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Vapor–liquid equilibria measurements of 1,1,1,2-tetrafluoroethane (HFC-134a) + 2,3,3,3-tetrafluoroprop-1-ene (HFO-1234yf) + isobutane (HC-600a) ternary system

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ABSTRACT

Vapor–liquid equilibrium data for the ternary mixture of 1,1,1,2-tetrafluoroethane (HFC-134a) + 2,3,3,3-tetrafluoroprop-1-ene (HFO-1234yf) + isobutane (HC-600a) were measured at temperatures from 283.15 K to 323.15 K using a recirculation apparatus. The uncertainties of the experimental data are estimated within 5 mK, 0.5 kPa, and 0.003 for the temperature, pressure, and the equilibrium liquid and vapor mole fractions, respectively. The experimental data were compared with the predicted data by the Peng Robinson equation of state with the vdW mixing rule and the Wong Sandler mixing rule, respectively. The binary interaction parameters of each binary mixture were determined from the experimental data in literature. The results show that the predicted data agree well with the experimental data. It indicates that the PR equation can be used to predict VLE data of ternary mixture of HFC-134a + HFO-1234yf + HC-600a. The proposed ternary mixture has potential to be refrigerant with low GWP and excellent thermodynamic performance.

1. Introduction

According to the European Union's F-Gas Regulations [1,2], GWP (Global Warming Potential) higher than 150 will be banned. Thus, it is important to find new refrigerants or mixtures with zero ODP (Ozone Depletion Potential) and low GWP. HFO-1234yf, jointly developed by Honeywell and DuPont, has a very low GWP of about 4, zero ODP, a very short atmospheric lifetime of 0.03 years, and its thermophysical properties are similar to those of HFC-134a [3,4]. Thus, it has been used as a replacement for HFC-134a in automotive air conditioners. However, HFO-1234yf has a smaller latent heat, which causes a large mass flow rate and a large pressure drop, and leads to a decrease in COP. To overcome this drawback, several binary mixtures of HFO-1234yf with HFCs, HCs, and CO_2 [5–10] and one ternary mixture of HFC-134a + HFO-1234yf + DME [11] have been studied. In which, HC-600a has much larger latent heat and excellent thermodynamic performance, but it is highly flammable and explosive, and HFO-1234yf is also mildly flammable. To decrease the flammability of HFO-1234yf + HC-600a binary mixture, non-flammable HFC-134a is proposed as a component of the mixture. Thus, in this work, the vapor–liquid equilibrium data for the ternary system of HFC-134a (1) + HFO-1234yf (2) + HC-600a (3) were measured at intervals of 10 K in the temperature range from 283.15 to 323.15 K.

2. Experimental

2.1. Chemicals

The purities and suppliers of HFC-134a, HFO-1234yf and HC-600a are summarized in Table 1. All samples were used without any further purification.

2.2. Apparatus

The VLE data for the ternary system of HFC-134a (1) + HFO-1234yf (2) + HC-600a (3) were measured by a recirculation apparatus, the details of which have been reported previously [6–9]. Thus, only a brief description is given here. As shown in Fig. 1, the apparatus consists of an equilibrium cell, a liquid thermostatic bath, an air thermostatic bath, a sampling system, and measurement

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Table 1
Suppliers and purities of the chemicals.

Component	Supplier	Purity/mass%
HFC-134a	Zhejiang Juhua, China	99.9
HFO-1234yf	Honeywell, USA	99.9
HC-600a	Nanjing Special Gas, China	99.9

instruments. To improve the stability of VLE measurement, the sampling loops outside the liquid thermostatic bath were guarded by the air thermostatic bath, which is operated at the same temperature of the liquid thermostatic bath.

The temperature was measured by a 25 Ω platinum resistance thermometer (Model WZPB-1, Kunming Temperature Instruments Ltd., China, calibrated by the manufacture). Its uncertainty was estimated to be 0.002 K. The temperature was measured by a 25-Ω platinum resistance thermometer with an uncertainty of 2 mK. The temperature fluctuation of the thermostatic bath is within 4 mK. The uncertainty of digital multimeter (Agilent 3458A) is estimated within 2 mK. Thus the standard temperature uncertainty is to be less than 5 mK. The pressure is measured by means of a pressure transducer (Model TS109, Kunshan Danrui sensor measurement and Control Technology Co., Ltd.), with an uncertainty of 0.5 kPa (0.025% of full scale of 2 MPa, calibrated by the manufacture). The mole fractions of the vapor and liquid phase were measured using a gas chromatograph (GC112A, Shanghai Precision and Scientific Instrument Ltd., China) equipped with a flame ionization detector (FID) and a Porapak-Q packed column (80/100 mesh, 2 m long, 3 mm diameter). It was calibrated with mixtures of known mole fraction of components that were prepared gravimetrically using an electronic balance (JA5003, 1 mg/500 g, Shanghai Hengping, China). The total uncertainty of the vapor and liquid phase mole fraction is estimated to be within 0.003 for the HFC-134a (1) + HFO-1234yf (2) + HC-600a (3) mixture.

2.3. Experimental procedure

The detail of the experimental procedure has been reported in our previous studies [6–9], it is only briefly described here. Firstly, the whole system was evacuated by a vacuum pump to remove all gases, then washed using HC-600a and evacuated again. After that, an appropriate amount of HC-600a, HFO-1234yf and HFC-134a were introduced into the cell in order of increasing vapor pressure at the same temperature. The magnetic pump was used to circulate the saturated vapor from the top of the cell through a four-way valve to the bottom of the cell. When the equilibrium was established at the desired temperature, the pressure in the equilibrium cell was measured and the vapor sample was taken into the gas chromatograph and the vapor phase mole fraction was measured immediately. After that, the four-port valve was switched to liquid phase circulation, and the liquid sample was taken into the gas chromatograph for liquid phase mole fraction measurement. The mole fractions of each phase were measured at least four times in succession, and the average values were considered as the results.

3. Result and VLE calculation

In this work, the PR equation of state [12] with vdW mixing rule and WS mixing rule [13] were used to calculate VLE data for HFC-134a (1) + HFO-1234yf (2) + HC-600a (3) system. The critical properties and the acentric factors of pure components were presented in Table 2 [14].

The vdW mixing rule can be written as

$$a = \sum_{i=1}^{2}\sum_{j=1}^{2} x_i x_j a_{ij} \tag{1}$$

$$A_{\infty}^{E},\ i = 1, 2;\ j = 1, 2 \tag{2}$$

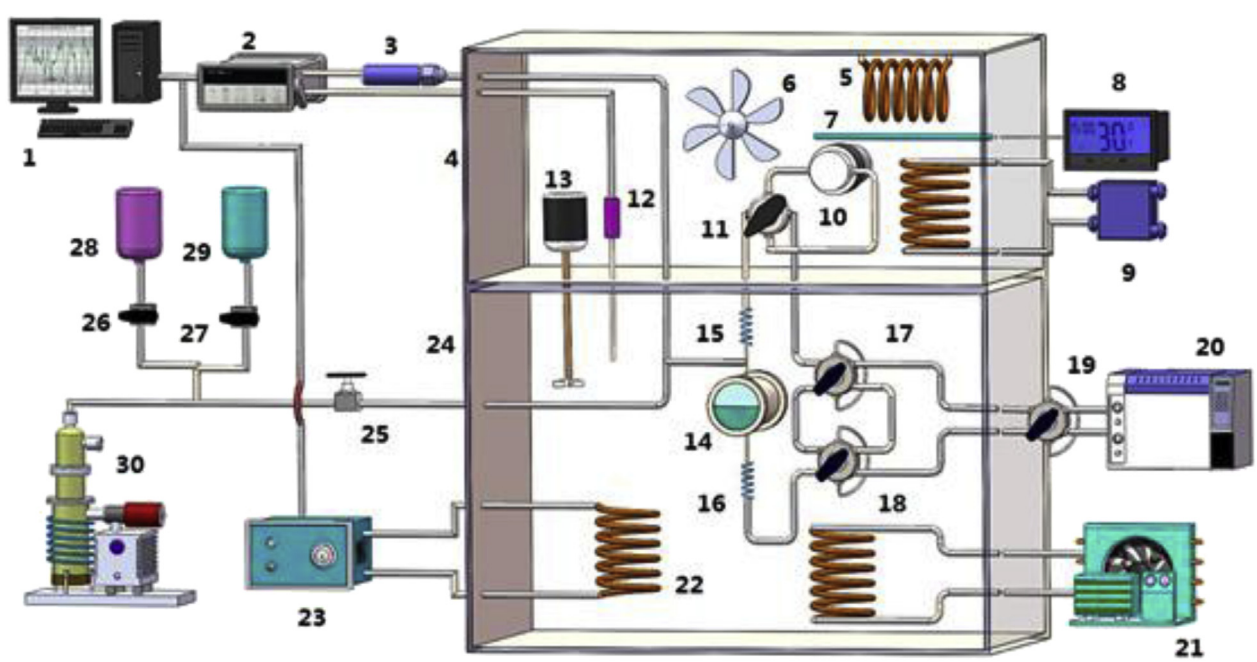

Fig. 1. Schematic diagram of the VLE experimental system. (1) computer; (2) data acquisition instrument; (3) pressure transducer; (4) air thermostatic bath; (5) heater A; (6) fan; (7) temperature sensor; (8) digital temperature controller; (9) semiconductor chiller; (10) magnetic; (11) four-port valve; (12) platinum resistance thermometer; (13) stirrer; (14) equilibrium cell; (15) liquid-phase heat exchanger; (16) vapor-phase heat exchanger; (17) six-port valve 1; (18)six-port valve 2; (19)six-port valve 3; (20) gas chromatograph; (21) refrigeration system; (22) heater B; (23) temperature controller; (24) liquid thermostatic bath; (25)–(27) valves; (28)sample cylinder A; (29)sample cylinder B; (30) vacuum pump.