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HFC+HC BLENDS AS REFRIGERANTS

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ABSTRACT

The blends formed mixing hydrofluorocarbons (HFC) and hydrocarbons (HC) can be classified as semi-natural refrigerants since, by optimizing the composition, it is in principle possible to adapt basic features such as GWP, TEWI and flammability to the industrial and environmental requirements. However, the energetic efficiency must be competitive with the traditional refrigerants. To evaluate the efficiency, it is necessary to know the actual thermodynamic properties. VLE measurements allow to regress the interaction parameters for selected equations of state so to calculate with good accuracy the thermodynamic properties. For this reason, VLE measurements were performed at the CNR-ITC for several of HFC+HC blends. The present paper summarizes the results obtained for VLE measurements and discuss the possibility to apply the mixtures studied in HVAC&R applications by analyzing their performance in ideal cycles, calculated on the base of selected models, in comparison with the traditional refrigerants.

1. INTRODUCTION

In an effort to identify suitable alternative refrigerants in HVAC&R applications, particular attention has been paid

Table 1. Characteristic properties of the fluids*

	NBT (°C)	Tc (°C)	Pc (kPa)	GWP** (100 years)
R245fa	14.90	154.05	3640	950
R236ea	6.19	139.29	3502	1200
R236fa	-1.44	124.92	3200	9400
R134a	-26.07	101.06	4059	1300
R143a	-47.24	72.71	3761	4300
R227ea	-16.45	101.65	2926	3500
R125	-48.09	66.02	3618	3400
R290	-42.09	96.67	4247	3
R600a	-11.67	134.67	3640	3
R22	-40.81	96.14	4990	1700
R502	-44.95	80.15	3918	4516
R410A	-51.36	71.36	4903	1975
R404A	-46.6	72.14	3735	3784
R407C	-43.9	86.05	4634	1652

* McLinden et al. (1998), ** IPCC (2001)

to the mixtures between HCs and HFCs, as underlined by the IUPAC project on halocarbons. Hydrocarbons are excellent refrigerants, both for their energetic efficiency and their environmental compatibility witnessed by zero ODP and negligible GWP. Unfortunately, they are highly flammable and for this reason they are not widely applied. On the other hand, HFCs are good refrigerants, but due to their high GWP they could be not considered long term alternatives to traditional refrigerants, especially in view of the application of the Kyoto Protocol. Mixtures of HCs and HFCs are promising since they can contemporarily reduce the flammability of HCs and the GWP connected to HFCs. But are these mixtures actually promising as refrigerants? Is their energetic efficiency and, consequently, their TEWI lower than that of the present refrigerants? In other words: are they actually a possible convenient alternative from both the environmental and the energetic point of view to the present refrigerants?

This paper try to give at least a partial answer to these questions by evaluating the performance of some HC+HFC blends in ideal refrigerant cycles calculated applying EoS with binary interaction coefficients calculated on the base

of experimental VLE measurements performed at our laboratories in Padova.

The experimental data on the thermodynamic properties of binary mixtures of HFCs and HCs are still limited and the evaluation of vapor-liquid equilibrium (VLE) behavior is crucial. Within the last years, an extensive research work has been performed by CNR-ITC in Padova to study the saturation properties of mixtures formed by hydrocarbons and their fluorinated derivatives. Several VLE measurements on HC+HFC systems were performed to give useful information for the selection of new refrigerants, but also to extend the knowledge on the thermodynamic behavior of hydrofluoropropanes when mixed with other HFCs or hydrocarbons. The systems considered in this paper are R125 + R290, R143a + R290, R290 + R134a, R290 + R227ea, R290 + R236fa, R290 + R236ea, R290 + R245fa, R134a + R600a, R600a + R236fa, R600a + R245fa.

The Carnahan-Starling-De Santis EoS and the Lemmon-Jacobsen model (applied in the REFPROP 6.0 database (McLinden, 1998)) with binary interaction parameters regressed on the base of the experimental VLE measurements, have been used to represent the thermodynamic properties of such blends. The models have been used to calculate the performance of the binary mixtures in ideal cycles so to predict their actual suitability as possible alternative refrigerants.

2. SUMMARY OF EXPERIMENTAL

Vapor-liquid equilibrium data for the systems 1,1,1,2-tetrafluoroethane (R134a) +isobutene (R600a) (Bobbo et al., 1998a), R600a + 1,1,1,3,3,3-hexafluoropropane (R236fa) (Bobbo et al., 1998a, 1,1,1-trifluoroethane (R143a) + propane (R290) (Stryjek et al., 1998), R290 + R134a (Stryjek et al., 1998), R290 + R245fa (Bobbo et al., 2000a), R290 + R236fa (Bobbo et al., 2000b), R290+ 1,1,1,2,3,3-hexafluoropropane (R236ea) (Bobbo et al., 2000c), R600a + 1,1,1,3,3-pentafluoropropane (R245fa) (Bobbo et al., 2001), R290 + 1,1,1,2,3,3,3-heptafluoropropane (R227ea) (Bobbo et al., 2002a) and pentafluoropropane (R125) + R290 (Bobbo et al., 2002b) have been measured in a vapor recirculation apparatus at isothermal conditions (Table 2).

The experimental set-up ha been allready described in detail (Bobbo et al., 1998) and used without modification. The VLE cell is endowed with glass windows and equipped with a magnetic pump used to force the vapor through the liquid. The composition of the phases is measured by a gas chromatograph connected on-line to the equilibrium cell, after careful calibration of the FID detector by means of gravimetrically-prepared mixtures. The uncertainty in composition measurement is estimated to be within ± 0.003 in mole fraction for both liquid and vapor phase. The uncertainty in temperature and pressure measurements is estimated to be ± 0.02 K and ± 1 kPa respectively.

The HC+HFC systems, due to the strong repulsive forces between the two different kind of molecules, show a strongly positive deviation from the Raoult's, often with the formation of an azeotrope. Figure 1 and 2 exemplify the typical behavior for the VLE of these mixtures. The higher is the difference between the saturation pressure of the

Table 2. number of experimental data and isotherms measured at CNR-ITC for the selected mixtures.

System	Reference	Nr. Exp. points	Isotherms (°C)
R134a+R600a	(Bobbo et al., 1998)	28	20.51, 30.57
R600a + R236fa	(Bobbo et al., 1998)	15	30.57
R143a + R290	(Stryjek et al., 1998)	17	10.03
R290 + R134a	(Stryjek et al., 1998)	16	10.03
R290 + R245fa	(Bobbo et al., 2000°)	32	25.03,40.07
R290 + R236fa	(Bobbo et al., 2000b)	49	9.98, 30.04, 50.11
R290+ R236ea	(Bobbo et al., 2000c)	43	9.97, 25.01, 40.06
R600a + R245fa	(Bobbo et al., 2001)	40	20.00, 30.00, 40.00
R290 + R227ea	(Bobbo et al., 2002a)	39	5.00, 20.00, 35.00
R125 + R290	(Bobbo et al., 2002b)	57	-15.00, 0.00, 15.00, 30.00

two pure fluids in the mixture, the closer to the low boiling compounds is the azeotropic composition and the higher is the glide of pressure at a given temperature (and then the isobaric glide of temperature).

Table 1 reports some basic characteristics of the compounds which form the binary mixtures here considered, together with those of some of the most important traditional and new generation refrigerants that will be considered as a reference to evaluate the performance of the HC+HFC systems.

2. DATA CORRELATION

In order to represent the thermodynamic properties of the cited systems, the VLE data have been regressed by means of two selected models: the Carnahan-Starling-De Santis (CSD) equation of state (De Santis et al., 1976) and the Lemmon-Jacobsen (LJ) model (Lemmon, 1996; Lemmon and Jacobsen, 1999; Lemmon and Tillner-Roth, 1999). The CSD has been chosen for its simplicity and good accuracy in representation of VLE for this family of mixtures. CSD EoS well represents the saturation boundaries of HC+HFC mixtures by simply applying the classical Van der Waals mixing rule: it requires only one binary interaction parameter, i.e. k_{12} , to be fitted for the data reduction.

The LJ model, which is the fundamental equation used in the Refprop 6.0 database, is a Free Helmholtz Energy (A) mixture model which allows the direct calculation of all the thermodynamic properties by proper differentiation. Thus, if the potential A is not well represented, errors can be generated in calculating other properties, such as enthalpy, entropy and density. The only use of VLE data to regress the interaction parameters in the Helmholtz energy equations allows a good representation of all the thermodynamic properties in the region close to the saturation boundaries.

Far from the saturation boundaries, even if the errors become a little greater, they should maintain a good consistency due to the theoretically grounded conception of the model. In fact, the model can work either in predictive or in correlative mode using up to five interaction parameters. The best results are clearly achieved in correlative mode, with interaction parameters obtained on the base of experimental data. However, this is not the case in the Refprop 6.0 for almost all the systems here considered. This reduces considerably the reliability of the calculations and can mislead in both the selection of proper alternatives to the traditional fluids and the design of the refrigerating plants components when these systems are considered. In this work, the interaction parameters used in the LJ mixture model have been regressed on the base of our experimental data. As an example of the influence of

Table 3: CSD and LJ model interactions parameters for the selected mixtures and deviations for pressure and vapour composition

System	CSD EoS			LJ Model						
	k_{ij}	AAD P [%]	AAD y	k_t	k_v	F_{ij}	β	γ	AAD P [%]	AAD y
R134a+R600a	0.1467	1.97	0.0102	0.8008	1.0123	-0.2988	1.0084	0.9833	0.38	0.0026
R600a + R236fa	0.1363	0.23	0.0030	0.8156	1.0172	0.0102	1.0405	0.9217	0.25	0.0030
R143a + R290	0.1207	0.68	0.0068	0.8293	1.0082	0.0559	1.0012	0.9429	0.47	0.0026
R290 + R134a	0.1535	0.50	-	0.8604	0.8749	-1.2345	0.9500	1.1150	0.57	n.a.
R290 + R245fa	0.1472	0.79	0.0026	0.8571	1.0153	-0.0999	1.0604	0.8196	0.71	0.0023
R290 + R236fa	0.1376	0.82	0.0030	0.8557	0.9077	-0.3632	1.1234	0.8865	0.60	0.0044
R290+ R236ea	0.1390	0.91	0.0033	0.8172	0.9121	0.9225	1.0555	0.8951	0.60	0.0034
R600a + R245fa	0.1413	0.58	0.0050	0.8172	1.0121	0.0584	1.0354	0.9223	0.51	0.0032
R290 + R227ea	0.1202	0.73	0.0029	0.8347	0.9446	0.0694	1.1044	0.8970	0.48*	0.0028*
R125 + R290	0.1397	0.39	0.0027	0.8921	0.8816	-1.8412	1	1	0.44	0.0051

the interaction parameters regressed on the base of experimental data, Figure 3 show the results of VLE calculations for the mixture R290(1)+R227ea(2).

For both models, the interaction parameters were regressed on the base of the VLE data measured at CNR-ITC by minimizing the objective function F:

$$F = \sum_{i=1}^{N_p} \left(\frac{\Delta P}{P_{\text{exp}}} \right)^2 \quad (1)$$

where N_p is the number of experimental points, P_{exp} is the experimental saturation pressure, P_{calc} is the calculated saturation pressure and $\Delta P = P_{\text{exp}} - P_{\text{calc}}$.

Table 3 summarises the interaction parameters regressed on the base of the VLE experimental data for both the CSD EoS and the LJ model.

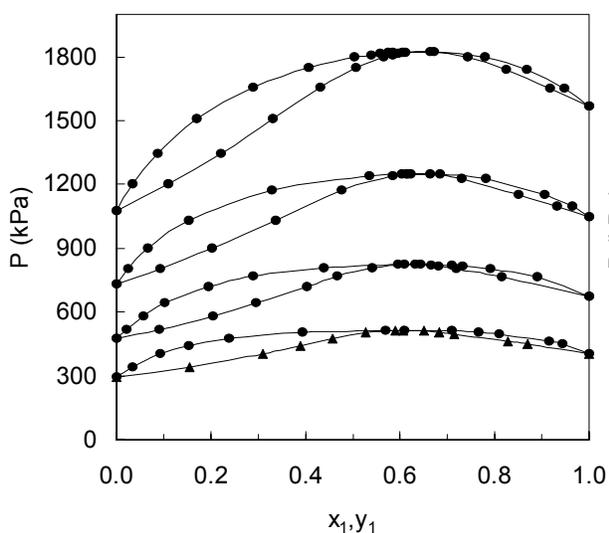


Figure 1: VLE experimental isotherms for the R125(1)+R290(2) system: (●) experimental; (—) trend line

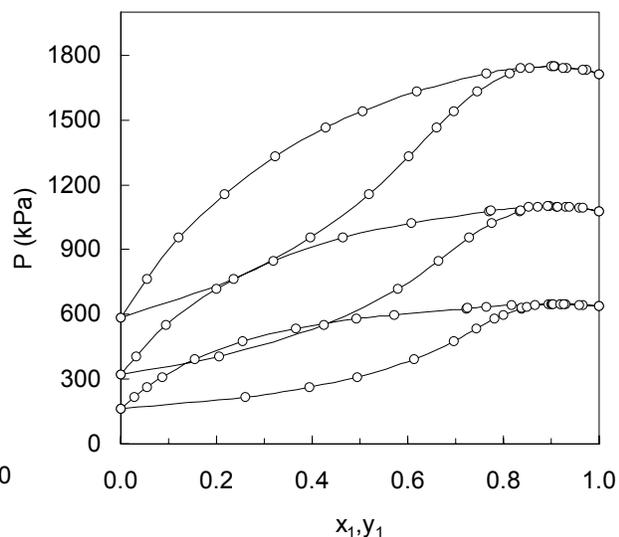


Figure 2: VLE experimental isotherms for the R290(1)+R236ea(2) system: (○) experimental; (—) trend line

3. EVALUATION OF CYCLES PERFORMANCE

All the systems have been studied as a working fluid in two standard refrigerating cycle, using the two models mentioned above to calculate their thermodynamic properties in the cycle. It is worth to underline again the importance of using, for any model, interactions parameters regressed on the base of experimental data, as already treated in detail elsewhere (Artico et al., 2002). Here it is enough to remind that the cycles conditions and performance can be strongly different applying the “ideal” interactions parameters (as in the case of the LJ model in Refprop 6.0 for most of the systems here considered) and the “experimental” interaction parameters. This is evident from Figure 4, where the boundary conditions and the ideal cycles calculated with the LJ model in predictive mode (Refprop 6.0) and in correlative mode are compared. E.g., the saturation pressures in the first mode are much lower than in the second one, influencing all the other thermodynamic properties and then the performance of the mixtures. However, the compressor work and the refrigerating effect have similar deviations and the effect on their ratio is quite small: therefore the Coefficient of Performance (COP) is relatively sensitive to these differences. On the contrary, the large deviations of the vapour density at the compressor suction determine a strong effect on the

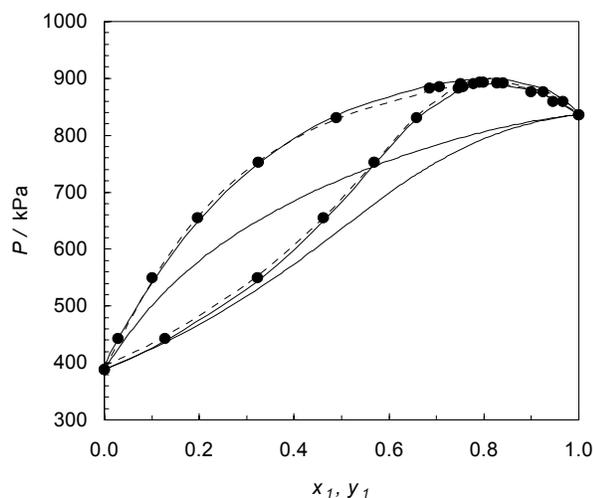


Figure 3. VLE for the R290 (1) + R227ea (2) system at 293.15 K: experimental data (\bullet), calculated curves with the CSD EoS (—) and the LJ EoS (---) in correlative mode (present work) and with the LJ EoS in predictive mode (Refprop 6.0) (bold —)

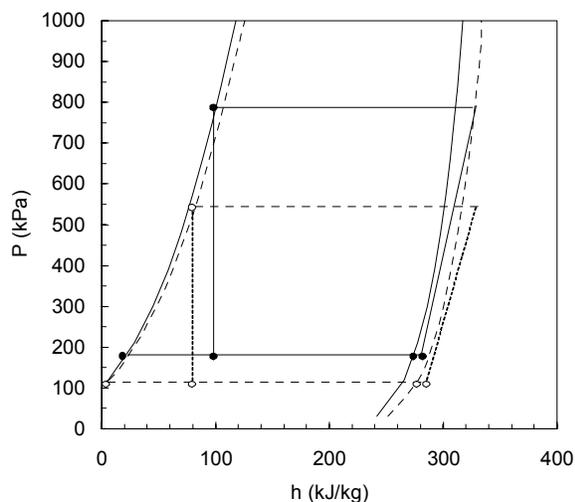


Figure 4: Ideal cycle calculated for the mixture R134a+R600a at equimolar composition with both the predictive mode (Refprop 6.0, ---) and the correlative mode (LJ model with interaction parameters regressed on the base of experimental VLE data, —).

volumetric refrigerating effect (VRE). This means that calculating the thermodynamic properties with the predictive mode can strongly mislead in dimensioning the compressor.

Two different single stage ideal cycles have been considered in order to evaluate the efficiency of the binary systems here considered in comparison with some reference fluids (R22, R134a, R502, R404A, R407C, R410A). The operative conditions of the cycles are summarized in Table 4. These conditions have been selected among the reference cycles considered in ASHRAE (2001), referring to the typical conditions for refrigeration and air conditioning.

Anyway, a problem arose in the calculations of the cycles with the LJ model by applying the subroutines available in Refprop 6.0, since in several cases it was not possible to calculate the temperature at the throttling valve outlet (and evaporator inlet). The problem is due to the non convergence in the calculation of the temperature in the two phase region by using the subroutines for the isoenthalpic transformation. This was particularly evident, for a given system, for the mixtures with a high concentration of the high boiling components.

However, the CSD EoS, with the interaction parameters shown in Table 3, gives very similar results in terms of cycles performance (COP and VRE) and then the following discussion will be based on the results obtained applying this EoS. The reader will have anyway the possibility to apply the interaction parameters presented in Table 3 with Refprop 6.0 using the mixing rule LM1 (Lemmon and Jacobsen model for mixtures) from the available options in the database.

Table 4: Operative conditions of the ideal cycles

Cycle	a	b
Evaporating Temperature ($^{\circ}\text{C}$)	-23	4
Condensing Temperature ($^{\circ}\text{C}$)	37	37
Superheating ($^{\circ}\text{C}$)	10	10
Sub cooling ($^{\circ}\text{C}$)	0	0
Isentropic Efficiency	0.8	0.8

Considering it is impossible to reproduce all the information derived by the cycles calculations, we will rely on the diagrams of COP and VRE shown in figure 5 and 6, where these parameters have been plotted as a function of the mass fraction for each system. In the same diagrams, it is possible to compare the performance of the mixtures with the reference fluids.

The COP has been calculated as the ratio between the compression work and the refrigerating effect including the

superheating (10 K) at the evaporator outlet. The VRE has been calculated as the product between the refrigerating effect including the superheating and the density of the fluid at the compressor inlet. The isentropic efficiency in the compression process is 0.8.

The curves in figure 5 and 6 has been plotted as a function of the mass fraction of the hydrocarbon (R290 or R600a) in the system considered. This allow to highlight the difference between mixtures with propane and mixtures with isobutane. The mixtures with R290 are drawn with a full line, while the mixtures with R600a with a dotted line.

Coefficient of performance (COP): the COP of R600a is higher than that of R290 in both the cycles here considered. This is due to the higher critical temperature of isobutane. For the systems where the HC has higher NBT than the HFC, by adding an HFC to the hydrocarbons, i.e. reducing the mass fraction of the HC, there is a slight decrease in the energetic efficiency up to a well defined mass fraction at which begin a sudden degradation of the COP up to a minimum. The change is in the correspondence of the azeotropic composition for the mixture considered. The

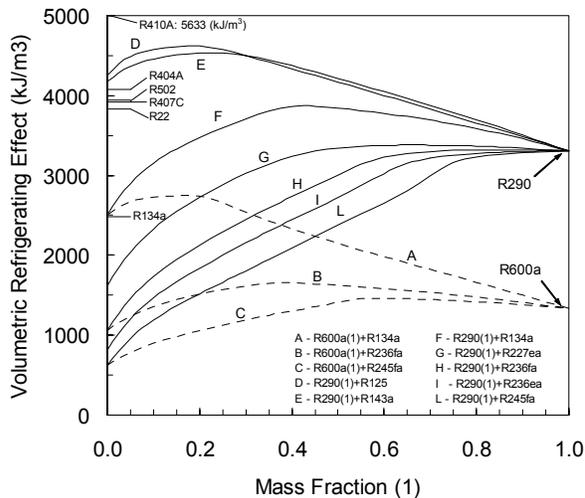
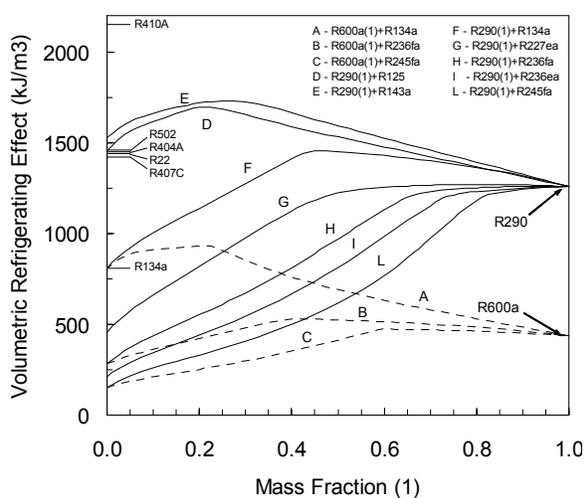
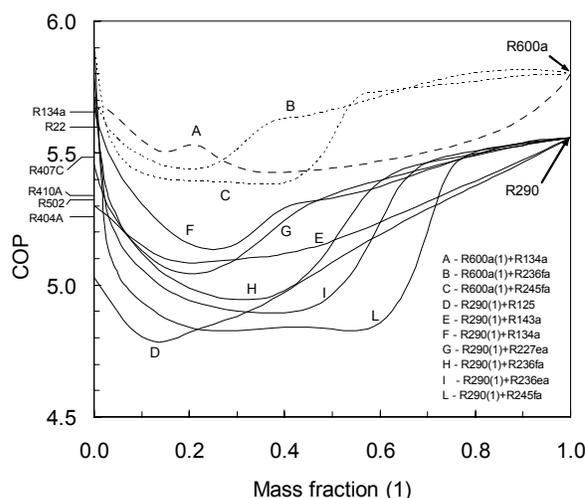
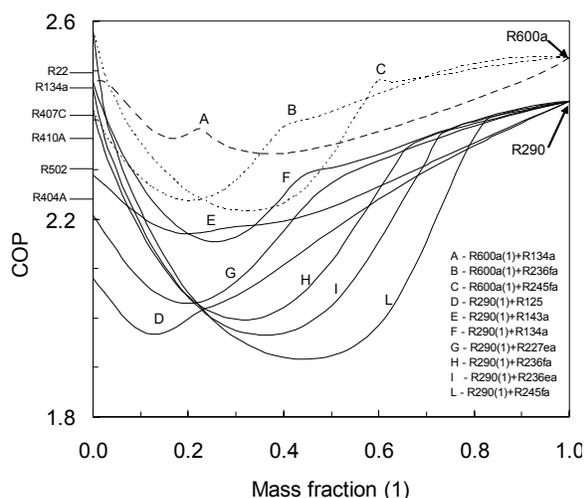


Figure 5. COP and Volumetric Refrigerating Effect (VRE) as a function of the mass fraction at the working conditions of cycle (a) in Table 4.

Figure 6. COP and Volumetric Refrigerating Effect (VRE) as a function of the mass fraction at the working conditions of cycle (b) in Table 4.

higher the difference in the boiling point between the HFC and the HC, the wider is the “depression” in the COP curve, which has a parabolic shape in this region. After that, the COP increases again up to the value of the pure HFC.

More regular behaviour, anyway with a minimum of COP at some specific composition, is shown by the mixtures of R600a with R134a and of R290 with R125 and R143a, for which the boiling temperature of the HC is higher than that of the HFC. In general, the mixtures including R600a show higher COP than the mixtures with R290: however, it must be considered that at the conditions of cycle (a), the evaporation pressures for high boiling mixtures are frequently below atmospheric pressure, depending on the mass fraction and on the boiling temperature of the two components.

In comparison with the reference fluids, it is clear that the mixtures with R290 could be competitive in terms of energetic efficiency only at mass fractions of propane higher than that of the azeotrope. Mixtures with R600a show a quite good values of COP, especially those with R245fa and R236fa, even if the problem of subatmospheric pressure in the evaporator must be considered.

Finally, the COP at the conditions of cycle (b) is much higher than for cycle (a). This is quite obvious due to the much lower difference between the evaporating and the condensing temperature.

Volumetric Refrigerating Effect (VRE): this parameter is strongly connected to the density of the working fluid. For both the operating conditions considered, it is evident that mixtures with R290 have a much higher VRE than those with R600a. This is due to the higher saturation pressure of R290 and thus its higher vapour density. The best performance are obtained by the systems formed by propane with R143a and R125, while a continue reduction of VRE is obtained by mixtures with the HFC having higher and higher NPT. Also in this case there is a sudden (even if less dramatic than for COP) change in the slope of the VRE curve in correspondence to the azeotropic composition, with a decrease of VRE towards the value for the pure HFC.

The VRE of R600a is around one third of that of R290 and this strongly influence the VRE of the mixtures, penalising those with R600a, at least at the higher mass fraction of R600a. This means that the dimensions of the compressor must be quite bigger, with consequent drawbacks in terms of costs and encumbrance.

The mixtures of R290 with R125 and R143a, in the low R290 mass fraction range, obtain VRE better than that of the reference fluids, except R410A which has a very high volumetric effect due to its high density. The volumetric effect of the other mixtures with R290 is generally lower than that of the reference fluids, a part from R134a. Anyway, in the high R290 mass fraction range, the VRE is acceptable in comparison with that of R404A, R502, R407C and R22. The mixtures with R600a has in any case a volumetric refrigerating effect much below that of the reference fluid.

6. CONCLUSIONS

The analysis performed in this paper showed that the performance of the HCs+HFCs blends here considered is satisfying, with reference to the performance of the reference fluids, only in some range of composition and as a compromise between different requirements. Mixtures with R600a show a relatively good COP, but their application could be limited due to the possible subatmospheric evaporating pressure and the poor volumetric refrigerating effect. Mixtures with R290 have acceptable COP and volumetric refrigerating effect only at relatively high mass fraction of propane. This is positive since it allows a reduction in the GWP of the HFC, but the mass fraction of the HFC in the mixture could be not enough to eliminate the flammability. Moreover, deeper considerations on the TEWI, it means the total equivalent warming impact, should be done considering that in same case the pure HFC has higher COP than the hydrocarbons and thus have a lower indirect effect, that is generally dominant on the direct effect represented by the GWP.

NOMENCLATURE

AAD	Absolute average deviation	NBT	Normal Boiling Temperature	(°C)
CSD	Carnahan Starling de Santis EoS	P_c	Critical Pressure	(kPa)
EoS	Equation of State	P	Pressure	(kPa)
GWP	Global Warming Potential	T_c	Critical Temperature	(°C)
k_{ij}	CSD interaction parameter	TEWI	Total Environmental Warming Impact	
$k_t, k_v, F_{ij}, \beta, \gamma$	LJ model interaction parameters	y	vapor mole fraction	

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