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Evaluation of non-azeotropic mixtures containing HFOs as potential refrigerants in refrigeration and high-temperature heat pump systems

ZHANG ShengJun*, WANG HuaiXin & GUO Tao

Department of Thermal Energy and Refrigeration Engineering, Tianjin University, Tianjin 300072, China

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With the increasing environmental concern on global warming, hydrofluoro-olefin (HFOs), possessing low GWP, has attracted great attention of many researchers recently. In this study, non-azeotropic mixtures composed of HFOs (HFO-1234yf, HFO-1234ze(z), HFO-1234ze(e) and HFO-1234zf) are developed to substitute for HFC-134a and CFC-114 in air-conditioning and high-temperature heat pump systems, respectively. The cycle performances were evaluated by an improved theoretical cycle evaluation methodology. The results showed that all the mixtures proposed herein were favorable refrigerants with excellent thermodynamic cycle performances. M1A presented lower discharge temperature and pressure ratio and higher COP_c than that of HFC-134a. The volumetric cooling capacity was similar to HFC-134a. It can be served as a good environmentally friendly alternative to replace HFC-134a. M3H delivered similar discharge temperature as CFC-114 did. And the COP_h was 3% higher. It exhibits excellent cycle performance in high-temperature heat pump and is a promising refrigerant to substitute for CFC-114. And the gliding temperature differences enable them to exhibit better coefficient of performance by matching the sink/source temperature in practice. Because the toxicity, flammability and other properties are not investigated in detail, extensive toxicity and flammability testing needs to be conducted before they are used in a particular application.

hydrofluoro-olefin, non-azeotropic mixtures, refrigerant, air-conditioning system, high-temperature heat pump

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1 Introduction

Accelerating phaseout of chlorine and bromine containing refrigerant gases in developed and developing countries was agreed at the 19th Meeting of the Parties to the Montreal Protocol due to the depletion of the ozone in the stratosphere.

The focus on the possible global climate change has led to the restricted emissions of CO_2 and five other GHGs (methane, nitrous oxide, sulfur hexafluoride, HFCs and PFCs) in developed countries required by the Kyoto Protocol. According to a survey by IPCC, HFCs atmospheric concentrations rise at a rate of 13%−17% per year between 2001 and 2003. Of all the emissions, mobile air conditioning (MAC) dominates other sectors, accounting for over 65% of all HFCs refrigerant emissions [1]. Thus, legislative action in Europe [2] phases out the use of the hydrofluorocarbon having GWP greater than 150 in MACs of newly manufactured vehicles from 2011.

Recently, fluorinated propane isomers, possessing low GWPs, become the limelight of research and development activities as a potential substitute for HFCs. Brown [3] predicted the critical temperatures, critical pressures, critical

*Corresponding author (email: zhangshengjun@tju.edu.cn)

densities, acentric factors, and ideal gas specific heats at constant pressure for eight fluorinated olefins using group contribution methods. From among the fluorinated propane isomers, the most promising HFC-134a replacement fluid was HFO-1234yf [4]. DuPont and Honeywell proposed the HFO-1234yf as the substitute for HFC-134a in MAC. And thermodynamic properties of HFO-1234yf were estimated by Thomas [5] using M-H EOS and the modeling results of the comparison between HFO-1234yf and HFC-134a showed that it behaved very similarly as HFC-134a did in refrigeration system. Its compatibility with motor, sealing materials and POE lubricants, the toxicity and the flammability properties was also investigated by Thomas. Park [6] studied the nucleate boiling heat transfer coefficients of HFC-134a and HFO-1234yf on a flat plain and low fin surfaces for the design and manufacture of high efficiency evaporators for HFO-1234yf. Four blends, AC-1, DP-1, JDH and fluid H possessing low GWP and using HFO-1234yf as ingredient [7], used to be considered as the substitutes for HFC-134a, but further consideration was ended because of their toxicity.

However, there are other potential applications for fluorinated propane isomers beyond the automotive industry. Calm [8] pointed out that although the current regulatory pressures addressed mobile air conditioners, future extension to other applications was almost certain. Brown [9] estimated the thermodynamic and transport properties of HFO-1234ze(z) and predicted the performance potential of HFO-1234ze(z) in high-temperature heat pump applications for the substitute for CFC-114. The comparison between eight fluorinated propane isomers and conventional refrigerant in idealized vapor compression refrigeration cycle for three applications, i.e. unitary air conditioning, automotive air conditioning and high-temperature heat pump, was conducted by Brown. The results showed that they were favorable refrigerants for these potential applications [10].

It is shown both theoretically and experimentally that non-azeotropic refrigerant mixtures have the potential to increase the COP by approximating the temperature variation of the working fluid during evaporation or condensation at constant pressures to the temperature profile of the source or sink fluid. And the use of non-azeotropic mixtures can not only reduce the irreversibilities of the components but also may help in the research for environmentally safe refrigerants [11–15].

There was little work concerning the use of non-azeotropic mixtures including HFOs (HFO-1234yf, HFO-1234ze(z), HFO-1234ze(e) and HFO-1234zf) in air-conditioning and high-temperature heat pump systems. Therefore, in this study cycle performance of HFOs and their mixtures, M1A (HFO-1234zf/HC-290, 60%/40% in the mass), M2A (HFO-1234ze(z)/HC-290, 30%/70% in the mass), M3A (HFO-1234ze(e)/HC-1270, 70%/30% in the mass) and M1H (HFO-1234ze(z)/HC-601, 95%/5% in the mass), M2H (HFO-1234ze(e)/HC-601b, 40%/60% in the mass), M3H (HFO-1234zf/HC-601, 65%/35% in the mass), were evaluated theoretically in an attempt to examine the possibility of substituting HFC-134a and CFC-114, respectively. Thermodynamic properties of HFOs were estimated with the group contribution techniques and P-T Equation of State (P-T EoS). The credibility of this method was presented. The cycle performance of the working fluids was compared in both systems.

2 Estimating thermodynamic properties

Although much work has been done regarding these fluorinated propane isomers, there are little thermodynamic data available in the open literature for R-1234yf and an even scarcer amount of data are available in the open literature regarding the others. Then estimating thermodynamic properties with equation of state coupled with group contribution techniques is a simple and accurate approach.

Brown [9] demonstrated the ease and accuracy of implementing such an approach by using several group contribution techniques coupled with the Peng-Robinson Equation of State contained in REFPROP 8.0. And the thermophysical property values of HFO-1234yf and HFO-1234ze(z) were predicted by knowing only their normal boiling point temperature (NBP) and their structural formula. Thomas [5] described a Martin-Hou Equation of state model for the calculation of thermophysical properties of HFO-1234yf. Zheng and Wang [16] showed the calculation accuracy of 33 refrigerants, including halocarbons, CFCs, HCFCs, and HFCs, using P-T EoS.

In this paper, methodologies illustrated by Zheng and Wang are used to estimate the thermodynamic property values of HFOs. The comparison between values generated by this methods for R1234yf and the published data is conducted.

2.1 Method of estimating the thermodynamic properties

The general form of P-T EoS [17] is shown as eq.(1) and the mixing rules are given from eqs. (9)–(13).

$$P = \frac{RT}{v-b} - \frac{a[T]}{v(v+b)+c(v-b)}, \tag{1}$$

$$a[T] = \Omega_a \left(R^2 T_c^2 / P_c\right) \alpha[T_r], \tag{2}$$

$$\alpha[T_r] = \left[1 + F\left(1 - T_r^{1/2}\right)\right]^2, \tag{3}$$

$$b = \Omega_b \left(RT_c / P_c\right), \tag{4}$$

$$c = \Omega_c \left(RT_c / P_c\right), \tag{5}$$

Table 1 Estimated fundamental thermodynamic parameters for fluorinated propane isomers

Refrigerant	Molecular formula	NBP (K)	T_c (K) [3]	P_c (kPa) [3]	C_p^0@T_r=0.8 (kJ·kg^{-1}·K^{-1})	ω [3]
HFO-1234yf	CF3CF=CH2	245.15	369.3	3435	0.818	0.28
HFO-1234ze(e)	CF3CH=CHF	254.15	384.4	3576	0.750	0.295
HFO-1234zf	CF3CH=CH2	251.65	389.7	3849	0.912	0.284
HFO-1234ze(z)	CF3CH=CHF	282.15	426.8	3970	0.890	0.333

$$\Omega_c = 1 - 3\zeta_c, \tag{6}$$

$$\Omega_b^3 + \left(2 - 3\zeta_c\right)\Omega_b^2 + 3\zeta_c^2\Omega_b - \zeta_c^3 = 0, \tag{7}$$

where Ω_b is the least cubic root of eq. (6), R is the universal gas constant of working fluid, T_r=T/T_c, T_c, P_c, F and ζ_c are demanded for evaluation of the fluids. The parameters of F, ζ_c are fitted as the function of ω (eccentric)

$$F = 0.452413 + 1.30982\omega - 0.295937\omega^2, \tag{8}$$

$$\zeta_c = 0.329032 - 0.076799\omega + 0.0211947\omega^2. \tag{9}$$

Mixing rules for refrigerant mixture are shown as

$$a_m = \sum_i \sum_j x_i x_j a_{ij}, \tag{10}$$

$$b_m = \sum_i x_i b_i, \tag{11}$$

$$c_m = \sum_i x_i c_i, \tag{12}$$

$$a_{ij} = \xi_{ij}(a_{ii}a_{jj})^{1/2}. \tag{13}$$

Thermodynamic properties can be predicted by P-T EoS by knowing values of critical temperature (T_c), critical pressure (P_c), acentric factor (ω), and ideal gas specific heat(C_p^0). For not-so-well-defined fluids, group contribution techniques are usually used to estimate these parameters based only on the fluid's NBP and its structural formula. T_c, P_c and ω were predicted by Brown showing a good accuracy [3]. And C_p^0 was estimated by using the group contribution method of Joback [18].

Table 1 provided estimates for the thermodynamic parameters T_c, P_c, ω, and C_p^0 of the fluorinated olefins based only on the refrigerants' NBPs and their structural formulas.

2.2 The reliability of the EoS

Since few data have been reported in the open literature for these refrigerants, a comparison based on the thermodynamic parameters from Table 1 and P-T EoS with open literature can be made only for R-1234yf. Comparisons of the P-T EoS predictions were made with the experimental data and estimated data by other researchers for liquid enthalpies, vapor enthalpies, vapor pressures and vapor densities. Deviations in the predicted values along with temperature and pressure were shown in Figures 1–4.

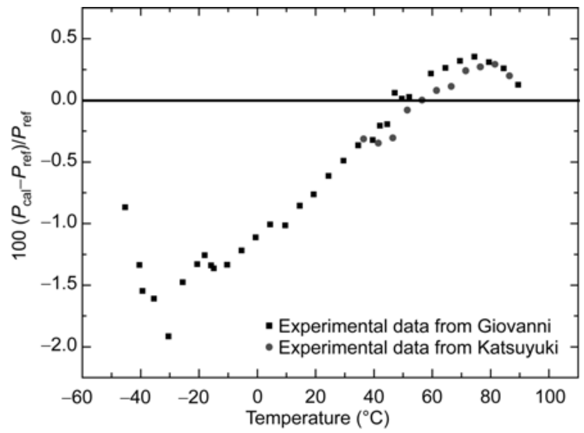

Figure 1 Deviations in the saturated vapor pressure predictions with P-T EoS from the experimental values by Di Nicola and Tanaka.

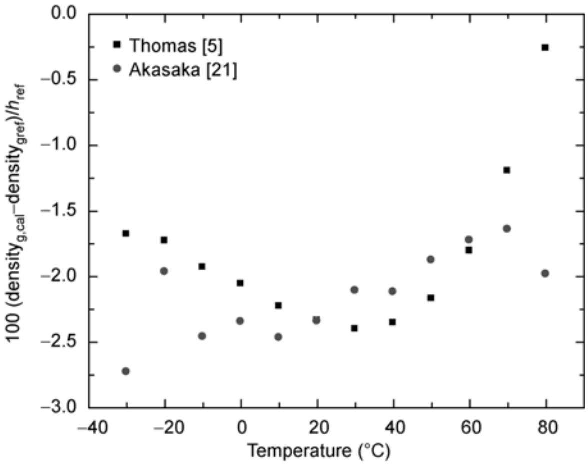

Figure 2 Deviations in vapor density values from the estimated values by Thomas and Akasaka.

The deviations of the saturated vapor pressure predictions with P-T EoS from the experimental values by Di Nicola [19] and Tanaka [20] were exhibited in Figure 1. Most of them were within ±1.5% and the average absolute deviation (AAD) was 0.61%.

Figure 2 presented the deviations of the present vapor density data from those estimated values by Thomas [5] and Akasaka [21]. With the increase of the temperature, the density prediction with P-T EoS tended to be close to the published ones. And the AAD was 1.9%.

Figures 3 and 4 showed deviations in the calculated h_f and h_g with P-T EoS from the values by Spatz [4] and Tho-

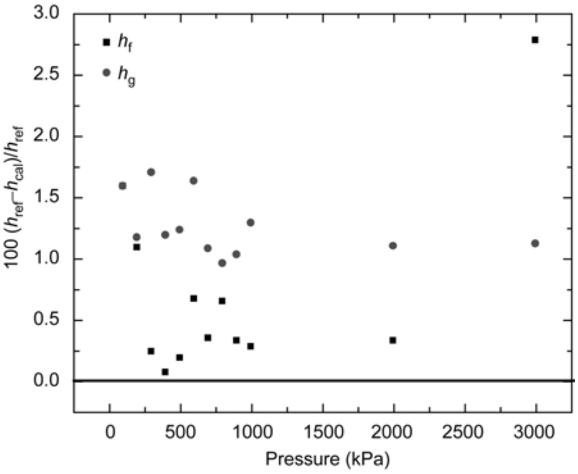

Figure 3 Deviations in vapor and liquid enthalpy values from the estimated values by Spatz.

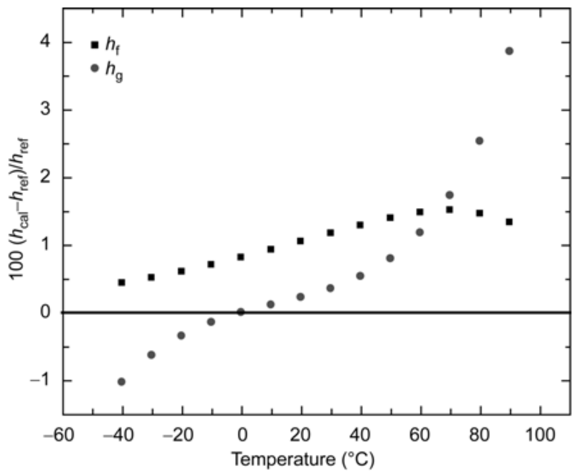

Figure 4 Deviations in vapor and liquid enthalpy values from the estimated values by Thomas.

mas [5]. The deviations of the PT prediction were mostly within 2%. And the AAD from Spatz and Thomas was 0.71%, 1.25% and 1.04%, 0.65%, respectively.

As indicated in the above-mentioned figures, the thermodynamic property values of R1234yf estimated by P-T EoS are favorable as compared with the known values.

3 Cycle performances of the fluids

The evaluation of the refrigerant cycle performance in an idealized vapor compression cycle is an effective approach to screen possible candidate refrigerants without resorting to time-consuming and detailed system modeling. In this study, thermodynamic performance of four pure fluorinated propane isomers and six mixed refrigerants containing the HFOs and conventional environmental-friendly refrigerants were evaluated in an attempt to examine the possibility of substituting for HFC-134a in air conditioning and for R114 in high-temperature heat pump. *COP* and volumetric capacity were used as the screening criteria and compressor discharge temperature and pressure ratio were considered as the factor for estimating the safety and volumetric efficiency of the compressor. An emphasis was given to those fluids that provided a similar volumetric capacity, so a compressor change could be avoided in the existing equipment.

An improved theoretical cycle evaluation calculation method [22] was used to analyze the cycle performance of refrigerants. Motor efficiency, mechanical efficiency and isentropic efficiency were assumed to be 0.85. The clearance volume was 0.03 and the shell heat loss rate was 0.02. The operation conditions assumed for air-conditioning system were evaporating temperature of 7.2°C, condensing temperature of 54.4°C, superheating degree of 11.1°C, subcooling degree of 8.3°C. Theoretical cycle performance of refrigerants in high-temperature heat pump was investigated with a fixed cycle temperature lift of 45°C and a variation of condensing temperature between 70°C and 110°C. The superheating and subcooling temperature was kept at 5°C. Basic thermodynamic and environmental properties of the fluids were given in Table 2. The gliding temperature difference (GTD) in Table 2 was calculated when the condensing temperature was 90°C.

3.1 Potential application of the HFOs in air conditioning

It was found in Figure 5 that R32 exhibited the highest q_{vc}, T_{dis} and the lowest COP_c, so it was not a good drop-in substitute for HFC-134a. HFO-1234yf, recommended by many researchers as the replacement for HFC-134a, provided that q_{vc} was 3.5% less than HFC-134a and 12°C lower than the compressor discharge temperature. Although the COP_c was 1.7% lower than HFC-134a, the pressure ratio was 7.8% lower than HFC-134a which may result in a higher compressor efficiency. The other HFOs yielded the q_{vc} much less than HFC-134a, which meant none of them could be considered as the replacement for HFC-134a as pure fluids.

Table 2 Basic thermodynamic and environmental properties of the selected fluids

Substance	Molar mass (kg/kmol)	T_c (°C)	P_c (MPa)	ODP	GWP 100 a	$T_{GTD,evap}$ (°C)	$T_{GTD,cond}$ (°C)
M1A	65.27	104.88	4.09	0	<10	3.08	2.64
M2A	54.04	104.76	4.21	0	<10	11.01	7.99
M3A	51.90	94.27	4.47	0	<10	4.50	4.25
M1H	110.82	157.01	4.31	0	<10	2.27	1.88
M2H	84.58	146.00	3.31	0	<10	6.04	3.35
M3H	86.04	149.98	3.65	0	<10	20.07	15.41

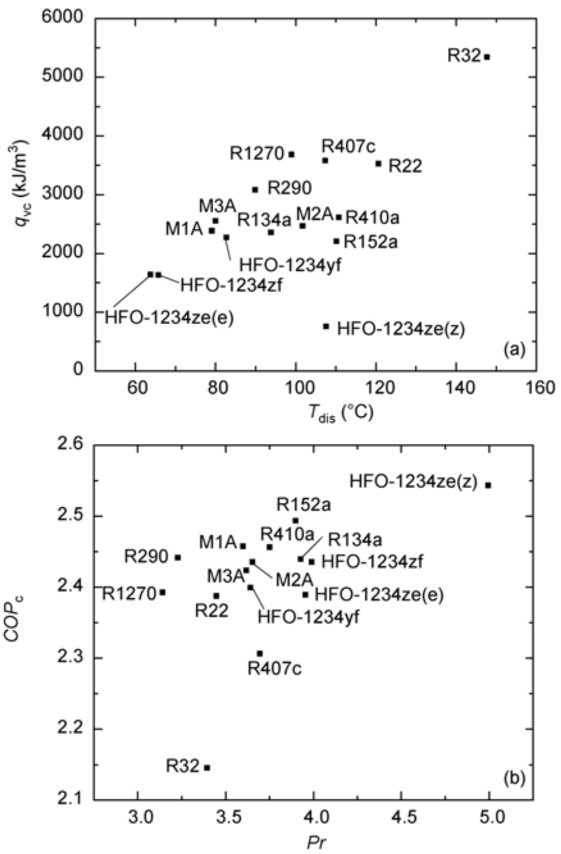

Figure 5 (a) q_{vc} and T_{dis} of the working fluids in air-conditioning condition; (b) COP_c and Pr of the working fluids in air-conditioning condition.

All the mixtures delivered similar q_{vc} as HFC-134a did. M1A presented higher COP_c and lower Pr than HFC-134a. The discharge temperature was about 15°C lower than HFC-134a. It presented the most favorable cycle performance. M2A exhibited 16°C higher discharge temperature and 8% lower Pr than HFC-134a. And its gliding temperature was the highest one. M3A showed the lower discharge temperature and lower Pr than HFC-134a. Although the COP_c of M2A and M3A were lower than HFC-134a, they had potential improvement by the temperature match. Thus, all the mixtures are favorable alternative for HFC-134a in air-conditioning system.

3.2 Potential application of the HFOs in high-temperature heat pump

It is identified by Figure 6 that among the fluorinated propane isomers, HFO-1234ze(z) delivered similar q_{vh} as CFC-114 and exhibited the highest COP_h. However, the discharge temperature was 17°C higher than that of CFC-114.

M1H and M3H exhibited almost the same q_{vh} as CFC-114 and higher COP_h and Pr than that of CFC-114. M1H showed the highest discharge temperature of the mixtures. When the condensing temperature was 90°C, the discharge temperature of M1H was 118°C, similar to HFO-1234ze(z), 20°C higher than that of CFC-114 and the COP_h was 5.6% higher than that of CFC-114. The COP_h of M2H was 1.2% less than that of CFC-114, but the discharge temperature was 3°C lower. M3H provided the discharge temperature of 101°C in the same condition. And the COP_h was 2.5% higher than that of CFC-114.

Overall, all the mixtures investigated herein showed favorable cycle performance and were appropriate replacement for CFC-114 in high-temperature heat pump with potential improvement in COP_h by the temperature match. M3H showed the best cycle performance comprehensively.

4 Other properties of the fluid

Thermodynamic properties are studied to prove that HFOs and their mixtures are favorable refrigerants to substitute for HFC-134a and CFC-114 in air-conditioning and high temperature heat pumps, respectively. Other characteristics, such as heat transfer coefficients, environmental impact, compatibility with common materials, toxicity and flammability, are also important for refrigerants to be inspected before they are used in particular applications.

The predicted heat transfer coefficients of HFO-1234yf vs. HFC-134a were −8% in condenser and +3% in evaporator. The miscibility in common lubricants showed that it was well miscible with POE. Thermal stability and compatibility with plastics, elastomers were tested to show that it could safely be used with good comparable degree of interaction [23]. The environmental performance was investigated to show that the ODP=0, GWP=4, atmospheric lifetime=11days. And no high GWP products were produced once it was broken down [24]. The flammability of HFO-1234yf was shown in Table 3. It was potential for A2L by ISO 817 classification and classified to class A2 by ASHARE standard. Due to the low BV (Burning Velocity) and high MIE (Minimum Ignition Energy), ASHARE SSPC 34 Flammability Subcommittee recommended to add a new A2L flammability classification group for HFO-1234yf, which is the lowest flammability classification [25]. It had low acute and chronic toxicity [23]. It is concluded that HFO-1234yf is the potential drop-in alternative refrigerant of HFC-134a in air conditionings.

The other HFOs have no ODP and almost certainly have a GWP on the order of probably no more than 10 or so based on values of HFO-1234yf. Although the toxicity, flammability and other physical and chemical properties are not investigated in detail, they are predicted to be little greater or lesser degrees than R1234yf. Thus, before HFO-1234ze(z), HFO-1234ze(e) and HFO-1234zf could be used in a particular application, extensive toxicity and flammability testing is highly recommended to be conducted. However, they could be very likely to replace CFC-114 in high-temperature heat pump.

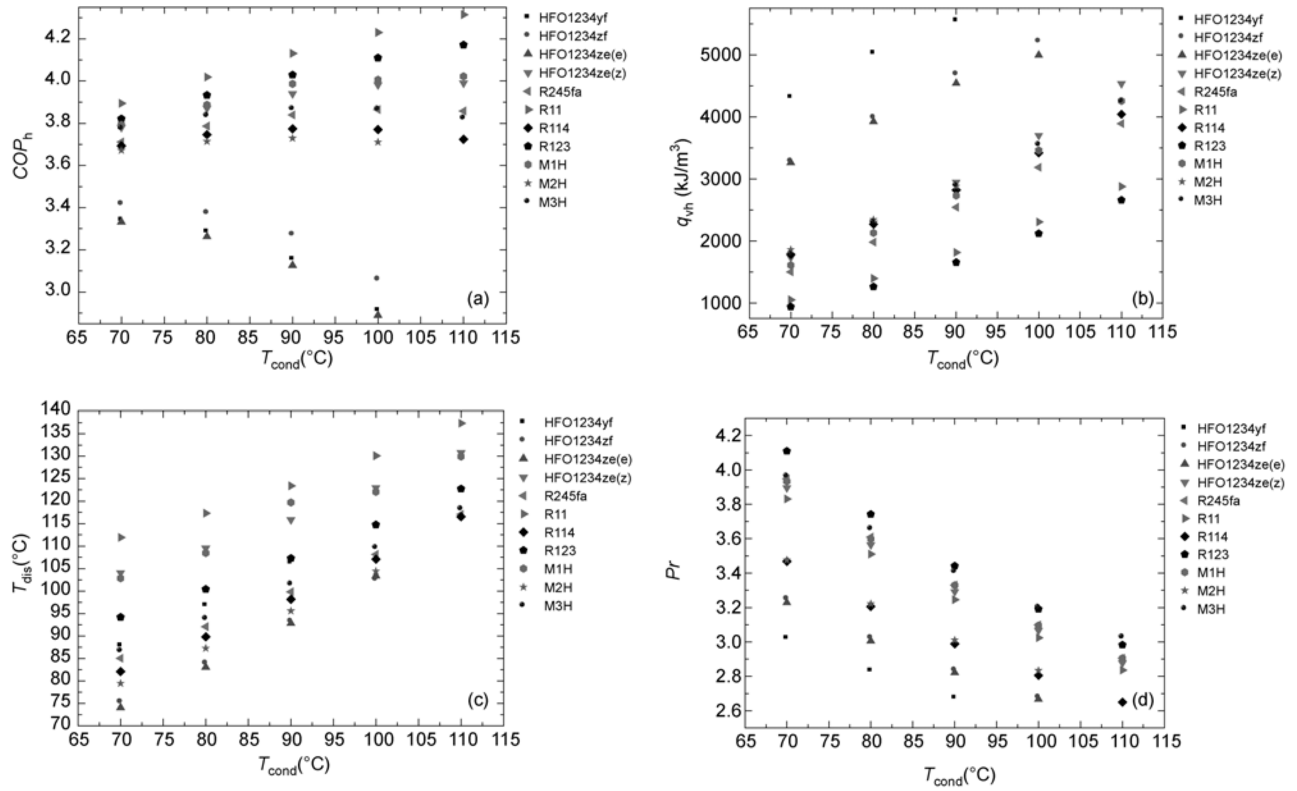

Figure 6 Variation of cycle performance with condensing temperature. (a) Variation of heating *COP* with condensing temperature, (b) variation of volumetric heating capacity with condensing temperature, (c) variation of discharge temperature with condensing temperature, (d) variation of pressure ratio with condensing temperature.

Table 3 Flammability of refrigerants [23]

Refrigerant	LFL$^{a)}$ (vol%)	UFL$^{a)}$ (vol%)	(UFL-LFL) (vol%)	MIE (mJ)	HOC (kJ/g)	BV (cm/s)
Propane	2.2	10.0	7.8	0.25	46.3	46
R152a	3.9	16.9	13.0	0.38	16.5	23
R32	14.4	29.3	14.9	30–100$^{b)}$	9.4	6.7
Ammonia	15.0	28	13.0	100–300$^{b)}$	18.6	7.2
HFO-1234yf	6.2	12.3	6.1	5000–10000$^{b)}$	10.7	1.5$^{c)}$

a) Flame limits measured at 21°C, ASTM 681-01; b) tests run in 12 liter flask to minimize wall quenching effects; c) HFO-1234yf BV measured by AIST, Japan.

5 Conclusion

The theoretical cycle performance of HFOs and their mixtures were investigated to substitute for HFC-134a and CFC-114 in air-conditioning and high-temperature heat pump systems, respectively, using an improved theoretical cycle evaluation methodology. And the following conclusions were drawn:

(1) P-T EoS was implemented in estimating the thermodynamic properties combined with group contribution method. The predicted results agreed well with the published data.

(2) The COP_c of M1A was 1.5% higher and the discharge temperature was 15°C lower than that of HFC-134a. The Pr was 9% lower than that of HFC-134a. Due to the excellent cycle and environmental performance, it was a good substitute for HFC-134a in air-conditioning system.

(3) M3H provided similar q_{vh} and discharge temperature as CFC-114 did and delivered 2.5% higher COP_h than that of CFC-114. It exhibited better cycle performance comprehensively in high-temperature heat pump system and was a potential refrigerant to substitute for CFC-114.

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