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Journal of Petroleum Science and Engineering

journal homepage: www.elsevier.com/locate/petrol

Experimental investigations on scaled-up methane hydrate production with surfactant promotion: Energy considerations



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ARTICLE INFO

Article history:

Received 31 October 2013

Accepted 15 June 2014

Available online 23 June 2014

Keywords:

methane hydrate
surfactant
scale-up
water spraying
hydrate formation
energy cost

ABSTRACT

An investigation about methane hydrate formation in a water spraying reactor with an inner volume of 25 L was carried out. In particular, the effects of changes in operational parameters were studied, in order to gain a deeper understanding of factors that play major roles on the energy cost of the whole process. Parameters changed include surfactant addition, spraying time, gas pressure, water amount loaded, and differential pressure on gas nozzles. Rapid methane hydrate formation is achieved only with sodium dodecyl sulfate surfactant promotion in all conditions tested, with reaction times in the range of few tens of minutes. Analysis of the energy cost for various steps of the process shows how reaction time (in the range used) and gas cooling work have quite negligible contribution, whereas gas compression work gives by far the main energy cost.

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1. Introduction

Gas hydrates are nanostructured crystalline materials in which small hydrophobic guest molecules are trapped inside cages of host water molecules (Sloan, 1998; Sloan and Koh, 2008). Growing interest has been focused in the last few years upon methane hydrate, due to the numerous possible applications in the broad area of energy and climate effects (Sloan, 2003; Sloan and Koh, 2008). Objects of specific investigations include:

- *Flow assurance*: oil and gas pipelines in cold regions/deep seas tend to form methane hydrates which eventually block the pipeline (Sloan, 2003; Di Profio et al. 2007). Research currently focuses on the search of hydrate inhibitors, especially low-dosage hydrate inhibitors (Masoudi and Tohidi, 2010; Seo and Kang, 2012).
- *Unconventional energy source*: the amount of energy stored in methane hydrate form in subcontinental margins and permafrost regions is estimated to be at least equal to the overall amount of the other fossil fuels combined, and the research about locating resources and methane production from hydrate bearing

sediments is increasing every year (Fitzgerald et al., 2012). Furthermore, methane, after its release, may be used in low environmental impact energy production devices such as molten carbonate fuel cells (Rossi and Nicolini, 2011).

- *Global climate issues*: methane is a much stronger green-house gas than carbon dioxide: a global warming of 4–8 °C in the geological past has been ascribed to an abrupt methane release from submarine hydrates (Kennett et al., 2003).
- *Separation of CO₂ from mixed gas in a form of gas hydrate*: it is expected to be an alternative method for capturing carbon at the extensive emission sites, or for purifying biogas or syngas (Li et al., 2010; Rufford et al., 2012); moreover simultaneous CO₂ sequestration and CH₄ production has also been recently considered (Bi et al., 2013).
- *Storage and transportation*: GH technology may be applied to the safe storage and transportation of natural gas in general.

In particular, storing and transporting natural gas as hydrate medium, especially from remote gas fields, is one of the most promising applications of gas hydrates. It is estimated that 70% of the total gas reserve is either too far from an existing pipeline or too small to justify a liquefaction facility (Sloan, 2003). Gudmundsson et al. (1992) and Gudmundsson and Børrehaug (1996) of Norwegian University of Science and Technology suggested that natural gas hydrates (NGH) are expected to become a new medium for energy storage and transport, and they also published a feasibility study showing a cost saving of 24% for the

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transport of natural gas in hydrates form compared to liquefied natural gas (LNG). The storage and transportation of gas as NGH is appealing for industrial utilization also because of its high safety for both human beings and the environment. In fact NGH do not explode in the event of a fire or tank rupture, but burn or release its contents slowly; NGH do not request heavy-duty containers for storing due to a much lower vapor pressure compared with normal pressure of loading of a pressurized tank; finally, NGH use water as storage medium, which has a “zero” environmental impact (Di Profio et al., 2007).

Despite mentioned advantages, the feasibility of using clathrate hydrates for industrial purposes has long been an intriguing subject of engineering studies (Lang et al., 2010; Okutani et al., 2008). Also the Japanese company Mitsui Engineering & Shipbuilding, which demonstrated operation of large scale plant and has been working on the improving the economics of transportation since 2001, is still searching to improve the GH production, and is currently focusing on development of methods for increasing the energy density improving pellet formation (Muruyama et al., 2011). Generally speaking, the main issues include the slow formation rates, unreacted interstitial water as a large percentage of the hydrate mass, economy of process scale-up (Lang et al., 2010). The lack of a technology that allows the production of hydrate in a continuous manner suitable for scale-up to industrial settings has hindered the utilization of gas hydrates in the possible applications. Hydrate formation is always the first process in any of these yet-to-be established technologies. Some authors proposed that the reduction of power consumption is of primary importance for the implementation of the NGH transport process, with more than 70% of power consumption attributable to hydrate formation in the absence of surfactants (Daimaru et al., 2007). Thus, how to efficiently form hydrates is a technical task research of rather wide interest (Lang et al., 2010).

Among the solutions to increase the hydrate formation rate and storage capacity, development and improvement of reactors have been carried out, with the aim of increasing mass transfer processes. Stirring reactors, and bubble towers have been tested, with some problems (Lang et al., 2010). Spraying reactor is a type of reactor for gas–liquid reactant systems, whose application for methane hydrate formation has been studied by Mori et al., who designed and modified spraying reactors (Ohmura et al., 2002; Tsuji et al., 2004). This kind of reactor has to be considered the best for applications to scaled-up systems, and a scaled-up spraying reactor for hydrate production was designed (Brinchi et al.,

2011; Rossi et al., 2012) and improved (Castellani et al., 2013) in CIRIAF lab.

In this work, this spraying reactor – already used in CIRIAF lab for CO₂ capture (Castellani et al., 2013) – was used to investigate methane hydrate production. The reactor is a scaled-up spraying reactor, with inner volume of 25 L, and 6 nozzles. The main objective of experimentation is to have a deeper understanding of the various contributions to the total energy cost necessary for the whole process of GH production, with the final aim of lowering energy costs of hydrate formation. The use of surfactants promotion is used. Their effects on chemical reaction rates and equilibria have been studied for many years, with special attention to room temperature conditions in aqueous media (Fendler, 1982). Currently, investigations of surfactant effects have enlarged to a wide broader temperature range also to exploit various applications (Di Michele et al., 2011, 2013; Weerasooriya et al., 2011). The previous studies reported that addition of surfactant to liquid water causes a drastic promotion on the rate of formation and occupancy of gas hydrates, and decreases the induction time, even in unstirred reactors (Zhong and Rogers, 2000; Di Profio et al., 2007, 2011; Ganji et al., 2007; Okutani et al., 2008; Lang et al., 2010; Ando et al., 2012; Lo et al., 2012). In particular, a recent investigation about a great number of surfactants (both commercial and not) of various charge type, various hydrophobic/hydrophilic balance, various chemical nature of the polar head group showed how negatively charged compounds are strong methane hydrate promoters (Di Profio et al., 2011). In the present work, anionic surfactant sodium dodecyl sulfate, SDS, was used which seems the best commercially available surfactant for improvement of methane hydrate formation (Di Profio et al., 2011).

2. Materials and method

2.1. The reactor

The general design of the present experiments followed that of previous tests performed in CIRIAF lab, in which CO₂ was used as guest gas (Castellani et al., 2013). Fig. 1 schematically shows the experimental setup used in the current investigation.

The main feature of the apparatus used for gas hydrate formation is a large volume: 25 L. Most laboratory reactors found in literature have a volume of about few hundreds mL, up to 1 L (Lang et al., 2010). A bigger size results in scale effects to be

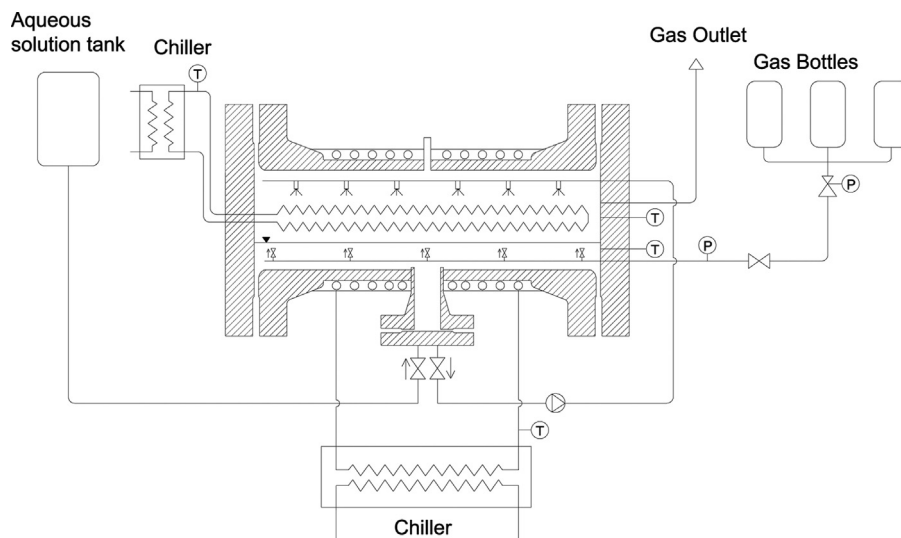


Fig. 1. Scheme of the reactor for gas hydrate production.

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