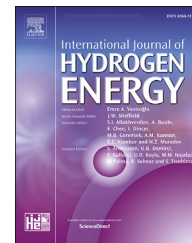




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Vapor-liquid equilibria for low temperature CH₄/H₂ mixture



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ABSTRACT

As a type of gas mixture mainly composed of methane, synthetic natural gas (SNG) needs accurate description for its vapor-liquid equilibria (VLE) in order to utilize it better. Particularly considering the hydrogen component is involved, current researches still lack of sufficient VLE data for CH₄/H₂ system at cryogenic temperature and low pressure which are common conditions for SNG industrial application. In this paper, a VLE experimental apparatus based on the static analytic method is used to measure VLE data for CH₄/H₂ system at seven different temperatures ranging from 100.1 K to 130.7 K and pressure below 3.5 MPa. Some commonly used equations are also used to analyze their reliability for predicting the VLE properties of the CH₄/H₂ system at cryogenic temperature and low pressure.

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Introduction

Hydrogen is commonly regarded as one of the most important types of energy in the future. But hydrogen itself is not abundant, it usually needs to be produced in some ways. As we all known, hydrogen appears in the product of the coal or biomass gasification process [1–3]. The product is usually called synthetic gas, and it may be further transformed into synthetic natural gas (SNG). Considering the large reserve of coal, producing hydrogen from coal gasification may be an important way of hydrogen production. Since synthetic gas and SNG are all hydrogen-containing mixtures, the vapor-liquid equilibria of these mixtures are of significance for the hydrogen industry. This paper will focus on the vapor-liquid equilibria of methane and hydrogen.

In view of the supplement for natural gas and positive effect in the clean and efficient use of coal, the interest in SNG

technology and its application has been increasing these years. The convenience of storage and transportation of SNG will be improved much if it is liquefied after its production. SNG is mainly composed of methane (95%–98%), hydrogen (1%), nitrogen and argon (1%–3%) after pre-treatment. Among those components, hydrogen shows obviously different thermal properties compared with other components. The existence of hydrogen, although not much, influences the thermal property of SNG and its further application in the liquefaction process [4,5]. Thus, acquiring the vapor-liquid equilibrium (VLE) data of the hydrogen-containing natural gas system, especially the CH₄/H₂ system is very important.

As the basis of thermal property, VLE data plays an important role in chemical and energy industry. And the knowledge of phase behavior is crucial to the design of industrial plants and processes. There has been experimental study of VLE data for CH₄/H₂ system since 1930s. In 1957, Benham et al. [6] measured VLE data for CH₄/H₂ system at the temperature of 116.5 K,

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144.3 K, 172.0 K, with pressure up to 27.6 MPa. In 1972, Sagara et al. [7] reported experimental VLE data for CH₄/H₂ system at five different temperatures ranging from 103.1 K to 173.65 K and pressure up to 10 MPa. In 1980, Tsang et al. [8] remarkably increased the VLE data of the CH₄/H₂ system at ten temperatures in the range of 92.3 K–180 K and pressure up to 140 MPa to form the mixture critical line. Later, Hong et al. [9] also measured the VLE data in the CH₄/H₂ system at ten different temperatures and up to critical pressure. Hu et al. [10] measured the isothermal vapor-liquid equilibrium data for CH₄/H₂/N₂ system using an experimental apparatus based on the static analytic method, with the temperature range from 100.0 K to 125.0 K, corresponding to pressure from atmospheric pressure up to 4.5 MPa. However, from previous researches, the VLE data at cryogenic temperature and low pressure ($T < 130$ K, $p < 5$ MPa) which has been the typical working condition of SNG liquefaction process and other utilization were still scarce. The purpose of this research is to expand VLE data especially for the cryogenic temperature and low pressure condition related to the SNG industry. Therefore, with temperature ranging from 100.1 K to 130.7 K, pressure from 0.237 MPa to 3.107 MPa, VLE data of the CH₄/H₂ system were presented and analyzed in this paper. And based on the obtained data, results of several theoretic equations used for the phase equilibrium analysis of the CH₄/H₂ system were discussed.

The commonly used methods in the experimental measurement for VLE data include distillation method, circulating

method, bubble point/dew point method and the more recently developed one, static analytic method with online gas chromatographic analysis, and so on. In this paper, a VLE experimental apparatus is designed and constructed based on the static analytic method. The main apparatus sketch and the constant temperature system is very similar to the one used by Gao et al. [11] in the measurement of the solubility data of CO₂ in CH₄ and CH₄/N₂ mixture except for the part of gas chromatographic analysis for vapor sample. Moreover, a better modified measurement system aiming at improving the accuracy of the collected data is added to the whole experimental apparatus system.

Experimental apparatus

Apparatus sketch

The VLE experimental apparatus used in the research is shown in Fig. 1. Based on the static analytic method with gas chromatographic analysis, a mixture of CH₄/H₂ gases is introduced into the cell and kept until it reaches the vapor-liquid equilibria. Afterwards, the liquid and vapor samples are drawn off to the chromatographic for the composition analysis. The experimental apparatus is mainly made up of two parts: constant temperature system and measurement system.

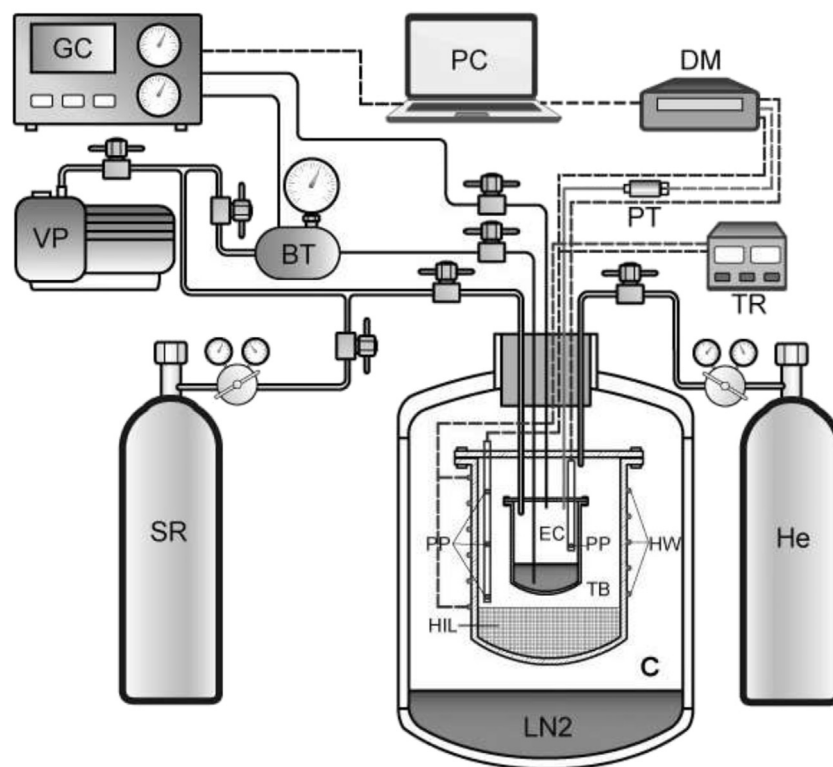


Fig. 1 – Static-analytic apparatus for VLE measurements. BT – buffer tank; C – cryostat; DM – digital multimeter; EC – equilibrium cell; GC – gas chromatograph; He – helium reserve; HIL – heat insulation layer; HW – heating wire; PP – platinum probe; PT – pressure transducer; SR – sample reserve; TB – thermostatic bath; TR – PID temperature regulator; VP – vacuum pump; PC – computer; LN₂ – liquid nitrogen.

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