



# Measurements of isothermal (vapor + liquid) equilibrium for the (propane + cis-1,3,3,3-tetrafluoropropene) system at temperatures from (253.150 to 293.150) K



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## ABSTRACT

cis-1,3,3,3-Tetrafluoropropene, with zero ozone depleting potential and low global warming potential, has received much attention in recent years. In this work, the (vapor + liquid) phase equilibrium for the (propane + cis-1,3,3,3-tetrafluoropropene) binary system was investigated using an apparatus based on the recirculation method over the temperature range from (253.150 to 293.150) K. The VLE results were correlated by the PR-VDW model and PR-HV-NRTL model. Both models can give satisfactory results and the PR-HV-NRTL model exhibits better performance than PR-VDW model on the VLE description for this binary system. No azeotropic behavior was observed during the experiment.

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## 1. Introduction

From an environmental perspective, switching from conventional refrigerants to low-GWP (global warming potential) refrigerants is the most important consideration for high temperature heat pumping, organic Rankine cycles (ORC), air-conditioning and refrigeration systems. R1234ze(Z) (cis-1,3,3,3-tetrafluoropropene), with the 10 days atmospheric life time and <1 GWP values [1], has been investigated in recent years. Brown et al. [2] predicted the critical temperature, critical pressure and acentric factor for R1234ze(Z) with group contribution methods, which gave 426.8 K, 3970 kPa and 0.333, respectively. Zhang et al. [3] evaluated non-azeotropic mixtures containing R1234ze(Z) as potential refrigerants in refrigeration and high-temperature heat pump systems. It was identified that among the fluorinated propene isomers, R1234ze(Z) delivered similar volumetric refrigerating capacity as R114 (1,2-dichloro-1,1,2,2-tetrafluoroethane) and exhibited the highest COP, although the discharge temperature was higher than that of R114. In the research of Hihara [4], the fundamental information about R1234ze(Z) was provided. Its compatibility with various materials and its flammability and toxicity had been clarified. Additionally, data on its thermodynamic and transport properties were presented. The critical temperature and critical pressure were 423.27 K and 3533 kPa in Hihara's work

[4]. Fedele et al. [5] measured the saturation pressure of R1234ze(Z) over the temperature range from 238.13 K to 372.61 K, and the NBP (normal boiling point) temperature and the acentric factor for R1234ze(Z) were estimated to be 282.73 K and 0.3257, respectively. Higashi et al. [6] measured the PpT properties, vapor pressures, saturated densities and the critical parameters for R1234ze(Z). The results of the critical parameters are as same as those of Hihara.

At the same time, many researchers had investigated the heat transfer characteristic and frictional pressure drop of R1234ze(Z) for both the condensation and evaporation. The pressure gradient of R1234ze(Z) was approximately three times greater than those of R1234ze(E) (trans-1,3,3,3-tetrafluoropropene) and the conventional refrigerant R134a (1,1,1,2-tetrafluoroethane), and the HTC (heat transfer coefficient) of R1234ze(Z) was approximately 2.6 times higher than those of R1234ze(E) and R134a according to Kondou et al. [7]. A similar conclusion was obtained from Longo et al. [8]. Then reported that the heat transfer coefficients of R1234ze(Z) were much greater than those of all the refrigerants now used in heat pump applications and the frictional pressure drop was similar to R600a. Fukuda et al. [9] and Petr and Raabe [10] also indicated the superiority of R1234ze(Z) in high-temperature heat pump and ORC applications. In conclusion, R1234ze(Z) is a potential low-GWP and low-flammable refrigerant with high HTC while its frictional pressure drop is higher than conventional refrigerants. R290 (propane) is also a natural working fluid, which is cheap and conveniently available. The drawback of

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R290 is flammability. Therefore, it may be an effective way to blend R1234ze(Z) with other R290 to integrate the advantages of both fluids and may achieve a better coefficient of performance [11,12].

(Vapor + liquid) equilibrium (VLE) of these interesting mixtures plays an important role in calculating and optimizing performance of vapor compression refrigeration cycles. In this paper, the VLE values for the {R290 + R1234ze(Z)} system will be presented at five temperatures ranging from 253.150 K to 293.150 K. The results will be correlated by two thermodynamic models: Peng–Robinson (PR) [13] equation of state (EoS) with Van der Waals (VDW) [14] mixing rule and PR EoS with non-random two liquids (NRTL) [15] activity coefficient model combined Huron–Vidal (HV) [16] mixing rule.

## 2. Experimental

### 2.1. Materials

R290 and R1234ze(Z) were supplied by Beijing AP BAIF Gases Industry Co., Ltd. and Beijing Yuji Science & Technology Co., Ltd. respectively. The purities of the chemicals stated as a mole fraction by the suppliers are 0.999 and 0.995, respectively. In order to eliminate the non-condensable gases, each sample underwent several cycles of freezing with liquid nitrogen, evacuation, and thawing. The fluids were used with no further purification. The critical temperatures, critical pressures, and acentric factors for R290 and R1234ze(Z) used in this work are taken from REFPROP 9.1 [17] and provided in Table 1.

### 2.2. Apparatus

The experimental set up used in this paper has been described in previous works [18–20], and used without modification, so only essential information is supplied here. The VLE was achieved in a cell with a magnetic pump used to force the vapor through the liquid. A 25  $\Omega$  standard platinum resistance thermometer and a Mensor Series 6000 digital pressure transducer were used to measure the temperature and pressure in the equilibrium cell. The combined standard uncertainties of the temperature and pressure measurement were estimated to be less than  $\pm 5$  mK and  $\pm 0.0005$  MPa, respectively. A gas chromatograph (Shimadzu GC2014) equipped with a thermal conductivity detector was used to analyze the compositions of the vapor and liquid phases. At least three analyses were performed for each sample until the deviations among them was less than 0.001. Taking into account the uncertainties from the calibration and the dispersion of analyses, the uncertainty on vapor and liquid mole fractions is estimated to be within  $\pm 0.005$  over the whole range of concentration.

### 2.3. Experimental procedure

First the pipeline and the equilibrium cell were evacuated and then purged by R1234ze(Z) for three times. Second, an amount of R1234ze(Z) was injected into the cell. When a desired temperature

**Table 1**  
Mole fraction purity, critical parameters ( $T_c, p_c$ ) and acentric factors  $\omega$  for R290 and R1234ze(Z).

Components	CAS No.	Mole fraction purity	$T_c/K^c$	$p_c/MPa^c$	$\omega^c$
R290 <sup>a</sup>	74-98-6	0.999 <sup>d</sup>	369.89	4.2512	0.1521
R1234ze(Z) <sup>b</sup>	29118-25-0	0.995 <sup>d</sup>	423.27	3.5330	0.3274

<sup>a</sup> Supplied by Beijing AP BAIF Gases Industry Co., Ltd.

<sup>b</sup> Supplied by Beijing Yuji Science & Technology Co., Ltd.

<sup>c</sup> Ref. [17].

<sup>d</sup> As stated by the supplier.

was reached and the fluctuations of the temperature and the pressure were less than  $\pm 3$  mK,  $\pm 100$  Pa, respectively, for more than 20 min in the cell, the equilibrium state was established. The saturated pressure of R1234ze(Z) was recorded. Then an estimated amount of R290 was injected into the equilibrium cell. When the same equilibrium state was achieved, the mole fractions of the vapor and liquid phases of {R290 + R1234ze(Z)} were collected by the gas chromatograph. The process above was repeated and the VLE measurements were accomplished.

## 3. Thermodynamic models

The experimental values were regressed by both PR-VDW model and PR-HV-NRTL model. The PR EoS is employed in the following form:

$$p = \frac{RT}{v-b} - \frac{a}{v(v+b) + b(v-b)}, \quad (1)$$

$$a = 0.457235 \times \frac{R^2 T_c^2}{p_c} \left[ 1 + (0.37464 + 1.54226\omega - 0.26992\omega^2) \left( 1 - \sqrt{T/T_c} \right) \right]^2, \quad (2)$$

$$b = 0.077796 \frac{RT_c}{p_c}, \quad (3)$$

where  $p$  is the pressure in Pa,  $v$  the mole volume in  $m^3 \cdot mol^{-1}$ ,  $T$  the temperature in K,  $R$  is the gas constant in  $J \cdot mol^{-1} \cdot K^{-1}$ ,  $p_c$  and  $T_c$  are the critical pressure and temperature, respectively, and  $\omega$  is the acentric factor of the pure component.

NRTL activity coefficient model is used to calculate the excess molar Gibbs energy, which is given by:

$$\frac{G^E}{RT} = \sum_i x_i \frac{\sum_j \tau_{ji} G_{ji} x_j}{\sum_i G_{ii} x_i}, \quad (4)$$

$$G_{ji} = \exp(-\alpha_{ji} \tau_{ji}), \quad (5)$$

where  $\tau_{ij}$ ,  $\tau_{ji}$  and  $\alpha_{ij} = \alpha_{ji}$  are adjustable parameters and  $\tau_{ii} = 0$ ,  $\alpha_{ii} = 0$ . The parameter  $\alpha_{ij}$  was used as a fixed value 0.3 in present work.

Activity coefficients obtained by NRTL model for a binary system are

$$\ln \gamma_1 = x_2^2 \left[ \frac{\tau_{21} G_{21}^2}{(x_1 + x_2 G_{21})^2} + \frac{\tau_{12} G_{12}}{(x_2 + x_1 G_{12})^2} \right], \quad (6)$$

$$\ln \gamma_2 = x_1^2 \left[ \frac{\tau_{12} G_{12}^2}{(x_2 + x_1 G_{12})^2} + \frac{\tau_{21} G_{21}}{(x_1 + x_2 G_{21})^2} \right]. \quad (7)$$

The Van der Waals mixing rule is expressed as

$$a_m = \sum_i \sum_j x_i x_j \sqrt{a_{ii} a_{jj}} (1 - k_{ij}), \quad (8)$$

$$b_m = \sum_i x_i b_i, \quad (9)$$

with  $k_{ij} = k_{ji}$  and  $k_{ii} = 0$ .

The HV mixing rule can be rewritten as

$$a = b \left[ \sum_i x_i \left( \frac{a_{ii}}{b_{ii}} \right) - \frac{g_{\infty}^E}{C} \right], \quad (10)$$

$$b = \sum_i x_i b_{ii}, \quad (11)$$

where  $C$  is a numerical constant with the value 0.623225 in PR EoS and  $g_{\infty}^E$  is the excess molar Gibbs energy calculated by NRTL activity coefficient model at infinite pressure.

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