

# High pressure vapor–liquid equilibria measurements and modeling of butane/ethanol system and isobutane/ethanol system



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## ABSTRACT

The high-pressure vapor–liquid equilibria of butane/ethanol system and isobutane/ethanol system were measured by a minimum continuous flow method using a process gas chromatography at 313–403 K. Experimental PXY data were compared with references. The modified RK EOS parameters were regressed and PXY data were also compared with PSRK EOS.

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## 1. Introduction

Bioethanol is considered to be one of the most promising alternatives to fossil fuels in the short and medium term. Several processes have been developed to produce bioethanol from corn or lignocellulosic biomass. Bioethanol production processes typically comprise four parts: pretreatment, scarification, fermentation and dehydration [1–4]. The development of energy-saving technology for each process is desired.

Many energy-saving dehydration processes for bioethanol production have been developed, such as membrane separation, zeolite adsorption and azeotropic distillation [5–9]. Horizoe et al. [10–12] developed an ethanol dehydration system with an extractive distillation using a light hydrocarbon solvent. To effectively develop this process, vapor–liquid equilibria at a wide range of conditions were needed.

Vapor–liquid equilibrium data of butane/ethanol system has been previously reported in the literature by Deak et al. [13], who measured a bubble point with the Cailletet apparatus at 323–523 K, and Soo et al. [14] who measured the composition of the vapor and liquid phases with ROLSI™ capillary samplers, analyzed by gas chromatography at 323–423 K. Kretschmer et al. [15], Holderbaum et al. [16] and Dahlhoff et al. [17] also reported the VLE of butane/ethanol system. Vapor–liquid equilibrium data of isobutane/ethanol system has been reported by Ouni et al. [18], who

measured VLE data at 313 K using the total pressure method. Furthermore, the Barker method was used to convert the measured PTZ data into PTXY data. In our dehydration process, operation temperature range is from 310 to 410 K, so VLE data of isobutane is still limited.

In this study, the high-pressure vapor–liquid equilibria of butane/ethanol system and isobutane/ethanol system were measured by the minimum continuous flow method using process gas chromatography at 313–403 K. Experimental PXY data were compared with reference data. The modified RK (Redlich–Kwong) EOS which is developed by Twu et al. [19,20] parameters were regressed and PXY data were also compared with PSRK (predictive Soave–Redlich–Kwong) EOS [21].

## 2. Measurements

### 2.1. Materials

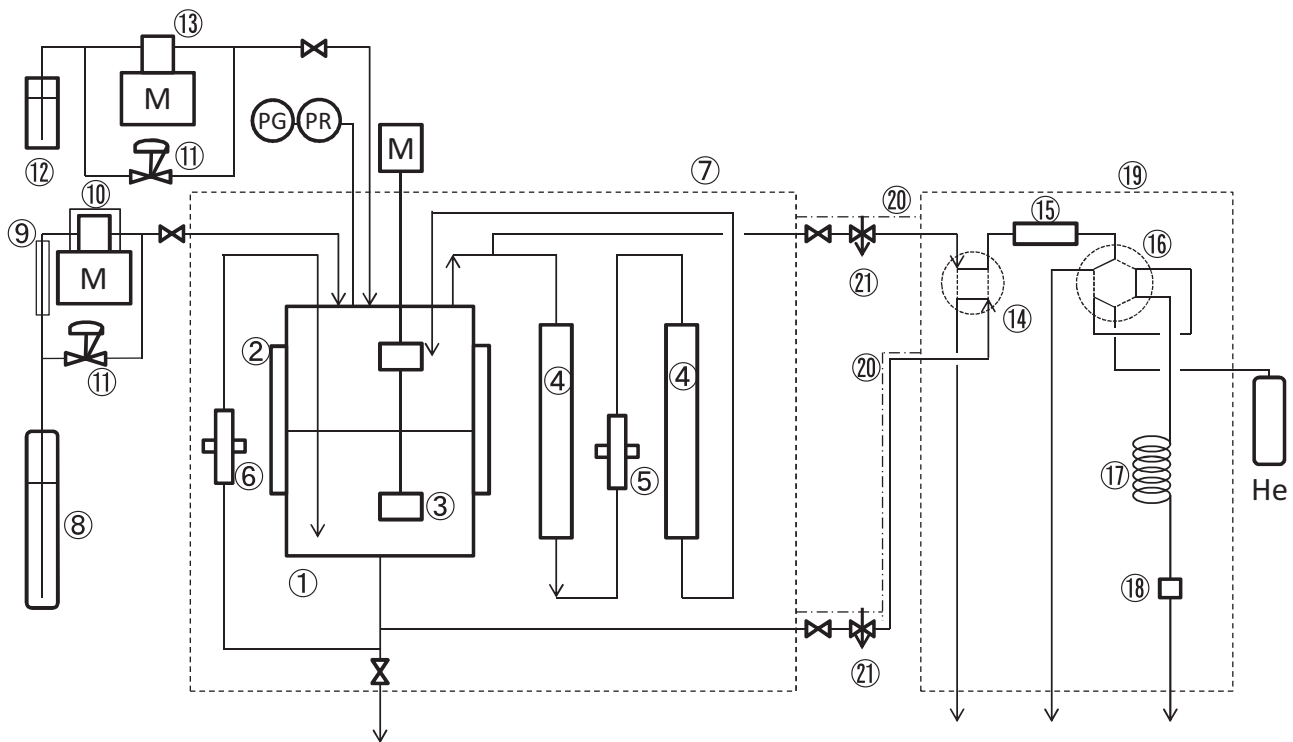
Table 1 lists the purity and supplier used in this study.

### 2.2. Apparatus and measuring procedure

A diagram of the vapor–liquid equilibrium apparatus is shown in Fig. 1. The apparatus comprised an equilibrium cell with a view window (950 cc), a liquid phase circulation magnetic pump, a vapor phase circulation magnetic pump and a process GC. Two gas buffers (both 950 cc) were placed before and after the vapor phase circulation magnetic pump to prevent pressure fluctuation. The equilibrium cell and circulation pump were immersed

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**Fig. 1.** Vapor–liquid equilibrium measurement apparatus. ① Equilibrium cell, ② window, ③ stirrer, ④ gas buffer, ⑤ gas phase circulation pump, ⑥ liquid phase circulation pump, ⑦ air bath, ⑧ butane bomb, ⑨ cooling jacket, ⑩ diaphragm pump, ⑪ back pressure regulator, ⑫ ethanol, ⑬ liquid pump, ⑭ 4-way valve, ⑮ mixing buffer, ⑯ 6-way valve, ⑰ column, ⑱ TCD detector, ⑲ process gas chromatography, ⑳ micro heater, ㉑ metering valve.

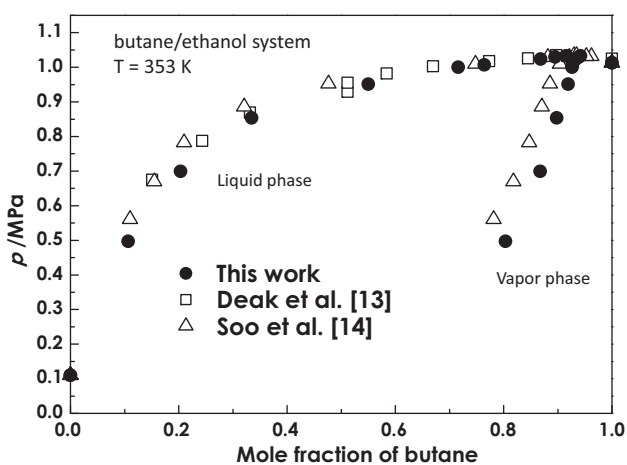
in a temperature-controlled air-bath which was controlled to within 0.1 °C with PID. View windows made from Quartz glass were attached to the cell to check the liquid level and condition. The temperature inside the equilibrium cell was measured with a platinum resistance probe (ChinoPt100 Class A). The pressure at equilibrium was measured by both a Bourdon gauge (GP35, NAGANO KEIKI Co., Ltd.), 0–35 MPa marked in 0.05 divisions and a digital gauge, 0–3 MPa (PAA-33XEi, Keller AG) with an accuracy of 0.1% of F.S.

The measuring procedure used is shown below. A desired amount of ethanol was fed into the equilibrium cell with a liquid pump (NP-KX500, Nihon Seimitsu Kagaku Co. Ltd.). CO<sub>2</sub> or butane was pressurized with a diaphragm pump (JZ104D-30V, Fuji Pump Co. Ltd.) and purged over three times to degas the liquid phase

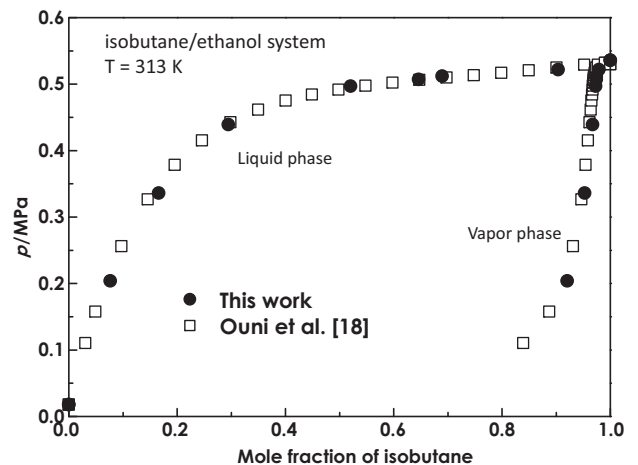
**Table 1**  
Properties and suppliers of materials.

Material	Purity (wt%)	Supplier
Butane	99.8	Toyoko Kagaku Co., Ltd. (Japan)
Isobutane	99.9	Toyoko Kagaku Co., Ltd. (Japan)
Ethanol	99.5	Wako pure chemical industries, Ltd. (Japan)

and replace the vapor phase. Equilibrium was achieved by agitation with a stirrer, and circulation of the vapor and liquid phases using a circulation pump. When the equilibrium was reached, the agitation and circulation ratio decreased. Small amounts of the vapor and liquid phases were continuously sampled with the aid of a metering valve and analyzed with a process gas chromatograph



**Fig. 2.** Phase equilibrium data of the butane/ethanol system compared with reference data at 353 K.



**Fig. 3.** Phase equilibrium data of the isobutane/ethanol system compared with reference data at 313 K.

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