



Experimental determination of hydrate phase equilibrium for different gas mixtures containing methane, carbon dioxide and nitrogen with motor current measurements



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ABSTRACT

Hydrate dissociation equilibrium conditions for carbon dioxide + methane with water, nitrogen + methane with water and carbon dioxide + nitrogen with water were measured using cryogenic sapphire cell. Measurements were performed in the temperature range of 275.75 K–293.95 K and for pressures ranging from 5 MPa to 25 MPa. The resulting data indicate that as the carbon dioxide concentration is increased in the gas mixture, the gas hydrate equilibrium temperature increases. In contrast, by increasing the nitrogen concentration in the gas mixtures containing methane or carbon dioxide decreased the gas hydrate equilibrium temperatures. Furthermore, the cage occupancies for the carbon dioxide + methane system were evaluated using the Van der Waals and Platteeuw thermodynamic theory with the Langmuir adsorption model and Peng-Robinson equation of state. The data demonstrated the increasing promoting effect of carbon dioxide with its concentration.

In addition, the motor current changes during the hydrate formation and dissociation processes were measured by keeping the rotation speed of the magnetic stirrer that was connected to a DC motor constant. The motor current measurements were reported and it showed that the hydrate plug formation and dissociation could be predicted by the changes in the motor current.

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1. Introduction

In recent years, the importance of gas hydrates has significantly increased in the energy sector. Large amounts of natural gas hydrate deposits were discovered beneath the permafrost areas and in deep oceanic sediments (Buffett, 2000; Kim et al., 2005; Sloan Jr. and Koh, 2008). These deposits present an enormous fuel resource (Makogon, 2010), $\sim 2.1 \times 10^{16} \text{ m}^3$ methane gas reserves were estimated (Kvenvolden, 1988), which is more than double the entire combined world reserves of oil, natural gas and coal (Makogon et al., 2007; Moridis, 2008; Sloan Jr. and Koh, 2008). Therefore, the oil and gas industry are now looking into commercially producing gas from these deposits, and preferably combine this production with the sequestration of carbon dioxide (Goel, 2006;

Eslamimanesh et al., 2012; Wood, 2015). In addition, gas hydrates have received growing attention because of their role in carbon dioxide capture to reduce carbon dioxide emissions (Dashti et al., 2015; Adeyemo et al., 2010), gas storage (Sun et al., 2003; Taheri et al., 2014) and transportation (Taheri et al., 2014), cool-energy storage (Wood, 2015; Xie et al., 2010) and water desalination (Eslamimanesh et al., 2012; Park et al., 2011). Thus such low temperature, high pressure reservoirs are a potential sink for anthropogenic carbon dioxide storage and climate change may be mitigated via this route (Kvamme et al., 2007). Secondly, gas hydrate formation presents the main flow assurance problem in the oil and gas industry (Sloan Jr. and Koh, 2008; Haghghi et al., 2009; Englezos, 1993). Here gas hydrate particles can agglomerate and build up gradually so that a large mass of hydrate is formed, which can block flowlines, valves, chokes and other production equipment (Sloan et al., 2010; Najibi et al., 2009).

Natural gas hydrates (clathrate hydrates) are solid ice-like, non-stoichiometric structures that consist of water and small gas

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Table 1
Review on the gas hydrate equilibrium for CH₄+CO₂, CH₄+N₂, CO₂+N₂ and water systems.

System	Reference	T/K	P/MPa
CH ₄ +CO ₂	(Unruh and Katz, 1949)	275.5–285.7	1.99–7.00
	(Adisasmito et al., 1991)	273.7–287.4	1.45–10.95
	(Dholabhai and Bishnoi, 1994)	277.56–284.84	3.41–17.90
	(Ohgaki et al., 1993)	280.3	3.04–5.46
	(Fan and Guo, 1999)	273.5–282.3	1.10–4.80
	(Servio et al., 1999)	273.5–283.1	1.7–5.070
	(Seo et al., 2001)	274.36–283.56	1.5–5.0
	(Beltrán and Servio, 2008)	275.14–285.34	1.92–7.47
	(Belandria et al., 2010)	279.1–289.9	2.96–13.06
	(Belandria et al., 2011)	277.9–285.5	2.72–8.27
	(Herri et al., 2011)	277.15	2.04–3.90
	(Sabil et al., 2014)	272.15–290.15	1.10–15.29
CH ₄ +N ₂	(Jhaveri and Robinson, 1965)	282.8–294.4	7.40–35.96
	(Mei et al., 1996)	273.2–279.8	2.64–32.42
	(Lee et al., 2006)	273.30–285.05	8.325–20.70
CO ₂ +N ₂	(Fan and Guo, 1999)	273.1–280.2	1.22–3.09
	(Kang et al., 2001)	273.75–284.25	1.565–32.308
	(Linga et al., 2007)	273.7	1.6–7.7
	(Bruusgaard et al., 2008)	275.00–283.00	2.0–22.4
	(Kim et al., 2011)	276.88–285.41	5.0–20.0
	(Herri et al., 2011)	273.40–281.10	5.60–6.10
	(Sfahi et al., 2012)	278.1–285.3	3.24–29.92
	(Sun et al., 2015)	273.4–278.4	5.28–17.53

molecules such as methane, carbon dioxide, nitrogen, ethane, propane or butane (Sloan Jr. and Koh, 2008; Delli and Grozic, 2014; Bishnoi and Natarajan 1996). Clathrate hydrates are classified into three categories based on the arrangement of the water molecules in the crystal structure and the size of the gas molecules (Carroll, 2014; Sloan Jr. and Koh, 2008); structure I (sI), structure II (sII), and hexagonal structure (sH) (Carroll, 2014; Sloan Jr. and Koh, 2008; Sloan, 2003). Typically, gas hydrates are formed and stable

Table 2
Gas mixtures used in this study.

Component Mol%	Mixture No.									
	1	2	3	4	5	6	7	8	9	10
CH ₄ %	100	90	84	80	90	80	69	64		
CO ₂ %		10	16	20					26	36
N ₂ %					10	20	31	36	74	64

under high pressure conditions at temperatures above the freezing point of water up to 25 °C if a gas hydrate former (i.e. a gas) and sufficient amount of water are available (Sloan Jr. and Koh, 2008).

In this context several experimental studies have reported hydrate equilibrium data for various gas mixtures, including for methane, nitrogen and carbon dioxide (Unruh and Katz, 1949; Adisasmito et al., 1991). Later, Ohgaki et al., (1993); Fan and Guo, (1999); Seo et al., (2000); Kang et al., (2001), Seo et al., (2001), Buruusgaard et al (Bruusgaard et al., 2008), and Sun et al., (2015), measured hydrate equilibrium data for CO₂ and CH₄ or N₂ while the N₂-CH₄ hydrate data were reported by Jhaveri and Robinson, (1965), Lee et al., (2006), and Mei et al., (1996). A summary of the experimental hydrate equilibrium data reported in the literature for the CH₄+CO₂, CH₄+N₂, CO₂+N₂ gas mixtures in the presence of water are listed in Table 1.

However, most of the existing experimental data are limited to low and medium pressure conditions, while gas hydrates naturally exist in a high pressure environment. Moreover, natural gas production from deep reservoirs requires hydrate prevention at high pressures: thus reliable and accurate hydrate equilibrium measurements are vital to formulate and validate thermodynamic models for predicting the hydrate forming conditions.

We thus measured hydrate equilibria for various gas mixtures (methane + carbon dioxide), (methane + nitrogen) and (nitrogen + carbon dioxide) for a wide range of temperatures and pressures.

2. Experimental methodology

2.1. Materials and gas preparation

Methane (purity 99.995 mol%), carbon dioxide (purity 99.9 mol %) and nitrogen (purity (99.99 mol%), all supplied by BOC Australia, were used as received or gas mixtures were prepared by mass balance as follows: empty 500 ml stainless steel bottles (Whitey DOT-3E1800 12EK082) were vacuumed using an Edwards Rotation pump (Model E2M2) for 30 min and then weighed using a high-precision electronic balance (Shimadzu model UW6200H, accuracy = 0.01 g). The vacuumed bottles were then filled with the gas (es) from the main cylinders and reweighed. The weight of the empty bottle was 1350 g. The weight difference was then converted

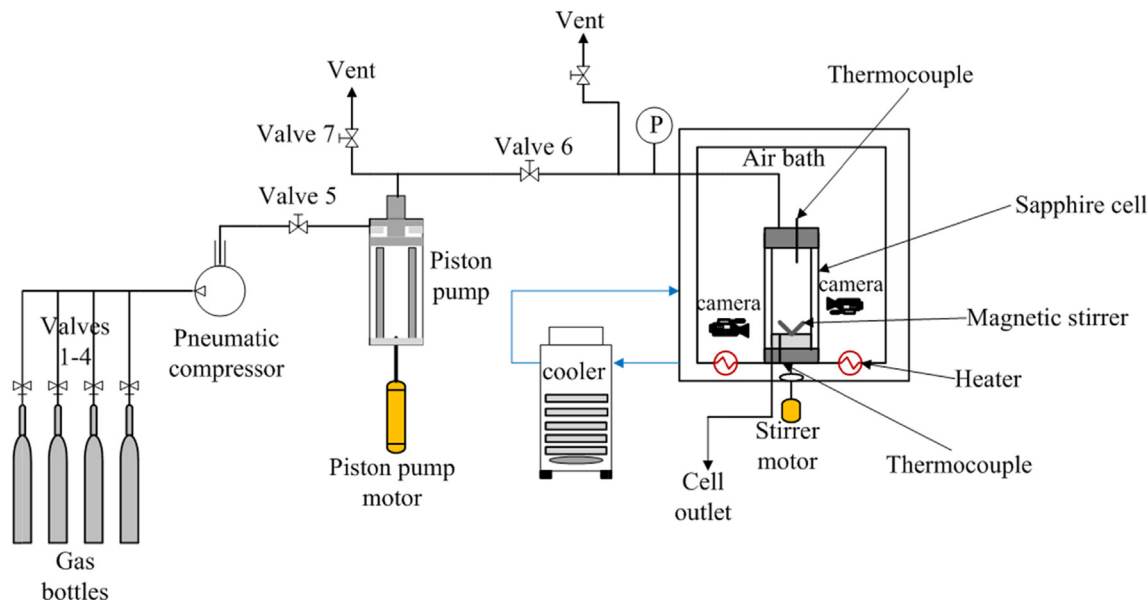


Fig. 1. Schematic of the PVT sapphire cell.

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