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The Status of Thermodynamic Data and Models for CF_3I and its Mixtures ⋆

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Abstract

There is a renewed interest in the use of the refrigerant CF_3I for refrigeration and air-conditioning applications, driven by its low global warming potential (similar to that of CO_2), low (but nonzero) ozone depletion potential, and its ability to suppress flammability. In this paper the existing thermodynamic data for this fluid are summarized. Furthermore, as CF_3I has been proposed as a component in mixtures with other refrigerants, the emphasis of this paper is on the survey of existing mixture data and an assessment of the suitability of existing thermodynamic modeling approaches and estimation schemes to model the properties of these mixtures. For mixtures where sufficient data exist, interaction parameters have been optimized.

1 Introduction

The compound CF_3I, or trifluoroiodomethane is a plausible refrigerant (named R-13I1 according to the ASHRAE 34 standard (ASHRAE, 2019c), or alternatively FIC-13I1) with three fluorines, one iodine, and one carbon, in a tetrahedral structure as shown in Fig. 1. The long and weak carbon-iodine bond makes the molecule unstable, which is beneficial from the standpoint of global warming potential (its atmospheric lifetime is on the order of days; see Table A-1 in WMO (2018)), but the presence of the iodine means that the molecule has a non-zero ozone depletion potential (Solomon et al., 1994; Zhang et al., 2020), a fact that has historically eliminated it from consideration in many analyses. A review of the evolution of refrigerants and refrigerant blends are discussed in a recent work McLinden and Huber (2020 (accepted).

The presence of a large number of halogens (F and I) in the molecule means that it can also serve as a good flame suppressant (Linteris et al., 2019), and there have been a number of studies that have considered the flammability of CF_3I-containing mixtures (Yang et al., 2015, 2012; Ural, 2003; Dlugogorski et al., 2002), and in particular, the ability of CF_3I to decrease the flammability of otherwise flammable refrigerant blends. CF_3I has also been proposed as more general flame suppressant (Yang et al., 1997) and as an electrical insulator to replace SF_6 (Zhao et al., 2017).

There are a few critiques of CF_3I as a working fluid that have been frequently leveled: 1) the molecule is not very stable 2) it is perhaps mildly toxic 3) it has a non-zero ozone depletion potential.

With regards to the first critique, experiments (Calderazzi and Colonna di Paliano, 1997) have indeed found that neat CF_3I is much less stable than other refrigerants, but it has been shown that its stability can be improved with the addition of stabilizers (phenols and epoxides) (Wilson et al., 2006), perhaps to a level that makes mixtures with CF_3I feasible.

The second critique pertains to toxicity. A study from the year 2000 found some inhalation sensitivity (Vinegar et al., 2000), but an exhaustive National Academies of Science report (NAS, 2004) concluded that "Overall, the toxicity of CF_3I is low", but reinforced the existing ruling that CF_3I was only to be used in unoccupied spaces. Nevertheless, in a 2019 addendum, CF_3I was added to ASHRAE Standard 34 with an A1 classification (ASHRAE, 2019a), indicating that the fluid was deemed to be of low

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toxicity and nonflammable. In a subsequent addendum (ASHRAE, 2019b), the CF_3I-containing R-466A blend (R-32/125/13I1 with mass composition 49.0/11.5/39.5) was added to the standard with an A1 classification.

The third critique pertains to the ozone depletion potential of the molecule. There are widely differing values reported for the ODP of CF_3I. Solomon et al. (1994) indicate that: "It is highly probable that the steady-state ozone depletion potential (ODP) of CF_3I for surface releases is less than 0.008 and more likely below 0.0001." The World Meteorological Organization (2018), on the other hand, lists the ODP of CF_3I as <0.09, with the note that this is an "upper limit reported by Brioude et al. (2010)" and "The derived ODPs…were shown to be strongly dependent on the region and season of the substance emission with the greatest values obtained for emissions in the Indian subcontinent". For reference, the currently phased out HCFC refrigerants have ODP that are on the order of the WMO's upper bound (Calm and Hourahan, 2007) on CF_3I's ODP (R-22: 0.050, R-142b: 0.070, etc.).

A recent analysis by Zhang et al. (2020) concluded that the very short atmospheric lifetime of CF_3I limited its transport to the stratosphere and that "almost all ozone depletion caused by CF_3I and CH_3I emissions occurs in the troposphere." The "conventional" value of ODP, i.e., the depletion of total column ozone, was calculated to be 0.019, but the depletion of stratospheric ozone was essentially zero. Those calculations were based on an "updated representation of tropospheric iodine chemistry" that considered additional reactions with iodine in the atmosphere and thus resulted in higher values of ODP compared to earlier studies. They found that the "the maximum tropospheric column ozone depletion by CF_3I …can reach 7% …on a regional basis" and were "mostly constrained to the lower troposphere in the Northern Hemisphere." Ozone in the troposphere also absorbs UV radiation, but it has negative health effects and is considered a pollutant. Zhang et al. (2020) propose a new metric, the "stratospheric ozone depletion potential (SODP)" that considers only the effects on stratospheric ozone and not tropospheric ozone. By this metric, the SODP of CF_3I is essentially zero.

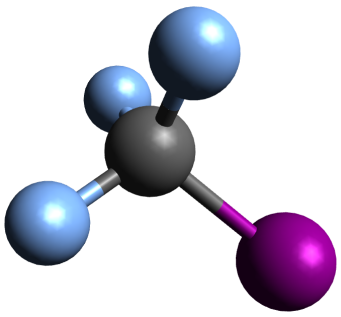

Fig. 1 3D structure of CF_3I from Avogadro (Hanwell et al., 2012)

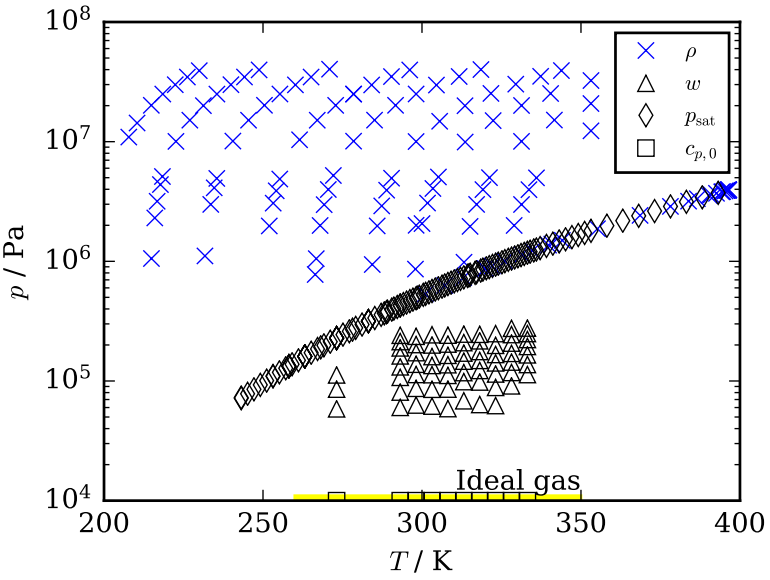

Fig. 2 A coverage diagram showing published data for CF_3I

Table 1 presents some characteristic values for CF_3I to help to orient the reader. Some values were taken from the equation of state of Lemmon and Span (2015), and others were taken from the literature. The unique identifiers are consistent with the values obtained from the NIST Chemistry Webbook.

Table 1 Characteristic values for CF_3I (GWP: Global warming potential; ODP: ozone depletion potential)

Parameter	Value	Reference
CAS	2314-97-8	
Standard InChI string	1S/CF3I/c2-1(3,4)5	
ASHRAE 34 Name	R-13I1	ASHRAE (2019c)
ASHRAE 34 Safety Class.	A1	ASHRAE (2019c)
GWP (100-year)	<1	Wuebbles (1995)
ODP upper limit	<0.09	Table A-1 of WMO (2018)
ODP	0.019	Zhang et al. (2020) (see above discussion)
SODP†	≈0	Zhang et al. (2020)
Normal boiling point	251.291 K	Lemmon and Span (2015); from REFPROP 10.0 (Lemmon et al., 2018)
Critical temp	396.44 K	Lemmon and Span (2015); from REFPROP 10.0 (Lemmon et al., 2018)

†: Stratospheric Ozone Depletion Potential as defined by Zhang et al. (2020); see discussion in text.

2 Pure Fluid Data and Models

A multiparameter equation of state for CF_3I was presented as an appendix to Lemmon and Span (2015). This equation of state was based upon a rather small body of experimental data from only a few research groups. Figure 2 shows the location of the existing experimental data in a $\log(p)$-T plane. This figure demonstrates the generally sparse coverage of the phase diagram. There are no overlapping datasets (important to assess reliability of the data). Therefore, a large part of the process of developing the equation of state for CF_3I came down to the experience of the equation of state developer, rather than just a straightforward fitting of existing experimental data points. The operator's expertise is needed to ensure reasonable extrapolation behavior in regions where no data exist.

This equation of state accurately represents the existing data (usually within fractions of a percent). Deviation plots between the presently available experimental data (exp) and the thermodynamic models implemented in NIST REFPROP 10.0 (RP) are shown in Fig. 3. The largest discrepancies are found for the saturated densities in the high-temperature region for T > 350 K.

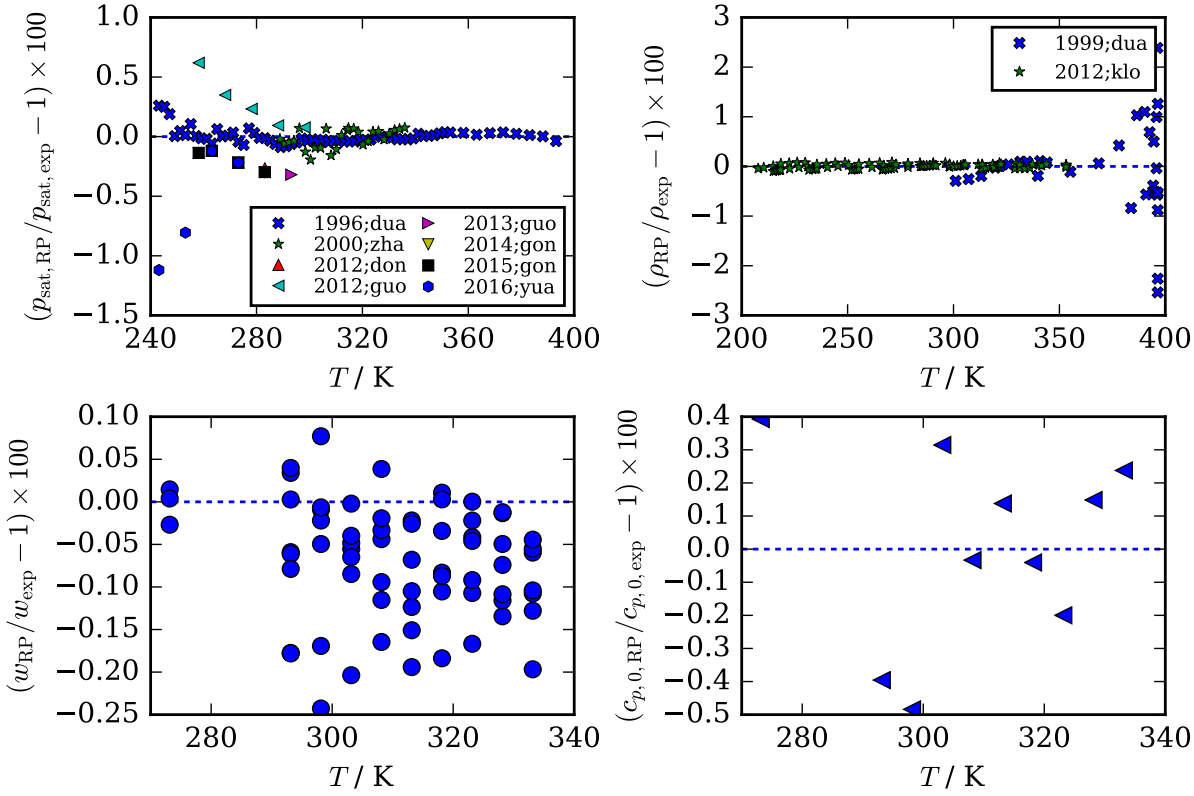

Fig. 3 Deviation plots for CF_3I for the experimental data for saturation pressure p (Guo et al., 2012; Yuan et al., 2016; Dong et al., 2012; Zhang et al., 2000; Gong et al., 2014; Guo et al., 2013; Duan et al., 1996; Gong et al., 2015), density ρ (Klomfar et al., 2012; Duan et al., 1999), speed of sound w (Duan et al., 1997) and ideal-gas specific heat $c_{p,0}$ (Duan et al., 1997) as a function of temperature from the equation of state (Lemmon and Span, 2015) as implemented in REFPROP 10 (Lemmon et al., 2018). For properties with multiple datasets, the labels for datasets are year and first three letters of the first author's name.

3 Mixture Data and Models

As highlighted above, CF_3I has found renewed interest based upon its capability for suppressing flammability and its low GWP. Unfortunately, the interest in CF_3I in this and prior decades has not resulted in a large body of experimental data in the open literature. The NIST ThermoDataEngine library contains a small number of data points for CF_3I-containing mixtures, of only a few types of mixtures, and only vapor-liquid equilibrium data.

Table 2 Data sources for VLE data considered in this study. The composition x_{CF_3I} refers to the liquid phase.

fluids	author	N_{data}	T / K	x_{CF_3I} / molar
CF_3I/CO_2	Yuan et al. (2016)	56	243.2 - 273.1	0.0 - 1.0
CF_3I/R-152a	Gong et al. (2015)	64	258.1 - 283.1	0.0 - 1.0
CF_3I/R-1234ze(E)	Guo et al. (2012)	100	258.1 - 298.1	0.0 - 1.0
CF_3I/propane	Gong et al. (2014)	56	258.1 - 283.1	0.0 - 1.0
CF_3I/isobutane	Guo et al. (2013)	64	263.1 - 293.1	0.0 - 1.0
CF_3I/nitrogen	Lim and Kim (1997)	48	293.2 - 313.2	0.8 - 0.9

Figure 4 shows the data available for mixtures containing CF_3I, and the references are summarized in Table 2. There is a narrow spectrum of data, only covering six references, one per binary pair. To ensure consistency and correctness of the measurements, it is preferred to have confirmatory measurements from at least one unaffiliated laboratory. In the case of the models implemented in REFPROP 10.0, there are a few different sources of mixture model parameters. For four mixtures (+R-152a, +propane, +isobutane, +nitrogen), the fitted parameters were obtained in the work of Bell and Lemmon (2016) and used without modification. For the mixture with R-1234ze(E), the parameters had been fitted in Bell and Lemmon (2016), but were not included in REFPROP 10.0 [1]; thus the model implemented in REFPROP 10.0 for this mixture is the estimation scheme of Lemmon and McLinden (2001). For the mixture with CO_2, the data had not yet been published at the time of publication of Bell and Lemmon, and were not included in the fitting campaign.

The thermodynamic models in REFPROP 10.0 (and also in CoolProp (Bell et al., 2014) and TREND (Span et al., 2019)) are of the multi-fluid Helmholtz-energy-explicit formulation. In this framework, multiparameter equations of state are used for the pure components, and an empirical mixture term and reducing functions are used to account for non-corresponding-states behaviors of the mixture. In the case here (because we are not considering binary departure functions explicitly), the four interaction parameters β_T, γ_T, β_V, and γ_V are the adjustable parameters. More information on this modeling approach is available in the literature (Kunz et al., 2007; Kunz and Wagner, 2012; Gernert and Span, 2016; Bell and Lemmon, 2016).

Overall, the models in REFPROP 10.0 provide a good representation of the data in a qualitative sense (identify azeotropes, etc.), but the models for the mixtures with R-1234ze(E) and CO_2 can be improved. In the case of R-1234ze(E), the improvement is simply to implement the parameters from Bell and Lemmon (2016) of $\beta_T = 1.00346$, $\gamma_T = 0.96604$, $\beta_V = 1$, $\gamma_V = 1$ for the mixture CF_3I + R-1234ze(E) (in that order; the β parameters are not symmetric)

For the mixture with CO_2, a novel parameter optimization approach was employed in this work in which nominal isotherms were identified from each dataset. For a given set of parameters under optimization, the nominal isolines were traced out with the algorithm of Bell and Deiters (2018). The objective function was the sum of the distances of each experimental data point from the isoline. This method avoids the challenges of carrying out the (frequently unreliable) blind phase equilibrium calculations in the work of Bell and Lemmon (2016), whereas the the isoline tracing is not prone to failure, even for more complicated mixture physics. This approach is in the spirit of the work of Tkaczuk et al. (2020). The fitted parameters for this mixture are $\beta_T = 1.013767$, $\gamma_T = 0.963166$, $\beta_V = 1$, $\gamma_V = 1$ for the mixture CO_2 + CF_3I (in that order).

A very important caveat of these models pertains to the complete lack of experimental data for density and caloric properties. Models fit to phase equilibrium data alone can result in significant (±20%) errors in density prediction, even for refrigerant blends with interaction parameters very close to 1.0, indicating a "symmetric" interaction.

[1] this oversight had its origins in the use of the fluid name `R1234ZE` for R-1234ze(E) in REFPROP 9.1, which was corrected to `R1234ZEE` in REFPROP 10.0

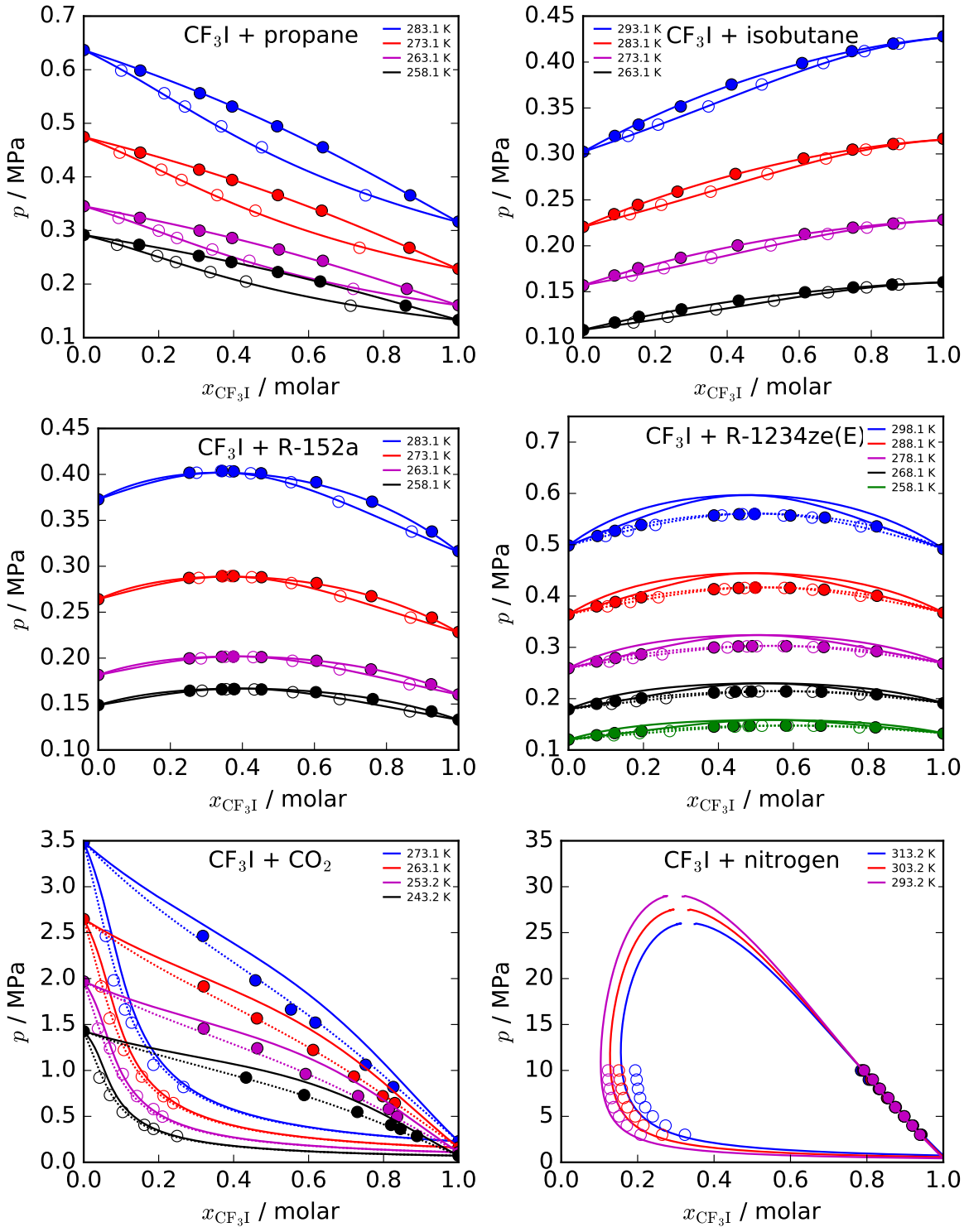

Fig. 4 Summary of experimental mixture VLE data in the open literature (see Table 2) for binary mixtures containing CF_3I (and where the second component is available in REFPROP 10.0). The solid curves indicate the isotherms calculated with the REFPROP 10.0 models (Lemmon et al., 2018) in concert with the isoline tracing routine of Bell and Deiters (2018). The updated models in this work are indicated by dashed curves; all model parameters are in Table 3. The filled markers are experimental saturated liquid points, and the open markers experimental saturated vapor points. The temperature is given by the color indicated in the legend.

Conclusions

There is, in general, a limited set of data available for mixtures with CF_3I. Therefore if the refrigeration industry continues to be interested in the use of this working fluid as a component in blends, additional

Table 3 Recommended binary interaction parameters (T.W.: fitted in this work). The order of fluids matters for the $\beta_{T,ij}$ and $\beta_{V,ij}$ parameters (e.g., $\beta_{T,ij} = 1/\beta_{T,ji}$). The pressure in the last column is the equimolar bubble-point pressure at 300 K (for model verification).

fluids	ref	$\beta_{T,ij}$	$\gamma_{T,ij}$	$\beta_{V,ij}$	$\gamma_{V,ij}$	$p_{300\ K}$ / MPa
R-744 + R-13I1	T.W.	1.01377	0.963166	1	1	3.32811
R-152a + R-13I1	Bell and Lemmon (2016)	1.00125	0.96404	1	1	0.65474
R-13I1 + R-1234ze(E)	Bell and Lemmon (2016)	1.00346	0.96604	1	1	0.59104
propane + R-13I1	Bell and Lemmon (2016)	1.00551	0.98807	1	1	0.79014
R-13I1 + isobutane	Bell and Lemmon (2016)	1.002	0.98459	1	1	0.46999
nitrogen + R-13I1	Bell and Lemmon (2016)	0.998772	1.30226	1	1	23.58994

experimental measurements will need to be carried out. Where data exist, the models in REFPROP 10 provide a good representation of the data; with additional model fine-tuning, the representation can be further improved. The publication of any existing unpublished data, particularly for mixtures, would be very valuable. According to the experimental data available in the open literature, the parameters in Table 3 are therefore recommended.

Acknowledgments

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