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Rheological study of CO_2 hydrate slurry in the presence of Sodium Dodecyl Sulfate in a secondary refrigeration loop

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ABSTRACT

Secondary refrigeration and thermal energy storage are promising solutions to enhance the performance of refrigeration systems and reduce the impact of refrigerants on the environment. To improve the energy efficiency of secondary refrigeration loops, phase change material (PCM) slurries with a high energy density, such as CO_2 hydrate slurries, can be used as a secondary refrigerant. In addition, hydrate-based processes could be an innovative option to capture CO_2 from flue gas. In both applications, the rheological properties of the CO_2 hydrate slurry have to be controlled. In the present study, CO_2 hydrate slurry in the presence of Sodium Dodecyl Sulfate (SDS) was studied in a dynamic flow loop. The results show that SDS used at concentrations of 1500–2000 ppm significantly decreases agglomeration and improves the flow properties of the slurry. Moreover, SDS helps decrease the viscosity of the CO_2-hydrate slurry at high fraction (> 10 vol%) and therefore could be suitable for use in industrial applications such as secondary refrigeration, in which hydrate slurries must be easy to handle.

1. Introduction

Over the past decade, gas hydrates have been the focus of attention in various fields such as refrigeration, gas transportation, water treatment and gas separation. Nowadays, refrigeration has a substantial impact on the environment and accounts for 8% of greenhouse gas (GHG) emissions: 80% of this impact is due to energy consumption and the remaining 20% is caused by refrigerant leakage, mainly Hydrofluorocarbon (HFC) fluids.

A number of international protocols have already begun to limit or prohibit the use of primary refrigerant fluids (Kyoto, 1997 or Montreal, 1985).

Due to the uncertainty surrounding the cost and availability of new refrigerants, secondary refrigeration could be considered as an alternative solution. This technology is effectively based on the use of an environmentally-friendly secondary fluid whose role is to transport cold energy from the place of production (engine room) to places of use (Guilpart et al., 2006). Thus, secondary refrigeration makes it possible to limit the amount of primary refrigerant used and to confine it. However, secondary refrigeration systems, unlike direct expansion ones (primary refrigeration), require additional heat exchangers and circulating pumps connected to the secondary loop that are responsible for exergy losses.

To overcome this problem, it is possible to use high energy density secondary fluids, such as phase change material (PCM) slurry (Zhang and Ma, 2012; Youssef et al., 2013), also called phase change slurry (PCS). In slurry systems, such as ice slurry (Ayel et al., 2003) or hydrate slurry (Fukushima et al., 1999; Fournaison et al., 2004), energy is stored during the phase change of the storage material (ice or hydrates) dispersed in a carrier liquid (continuous phase). PCM slurries have a higher energy density than single-phase secondary refrigerants due to both the sensible and latent heat capacities of the PCM.

Clathrate hydrates are crystalline structures that form by trapping guest molecules (e.g. CO_2, CH_4) (Sloan, 1998; Sloan and Koh, 2008). Some gas hydrates have a high dissociation enthalpy around 500 kJ kg_{water}^{-1} (Marinhas et al., 2006) higher than that of ice (333 kJ kg^{-1}). In the present work, the PCS is composed of CO_2 hydrate particles dispersed in an aqueous solution of Sodium Dodecyl Sulfate (SDS). One of the advantages of CO_2 hydrate slurry is that mechanical processes such as scraped- or brushed-surface heat

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Nomenclature

N	Number of moles of gas, mol	μ	Viscosity, Pa s
τ	Shear stress, Pa	Q	Volumetric flow rate, l h^{-1}
τ_0	Yield stress, Pa	R	Radius, m
$\dot{\gamma}$	Shear rate, s^{-1}	L	Length, m
n	Behavior index, (–)	u	Flow velocity, m s^{-1}
k	Consistency index, (Pa s^n)	ΔP	Pressure drop, Pa
		ϕ_s	Hydrate volume fraction, (–)

exchangers are not required to produce it, unlike ice slurry. It also forms at temperatures higher than 273 K which makes it suitable for air-conditioning applications (Zhang and Ma, 2012). But whatever the type of secondary refrigeration application, slurry flow properties are of paramount importance to assess the overall feasibility of the process.

Previous studies performed at Irstea have shown that CO_2 hydrate slurry in the aqueous phase can agglomerate in a dynamic loop (Delahaye et al., 2008, 2011; Jerbi et al., 2013), and even form plugs as in pipelines (Sum et al., 2011). Depending on whether the loop system studied does or does not have a stirred-tank reactor, hydrates can agglomerate and form plugs as from a high hydrate fraction of 20 vol% (with a stirred tank reactor) (Jerbi et al., 2013) or from a small hydrate fraction of 5–10 vol% (Delahaye et al., 2008; Jerbi et al., 2010).

Adding various chemical additives to the water before gas hydrate formation can substantially impact the thermodynamic equilibrium (Mohammadi and Richon, 2009; Trueba et al., 2011), the formation/dissociation kinetics (Ribeiro Jr and Lage 2008; Yoslim et al., 2010) and the physico-chemical properties such as wettability or adhesion force on hydrate particles (Zerpa et al., 2011, Aman et al., 2013). Among the additives tested on gas hydrates, SDS, along with tetrahydrofuran (THF), is one of the most studied and cited in the literature (Kumar et al., 2015). This anionic surfactant, SDS, has been found to enhance hydrate formation kinetics and the amount of hydrate formed with pure gas or gas mixtures (Ricaurte et al., 2014) in bulk or in porous media (Dicharry et al., 2013), even at very low dosage such as a few hundred ppm (Gayet et al., 2005). However, the action mechanism of SDS is not yet fully understood and has been debated in the literature for hardly more than 15 years. Interestingly, it has been suggested that SDS may have anti-agglomerant properties on hydrate particles (Zhang et al., 2007b; Torre et al., 2012), but no direct evidence of the "anti-agglomerant effect of SDS" has been provided to date in the literature for CO_2 hydrates.

This work presents a rheological study of CO_2 hydrate slurry in the presence of SDS carried out in a dynamic flow loop in order to observe the influence of SDS on slurry viscosity and agglomeration. The rheological behavior of CO_2 hydrate slurry with SDS has not yet been studied in the literature (see Table 3). This behavior is characterized in the present work by applying the capillary viscometer method (based on pressure drop vs. flow measurements), and the Herschel-Bulkley model is used in a first approach to represent the apparent viscosity of the slurry.

2. Materials and methods

2.1. The dynamic loop

A dynamic loop, described in previous works (Delahaye et al., 2008, 2011; Clain et al., 2012), is used to produce CO_2 hydrate slurry and to characterize its rheological properties. The loop is mainly composed of stainless steel pipes with an internal diameter of 8 mm (external diameter of 10 mm) and a total length of 2 m. A scheme of the apparatus is shown in Fig. 1. The total volume of the loop is 2.65 10^{-4} m^3. The loop is located in a cold room (6 m^3) whose temperature is controlled by PID. Temperature and pressure are maintained within the range of 268–293 K and up to 3.5 MPa respectively. The dynamic loop is also composed of a glass tube (with an inner volume of around 3 10^{-5} m^3) for detecting the formation of hydrate particles and visualizing hydrate slurry flow. The loop is equipped with a differential pressure gauge (ABB 265 DS, up to 0.02 MPa, ± 0.04%) to measure the pressure drops caused by fluid flow on a straight line of the loop (0.5 m), and with a pump (AxFlow GC-M25, maximum flow rate=0.17 m^3 h^{-1}) and an electromagnetic flowmeter (IFM6080K-type Variflux, ± 0.5%) to control and measure fluid flow. The device is equipped with 6 T-type thermocouples (± 0.3 K) and 2 pressure gauges (range: 0–5.0 MPa, accuracy 0.05%) (cf. Table 1).

2.2. Gas injection

A syringe pump (1000D ISCO) is used to control the CO_2 injected into the dynamic loop to form the CO_2 hydrate. Initially, gas is directly injected into the syringe pump which consists of a cylinder with a total volume of around 1000 cm^3. Pressure, volume and temperature are then used to calculate the number of moles of gas inside the syringe pump, $N_{gas}^{pump,\ i}$, based on a real gas equation. Afterwards, when gas is injected into the dynamic loop, the pressure in the syringe pump decreases (at a constant volume) and the number of moles of gas remaining in the syringe pump, $N_{gas}^{pump,f}$, can also be determined. The number of moles of gas injected into the loop, N_{gas}^{i}, is the difference between the initial and the final number of moles of gas in the syringe pump.

$$N_{gas}^{i} = N_{gas}^{pump\ i} - N_{gas}^{pump\ f} \quad (1)$$

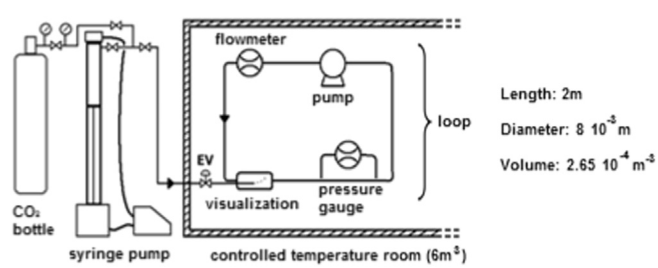

Fig. 1. Scheme of the dynamic loop.

Table 1
Detailed information on the equipment.

Name	Description	Range	Uncertainty
Pressure gauge	ABB 265 DS	0–0.02 MPa	± 0.04%
Pump	AxFlow GC-M25	0–0.17 m^3 h^{-1}	(–)
Electromagnetic flowmeter	IFM6080K-type Variflux	0.01–220 m^3 h^{-1}	± 0.5%
Thermocouples	T-type	3–673 K	± 0.3 K
Pressure gauges	(–)	0–5.0 MPa	± 0.05%

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