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An Assessment of Thermodynamic Models for HFC Refrigerant Mixtures Through the Critical-Point Calculation

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Abstract An assessment of thermodynamic models for HFC refrigerant mixtures based on Helmholtz energy equations of state was made through critical-point calculations for ternary and quaternary mixtures. The calculations were performed using critical-point criteria expressed in terms of the Helmholtz free energy. For three ternary mixtures: difluoromethane (R-32) + pentafluoroethane (R-125) + 1,1,1,2-tetrafluoroethane (R-134a), R-125 + R-134a + 1,1,1-trifluoroethane (R-143a), and carbon dioxide (CO_2) + R-32 + R-134a, and one quaternary mixture, R-32 + R-125 + R-134a + R-143a, calculated critical points were compared with experimental values, and the capability of the mixture models for representing the critical behavior was discussed.

Keywords Critical point · Mixture model · Multicomponent mixture · Helmholtz energy equation of state · Numerical calculation

1 Introduction

An assessment of thermodynamic models for hydrofluorocarbon (HFC) refrigerant mixtures based on Helmholtz energy equations of state is made through critical-point calculations for ternary or quaternary mixtures. Such assessments are rarely discussed in the literature. Since experimental critical point data are usually not used in developing mixture models, comparisons of the critical points calculated from a model with experimental values can be used to assess the capability of the model in the high-pressure region.

The mixture models discussed here can be used to estimate properties of multicomponent mixtures from those of their binary constituents. However, this approach can

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lead to a substantial error, because the properties of multicomponent mixtures are not necessarily the sum of those of the binary subsystems. In particular, the error becomes larger near the critical point. An assessment of calculated critical points highlights the magnitude of the error.

This work attempts to determine the critical points of three ternary refrigerant mixtures: difluoromethane (R-32) + pentafluoroethane (R-125) + 1,1,1-trifluoroethane (R-134a), R-125 + R-134a + 1,1,1,2-tetrafluoroethane (R-143a), and carbon dioxide (CO_2) + R-32 + R-134a, and one quaternary refrigerant mixture: R-32 + R-125 + R-134a + R-143a. Blends of refrigerants of R-32, R-125, and R-134a are used as R-407 for replacing chlorodifluoromethane (R-22), and those of R-125, R-134a, and R-143a are used as replacements for R-502. Blends of HFCs and low-GWP and nonflammable substances such as CO_2 are promising [1] because HFCs have a far from negligible GWP and flammability.

Several mixture models have been developed for these HFC mixtures. Lemmon and Jacobsen [2] developed a model for binary HFC mixtures including R-32, R-125, R-134a, and R-143a. This model (LJ model) can be extended to multicomponent HFC mixtures as the sum of their binary constituents. Lemmon and Jacobsen [3] also developed a generalized mixture model (GLJ model) applicable to a wide variety of binary mixtures containing CO_2, cryogens, and hydrocarbons. The GLJ model, which is applied only to systems where experimental data are not available, can also be extended to multicomponent mixtures. The GLJ model can be coupled with the LJ model, and these models can be applied to multicomponent mixtures including HFCs, CO_2, cryogens, and hydrocarbons, because the GLJ model and LJ model use a common equation to determine the reducing parameters required for calculations of residual contributions and excess contributions to the Helmholtz free energy.

On the other hand, Tillner-Roth et al. [4] developed a different mixture model for HFC mixtures. The model (TR model) is also applicable to multicomponent HFC mixtures. In particular, this model has a special ternary parameter for R-32 + R-125 + R-134a mixtures to obtain better agreement with experimental values. However, the use of this model in combination with the LJ model or GLJ model may not be permitted because the model uses a different equation to determine the reducing parameters.

The equations of state for the pure components used in each mixture model are tabulated in Table 1. All equations of state are explicit in the Helmholtz free energy. For R-125 and R-143a, the LJ model and TR model incorporate different equations of state.

Table 1 Pure-component equations of state used in mixture models

Component	Mixture model	Equation of state
R-32	LJ, TR	Tillner-Roth and Yokozeki [5]
R-125	LJ	Lemmon and Jacobsen [6]
	TR	Sunaga et al. [7]
R-134a	LJ, TR	Tillner-Roth and Baehr [8]
R-143a	LJ	Lemmon and Jacobsen [9]
	TR	Li et al. [10]
CO_2	GLJ	Span and Wagner [11]

2 Calculation Method

Many calculations of the critical point of mixtures using cubic equations of state have been successfully attempted over the past three decades. However, few attempts employing Helmholtz energy equations of state have been reported. Lemmon [12] described a method applying Helmholtz energy equations to the critical-point criteria expressed in terms of the Gibbs free energy. Using this method, Lemmon et al. [13] calculated the critical points of nitrogen + oxygen, nitrogen + argon, and oxygen + argon binary mixtures. Tillner-Roth et al. [4] reported the calculation results for the critical points of binary and ternary hydrofluorocarbon (HFC) mixtures. Tillner-Roth et al. also used the criteria expressed in terms of the Gibbs free energy. REFPROP [14], which is widely used software for calculating reference fluid thermodynamic and transport properties, can be used to estimate the critical point of mixtures. However, the values for binary mixtures come indirectly from ancillary equations and are estimated for multicomponent mixtures. For rapid calculations, REFPROP uses correlations for the binary critical curves that are based on full calculations from the equations of state.

This work uses the critical-point criteria expressed in terms of the Helmholtz free energy presented by Akasaka [15]. The criteria for binary mixtures are

$$L = \begin{vmatrix} A_{11} & A_{12} \\ A_{12} & A_{22} \end{vmatrix} = 0 \tag{1}$$

and

$$M = \begin{vmatrix} A_{11} & A_{12} \\ \frac{\partial L}{\partial N_1} & \frac{\partial L}{\partial N_2} \end{vmatrix} = 0, \tag{2}$$

where A is the Helmholtz free energy, N_i is the mole number of the ith component, and A_{11}, A_{22}, and A_{12} are the second-order partial derivatives of A with respect to mole numbers, which are defined as

$$A_{11} = \left(\frac{\partial^2 A}{\partial N_1^2}\right)_{T,V,N_2}, \quad A_{22} = \left(\frac{\partial^2 A}{\partial N_2^2}\right)_{T,V,N_1}, \quad \text{and } A_{12} = \left(\frac{\partial^2 A}{\partial N_1 \partial N_2}\right)_{T,V}.$$

All derivatives appearing in the criteria can be calculated directly from mixture models for Helmholtz energy equations of state. Equations 1 and 2 can be readily extended to multicomponent mixtures. The criteria for n-component mixtures are

$$L = \begin{vmatrix} A_{11} & A_{12} & \cdots & A_{1,n} \\ A_{21} & A_{22} & \cdots & A_{2,n} \\ \vdots & \vdots & \ddots & \vdots \\ A_{n,1} & A_{n,2} & \cdots & A_{n,n} \end{vmatrix} = 0 \tag{3}$$

and

$$M = \begin{vmatrix} A_{11} & A_{12} & \cdots & A_{1,n} \\ A_{21} & A_{22} & \cdots & A_{2,n} \\ \vdots & \vdots & \ddots & \vdots \\ A_{n-1,1} & A_{n-1,2} & \cdots & A_{n-1,n} \\ L_1 & L_2 & \cdots & L_n \end{vmatrix} = 0, \tag{4}$$

where

$$A_{i,j} = \left(\frac{\partial^2 A}{\partial N_i \partial N_j}\right)_{T,V} \quad \text{and} \quad L_i = \left(\frac{\partial L}{\partial N_i}\right)_{T,V,N_{j\neq i}}.$$

The notation $N_{j\neq i}$ indicates that all mole numbers except N_i are held constant. The derivatives appearing in Eqs. 3 and 4 should be calculated numerically, because analytical derivations of the derivatives are extremely involved. Using a seven-point formula, A_i and $A_{i,j}$ can be numerically estimated as

$$A_i = \left(\frac{\partial A}{\partial N_i}\right)_{T,V,N_{j\neq i}} = \frac{\Delta A}{60\Delta N}$$

and

$$A_{i,j} = \left(\frac{\partial A_j}{\partial N_i}\right)_{T,V,N_{j\neq i}} = \frac{\Delta A_j}{60\Delta N},$$

where ΔN is the step size for mole numbers, and ΔA and ΔA_j are calculated as

$$\begin{aligned}\Delta X = {} & 45X[N_i + \Delta N] - 9X[N_i + 2\Delta N] + X[N_i + 3\Delta N] \\ & -45[N_i - \Delta N] + 9X[N_i - 2\Delta N] - X[N_i - 3\Delta N],\end{aligned}$$

where X stands for A or A_j, and

$$X[N_i + k\Delta N] = X(T, V, N_1, N_2, \ldots, N_i + k\Delta N, \ldots, N_n).$$

This study set the size for ΔN to 10^{-3} mole.

A simple Newton–Raphson method was used to find a root of the simultaneous equations given by Eqs. 3 and 4. This method improves an approximation of T_c and V_c according to the equations,

$$T_\mathrm{c}^{(k+1)} = T_\mathrm{c}^{(k)} - \frac{1}{\Delta}\left[L\left(\frac{\partial M}{\partial V}\right)_{T,N_i} - M\left(\frac{\partial L}{\partial V}\right)_{T,N_i}\right] \tag{5}$$

and

$$V_\mathrm{c}^{(k+1)} = V_\mathrm{c}^{(k)} - \frac{1}{\Delta}\left[L\left(\frac{\partial M}{\partial T}\right)_{V,N_i} - M\left(\frac{\partial L}{\partial T}\right)_{V,N_i}\right], \tag{6}$$

where

$$\Delta = \left(\frac{\partial L}{\partial T}\right)_{V,N_i}\left(\frac{\partial M}{\partial V}\right)_{T,N_i} - \left(\frac{\partial L}{\partial V}\right)_{T,N_i}\left(\frac{\partial M}{\partial T}\right)_{V,N_i}.$$

Although analytical derivations of the derivatives appearing in Eqs. 5 and 6 are possible, they are very complicated for multicomponent mixtures. Numerical differentiation gives sufficiently accurate values for the derivatives. The step sizes for T and V were set to $10^{-5}T$ and $10^{-4}V$, respectively. Weighted averages of the critical temperatures and molar volumes of the pure components were used as initial estimates. If the iteration starting from the weighted averages failed, the reducing parameters or experimental values were used as initial estimates. The Euclidean norm was used for the convergence condition. The iteration continues until the condition,

$$\sqrt{L^2 + M^2} < \varepsilon$$

is satisfied. The convergence tolerance ε was set to 10^{-6}. Most calculations converged within 20 iterations.

The stability test is obligatory after a root is found, because some roots may not correspond to the global minimization of free energy. This work performed the test using the following conditions presented by Sadus [16]:

$$\left(\frac{\partial p}{\partial \underline{V}}\right)_T < 0, \tag{7}$$

$$\left(\frac{\partial^2 \underline{A}}{\partial x_1^2}\right)_{T,V}\left(\frac{\partial^2 \underline{A}}{\partial \underline{V}^2}\right)_T - \left(\frac{\partial^2 \underline{A}}{\partial x_1 \partial \underline{V}}\right)^2 > 0, \tag{8}$$

and

$$\left(\frac{\partial^2 \underline{A}}{\partial x_1^2}\right)_{T,V} > 0, \tag{9}$$

where p is the pressure, $\underline{V}$ is the molar volume, $\underline{A}$ is the molar Helmholtz free energy, and x_1 is the mole fraction of the first component.

3 Results and Discussion

3.1 R-32 + R-125 + R-134a Mixtures

Akasaka [15] investigated the capabilities of the LJ model and TR model for R-32 + R-125 and R-125 + R-134a mixtures. The LJ model can well represent the critical behavior of both binary mixtures. The TR model can predict successfully the critical curve of R-32 + R-125 mixtures. However, the model shows less capability for R-125 + R-134a mixtures. To complete the calculations for three binary constituents of R-32 + R-125 + R-134a mixtures, the critical point of R-32 + R-134a mixtures was calculated in this work. Figure 1 shows the results with experimental values by Kordikowski et al. [17], Nagel and Bier [18], and Higashi [19]. Both models can

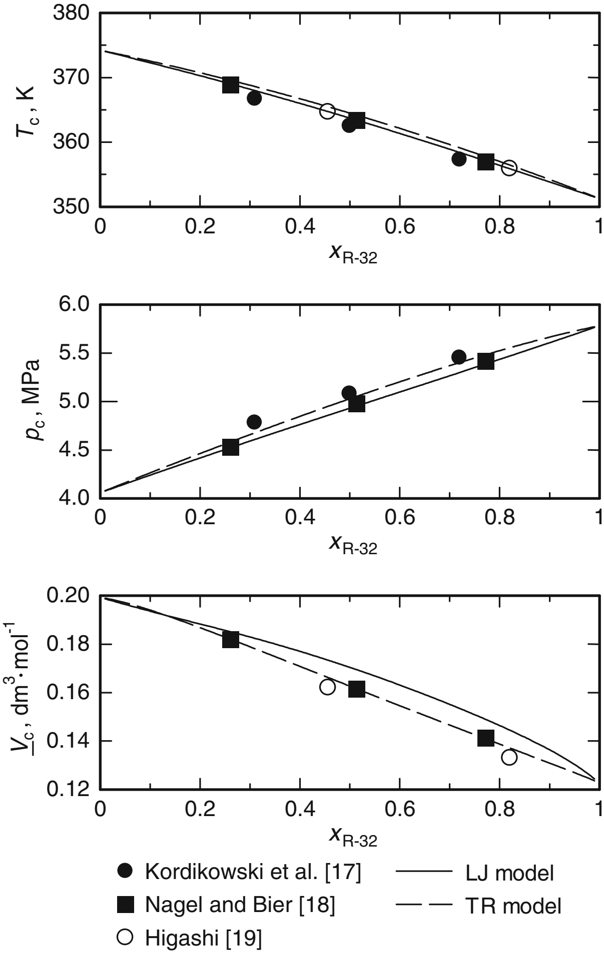

Fig. 1 Critical temperatures, pressures, and molar volumes of R-32 + R-134a mixtures calculated using the LJ model and TR model

Table 2 Mole fractions of R-32 (1) + R-125 (2) + R-134a (3) mixtures investigated experimentally

Mixture	Author(s)	x_1	x_2	x_3	Designation
(a)	Nagel and Bier [18]	0.4295	0.0868	0.4837	
(b)	Nagel and Bier [18]	0.3349	0.3292	0.3359	
(c)	Nagel and Bier [18]	0.1938	0.6072	0.1990	
(d)	Higashi [20]	0.3464	0.3003	0.3533	R-407A
(e)	Higashi [20]	0.3811	0.1796	0.4393	R-407C
(f)	Higashi [20]	0.4025	0.1047	0.4928	R-407E

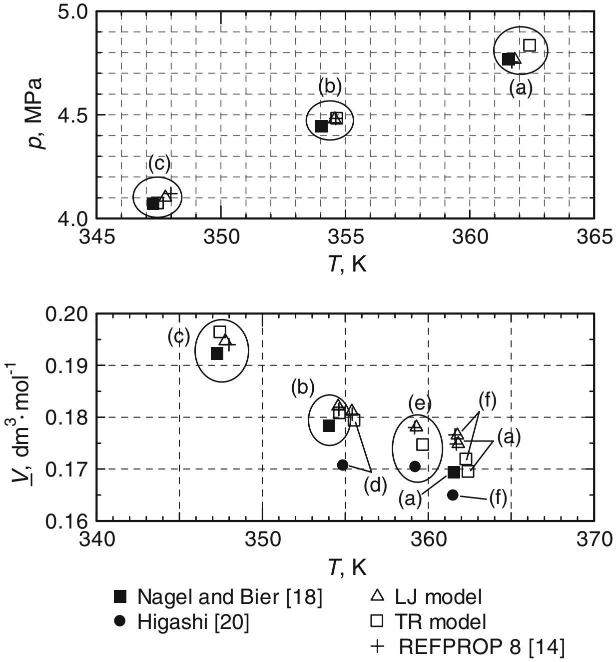

Fig. 2 Experimental and calculated values for the critical temperatures, pressures, and molar volumes of R-32 + R-125 + R-134a mixtures

predict T_c and p_c reasonably. The TR model is superior to the LJ model in the prediction of $\underline{V}_c$.

Table 2 lists mole fractions of the ternary mixtures investigated experimentally by Nagel and Bier [18] and Higashi [20]. The critical parameters of six mixtures with different compositions are the only ones that so far have been measured. All experimental data were obtained from the direct observation of the disappearance or reappearance of the meniscus. The critical points calculated using the LJ model and TR model for the six mixtures are plotted on $T - p$ and $T - \underline{V}$ diagrams in Fig. 2, along with the experimental values. On the diagrams, the same composition data are circled by dotted circles or indicated by solid lines. For comparison, the figure includes estimations from REFPROP.

Both the LJ model and TR model can predict T_c and p_c with high accuracies. The deviations are almost within 0.2 % for T_c and 0.8 % for p_c. On the other hand, the predictions for $\underline{V}_\mathrm{c}$ show larger deviations, which are about 3 % and sometimes exceed 5 %. However, in general, the experimental error in measurements of $\underline{V}_\mathrm{c}$ is considerably larger than that in T_c or p_c. If direct observation is used for an experimental determination of the critical point, the experimental error in $\underline{V}_\mathrm{c}$ can be estimated to be at least ±2 % [18]. Therefore, the predictions for $\underline{V}_\mathrm{c}$ by the LJ model and TR model are reasonable. It can be concluded that both the LJ model and TR model are capable of predicting the critical points of the ternary mixtures. For the ternary mixture with the special parameter, the $\underline{V}_\mathrm{c}$ predictions by the TR model are more accurate than those by the LJ model.

3.2 R-125 + R-134a + R-143a Mixtures

Akasaka [15] investigated the capabilities of the LJ model and TR model for R-125 + R-143a mixtures. Both models can be used to predict T_c of the mixtures accurately. Since experimental data for $\underline{V}_\mathrm{c}$ of the mixtures are limited and scattered, an assessment of the predictions for $\underline{V}_\mathrm{c}$ was not made. This work calculated the critical points of R-134a + R-143a mixtures to complete the calculations for three binary constituents of R-125 + R-134a + R-143a mixtures. Figure 3 shows the results and experimental values by Nagel and Bier [21] and Y. Higashi, Private communication. The predictions for T_c, p_c, and $\underline{V}_\mathrm{c}$ by the LJ model show good agreement with experimental values. The TR model can predict T_c and p_c well, but the predictions for $\underline{V}_\mathrm{c}$ deviate from the experimental values.

Table 3 lists mole fractions of the ternary mixtures investigated experimentally by Nagel and Bier [21] and Fujiwara et al. [22]. Only two mixtures have been investigated. The critical points calculated using the LJ model and TR model are plotted on $T-p$ and $T-\underline{V}$ diagrams in Fig. 4. The critical points calculated using the LJ model show excellent agreement with the experimental values. The maximum deviations are 0.2 % for T_c, 1.2 % for p_c, and 1.3 % for $\underline{V}_\mathrm{c}$. The TR model can predict T_c and p_c accurately. Although the predictions for $\underline{V}_\mathrm{c}$ by the TR model deviate from the experimental values, the deviations are acceptable. Therefore, both models can represent well the critical behavior of the ternary mixtures.

3.3 CO_2 + R-32 + R-134a Mixtures

The GLJ model uses three adjustable parameters to represent characteristics of a binary mixture. These parameters, ζ_{ij}, ξ_{ij}, and F_{ij} are usually determined by fitting experimental data. The parameters ζ_{ij} and ξ_{ij} modify the reducing parameters and have a great effect on VLE and $p-V-T$ properties. The parameter F_{ij} determines the contribution to the Helmholtz free energy from mixing. Lemmon and Jacobsen [3] presented recommended sets of these parameters for various binary mixtures.

Unfortunately, a recommended parameter set is not available for CO_2 + R-32 mixtures and CO_2 + R-134a mixtures. This work determined parameter sets by fitting available experimental data for CO_2 + R-32 mixtures [1] and for CO_2 + R-134a mix-

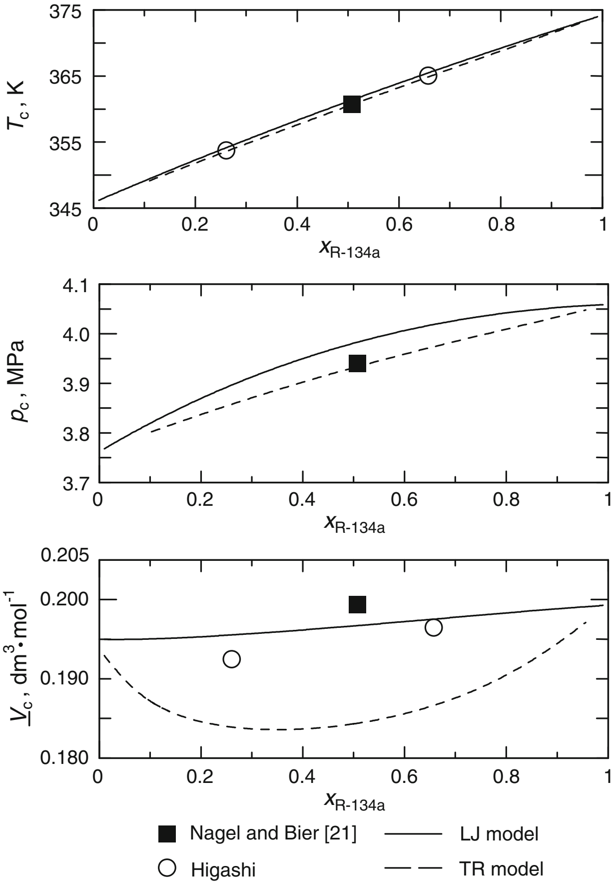

Fig. 3 Critical temperatures, pressures, and molar volumes of R-134a + R-143a mixtures calculated using the LJ model and TR model

Table 3 Mole fractions of R-125 (1) + R-134a (2) + R-143a (3) mixtures investigated experimentally

Mixture	Author(s)	x_1	x_2	x_3	Designation
(a)	Nagel and Bier [21]	0.1689	0.6684	0.1627	
(b)	Fujiwara et al. [22]	0.3578	0.0383	0.6039	R-404A

tures [23]. Table 4 shows parameter sets determined in this work, as well as those built in REFPROP. For both mixtures, this work set nonzero values to the parameters ξ_{ij} and F_{ij}. Figures 5 and 6 show T_c and p_c calculated using these different two-parameter sets with experimental values by Kordikowski et al. [17,24]. Clearly, for CO_2 + R-32 mixtures the parameter set determined in this work has an advantage in the predictions for T_c and p_c over the parameter set built in REFPROP. For CO_2 + R-134a mixtures, the parameter set determined in this work improves the accuracy of T_c over the whole range of composition, and that of p_c in CO_2-rich compositions.

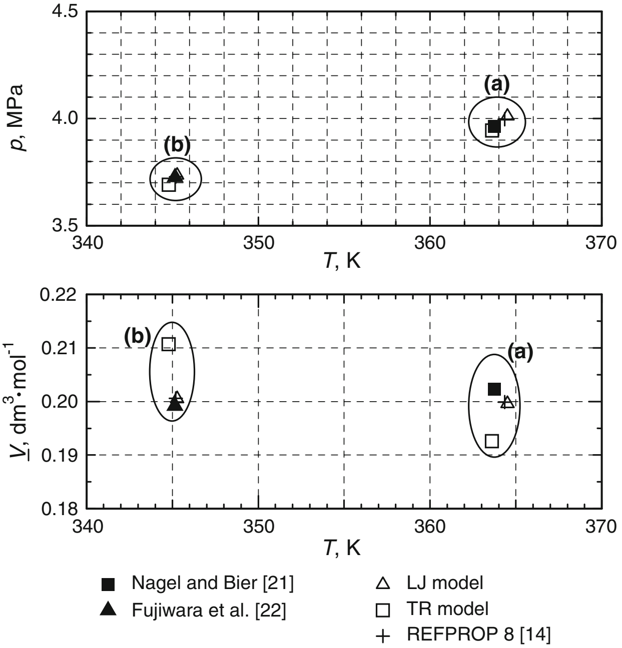

Fig. 4 Experimental and calculated values for the critical temperatures, pressures, and molar volumes of R-125 + R-134a + R-143a mixtures

Table 4 GLJ model parameter sets determined in this work and those built in REFPROP for CO_2 + R-32 and CO_2 + R-134a mixtures

	ζ_{ij} (K)	ξ_{ij} ($m^3 \cdot mol^{-1}$)	F_{ij} (−)
CO_2 + R-32			
This work	−8.175	-1.9161×10^{-5}	0.3697
REFPROP	−3.120	0	0
CO_2 + R-134a			
This work	−5.599	-7.9932×10^{-6}	0.3690
REFPROP	1.293	0	0

Kordikowski et al. [17] reported experimental data for the critical points of the ternary mixtures with nine different compositions. They used the acoustic method, which is not constrained by the subjectivity of the experimenter. Table 5 lists the mole fractions of the mixtures investigated. The compositions of the mixtures are classified into the following three groups: the first group of mixtures, (a), (b), and (c), contain equimolar amounts of R-32 and R-134a, and the second group of mixtures, (d), (e), and (f), contain larger amounts of R-32 than R-134a. The final group of mixtures, (g), (h), and (i), contain larger amounts of R-134a than R-32. For all mixtures, CO_2 is more abundant, and its mole fraction varies from 0.60 to 0.84.

The critical points of these mixtures calculated using the LJ model and GLJ model are plotted on $T-p$ diagrams in Fig. 7. On the diagrams, the same composition data

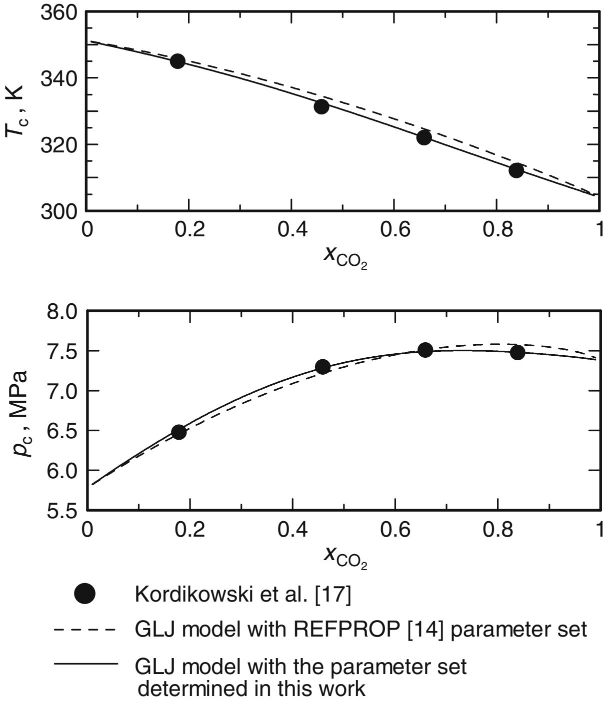

Fig. 5 Critical temperatures and pressures of CO_2 + R-32 mixtures calculated using the GLJ model

are connected with solid, dotted, and dotted-dashed lines. The parameter set determined in this work improves the prediction accuracies for T_c and p_c of all mixtures except for the mixture (g). The most successful predictions for T_c are obtained from the second group of mixtures, and the most accurate predictions for p_c are taken from the first group of mixtures. The prediction accuracies for T_c and p_c of the final group of mixtures are inferior to the first and second group mixtures. This suggests that the parameter set for CO_2 + R-134a mixtures used here is less optimized than that for CO_2 + R-32. The REFPROP estimations for T_c are in good agreement with the experimental values, but the estimations for p_c deviate considerably.

3.4 R-32 + R-125 + R-134a + R-143a Mixtures

The quaternary mixtures have six binary constituents. Lemmon and Jacobsen [3] give recommended parameter sets for the LJ model except for the R-32 + R-143a mixture. The prior calculations have shown that each of the parameter sets is valid for the representation of critical behavior of the corresponding binary constituents. Although a recommended parameter set for R-32 + R-143a mixtures has not been presented in the literature, an estimated parameter set is available in REFPROP. This work incorporated the REFPROP parameter set without any modification. No further optimization was done because experimental data for VLE or $p-V-T$ properties are not available

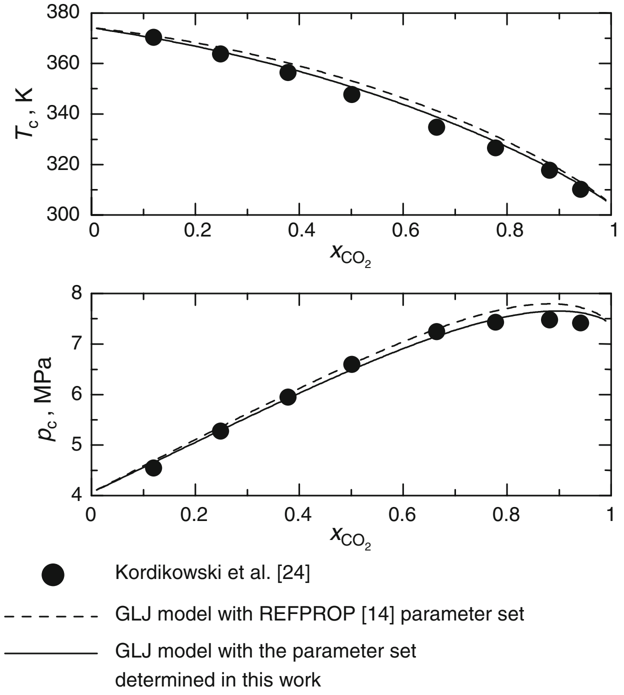

Fig. 6 Critical temperatures and pressures of CO_2 + R-134a mixtures calculated using the GLJ model

for R-32 + R-143a mixtures. Fröba et al. [25] reported one experimental value for T_c of the quaternary mixtures determined by the dynamic light scattering method. Table 6 lists mole fractions of the mixture investigated, as well as experimental and calculated values for T_c. The calculated value shows good agreement with the experimental value. To further assess the critical behavior of the LJ model for the quaternary mixtures, other experimental data with different compositions are desired.

Table 5 Mole fractions of CO_2 (1) + R-32 (2) + R-134a (3) mixtures investigated experimentally

Mixture	x_1	x_2	x_3
$x_1 > x_2 = x_3$			
(a)	0.66	0.17	0.17
(b)	0.76	0.12	0.12
(c)	0.80	0.10	0.10
$x_1 > x_2 > x_3$			
(d)	0.70	0.22	0.08
(e)	0.76	0.17	0.07
(f)	0.84	0.12	0.04
$x_1 > x_3 > x_2$			
(g)	0.60	0.12	0.28
(h)	0.70	0.09	0.21
(i)	0.81	0.06	0.13

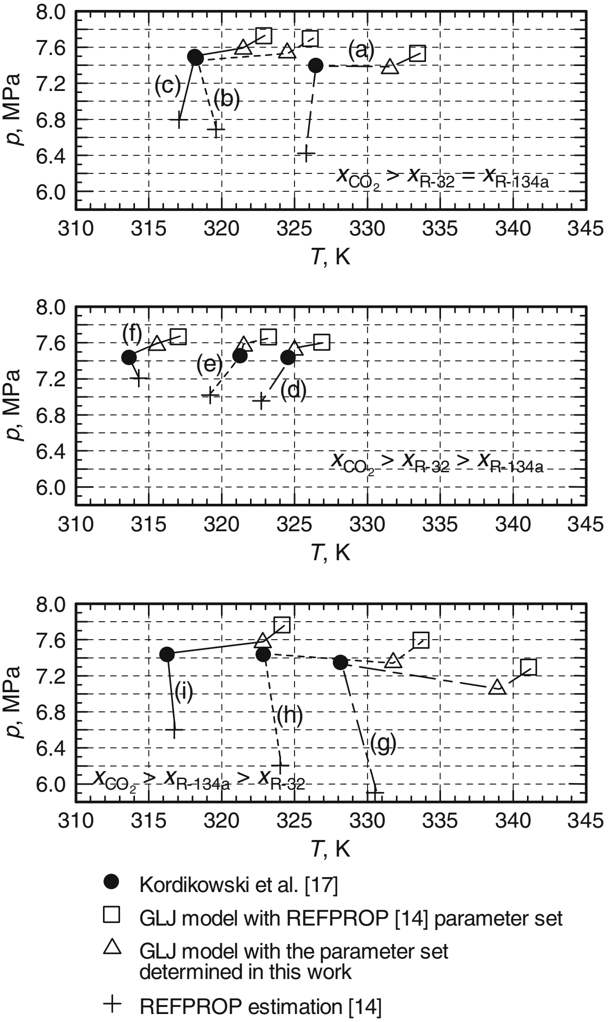

Fig. 7 Experimental and calculated values for the critical temperatures and pressures of CO_2 + R-32 + R-134a mixtures

Table 6 Experimental and calculated values for the critical temperature of R-32 (1) + R-125 (2) + R-134a (3) + R-143a (4) mixtures

	Mole fraction of investigated mixture			
Author	x_1	x_2	x_3	x_4
Fröba et al. [25]	0.243	0.210	0.253	0.294
	Experimental and calculated critical temperature (K)			
	Experimental	LJ model	REFPROP	
	352.05	351.08	351.46	

4 Conclusion

This paper assessed the capability of the LJ, GLJ, and TR models through critical-point calculations for three ternary and one quaternary HFC mixtures.

For R-32 + R-125 + R-134a mixtures and R-125 + R-134a + R143a mixtures, the LJ model and TR model can predict the critical point parameters reasonably well. Deviations of predicted critical temperatures and pressures from experimental values are within 1 %. For R-32 + R-125 + R-134a + R-143a mixtures, the predicted critical temperatures by the LJ model agree with experimental values within 0.3 %. For CO_2 + R-32 + R-134a mixtures, the GLJ model coupled with the LJ model shows sufficiently accurate predictions for the critical temperature and pressure. If more optimized parameter sets of the GLJ model for the CO_2 + R-32 and CO_2 + R-134a mixtures are used, it can be expected that the predictions become more accurate.

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