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Investigation of hydrate plugging in natural gas+diesel oil+water systems using a high-pressure flow loop

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ABSTRACT

To investigate hydrate plugging processes and hydrate plugging mechanisms, a high-pressure flow loop was newly designed and constructed where hydrate plugging experiments were performed from natural gas+diesel oil+water systems for a range of water cuts (30–100%) and initial liquid flow rates (1600–2400 kg h^{-1}). Based on the experimental data of hydrate morphology and flow parameters, hydrate formation and distribution characteristics in the flow loop were analyzed and two hydrate plugging processes together with the corresponding hydrate plugging mechanisms were proposed. For gradual hydrate plugging, the plugging process can be divided into four stages. Formation and growth of a hydrate deposition layer is the governing plugging mechanism. For rapid hydrate plugging, the plugging process can also be divided into four stages. Liquid stratification and a sharp increase in viscosity is the governing plugging mechanism for rapid hydrate plugging. In addition, silt-like hydrates and flocculent-like hydrate deposition layer were observed in gradual plugging experiments, whereas slurry-like hydrates with no obvious deposition layer were observed in rapid plugging experiments.

1. Introduction

Natural gas hydrates (NGH) are crystalline solids composed of water and gas molecules such as methane, ethane, propane, and carbon dioxide (Sloan et al., 2003). NGH are easy to form when the ambient temperature is relatively low and the ambient pressure relatively high (Sloan et al., 2010). Since the first explicit hydrate plugging incident was identified in 1934 (Hammerschmidt, 1934), NGH have always been found during the low-temperature, high-pressure process of oil exploitation and transportation (Sloan et al., 1998). In addition, with the development tendency of oil industry moving towards deep sea and ultra-deep sea, hazards caused by hydrate plugging are now posing a severe threat to the subsea flow assurance (Sohn et al., 2015; Li et al., 2015). The average expense for subsea hydrate prevention is approximately $ 1,000,000 per mile (Jassim et al., 2010). Considering such situation, many investigations have been conducted on hydrate prevention strategies. Among these strategies, thermodynamic inhibition and kinetic inhibition are most commonly used both in laboratory scale and industrial scale. Thermodynamic inhibition prevents hydrate formation by injecting the thermodynamic hydrate inhibitors (THIs), which can shift the hydrate equilibrium curve to a lower temperature and higher pressure condition (Karamoddin et al., 2014; Kim et al., 2015). Instead of THIs, kinetic inhibition uses kinetic hydrate inhibitors (KHIs) and anti-agglomerants (AAs) to slow down the nucleation and growth process of hydrate particles and to prevent the hydrate particles from agglomerating, respectively (Huo et al., 2001; Joshi et al., 2013; Sun et al., 2015). KHI and AA are collectively called the low dosage hydrate inhibitors (LDHI). Compared to THI, LDHI has the advantages of being economical and eco-friendly. More detailed information about LDHI can be found elsewhere (Kelland et al., 2006; Villano et al., 2011; Zhao et al., 2015).

Hydrate formation and flow characteristics are of great importance to plug prevention in offshore operations. Actually, researchers have performed a great deal of work and made many achievements on it (Taylor et al., 2007; Fidel-Dufour et al., 2005; Webb et al., 2014; Peng et al., 2012; Ding et al., 2016). However, much work still needs to be conducted to investigate the process of hydrate plugging. Until now, most related studies focus on the changing trend of flow parameters (e.g., temperature, pressure, flow rate, pressure drop) (Lorenzo et al., 2014; Chen et al., 2015; Grasso et al., 2014), and very few literatures involve hydrate morphology evolvements and liquid stratification phenomena during the plugging process. Greaves et al. (2008) carried out experiments on hydrate formation from water-in-oil (W/O) emulsions in an autoclave cell and found that hydrate formation and dissociation may turn W/O emulsions into o/W/O (oil droplets within water drops dispersed in oil) or even oil-in-water (O/W) emulsions.

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Nomenclature

D	inner diameter of the flow loop m
h_f	frictional head loss m
g	gravity m s^{-2}
L	length of the flow loop m
M_G and M_W	molar mass of natural gas and water, respectively kg mol^{-1}
n_G and n_{HW}	mole numbers consumed by hydrate formation for natural gas and water, respectively mol
v	mean velocity in the flow loop m s^{-1}
V_W and V_D	volume of water and 0# diesel oil that loaded in the flow loop, respectively m^3
ΔP	pressure drop along the flow loop Pa
λ	frictional coefficient -
ρ	fluid density kg m^{-3}
ρ_W and ρ_D	density of water and 0# diesel oil, respectively kg m^{-3}
Φ	hydrate mass fraction in the flow loop %
$\Phi_{deposit}$	hydrate mass fraction at the stable point %

Chen et al. (2013) observed five types of gas hydrate morphologies (clumpy-like, slush-like, flocculent-like, slurry-like, and powder-like) in water + diesel systems when screening and compounding AAs. However, the hydrate morphologies mentioned above were observed in a series of sapphire cell experiments. Therefore, the five morphologies could not represent the real conditions of hydrates in flowing pipelines precisely. In the flow loop experiment of CH_3CCl_2F hydrate slurry, Wang et al. (2008) found that hydrates would transit from slurry-like to slush-like with the increase of hydrate volume fraction. A similar phenomenon was observed in the flow loop experiment of tetrahydrofuran (THF) hydrate slurry conducted by Wang et al. (2010). However, in these two experiments, CH_3CCl_2F hydrate and THF hydrate were formed in tap water rather than a mixture of water and oil. Consequently, the experimental phenomenon should not be used directly to interpret the plugging mechanism in oil transmission lines.

The hydrate plugging mechanism is another research hotspot when handling hydrate risks. It is commonly accepted that hydrate plugging usually results from the combined action of several plugging mechanisms and the governing plugging mechanism for different systems differs a lot. For oil dominated systems, where water is well dispersed in the oil as droplets, hydrates initially form and grow on the surface of water droplets. As the hydrates growing process continues, more internal water in the droplet is consumed and the hydrate shell become thicker and thicker. Eventually, the entire water droplet would convert into a hydrate particle (Larsen et al., 2001). Hence, when there are enough hydrate particles in the bulk phase, the capillary liquid bridge force between different particles will lead to agglomeration and finally result in a plugging condition (Camargo et al., 2002; Zerpa et al., 2011). However, very little research has reported pipeline plugging directly caused by hydrate particle agglomeration. In addition to agglomeration, particle bedding and wall adhesion are the two other main plugging mechanisms in oil dominated systems (Grasso et al., 2015; Sjöblom et al., 2010; Nicholas et al., 2009). For high water cut systems and partially dispersed systems, the hydrate plugging mechanisms are still under research and no definite conclusions can be drawn (Joshi et al., 2013). Chen et al. (2015) investigated the hydrate plugging mechanism in systems of different water cuts by a series flow loop experiments. The experimental results demonstrated that the formation, shrinking and deposition of an oil-in-hydrate network could directly lead to plugging conditions in high water cut systems. A partially dispersed system is defined as a system where water partially exists as free water and partially disperses in the oil phase. Through flow loop experiments, Vijayamohan et al. (2014) noted that the growth of hydrate sheet on the pipe wall coupled with the bedding of hydrates formed in the bulk phase is the governing plugging mechanism in partially dispersed systems.

In the present work, a high-pressure flow loop with visual windows was newly constructed. Through this flow loop, hydrate plugging experiments in natural gas+diesel oil+water systems were performed for a range of water cuts (30–100%) and initial flow rates (1600–2400 kg h^{-1}). Based on the experimental results, two types of plugging conditions in multiphase flow systems were identified. Firstly, the study on plugging processes and plugging stages of the two conditions were given. Then, hydrate morphology evolvements and liquid stratification phenomena during the plugging processes were analyzed. Finally, based on plugging processes and the calculated hydrate mass fractions, two plugging mechanisms were put forward for each plugging condition.

2. Experimental section

2.1. Flow loop system

To investigate the process and mechanism of pipeline hydrate plugging, a high-pressure flow loop, which is shown in Fig. 1, was newly designed and constructed by the Shandong Key Laboratory of Oil-Gas Storage and Transportation Safety in China University of Petroleum (East China). The main body of the flow loop is composed of several stainless steel double pipes with an inner diameter of 26 mm and a total length of 24 m.

Together with a 21 L mixing tank and the high-pressure hoses used for connecting, the whole volume of the flow loop system is approximately 40 L. The design pressure of the system is 15 MPa and the design temperature ranges from 253.15 to 373.15 K. In order to observe the plugging process conveniently and directly, a circular visual window (Φ65 mm) and a rectangle visual window were equipped on the flow loop. In the present work, hydrate morphological evolvements were all observed through the circular visual window due to the vision-field limitations of the rectangle visual window. In experiments, the system temperature was controlled by two water chilling units and a magnetic centrifugal pump (circulating pump rather than booster pump) with a rated flow rate of 3000 kg h^{-1} was applied to circulate the mixture in the flow loop. In addition, flow parameters such as temperature, pressure, flow rate, and pressure drop can be collected by different sensors installed on the flow loop and then recorded by a PC-based data acquisition system during the experiments. The schematic diagram of the flow loop system is shown in Fig. 2.

2.2. Experimental materials

In this work, a mixture of natural gas, 0# diesel oil and tap water is

Fig. 1. High-pressure flow loop at China University of Petroleum (East China).