



Interfacial tension between (methane + ethane + propane) gas mixture and water from 283.2 K to 298.2 K under up to 10 MPa



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ARTICLE INFO

Article history:

Received 7 November 2016

Received in revised form 9 January 2017

Accepted 11 January 2017

Available online 13 January 2017

Keywords:

Interfacial tension

Natural gas

Clathrate hydrate

Energy storage and transportation

Flow assurance

ABSTRACT

This paper reports a study of the interfacial tension between natural gas (methane + ethane + propane gas mixture) and water. The composition of the (methane + ethane + propane) gas mixture was 89.95:7.05:3.00 and 95.0:3.5:1.5, in mole ratio. These measurements were performed using the pendant drop method at pressure and temperature conditions of up to 10 MPa and between 283.2 K and 298.2 K, respectively. These conditions were chosen to eliminate the possibility of clathrate hydrate formation. Interfacial tension in the (natural gas + water) system decreased with increasing pressure, but no temperature dependence was found for any of the at all conditions. The interfacial tension gradient with respect to pressure was found to depend on the composition of gas phase. The interfacial tension in a (natural gas + water) system was found to be smaller than that of a (methane + water) system.

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1. Introduction

The International Energy Agency (IEA) [1] predicts that world energy consumption will continue to increase until 2040. The reduction of greenhouse gas emissions is one of the most important problems to be solved and, for this reason natural gas has an important role as an environmentally friendly energy resource. The multiphase flow dynamics of hydrocarbons and water in the oil and gas pipeline and the multiphase flow dynamics strongly depend strongly on the pressure and temperature conditions. Interfacial tension is one of the fundamental thermodynamic state functions, which controls the dynamics of multiphase flows in such mixed (gas + oil + water) flows.

Liquefied natural gas (LNG) is the most common medium for storage and transportation of natural gas. However LNG forming conditions require unfavourable operating conditions at low temperature. To solve this problem, the storage and transportation of natural gas via clathrate hydrate have been proposed [2–4]. Clathrate hydrate (hereafter referred to as simply hydrate) is a crystal compound, where water molecules consisting of hydrogen-bonded cage encloses a guest molecule. Hydrate can contain natural gases which would normally occupy a volume of 160 m³ in their gaseous form, within just 1 m³ of hydrate [5]. Natural gas hydrate forming conditions usually are at low temperature and

high pressure, but, once formed natural gas hydrate may be stored at 253 K under atmospheric pressure. This is due to the self-preservation effect [6–9]. For the transportation and storage of natural gas, the total cost of using natural gas hydrate is comparable with LNG. Additionally, natural gas hydrate is a fundamentally safe substance, with the chance that hazardous situations will occur far less frequently because of its crystal solid structure with water molecules and a flame spread speed much slower than that of natural gas [10,11]. It is generally known that hydrate preferentially forms at the interface between water and guest substances [12–14]. Increasing the interfacial area is key to designing efficient processes and various method have been examined, for example, the bubbling method and the spraying method [15,16]. To this end, accurate measurements of the interfacial tension between natural gas and water are helpful.

On the other hand, natural gas hydrate blocks oil and gas pipelines at low temperature and high pressure [17]. This occurs because of the emulsification of oil, water and natural gas mixture. Here an understanding of the interfacial tension between natural gas and water is also useful.

As mentioned above, the interfacial tension is the one of the most important thermodynamic properties to control hydrate formation and to understand multiphase flow dynamics such as the pipeline.

Sachs and Meyn [18] measured the interfacial tension between water and methane which is the main component of natural gas, using the pendant drop method at 298.15 K and at pressures up

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to 46.8 MPa, which are methane hydrate forming conditions. They showed that accurate measurement values may be successfully obtained using the pendant drop method. Yasuda et al. [19] measured the interfacial tension between methane and water at conditions from 278.15 K to 298.15 K, up to 10 MPa, which is around hydrate forming conditions. They revealed that the interfacial tension decreased with increasing temperature and temperature dependency could not be observed for these conditions. Khosharay and Varaminian [20] measured the interfacial tension of (methane + water), (ethane + water), and (propane + water) from 284.15 K to 312.15 K and for pressures up to 6 MPa. They reported that the interfacial tension in (ethane + water) and (propane + water) systems decreases with increasing pressure, as with the (methane + water) system.

Literature data of interfacial tension in (methane + water), (ethane + water), and (propane + water) systems have been reported but the interfacial tension for systems with a (light hydrocarbon mixture + water) has so far not been reported. In this study, we performed experimental measurements of the interfacial tension between natural gas (methane + ethane + propane) and water using the pendant drop method. The composition of the (methane + ethane + propane) gas mixture was 89.95:7.05:3.00 and 95.0:3.5:1.5 in mole ratio. Various experimental pressures from 1 MPa to 10 MPa and temperatures from 283.2 K to 298.2 K were tested. These conditions were chosen to eliminate the possibility of hydrate formation. In comparison with the literature data on methane-water systems, the effect of ethane and propane on the interfacial tension is discussed.

2. Experimental

2.1. Materials

The specifications and sources of the compounds used in the measurements are summarized in Table 1. A gas cylinder containing natural gas ($\text{CH}_4:\text{C}_2\text{H}_6:\text{C}_3\text{H}_8 = 89.95:7.05:3.00$, mole percentage) was purchased from Tomoe Shokai Co., Ltd. and by mixing natural gas and methane gas purchased from Takachiho Chemical Industrial Co., Ltd. A natural gas with composition ($\text{CH}_4:\text{C}_2\text{H}_6:\text{C}_3\text{H}_8 = 95.0:3.5:1.5$, mole percentage) was created. Deionized and distilled water prepared in our laboratory (model WG 222, Yamato Scientific Co., Ltd.) was used in all experiments. The electrical conductivity of the water was less than $0.5 \times 10^{-4} \text{ S}\cdot\text{m}^{-1}$. To confirm the reliability of the sample water used in this study, values of interfacial tension of water under a pressure of 0.02 MPa were determined and compared to the results obtained from the IAPWS reported correlation [21].

2.2. Apparatus and procedure

Fig. 1 shows the schematic illustration of the experimental apparatus used in this study. The apparatus and the details of the procedure were explained in a previous study by Akiba and Ohmura [22]. The apparatus has an inner volume of 25 cm^3 and was fitted with two glass windows at the front and back to allow

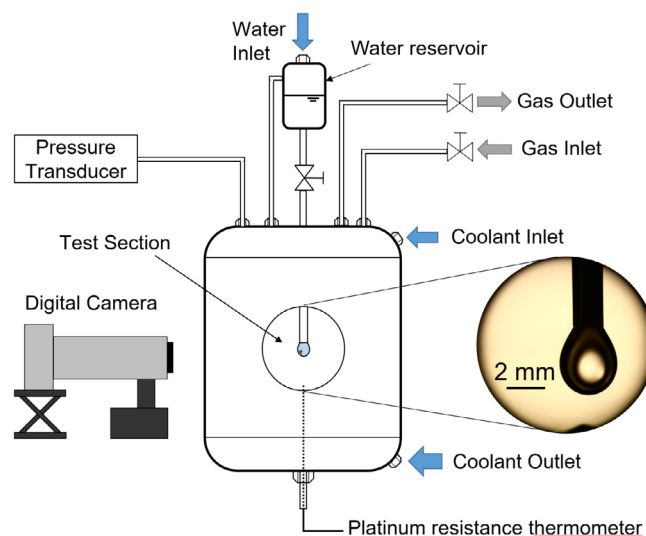


Fig. 1. Schematic illustration of experimental apparatus.

visual observation of the pendant drop. A cylindrical tube, made of stainless steel 1.583 mm in diameter, was inserted vertically into the test section to hold the pendant drop. The evacuated test section was filled with natural gas at the prescribed pressure which was measured using an FP101 pressure transducer supplied by Yokogawa Electric Corporation, which has a 95% uncertainty of 0.03 MPa. The apparatus temperature was controlled by circulating ethylene glycol aqueous solution and measured by a Class B Pt-wire thermometer supplied by Ichimura Metal Co., Ltd., with a 95% measurement uncertainty of 0.2 K. It took approximately 15 min to reach a constant value for the interfacial tension, with the interfacial tension measured every 1 min. The hanging droplet was maintained for 2 h to ensure that the droplet had reached steady state. Thereafter, the shape of each droplet was captured every 30 min for 2 h using a digital camera (EOS 50 D, Canon, Inc). We calculated the arithmetic average of the interfacial tension from the captured images and regarded these as the representative interfacial tension values for these measurements. The measurements of the interfacial tension were tested for four temperature conditions ($T = 283.2 \text{ K}, 288.2 \text{ K}, 293.2 \text{ K}, 298.2 \text{ K}$). The experimental pressures were determined to have not exceed the equilibrium pressure of natural gas hydrate for each of the temperature conditions and allowed us to avoid disruption caused by hydrate crystal formation. The equilibrium conditions of natural gas hydrate were predicted by CSMGem [23].

In this study, the conventional method of the selected plane was used to deduce the interfacial tension values which were calculated using following equations.

$$\gamma = \frac{\rho g d_e^2}{H} \quad (1)$$

$$\frac{1}{H} = f\left(\frac{d_s}{d_e}\right) \quad (2)$$

Table 1
The sample provenance and mass fraction purity of chemicals used in this work.

Sample	Source	Purity	Purity analysis
Methane + ethane + propane gas	Tomoe Shokai Co., Ltd.	$\text{CH}_4:\text{C}_2\text{H}_6:\text{C}_3\text{H}_8 = 89.95:7.05:3.00$, mole percentage with $\pm 0.001\%$ ($k = 2$)	Measuring mass by precision balance
Methane + ethane + propane gas	Laboratory made	$\text{CH}_4:\text{C}_2\text{H}_6:\text{C}_3\text{H}_8 = 95.0:3.5:1.5$, mole percentage with $\pm 0.1\%$ ($k = 2$)	Measuring mass by micro gas chromatograph
Methane gas	Takachiho Co., Ltd	99.99, mole percentage with $\pm 0.001\%$ ($k = 2$)	Measuring mass by precision balance
Water	Laboratory made	Electrical conductivity was less than $0.5 \times 10^{-4} \text{ S}\cdot\text{m}^{-1}$	Measuring electrical conductivity

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