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Critical Parameters Measurements of Four HFE + HFC Binary Systems: Pentafluoromethoxyethane (HFE-245Mc) + Pentafluoroethane (HFC-125), + 1,1,1,2-Tetrafluoroethane (HFC-134a), + 1,1,1,2,3,3,3-Heptafluoropropane (HFC-227ea), and + 1,1,1,2,3,3-Hexafluoropropane (HFC-236ea)

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Critical parameters measurements on four hydrofluoroether (HFE) + hydrofluorocarbon (HC) binary systems—pentafluoromethoxyethane ($CF_3CF_2OCH_3$, HFE-245mc) + pentafluoroethane (CHF_2CF_3, HFC-125), + 1,1,1,2-tetrafluoroethane (CH_2FCF_3, HFC-134a), + 1,1,1,2,3,3,3-heptafluoropropane (CF_3CHFCF_3, HFC-227ea), and + 1,1,1,2,3,3-hexafluoropropane (CHF_2CHFCF_3, HFC-236ea)—have been carried out with uncertainties of ±10 mK in temperature, ±0.5 kPa in pressure, ±1 kg m^{-3} in density, and ±0.5% in composition. The experimental data were correlated by equations proposed by Higashi.

1. Introduction

Chlorofluorocarbons (CFCs) have been utilized extensively as refrigerants, blowing agents, and cleaning solvents because of their chemical stability and physical properties. However, they were identified as contributing to ozone layer depletion and global warming and were abolished. They were replaced by alternative CFCs (hydrochlorofluorocarbons (HCFCs) hydrofluorocarbons (HFCs)). However, they too are required to be abolished by 2020. Furthermore, the HFCs have been also identified as greenhouse gases in the Kyoto Protocol and will be banned soon. The development of new alternatives has thus become a matter of urgency.

Hydrofluoroethers (HFEs) are environmentally benign compounds having zero ODP and low GWP and are expected to be the new alternatives.[1,2] The Research Institute of Innovative Technology for the Earth (RITE) synthesized and evaluated about 150 HFEs.[3,4] Of these, trifluoromethoxymethane (CF_3OCH_3, HFE-143m) and pentafluoromethoxyethane ($CF_3CF_2OCH_3$, HFE-245mc) were found to be possible alternatives for dichlorodifluoromethane (CCl_2F_2, CFC-12) and 1,2-dichloro-1,1,2,2-tetrafluoroethane ($CClF_2CClF_2$, CFC-114), respectively. Unfortunately, they are slightly combustible (ASHRAE class 2),[5] and for safety reasons, the HFEs are expected to be used in binary mixtures with inflammable HFCs.

The critical parameters (critical temperature, pressure, and density) are the most important physical properties for the development of an equation of state to calculate and estimate the thermodynamic properties of fluids in industry. However, reliable information on the thermophysical properties of these mixtures has not yet been reported. In previous papers, we described an apparatus for the precise measurement of critical properties of fluids and its application to the newly synthesized HFEs, hydrofluoroketones, and a hydrofluoroamine[6,7] and critical parameters of four binary mixtures of HFE-245mc + pentafluoroethane (CHF_2CF_3, HFC-125), + 1,1,1,2-tetrafluoroethane (CH_2FCF_3, HFC-134a), + 1,1,1,2,3,3,3-heptafluoropropane (CF_3CHFCF_3, HFC-227ea), and + 1,1,1,2,3,3-hexafluoropropane (CHF_2CHFCF_3, HFC-236ea).[8] In this work, the apparatus was used to measure the critical parameters of the binary mixtures of HFE-245mc and HFC-125, HFC-134a, HFC-227ea, and HFC-236ea.

2. Experimental Section

Materials. Table 1 summarizes the sample codes, molecular formulas, molar-based purities, and normal boiling points for all of the compounds used in this study. They were all supplied by RITE. Their molar-based purities were analyzed by gas chromatography (Hewlett-Packard, model HP-6890; thermal conductivity detector). Previously reported critical parameters of these compounds are given in Table 2 together with our experimental results. Procedures for the critical parameter measurements of pure substances were described in the previous papers.[6,7]

Apparatus. The critical parameters of the mixtures were measured by observing the behavior of the meniscus at the vapor−liquid interface in an optical cell. Figure 1 is a schematic representation of the experimental apparatus. The apparatus is composed of four main parts: (A) a rectangularly shaped optical cell (ca. 5 cm^3 in volume), (B) two variable-volume vessels to control the inner volume of

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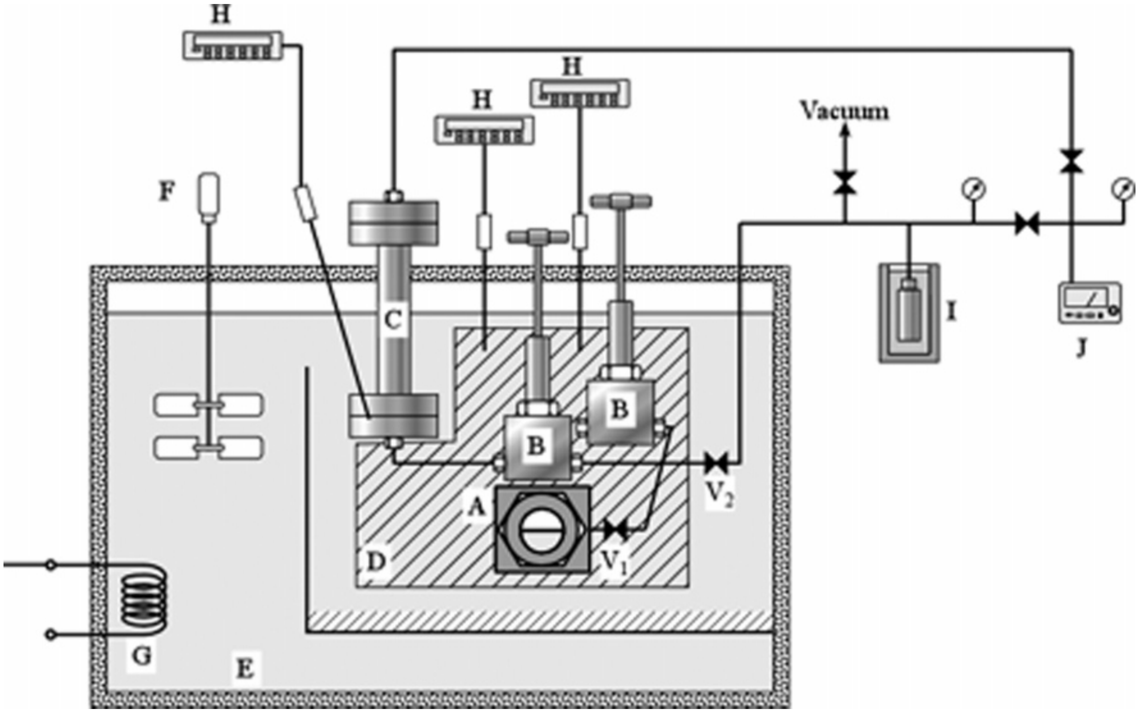

Figure 1. Experimental apparatus. A, Optical cell; B, variable-volume vessel; C, differential null-pressure detector; D, aluminum blocks; E, constant-temperature oil bath; F, impeller; G, temperature controller; H, platinum resistance thermometer; I, cold trap; J, quartz crystal pressure gauge; V_1, cutoff valve; and V_2, separation valve.

Table 1. Compounds Used in This Study

sample code	molecular formula	name	CAS RN	purity	T_b/K
HFE-245mc	$CF_3CF_2OCH_3$	pentafluoromethoxyethane	22410-44-2	99.99	279.05
HFC-125	CHF_2CF_3	pentafluoroethane	354-33-6	99.8	224.65
HFC-134a	CH_2FCF_3	1,1,1,2-tetrafluoroethane	811-97-2	99.99	246.60
HFC-227ea	CF_3CHFCF_3	1,1,1,2,3,3,3-heptafluoropropane	431-89-0	99.8	257.65
HFC-236ea	CHF_2CHFCF_3	1,1,1,2,3,3-hexafluoropropane	431-63-0	99.4	277.65

Table 2. Critical Parameters of HFE-245mc

	author	T_C/K	P_C/MPa	ρ_C/kg m^{-3}	reference
HFE-245mc	Sako et al.	406.80 ± 0.03	2.887 ± 0.002	499 ± 10	9
($CF_3CF_2OCH_3$)	Tsuge et al.	406.80 ± 0.03	2.8863 ± 0.0015		10
	Yoshii et al.	406.83 ± 0.017		509 ± 3	11
	this work	406.82 ± 0.01	2.885 ± 0.0005	491 ± 1	

the apparatus, (C) a differential null-pressure detector, and (D) aluminum blocks that acted as thermal masses to minimize temperature fluctuations. The optical cell was connected to the two variable-volume vessels and the differential null-pressure detector by a valve (V_1). The central axis of these vessels and the detector were adjusted to be at the same level. The temperature of the oil bath was controlled to within ±3 mK in the range of (300 to 450) K. Under these conditions, the uncertainty in the critical temperature is estimated to be ±10 mK. The uncertainties in pressure, density, and composition were estimated to be less than ±0.5 kPa, ±1 kg m^{-3}, and ±0.5% (molar base), respectively. The apparatus was specially designed for critical parameter measurements with small sample sizes and needs only (5 to 6) mL for a single experiment. A detailed description of the experimental apparatus was given in the previous papers.[6−8]

Procedure. To prepare the desired composition of mixtures for investigation, the apparatus shown in Figure 2 was used. From stainless steel sample cylinders A (containing sample 1) and B (containing sample 2), samples were transferred first to stainless steel cylinder C and then to glass cylinder D separately by using hot water and liquid nitrogen and then mixed. The initial composition of the sample mixture was calculated from the weight ratio of pure samples in cylinder D. The sample mixture in cylinder D was transferred to glass cylinder E and loaded into the optical cell situated in a constant-temperature air bath at 233 K under vacuum. After (80 to 90)% of the optical cell was filled with the liquid mixture, the cell was disconnected from the apparatus at valve V_1 and connected to the critical parameters measurement apparatus shown in Figure 1. A detailed description of the sample mixture preparation will be found in the literature.[8]

After the optical cell was connected to the main apparatus (Figure 1, at V_1), the remaining part of the apparatus was evacuated. Valve V_2 was closed, and the temperature was raised to the desired value. After the temperature fluctuations became less than ±5 mK, V_1 was opened to fill the apparatus. After the pressure fluctuations became less than ±0.1 kPa, they were recorded, V_1 was closed, and the temperature was raised in 10 K increment to near the critical temperature.

Near the critical temperature, when critical opalescence began to appear, the temperature increment was decreased from (10 to 5) mK. The density inside the optical cell was adjusted by the variable-volume vessels to give an equally strong critical opalescence in both the gas and liquid

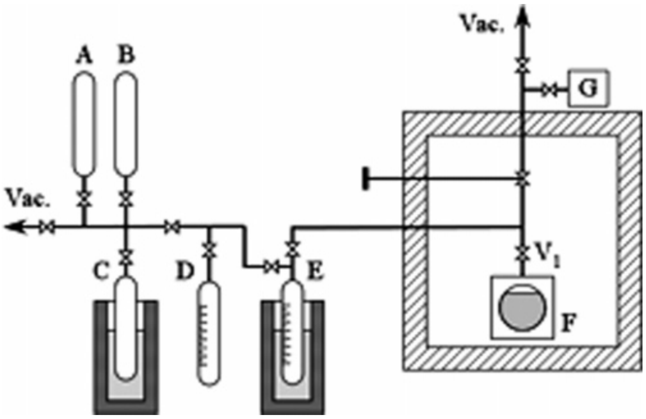

Figure 2. Apparatus for sample loading. A and B, Stainless steel cylinder for storing components 1 and 2; C, stainless steel cylinder for purification; D and E, glass cylinder for purification and volume measurement; F, optical cell; and G, pressure gauge.

phases, and the temperature and pressure were recorded. These procedures were repeated with a fine adjustment of the position of the meniscus until the temperature exceeded the critical temperature T_{CH} at which the meniscus disappeared, while keeping equally strong critical opalescence in both the gas and liquid phases. In all of the procedures described above, V_1 was always closed except for the time when the temperature fluctuation was smaller than 5 mK.

Once the temperature exceeds T_{CH}, the temperature is set to 5 K higher than T_{CH} and kept constant for (4 to 20) h with V_1 open to achieve identical compositions in the optical cell and the null-pressure detector. After the equilibration of the composition, the temperature was decreased slowly to observe the reappearance of the meniscus at temperature T_{CL}. Then at a temperature between T_{CH} and T_{CL} V_1 was closed, and another determination of the critical temperature was conducted. The critical pressure was calculated from a linear interpolation of the $p-T$ curve near the critical point. A detailed description of the experimental procedures can be found in previous work.[8]

After the critical parameters measurements, the samples inside the optical cell and the null-pressure detector were trapped in the cold trap (Figure 1, P) separately and weighed. The critical density was then determined from the mass of the sample and the known internal volume of the optical cell. The composition of the samples in the null-pressure detector and the optical cell were analyzed by gas chromatography (Hewlett-Packard, model HP-6890; thermal conductivity detector).

3. Results and Discussion

Critical parameters of HFE-245mc are summarized in Table 2 together with data by Yoshii et al.[9] and us.[10] Compared with the reported data, the critical temperature and pressure are almost identical. However, differences in the critical density values are rather large. These data were taken by expanding the excess amount of HFE-245mc into the expansion vessel to achieve the critical density. However, as described in the Experimental Section, our data were taken after cautiously adjusting the position of the meniscus with the variable-volume vessels. Therefore, we believe that the present data are more accurate than the previously reported results. Furthermore, the determination of the critical density is very difficult by its very nature. Near the critical point, the density changes greatly with only minute changes in temperature and/or pressure. To determine which value is correct, reports of other researchers should be awaited. Critical parameters of HFCs measured in this study and comparison with the literature data can be found in the literature.[8]

Figure 3 shows the critical loci of the four binary systems investigated in this study. Numerical values are tabulated in Tables 3−6. Similar to the case of the HFE-143m binary mixture systems previously reported,[8] the critical density data showed a wider spread compared with the critical temperature and pressure. The larger the difference in boiling point as well as critical temperature, the wider the data spread. It is presumably due to the same reason described above.

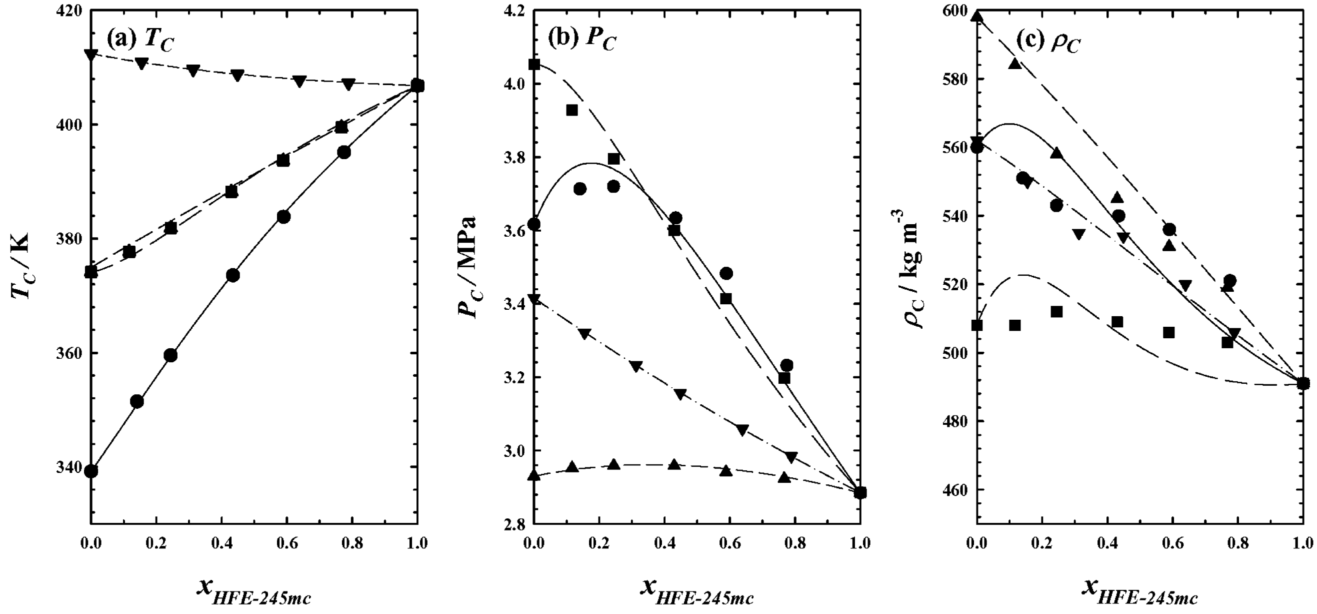

Figure 3. Critical parameters of the binary mixtures of HFE-245mc and HFCs. (a) Critical temperature, (b) critical pressure, and (c) critical density for ●, HFC-125; ■, HFC-134a; ▲, HFC-227ea; and ▼, HFC-236ea. Lines are the correlated results from eqs 1 to 6: —, HFC-125; – – –, HFC-134a; - - -, HFC-227ea; and —·—·—, HFC-236ea.

Table 3. Critical Parameters of HFE-245mc (1) + HFC-125 (2) Systems

x_1[a]	T_C/K	P_C/ MPa	ρ_C/ kg m^{-3}	x_1[b] (optical cell base deviation/%)[d]	x_1[c] (optical cell base deviation/%)[e]
0.0000	339.20	3.617	560	0.0000 (0.00)	0.0000 (0.00)
0.1408	351.46	3.714	551	0.1430 (−1.56)	0.1400 (0.57)
0.2438	359.55	3.720	543	0.2462 (−0.98)	0.2389 (2.01)
0.4346	373.59	3.634	540	0.4285 (1.40)	0.4309 (0.85)
0.5897	383.83	3.483	536	0.5914 (−0.29)	0.5878 (0.32)
0.7753	395.14	3.232	521	0.7723 (0.39)	0.7774 (−0.27)
1.0000	406.82	2.885	491	1.0000 (0.00)	1.0000 (0.00)
			AAD/%	(0.66)	(0.57)

[a] Mole fraction in the optical cell. [b] Mole fraction calculated from the weight ratio. [c] Mole fraction in the null-pressure detector and variable-volume vessels. [d] $100(x_{\text{optical cell}} - x_{\text{feed}})/x_{\text{optical cell}}$. [e] $100(x_{\text{optical cell}} - x_{\text{null pressure detector}})/x_{\text{optical cell}}$.

Table 4. Critical Parameters of HFE-245mc (1) + HFC-134a (2) Systems

x_1[a]	T_C/K	P_C/ MPa	ρ_C/ kg m^{-3}	x_1[b] (optical cell base deviation/%)[d]	x_1[c] (optical cell base deviation/%)[e]
0.0000	374.13	4.053	508	0.0000 (0.00)	0.0000 (0.00)
0.1167	377.73	3.929	508	0.1173 (−0.51)	0.1173 (−0.51)
0.2446	381.88	3.796	512	0.2444 (0.08)	0.2451 (−0.20)
0.4299	388.22	3.601	509	0.4139 (3.72)	0.4290 (0.21)
0.5886	393.69	3.414	506	0.5874 (0.20)	0.5838 (0.82)
0.7674	399.47	3.197	503	0.7610 (0.83)	0.7657 (0.22)
1.0000	406.82	2.885	491	1.0000 (0.00)	1.0000 (0.00)
			AAD/%	(0.76)	(0.28)

[a] Mole fraction in the optical cell. [b] Mole fraction calculated from the weight ratio. [c] Mole fraction in the null-pressure detector and variable-volume vessels. [d] $100(x_{\text{optical cell}} - x_{\text{feed}})/x_{\text{optical cell}}$. [e] $100(x_{\text{optical cell}} - x_{\text{null pressure detector}})/x_{\text{optical cell}}$.

Table 5. Critical Parameters of HFE-245mc (1) + HFC-227ea (2) Systems

x_1[a]	T_C/K	P_C/ MPa	ρ_C/ kg m^{-3}	x_1[b] (optical cell base deviation/%)[d]	x_1[c] (optical cell base deviation/%)[e]
0.0000	375.00	2.930	598	0.0000 (0.00)	0.0000 (0.00)
0.1823	381.10	2.952	584	0.1832 (−0.49)	0.1819 (0.22)
0.3060	385.17	2.959	558	0.3068 (−0.26)	0.3083 (−0.75)
0.4769	390.66	2.958	545	0.4794 (−0.52)	0.4783 (−0.29)
0.6944	397.47	2.941	531	0.7105 (−2.32)	0.6890 (0.78)
0.8132	401.10	2.923	519	0.8593 (−5.67)	0.8124 (0.10)
1.0000	406.82	2.885	491	1.0000 (0.00)	1.0000 (0.00)
			AAD/%	(1.32)	(0.31)

[a] Mole fraction in the optical cell. [b] Mole fraction calculated from the weight ratio. [c] Mole fraction in the null-pressure detector and variable-volume vessels. [d] $100(x_{\text{optical cell}} - x_{\text{feed}})/x_{\text{optical cell}}$. [e] $100(x_{\text{optical cell}} - x_{\text{null pressure detector}})/x_{\text{optical cell}}$.

From the tables, it is also clear that, as described in the Experimental Section, there are compositional differences between samples recovered from the optical cell and null-pressure detector. The larger the boiling point and critical temperature difference, the larger the average absolute deviation (AAD) of the composition. This fact suggests the existence of a small temperature difference between the optical cell and null-pressure detector that results in composition fluctuations. However, because the AADs are smaller than 1% for all four systems, there will be no effect on the critical parameters measurements. The tendency that the larger the difference in the normal boiling point the wider the AAD of the composition difference is similar to that observed with the HFE-143m binary mixture systems.[8]

Table 6. Critical Parameters of HFE-245mc (1) + HFC-236ea (2) Systems

x_1[a]	T_C/K	P_C/ MPa	ρ_C/ kg m^{-3}	x_1[b] (optical cell base deviation/%)[d]	x_1[c] (optical cell base deviation/%)[e]
0.0000	412.41	3.416	562	0.0000 (0.00)	0.0000 (0.00)
0.1546	410.97	3.322	550	0.1515 (2.01)	0.1546 (−0.00)
0.3124	409.73	3.233	535	0.3085 (1.25)	0.3121 (0.10)
0.4485	408.82	3.157	534	0.4427 (1.29)	0.4488 (−0.07)
0.6388	407.86	3.059	520	0.6317 (1.11)	0.6388 (−0.00)
0.7888	407.29	2.985	506	0.7826 (0.79)	0.7887 (0.01)
1.0000	406.82	2.885	491	1.0000 (0.00)	1.0000 (0.00)
			AAD/%	(0.92)	(0.03)

[a] Mole fraction in the optical cell. [b] Mole fraction calculated from the weight ratio. [c] Mole fraction in the null-pressure detector and variable-volume vessels. [d] $100(x_{\text{optical cell}} - x_{\text{feed}})/x_{\text{optical cell}}$. [e] $100(x_{\text{optical cell}} - x_{\text{null pressure detector}})/x_{\text{optical cell}}$.

Table 7. Correlated Results

system	parameters	AAD/%
HFE-245mc + HFC-125	$\Delta_T = -13.9042$	T_C: 0.07
	$\Delta_P = 0.9202$	P_C: 1.03
	$\Delta_V = -46.9519$	ρ_C: 1.72
HFE-245mc + HFC-134a	$\Delta_T = -13.7293$	T_C: 0.11
	$\Delta_P = 0.5782$	P_C: 0.93
	$\Delta_V = -53.9771$	ρ_C: 1.25
HFE-245mc + HFC-227ea	$\Delta_T = -1.4020$	T_C: 0.01
	$\Delta_P = 0.1026$	P_C: 0.04
	$\Delta_V = -5.5964$	ρ_C: 0.80
HFE-245mc + HFC-236ea	$\Delta_T = -1.4767$	T_C: 0.01
	$\Delta_P = 0.0239$	P_C: 0.01
	$\Delta_V = -7.1871$	ρ_C: 0.38

Higashi proposed the following equations to correlate the critical loci of binary HFC mixtures:[12,13]

$$T_{\text{Cm}} = \theta_1 T_{\text{C1}} + \theta_2 T_{\text{C2}} + 2\theta_1\theta_2\Delta_T \tag{1}$$

$$V_{\text{Cm}} = \theta_1 V_{\text{C1}} + \theta_2 V_{\text{C2}} + 2\theta_1\theta_2\Delta_V \tag{2}$$

$$P_{\text{Cm}} = \theta_1 P_{\text{C1}} + \theta_2 P_{\text{C2}} + 2\theta_1\theta_2\Delta_P \tag{3}$$

$$\rho_{\text{Cm}} = \frac{M_{\text{m}}}{V_{\text{Cm}}} \tag{4}$$

$$M_{\text{m}} = x_1 M_1 + (1 - x_1)M_2 \tag{5}$$

$$\theta_i = \frac{x_i {V_{\text{C}i}}^{2/3}}{\sum_{j=1}^{2} x_j {V_{\text{C}j}}^{2/3}} \qquad i = 1, 2 \tag{6}$$

where $T_{\text{C}i}$, $V_{\text{C}i}$, and $P_{\text{C}i}$ are the critical temperature, critical molar volume, and critical pressure of component i, respectively, x is the mole fraction, M_i is the molecular weight of component i, ϑ_i is the surface ratio of component i given by eq 6, and Δ_T, Δ_V, and Δ_P are the fitting parameters for critical temperature, volume, and pressure, respectively. Subscript m means mixture. Experimental data obtained in this study are correlated with the equations.

Correlated results are shown as lines in Figure 3 and summarized in Table 7. As shown in the Figure and Table, the AADs of the critical parameters are less than ±1 %. However, as mentioned above, the AADs of the critical density are much greater than that of the critical temperature.

4. Conclusions

We have measured the critical parameters of four binary systems—HFE-245mc + HFC-125, + HFC-134a, + HFC-

227ea, and + HFC-236ea with uncertainties of ±10 mK in temperature, ±0.5 kPa in pressure, ±1 kg m^{-3} in density, and ±0.5% in composition. The experimental data spread of the critical density was larger than that of the critical temperature and pressure presumably because of the difficulty in obtaining density measurements near the critical condition where the density changes sharply with only minor changes in temperature and pressure. Experimental results were correlated with equations proposed by Higashi. The maximum AAD was less than 1% for all critical parameters.

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