



Impact of acetone on phase boundary of methane and carbon dioxide mixed hydrates



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ABSTRACT

In this work, the hydrate phase boundary of a gas mixture consisted of 29.9 mol% CH₄ and 70.1 mol% CO₂ is experimentally studied in the presence of 1–7 mol% of an aqueous acetone solution. Results indicated that acetone acts as a weak inhibitor on this gas mixture. In addition, enthalpy of hydrate dissociation for this system is reported. Based on the results, the inhibition effect of acetone is found to be decreasing with the increase of acetone concentration while the enthalpy of hydrate dissociation is increasing with the increase of acetone concentration within the studied range.

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1. Introduction

Gas hydrates are crystalline complex structures consist of hydrated polyhedron lattice, which acts as a host and entrapped guest molecules. Guest molecules, generally, may carry strong hydrophobic characteristics and none or weak polarity, which leading to immiscibility with water [1]. Still, some water soluble hydrocarbon can form clathrate hydrate due to minimization of Gibbs free energy of the guest and host molecules at high pressure and relatively low temperatures [2].

Increasing energy demand across the globe in the 21st century makes non-conventional fuel resources to become economically interesting. This includes the development of high carbon dioxide (CO₂) gas reservoir where CO₂ content can be more than 50 mol% of the gas originally in place. For example, K5 field located offshore of Sarawak, Malaysia with 25.65 trillion SCF gas reserve, shows evidence of bearing more than 70 mol% CO₂ [3]. In addition, biogas and landfill gas that can be classified as renewable energy resources, mostly consist of 45–60 vol% methane (CH₄) and 40–60 vol% CO₂ with traces of some other gases [4,5]. Economically, the high CO₂ gas requires an efficient CO₂ separation module to make these

resources marketable or even suitable as feedstock for manufacturing processes [3]. On the other hand, international regulations, such as Kyoto Protocol, emphasize on the reduction of greenhouse gases emission, particularly CO₂, to the atmosphere [6,7]. Therefore, both economic and environmental aspects are pointing to a need for a clean CO₂ separation process prior to the utilization of these new resources.

The common gas separation technologies such as chemical absorption, adsorption, cryogenic distillation and membrane separation, are incapable, both economically and technically, to capture the large amounts of CO₂ from gas streams. Some critical problems are associated with large energy consumption, corrosion, foaming, and low capacity. For example, estimations showed that the deployment of absorption capturing technology in flue gas cleaning from a modern power plant can reduce the thermal efficiency from approximately 45% to approximately 35% [8]. Therefore, development of new and existing capturing technologies should lower the energy requirements in order to minimize the overall costs as well as environmental impacts [9].

Utilization of gas hydrate as a separation technology is one of the alternatives. Separation of CO₂ from N₂ by forming gas hydrate in the presence of tetrahydrofuran (THF) was reported by Kang and Lee [10]. According to their investigation, utilization of hydrate technology is less energy intensive compared to common capturing

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process due to the moderate temperature condition of 273–283 K. In addition, using THF can decrease hydrate formation pressure and consequently decrease the overall energy consumption for the process [10]. There are several publications in the open literature that suggest hydrate formation process for separation of CO₂ from N₂, H₂ and some other gases. Eslamimanesh et al. (2012) presented a comprehensive survey of experimental studies dealing with separation of gases by hydrate formation technology [11]. However, separation of CO₂ from CH₄ received less attention in these studies. Seo et al. (2000) showed that separation of CO₂ from CH₄ is possible through hydrate formation process [12]. Both CO₂ and CH₄ are good gas hydrate former and their gas hydrate phase boundaries are relatively close to each other. This makes their separation through hydrate formation process more challenging. A suitable promoter to enhance the separation may be the key. THF as the most famous promoter is shown good promotion effects for both gases [13,14]. Consequently, it cannot be a good candidate to enhance their separation.

Acetone, as a common solvent, can be a good candidate for this purpose. Acetone has shown a thermodynamic promotion effect on methane hydrate formation at low concentration [14,15]. The promotion effects of acetone on methane hydrate is first observed by Ng and Robinson when they were looking for a new inhibitor for methane hydrate [16]. Acetone is a polar compound that is liquid at room temperature. At first glance, acetone seems to be an inhibitor, as it has many characteristics in common with alcohols. But, at low concentrations, it thermodynamically promotes the methane hydrate formation condition. The maximum promotion effect was observed at around 6 mol% concentration [17]. This promotion effect is converted to inhibition when acetone concentration is higher than 30 mol% [15]. On the other hand, acetone showed thermodynamic inhibition effect for carbon dioxide hydrate at all concentration [18]. Therefore, as acetone has different effects on pure CH₄ and CO₂ hydrates at low concentrations, it may show some selectivity on separation of these gases by hydrate formation process. Yet, prior to study the separation efficiency, the equilibrium phase boundary for such system should be studied and analyzed first. Acetone and water are well known for non-ideal behaviors and hence predictions of hydrate formation in such system through thermodynamic modeling have a considerable error. Thus, in this work, the phase boundary of gas hydrate for a system of water + acetone + CO₂ + CH₄ is experimentally measured and reported.

2. Experimental section

2.1. Material

Acetone with purity of 99.8% was purchased from Merck Millipore. A pre-mixed CH₄ and CO₂ gas mixture was purchased from Air Products Singapore Pte. Ltd.. The gas mixture ratio as specified by the supplier is 29.99 mol% CH₄ and 70.01 mol% CO₂. This CO₂–CH₄ ratio is selected in order to simulate the K5 field gas composition and the pre-mixed gas was used without any further purification. Deionized water was used to prepare aqueous acetone solutions.

2.2. Apparatus

A high-pressure cell, manufactured by Dixon FA engineering Sdn. Bhd. is used for the measurement of gas hydrate phase boundary. The schematic of the experimental rig is depicted in Fig. 1. The equilibrium cell is made of stainless steel and has an internal volume of 500 ml. The cell is equipped with a PT-100 platinum thermometer with the accuracy of ± 0.15 °C. A GP-M250 Keyence pressure transducer with the accuracy of $\pm 1.0\%$ full span

is used to measure the pressure inside the cell. In addition, a magnetic stirring system consisted of a 2-bladed pitch impeller and a 400 rpm motor is used to agitate liquid in the cell. The cell is immersed inside a thermostatic bath. The bath temperature is controlled by a PID controller with an accuracy of ± 0.3 °C. In addition, the bath temperature set point is programmable through data acquisition system. Pressure and temperature data is recorded every second.

2.3. Procedure

The -hydrate equilibrium points were measured by employing an isochoric method. The cell was washed using distilled water and dried. The extra air was removed from the cell using a vacuum pump. 100 cm³ of aqueous acetone solution at the desired concentration was fed into the cell through liquid injection point and gas was purged three times to ensure complete removal of air from the system. Then, gas was introduced to the cell until the desired pressure is achieved at room temperature. The stirring system was turned on and then the temperature was decreased to 273 K. System was kept at this temperature for 4 h to ensure gas hydrate formation. After that, the system was warmed stepwise to 293 K. At each step, the temperature was held constant for 30–240 min, according to the set point temperature and expected equilibrium temperature. The hydrate dissociation condition was determined through P-T diagram, as described by Tohidi et al. [19].

To produce the phase boundary for CO₂ + CH₄ + water + acetone system, four different acetone concentrations were studied at three different pressures. In addition, to validate the accuracy of the experimental setup, methane hydrate phase boundary in the presence of deionized water at three different pressures was measured and compared with literature data.

3. Results and discussion

3.1. Validation of the new setup

The pressure-temperature profile for CH₄-water system as a sample of the experimental procedure followed in this work is depicted in Fig. 2. The cooling step is set to be fast to shorten the experimental time. The rapid pressure drop is due to the hydrate formation. The heating part consisted of a fast heating step followed by several slow steps. The location of the change in the P-T slope, as shown in Fig. 2, is taken as the hydrate equilibrium point. The methane hydrate equilibrium point shown in Fig. 2 is 5.808 MPa and 281.35 K. In Fig. 3, CH₄-water equilibrium data points are included. As shown in this figure, the measured data coincides with pure methane hydrate data reported by Nakamura et al. [20].

3.2. CH₄ + CO₂ + acetone + water hydrates equilibrium data

The equilibrium points of CH₄ + CO₂ + acetone + water hydrates are tabulated in Table 1. Four different aqueous solutions of 1, 3, 5 and 7 mol% of acetone are studied in this work. In addition, blank deionized water is studied as reference condition. For each solution, the hydrate dissociation condition is reported for three to four different pressures between 2.68 and 5.36 MPa. In each experiment, the pre-mixed gas mixture of 29.9 mol% CH₄ and 70.1 mol% CO₂ is used as the feed gas. In each experiment, based on starting pressure and temperature condition the final concentration of gas and liquid phase composition is changed. In Table 1, the CO₂ and CH₄ concentration in gas phase is calculated by vapor-liquid equilibrium (VLE) calculations at equilibrium pressure and

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