



# Isothermal (vapour + liquid) equilibrium measurements and correlation for the {n-butane (R600) + 1,1,1,3,3-pentafluoropropane (R245fa)} system at temperatures from (303.150 to 373.150) K



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## ABSTRACT

The isothermal VLE for the {n-butane (R600) + 1,1,1,3,3-pentafluoropropane (R245fa)} system was measured at five temperatures from (303.150 to 373.150) K based on the static-analytical method. The experimental values were correlated by the Peng–Robinson equation of state with the Huron–Vidal mixing rule involving the non-random two-liquid (NRTL) activity coefficient model. The results show that the experimental values are in good agreement with the calculated value. The maximum average absolute relative deviation of pressure and vapour phase mole fraction are 0.35%, 0.0047, respectively. Obviously azeotropic behaviour can be observed for the binary system.

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## 1. Introduction

Organic compound mixtures have been widely used as working fluids for power generation cycles, refrigeration and heat pumps [1–3]. Hydrocarbons (HCs), hydrofluorocarbons (HFCs) and their mixtures have good thermophysical properties and environmental friendliness (zero ODP and low GWP), so they have been used in organic Rankine cycle and refrigeration systems [4–6]. To analyse the performance of these systems, knowledge of the (vapour + liquid) equilibrium (VLE) of these mixtures is necessary.

A significant quantity of VLE values for (HC + HFC) system at temperatures below 323.15 K are available [7–11]. Some works [12,13] on the VLE at temperatures above 353.15 K were published. The VLE for the {1,1-difluoroethane (R152a) + R600} system was measured by Jihoon *et al.* [12] at temperatures from 313.15 K to 363.15 K. Elise *et al.* [13] obtained isothermal VLE data for the {R245fa + isopentane (R601a)} and {R365mfc + R601a} systems with temperature from 363 K to 413 K and pressure from 0.4 to 2.9 MPa.

In present work, the VLE for the {n-butane (R600) + 1,1,1,3,3-pentafluoropropane (R245fa)} system at  $T = (303.150 \text{ to } 373.150) \text{ K}$  was measured using a static-analytic method. Then the results were correlated using the Peng–Robinson EoS [14] with

the Huron–Vidal (HV) mixing rule [15] involving the non-random two-liquid (NRTL) activity coefficient model [16].

## 2. Experimental

### 2.1. Materials

R600 was provided by Beijing AP BAIF Gases Industry Co. Ltd with a declared mole fraction purity of 0.998. R245fa was supplied by Zhejiang Lantian Environmental and Chemical Engineering Co. Ltd with a declared mole fraction purity of 0.990. Both materials were used without any further purification. table 1 gives the purity, critical temperature, critical pressure, and acentric factor for R600 and R245fa [17].

### 2.2. Equilibrium apparatus

The VLE measurements of the binary mixtures were carried out by a static-analytic method. The apparatus used in this work had been described in detail in our previous work [18].

A schematic of the apparatus is shown in figure 1. The equilibrium cell (150 mL) was immersed into the isothermal oil bath which was cooled by cooling water and heated by an electric heater. To obtain uniform temperature distributions in the liquid bath, a stirrer with a speed controller was used. Heat losses were reduced by a vacuum vessel installed outside the constant

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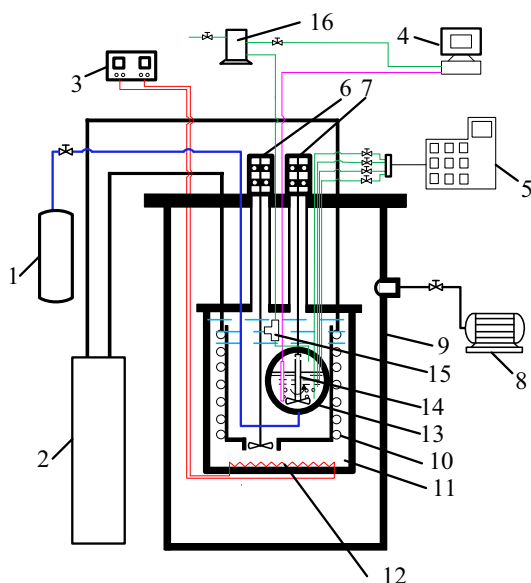
E-mail address: [gongmq@mail.ipc.ac.cn](mailto:gongmq@mail.ipc.ac.cn) (M. Gong).

**TABLE 1**  
Mole fraction purity, critical parameters ( $T_c$ ,  $p_c$ ) and acentric factors  $\omega$  for R600 and R245fa [18].<sup>a</sup>

Fluids	CAS no.	Supplier	Mole fraction purity <sup>b</sup>	$T_c$ /K	$p_c$ /MPa	$\omega$
R600	106-97-8	Beijing AP BAIF Gases Industry Co. Ltd	0.998	425.13	3.796	0.201
R245fa	460-73-1	Zhejiang Lantian Environmental and Chemical Engineering Co. Ltd	0.990	427.16	3.651	0.3776

<sup>a</sup> Both of the materials were used without further purification.

<sup>b</sup> Supplier purity (mole fraction) stated by the supplier.



**FIGURE 1.** A schematic diagram of the experimental apparatus. 1. Induction system; 2. Cooling water system; 3. Digital controller; 4. Temperature and pressure acquisition system; 5. GC2014; 6, 7. Agitator motor; 8. Vacuum pump; 9. Vacuum vessel; 10. Cooling coil; 11. Isothermal liquid bath; 12. Electric heater; 13. Equilibrium cell; 14. Stirrers in the cell; 15. Pressure transducer (GE UNIK5000); 16.  $N_2$ -filled system.

temperature liquid bath. By adjusting the power of the electric heater, a Shimaden SR 253 digital controller controlled the bath temperature. A  $25\ \Omega$  standard platinum resistance thermometer (Pt 25) was used to measure the temperatures in the cell. The Pt 25 has been calibrated by the Cryogenic Metrology Station of the Chinese Academy of Sciences based on the 1990 International Temperature Scale (ITS 90) with an uncertainty of  $\pm 5\ \text{mK}$  ( $k = 2$ ). The combined standard uncertainty of the temperature measurement is estimated to be within  $\pm 5\ \text{mK}$ .

The pressure measurements were made using a digital pressure transducer (Mensor CPT6100) and a pressure transmitter (GE UNIK5000). The pressure in the  $N_2$ -filled system was measured by the Mensor CPT6100. The uncertainty of digital pressure transducer is 0.01% FS with two full scales of 1.5 MPa and 3.0 MPa. The pressure difference between the equilibrium cell and the  $N_2$ -filled system was measured by the transmitter. The uncertainty of the transmitter is 0.04% FS with the range of (0 to 100) kPa and  $T = (233.15\ \text{to}\ 393.15)\ \text{K}$ . The pressure in the equilibrium cell can be obtained by adding above two pressures together. The combined standard uncertainty of pressure measurement is estimated to be less than  $\pm 500\ \text{Pa}$ .

A Shimadzu GC2014 gas chromatograph equipped with a thermal conductivity detector (TCD) was used to measure the vapour and liquid mole fractions in the equilibrium state. A six-port-automatic-sample-injection (SPASI) valve (figure 2) was used to sample accurately. In the pushing mode, the sample quantitative loop was connected to the equilibrium cell by the capillaries. The sample in

the equilibrium cell was introduced to the sample quantitative loop through the capillaries. In the sample mode, rotation of the rotor  $60^\circ$ , the sample quantitative loop was connected to the carrier gas helium and the chromatographic column and the sample in the loop was carried by helium into the chromatographic column and analysed. Compared to the volume of the cell, the volume of the quantitative loop is tiny and can be neglected and the large influence of sampling on the equilibrium state in the cell can be avoided. The sample method prevents the carrier gas from contaminating the equilibrium cell. The GC was calibrated with the mixtures obtained by the gravimetric method. The uncertainties of vapour and liquid phase composition measurements are estimated to be within  $\pm 0.005$ .

### 2.3. Equilibrium procedure

Experiments were performed by the following procedure. The system was evacuated at first to remove the remaining gas at room temperature. To purge the total system, R245fa was then driven into and pumped out from the cell at least three times. Then, R245fa was added to the cell and the temperature of the Shimaden SR 253 was changed to the desired value. After the steady state value was reached and maintained for at least 1 h and the temperature fluctuation was within  $\pm 3\ \text{mK}$  for at least 10 min, the saturated vapour pressure values of R245fa were recorded. After that, a proper amount of R600 was charged into the cell and mixed sufficiently with R245fa. After the desired temperature was reached for at least 1 h with the fluctuation less than  $\pm 3\ \text{mK}$  for more than 10 min, the equilibrium condition was considered to be reached. Then the pressure was recorded and the vapour and liquid compositions at the equilibrium state were analysed by the GC. At least three samples with a repeatable mole fraction less than 0.002 for each phase were measured and the average value was recorded. The VLE values for other concentrations were obtained by repeating this process by supplying a more adequate amount of R600. Finally, the cell was evacuated and pure R600 was measured to obtain its saturated vapour pressures.

## 3. Results and discussions

Table 2 contains the experimental results, the calculated value from REFPROP 9.0 [17] and the literature data [18–23,13] of saturated vapour pressures of pure R600 and R245fa. The symbols  $p_{\text{exp}}$ ,  $p_{\text{ref}}$  and  $p$  are the saturated vapour pressures from the experiment, calculation and literatures, respectively. The  $\Delta p$  and  $\Delta p/p_{\text{exp}}$  are the absolute deviation and relative deviation between  $p_{\text{exp}}$  and  $p_{\text{ref}}$ . The experimental values are basically in agreement with the calculated and literature results.

The VLE values for the binary system of (R600 + R245fa) were measured at  $T = (303.150, 323.150, 343.150, 363.15\ \text{and}\ 373.150)\ \text{K}$ . The PR–HV–NRTL model was used to correlate the VLE experimental data.

The PR EoS [14] is expressed as follows:

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

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