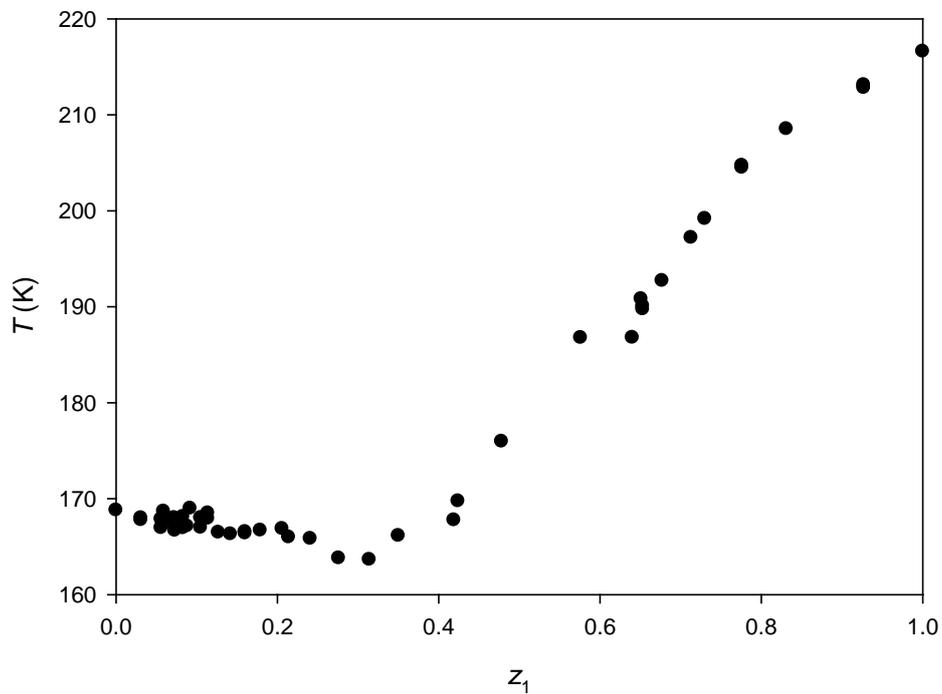


Figure 6. SLE for the R744 (1) + R1234ze(E) (2) binary pair.

4. Discussion

In many cases, using a binary system can be a good solution to overcome the main issues of new refrigerants. In the case of R1234yf and R1234ze(E), one of the main limitations lies in their flammability.

For this reason, after measuring the vapor pressures, the superheated vapor data and the triple points of pure fluids, in this paper, we focused on the SLE and the VLE behavior of R744 + R1234yf and R744 + R1234ze(E). In fact, to consider R744 as a refrigerant helps in terms of both avoiding flammability and keeping GWP low. In addition, since the main limitations of carbon dioxide as a refrigerant are related to its low critical temperature (very often, it is used as working fluid in trans-critical cycles) and its high triple point (solidification occurs at too high temperatures), to blend it with HFOs would provide improvements for all of these issues.

From the elaboration of the P - T curves, it is possible to derive the Vapor–Liquid Equilibrium (VLE) behaviour of the binary system. As explained elsewhere [34], the flash method with the Carnahan-Starling-DeSantis equation of state was used to derive the VLE data from the isochoric measurements. Keeping T , z_1 , and the number of moles equal to the experimental values during the calculation, the flash method provides the calculated values of mole fractions of the liquid phase (x_1) and vapor phase (y_1) and pressure for each isochoric point. This was obtained by ensuring the isofugacity conditions and minimizing the difference between the calculated volume and the measurement cell volume, determined from the gravimetric calibration.

In Figure 7, the VLE of R744 + R1234ze(E) binary system is reported together with the results from molecular Gibbs Ensemble Monte Carlo simulations [35].

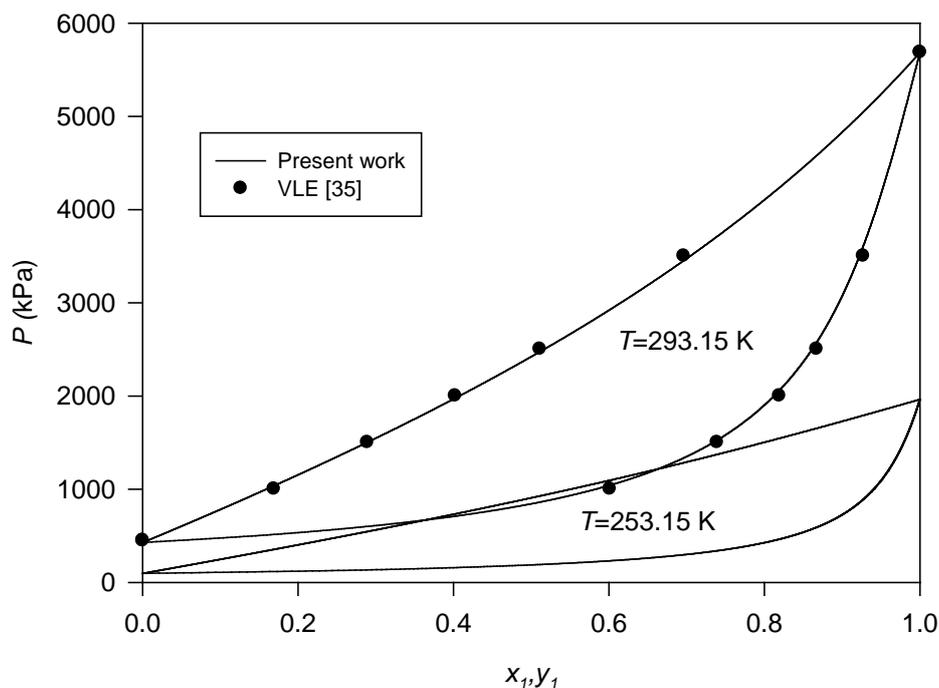


Figure 7. VLE for the R744 (1) + R1234ze(E) (2) binary pair.

From the figure, it is possible to notice that the system is zeotropic and shows an ideal behavior in terms of Raoult's law. The same results were achieved for the R744 + R1234yf binary system.

Furthermore, for R744 + R1234ze(E), the literature shows [35] that they are in good agreement with the values calculated from the isochoric apparatus.

From the T - x data (Figures 5 and 6), it is evident that for the R744 + R1234yf binary system, it was not possible to detect a eutectic point. This was probably due to the very far from each other triple point temperatures of the two system constituents, which probably caused a eutectic point at a very low carbon dioxide concentration region. For the R744 + R1234ze(E) binary system, the eutectic point was estimated at $x_1 = 0.30 \pm 0.05$ and $T = 164 \pm 1$ K.

The SLE data obtained confirm that the binary systems under analysis can be potentially adopted in plants at very low temperatures without any solidification problem.

In general, all the performed measurements confirm that the studied pure fluids and binary systems are valid options for the replacement of non-environmentally friendly working fluids.

5. Conclusions

Because of their low GWP, R1234yf and R1234ze(E) are very promising HFOs. They can be adopted in many domestic applications, but their thermophysical properties need to be estimated. In this paper, a summary of the vapor pressure and of the superheated vapor region data is reported together with the triple point and the SLE of their binary mixtures with carbon dioxide that can be attracting options for low-temperature applications. The results confirm that the two fluids are promising substitutes of R134a in domestic refrigeration.

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