Journal of Petroleum Science and Engineering ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at ScienceDirect

Journal of Petroleum Science and Engineering

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

Occurrence characteristics of gas hydrates formed from seepage gas in sandy deposits

Xiao-Hui Wang [a], Ji-Sheng Sun [a,b], Li-Yu Jiang [a], Yi-Fei Sun [a], Chang-Yu Sun [a,*], Guang-Jin Chen [a]

[a] State Key Laboratory of Heavy Oil Processing, China University of Petroleum, Beijing 102249, China
[b] SINOPEC Safety Engineering Institute, Qingdao 266071, China

ARTICLE INFO

Article history:
Received 16 June 2016
Received in revised form
2 September 2016
Accepted 6 September 2016

Keywords:
Gas hydrates
Sandy sediment
Seepage gas
Percolation model

ABSTRACT

To better understand the process that hydrocarbon gas moves through permeable conduits into the gas hydrate stability zone, a hydrate formation system, mainly composed by a tube reactor, was designed and constructed. The influence factors, such as pressure, temperature, and gas flux, have been investigated. Here also providing a method to measure the induction time of hydrates formation in sandy sediments by analyzing the gas flow rate and pressure drop. Either increasing pressure or reducing temperature can shorten the induction time and improve hydrate formation rate. A percolation model was established to investigate the variation of physical properties of the sediments caused by hydrates formation from seepage gas. The results show that hydrate does not uniformly generate in the whole space of the sediments, but quickly nucleate and accumulate in part of the pore space. With the formation of hydrate, the channel of gas migration could be blocked, and the porosity and permeability of sediments would reduce accordingly.

1. Introduction

Gas hydrates are ice-like solid formed by a way that gas molecules enclosed in cavities of water molecules under favorable conditions, and widely distributed in marine sediments or the permafrost zone. Gases, such as ethane, carbon dioxide and propane can form gas hydrates, while methane is the most common gas that is abundantly found in naturally occurring gas hydrate sediments. As the organic carbon storage is twice as much as the world's proven mineral fuel, gas hydrates are also called future energy (Boswell and Collett, 2011). There is much interest in exploiting this kind of energy source, but many unknown aspects are needed to be addressed. To develop or improve methane recovery methods, it is important to mimic natural conditions in a laboratory and study accumulation behaviors of methane hydrates in host sediments (Eaton et al., 2007). According to the drilling results, gas hydrate-bearing cores are mainly composed of sandstone, mudstone, oil shale and siltstone, which supply pore space or fractures for hydrate accumulation (Li et al., 2014; Saw et al., 2013; Komatsu et al., 2015; Chong et al., 2016; Heeschen et al., 2016). Besides temperature and pressure, the physical properties of rocks, such as, porosity and permeability, may affect the formation and distribution of gas hydrates. Meanwhile, the variations of physical properties may also be sufficient to change local thermodynamic equilibrium conditions, causing gas hydrates not form within hydrates stability zone (Zhang and Han, 2010). The occurrence of gas hydrates in coarse grained sandy deposits is owing to a gas source close to the deposits and migration pathways conduct gas into the deposits (Ruppel et al., 2008), such as, site NGHP-01-05 and NGHP-01-07 in the Krishna–Godavari Basin (Shankar et al., 2013). Under the function of driving forces, such as, buoyancy, geopressure and hydrodynamics, gas can move upward from the deeper source into gas hydrates stability zone through some permeable conduits. Hence, the faults and fractures are important pathways for gas migration and beneficial for hydrates accumulation.

It is a complicated process for the formation of gas hydrates from seepage gas, which not only relates to solid, liquid, gas phase and mutual phase transition, but also accompanied with the variation of physical, chemical and mechanical properties. All these will lead to the variation of physical properties of the reservoir (Su et al., 2012; Wang et al., 2015a). Unlike the seepage process of water or oil in porous media, it is a special kind of fluid mechanics problem for the formation of gas hydrates during gas migration, because the temperature, pressure, porosity and permeability of porous sediments would change gradually (Klauda and Sandler, 2003). The presence of gas hydrate in the sediments significantly affects the bulk physical properties. A gradual reduction of

* Corresponding author.
E-mail address: cysun@cup.edu.cn (C.-Y. Sun).

http://dx.doi.org/10.1016/j.petrol.2016.09.012
0920-4105/© 2016 Published by Elsevier B.V.

permeability with increasing hydrate saturation was reported (Delli and Grozic, 2014). In addition, due to the memory effect for hydrate nucleation, the dissociated water is much easier to combine with gas to regenerate hydrate than directly from free water, making this process more complex (Buchanan et al., 2005; Zeng et al., 2006). Therefore, the regeneration of hydrates has an important impact on gas seepage and properties of hydrate bearing sediments (Freij-Ayoub et al., 2007; Su et al., 2012). Natural gas hydrates-cores are rare, costly, heterogeneous, technically difficult and almost always show some degree of damage (Choi et al., 2014). In addition, the original characteristics of the natural sample can be easily altered by pressure and temperature changes during the lengthy processes of coring, transportation, and storage, even with newly developed pressure core techniques. As an alternative, laboratory formed hydrate-bearing sediments are often used to develop the technologies of its exploration and exploitation. Kono et al. (2002) emphasized that the formation and decomposition of hydrates within sediments are important and significant, since these synthetic samples could almost exactly reproduce the actual hydrates existing in mother nature either in permafrost or in deep-sea sediments. There are a number of different techniques to form gas hydrates in sediments, which accurately mimicking gas- rich or gas- lean hydrate formation conditions. Normally, hydrates in the laboratory are formed in a sealed vessel by guest molecules and liquid water, then increasing the pressure until crystalline hydrates form (Waite et al., 2004; Seol and Kneafsey, 2009; Lee et al., 2010; Heidaryan et al., 2010a; Jarrahian and Heidaryan, 2014a; Kakati et al., 2014,2015). A technique developed by Stern et al. (2004) was performed in which hydrates were formed from powdered ice and pressurized guest circumstance. To analyze the stress strain behavior of hydrates-bearing deposits, methane hydrate samples were manufactured in a closed reactor using methane and ice powder as raw materials (Yu et al., 2011; Li et al., 2013). Yang et al. (2012) and Yuan et al. (2013) proposed a method to control hydrate spatial distribution by mixing frozen sand with cold water. Raw dry quartz sands are wetted until saturated with distilled water, then the temperature decreases to the predetermined temperature required for the gas hydrates synthesis (Li et al., 2012a, 2012b). Most practices to form CH_4 hydrate in sands by an excess-gas method or ice-seeding method, where hydrate preferentially nucleates at water-gas interfaces and eventually coats the surface of sediment particles or cements particle contacts (Choi et al., 2014). The hydrate-formation process with these methods can be relatively fast, but hydrate pore habit appears different from those found in natural sandy sediments. There are still many uncertainties in determining the amount of natural gas trapped in hydrates and the distribution of hydrates in nature. To well interpret the logging data and develop suitable technologies to recover this kind of energy source, it is meaningful to study the occurrence of gas hydrates in coarse grained sandy deposits. However, most of these studies were either limited to homogeneous media or assumed that the hydrate-bearing sediments are homogeneous distributed, which can seldom reflect the distribution and accumulation process of gas hydrates in actual situation. To better understand the process that hydrocarbon gas moves through permeable conduits into the gas hydrate stability zone, a hydrate formation system, mainly composed by a tube reactor, was designed and constructed. During the experiment, the pressure in the reactor was kept constant, gas flux and pressure drop between the inlet and outlet were recorded simultaneously. The induction time of hydrate formation in sandy deposits can be figured out through the curves of gas flux and pressure difference. Furthermore, a percolation model was established and has the capacity to evaluate the variations of porosity and permeability of sediments during hydrate formation from seepage gas.

2. Experimental

2.1. Apparatus

An experimental device was designed and built to simulate the occurrence of gas hydrates in coarse grained sandy deposits when natural gas moves through permeable conduits into the gas hydrate stability zone, and the schematic diagram is shown in Fig. 1. It mainly consists of five parts: a spiral tube reactor, water bath, pressure control system, gas drying system, and data acquisition system. The spiral tube reactor, with maximum working pressure of 16 MPa, was made by Jiangsu Haian petroleum research Ltd and composed of a pipeline (Φ10 mm × 2000 mm), so it can be easily placed into the water bath. Ethylene glycol solution is acted as the cooling/heating media to maintain the temperature of water bath, which can range from 263.2 to 373.2 K with an uncertainty of 0.1 K. The pressure control system includes methane gas cylinder, a pressure regulator, a back pressure valve, and two pressure gauges. Pressures are monitored by two pressure transducers with an accuracy of 0.02 MPa, which are mounted at the inlet and the

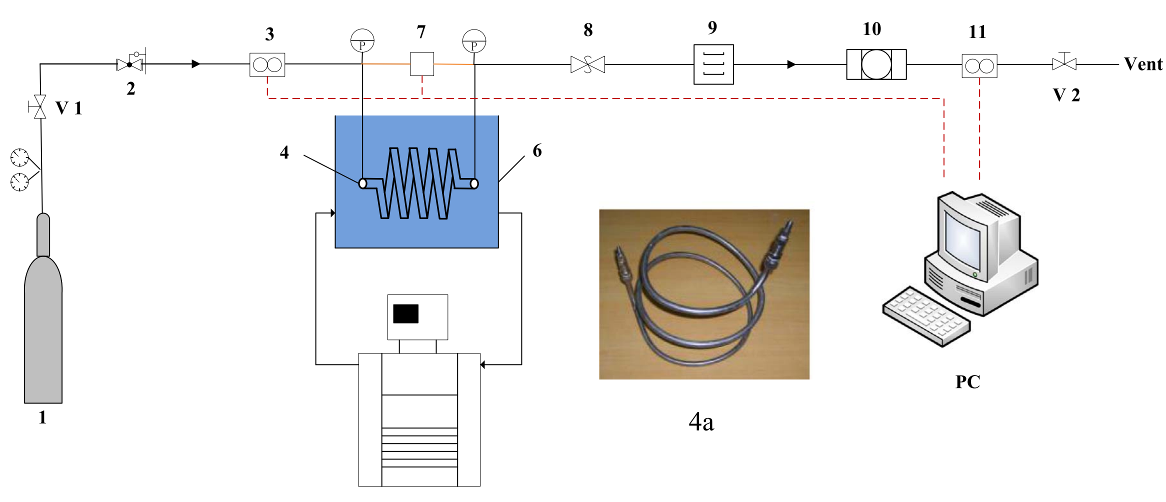

Fig. 1. Schematic diagram of experimental apparatus. 1 - gas cylinder, 2 - pressure reducing valve, 3, 11 - gas mass flowmeter, 4 - spiral thin tube reactor, 4a – picture of the tube reactor, 5 - chiller, 6 - water bath, 7 - Differential Pressure Gauge, 8 - back pressure valve, 9 - dryer, 10 - filter, P - pressure gauge, PC – computer.

Please cite this article as: Wang, X.-H., et al., Occurrence characteristics of gas hydrates formed from seepage gas in sandy deposits. J. Petrol. Sci. Eng. (2016), http://dx.doi.org/10.1016/j.petrol.2016.09.012