



# The investigation on the vapor + liquid equilibrium for the ternary mixture isobutene (R600a) + 1,1-difluoroethane (R152a) + 1,1,2,2-tetrafluoroethane (R134) at temperatures from 253.150 to 273.150 K

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## ABSTRACT

In this work, the vapor + liquid phase equilibrium for the ternary system isobutane + 1,1-difluoroethane + 1,1,2,2-tetrafluoroethane was measured using an apparatus based on the recirculation method at temperatures range from 253.150 to 273.150 K. The uncertainties of the temperature, pressure and composition were less than  $\pm 5$  mK,  $\pm 0.0005$  MPa and  $\pm 0.005$ , respectively. PR-VDW and PR-MHV2-NRTL models were employed to describe the VLE properties of system concerned. Both models can give satisfied results, and PR-MHV2-NRTL model exhibited better performance than PR-VDW model on the VLE description. A saddle point azeotropic behavior was found, and the azeotropic compositions were calculated by both methods. With the regressed binary interaction parameters from ternary VLE data, the binary VLE were predicted, and the results agree well with the experimental data.

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## 1. Introduction

Mixed refrigerants consisting of hydrocarbon (HCs) and hydrofluorocarbon (HFCs) attract more and more attentions since they can integrate the advantages of the single fluid and may achieve a better coefficient of performance [1–5]. Apparently, vapor + liquid equilibrium data of these mixtures play an important role in calculating and optimizing performance of vapor compression refrigeration cycles.

Isobutane (R600a), 1,1-difluoroethane (R152a) and 1,1,2,2-tetrafluoroethane (R134) are promising fluids which can be used as refrigerants or part of the mixed refrigerants [6,7]. The vapor + liquid equilibrium behaviors of the binary systems including R600a + R152a, R600a + R134 and R152a + R134 have been studied by Dong [8,9] and Lim et al. [10], and two positive azeotropes and one negative azeotrope were found. In this paper,

the vapor + liquid equilibrium for R600a + R152a + R134 ternary system was measured with an apparatus based on the recirculation method. The VLE data were correlated by two thermodynamics models: Peng–Robinson (PR) [11] equation of state (EoS) with Van der Waals (VDW) [12] mixing rule and PR EoS with non-random two liquids (NRTL) [13] activity coefficient model combined modified Huron–Vidal second-order (MHV2) [14,15] mixing rule. A saddle point azeotropy behavior was found at the experimental temperature range from 253.150 to 273.150 K.

## 2. Experimental section

### 2.1. Materials

R134 was supplied by Nanjing Yuji Tuohao Co. with a declared mole fraction purity of 0.995. R600a and R152a were provided by Beijing AP BAIF Gases Industry Co., Ltd. with the declared mole fractions of 0.999. All samples were used without further purification. The critical temperatures, critical pressures, and acentric factors for R134, R600a and R152a used in this work are taken from REFPROP 9.1 [16] and provided in Table 1.

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Nomenclature			
<i>Abbreviations</i>		$f$	fugacity coefficient
AAD	The average absolute deviations	$G$	excess molar Gibbs energy
AARD	The average absolute relative deviations	$k_{ij}$	binary interaction parameter between components $i$ and $j$
$c_1, c_2$	PR EoS parameter	$n$	number of the components
EoS	equation of state	$N$	number of experimental points
HCS	hydrocarbons	$p$	pressure
HFCs	fluorohydrocarbons	$R$	universal gas constant
MHV2	modified Huron–Vidal second-order	$T$	temperature
NRTL	non-random two liquids	$x$	liquid phase composition
PR	Peng–Robinson	$y$	vapor phase composition
R600a	isobutane	<i>Greek letters</i>	
R134	1,1,2,2-tetrafluoroethane	$\mu$	chemical potential
R152a	1,1-difluoroethane	$\omega$	the acentric factor of the pure component
RD	the relative deviations	$\tau$	binary interaction coefficients in NRTL activity model
VDW	Van der Waals	$\alpha$	coefficients in NRTL activity model
VLE	vapor liquid equilibrium	<i>Subscripts</i>	
<i>Symbols</i>		$c$	the critical parameter
$a$	attractive parameter in the EoS	$i, j$	component index
$a_m$	attractive parameter of the mixture	$m$	mixture
$a_{ij}$	cross parameter of an EoS	$r$	reduced parameter
$b$	covolume in the EoS	<i>Superscripts</i>	
$b_m$	covolume of the mixture	$V$	vapor phase
$C$	Antoine equation parameter	$L$	liquid phase

**Table 1**

Mole fraction purity, critical parameters ( $T_c$ ,  $p_c$ ) and acentric factors  $\omega$  for R600a, R152a and R134.

Components	CAS No.	Mole fraction purity	$T_c/K^c$	$p_c/MPa^c$	$\omega^c$
R600a <sup>a</sup>	75-28-5	0.999	407.85	3.6400	0.18600
R152a <sup>a</sup>	75-37-6	0.999	386.41	4.5168	0.27521
R134 <sup>b</sup>	359-35-3	0.995	391.74	4.6400	0.29300

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

<sup>b</sup> Supplied by Nanjing Yuji Tuohao Co.

<sup>c</sup> Reference [16].

## 2.2. Apparatus

The VLE measurements were conducted in an apparatus based on recirculation method which is achieved by driving the vapor phase at the top of the equilibrium cell into the liquid phase at the bottom. Detailed description about the experimental setup has been introduced in previous work [17–19]. The temperatures were measured by a 25  $\Omega$  standard platinum resistance thermometer, which was calibrated by the Cryogenic Metrology Station of the Chinese Academy of Sciences based on the 1990 International Temperature Scale (ITS90) with an uncertainty less than  $\pm 3$  mK. The temperatures in the cell were collected by a Guildline 6622A resistance/thermometry bridge. The combined standard uncertainty of the temperature measurement was estimated to be less than  $\pm 5$  mK. The pressures in the cell were measured by a Mensor Series 6000 digital pressure transducer which has two full scales of (2.5 and 1.25 MPa) with uncertainties of ( $\pm 0.0005$  and  $\pm 0.00025$  MPa), respectively. The second scale was used in this work, and the combined standard uncertainty of the pressure measurement was estimated to be  $\pm 0.0005$  MPa. The compositions of the vapor and liquid phases were analyzed by a gas chromatograph (Shimadzu GC2014) equipped with a thermal conductivity

detector. At least three analyses were performed for each sample to ensure deviations in mole fractions within  $\pm 0.0005$ . 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

Experiments for the R600a + R152a + R134 ternary system were performed by the following procedure. The cell and the recirculation tubes were evacuated at first to remove the residual impurities. Because of the similar saturated vapor pressure among the three single fluids, it is flexible to charge the three fluids into the cell in any order. After the desired temperature was reached and the temperature fluctuation in the cell was less than  $\pm 3$  mK and the pressure fluctuation was less than  $\pm 200$  Pa for at least 20 min, the equilibrium state was considered to be established. Then the vapor and liquid mole fractions were measured by the gas chromatograph at least three times respectively, and the average value was recorded. The VLE data for other compositions were measured by repeating the above procedure by supplying certain amount of the other two fluids step by step.

## 3. Thermodynamics models

PR-VDW model and PR-MHV2-NRTL model were employed to correlate the experimental data.

The PR EoS is expressed in the following form:

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

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