



Full Length Article

Hydrate formation in sediments from free gas using a one-dimensional visual simulator



Li-Tao Chen^{a,b}, Nan Li^a, Chang-Yu Sun^{a,*}, Guang-Jin Chen^a, Carolyn A. Koh^c, Bao-Jiang Sun^{b,*}

^aState Key Laboratory of Heavy Oil Processing, China University of Petroleum, Beijing 102249, China

^bSchool of Petroleum Engineering, China University of Petroleum, Qingdao 266580, China

^cCenter for Hydrate Research, Colorado School of Mines, Golden, CO 80401, United States

HIGHLIGHTS

- A one-dimensional visual hydrate simulator was built to study NGH formation.
- Hydrate grows from film formation, thickening, water adsorption intermittently.
- A colloidal solution formed from hydrate and salt water.
- Massive hydrate deposits in coarse sand and silt but not in clay marine sediment.
- Perturbation from flow or temperature can accelerate hydrate formation process.

ARTICLE INFO

Article history:

Received 25 November 2016

Received in revised form 2 February 2017

Accepted 14 February 2017

Available online 24 February 2017

Keywords:

Hydrate
Sediments
Formation
Morphology
Free gas
Electrical resistivity

ABSTRACT

To study natural gas hydrate formation in sediments, a one-dimensional visual hydrate simulator with three visual sapphire tubes and twelve electrode tips was designed and constructed. Using this device, the hydrate formation process was experimentally studied in coarse sands, silts, and natural clay marine sediments from free methane gas. During experiments, each of which lasting up to three weeks, a hydrate film formed initially at the gas-water interface and spread out to form a film network in sediments. The hydrate film grew in thickness by adsorbing water, where film thickening and water adsorption occur intermittently. The salting-out effect and the osmosis effect play a critical role in the hydrate film thickening-water adsorption process, and the perturbation arising from the fluctuation such as fluid flow and temperature may accelerate the hydrate formation. A colloidal solution formed from hydrate and salt water was also observed, which may be a new form of existence for marine natural gas hydrate. Electrical resistivity of the sediments initially decreased as hydrate formation increased the remaining water salinity, and then increased when additional hydrate formation restricted the water distribution in the sediment. Among three studied sediments, massive hydrate deposits were observed in coarse sand and silt but not in clay marine sediment, while fractures appeared when massive hydrates formed in sediments.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

Natural gas hydrates are crystalline compounds comprised of water and small guest molecules (e.g. methane or other light hydrocarbons), which are trapped in cages of a hydrogen-bonded water framework, at low temperature and high pressure conditions [1]. Natural gas hydrates are widely distributed in marine sediments and permafrost regions. Although estimates of the amount of gas trapped in hydrate-bearing sediments varies widely, natural gas hydrates still present a significant resource and may be

considered as a viable option for the future supply of natural gas [2]. Therefore, there is significant interest in studying natural gas hydrate-bearing sediments due to the vast energy potential these naturally occurring deposits may provide. Current understanding of the phenomena involved in gas hydrate formation and the physical properties of hydrate-bearing sediments have been reviewed previously [3,4].

When free gas is present in sediments, a hydrate film may form initially at the gas-water interface [5]. The hydrate film can surround sediment grains. A two-step model for hydrate growth in porous media has been proposed by Jin et al. [6]. In Jin's model, first, the hydrate film forms at the gas-water interface and then collapses inducing cracks on the film surface. Second, water enters

* Corresponding authors.

E-mail addresses: cysun@cup.edu.cn (C.-Y. Sun), sunbj@upc.edu.cn (B.-J. Sun).

the hydrate phase through the cracked film, resulting in further hydrate growth. Previous experiments have shown that hydrate formation can lead to cementing of coarse-grained sediment, which has been suggested to lead to fractures in the sediment [7]. Zhao et al. [8] and Song et al. [9] also showed from magnetic resonance imaging that hydrate formation in sediments is initially 'pore filling' continues to grow for around 1080 minutes. In more recent work, hydrate was considered to be 'load bearing' when the saturation was above 0.4 in a natural hydrate bearing sediment sample [10]. It has been suggested that hydrate film formation can occur within a very short time, while hydrate film thickening (massive hydrate formation) occurs over a much longer period. Film formation only determines the initial hydrate position, while hydrate film thickening determines the final position of massive hydrate [11–16]. Therefore, long term experiments are needed to study the hydrate film thickening or massive hydrate formation since significant water migration/redistribution may occur due to hydrate formation, which in turn affects massive hydrate formation [17,18]. In hydrate formation studies, the variation of temperature can be used to detect the appearance of hydrate [19–23], while the electrical conductivity [24–27], acoustic velocities [26,28,29], and thermal conductivity [30,31], can be a good indicator of the amount of hydrate that has formed.

Fractures in sediments are the best tunnel for free gas to migrate and therefore they are the best location to form massive hydrate. In fact, fracture-hosted methane hydrate deposits are commonly observed in nature [32]. However, laboratory observations of fractures in sediments caused by hydrate formation have been limited so far. Considering the influence of sediment type on hydrate formation, smaller grain sediments can lead to a faster hydrate formation rate [33]. Heesch et al. [34] investigated on gas hydrate growth using glass beads, silica powder, and clay minerals and found that there was a strong particle size effect on the kinetics of methane hydrate formation. Ruffine [35] used three mixtures of clays and sand as geologic matrix to study the influence of clay content on the hydrate morphology. Visual observations showed that the morphology shifts from disseminated through massive to mossy hydrates with increasing proportion of clays. Chong et al. [36] examined the effect of silica sand grain size on methane hydrate formation including four sizes of silica sands ranging from sand–silt cut off size (0.063 mm) to granular pebble (3.0 mm). Saw et al. [37] investigated the effect of bentonite clay on methane hydrate formation and dissociation in synthetic seawater of salinity 3.55% of total dissolved salts.

In summary, there is limited understanding of hydrate formation in sediments, especially over long time scales. Furthermore, the mechanism of massive hydrate formation is still unclear. In this work, in order to study hydrate formation and accumulation along with the fluid migration path (ideally in the vertical direction), namely one dimension, a one-dimensional visual hydrate simulator (ODVHS) with a height-to-diameter ratio of 40 was designed and constructed. Eight long-term (3 weeks each) experiments on hydrate formation and accumulation from upward migrating free methane gas in coarse sands, silts, and clay marine sediments were performed in order to reveal the mechanism of massive hydrate formation in sediments. Visual observations of hydrate formation were made through the transparent sapphire tubes. From detailed analyses of the visual observations and electrical resistivity data, we propose a new hydrate formation mechanism in sediments based on the long time scale experiments. The phenomenon of hydrate adsorbing water and the formation of a hydrate containing colloidal solution are reported here for hydrated sediment systems. Surrounding of sediment by hydrate and the occurrence of fractures in surrounded sediments were observed. It was also found that turbulence could accelerate hydrate formation.

2. Experimental section

2.1. Apparatus

The one-dimensional visual hydrate simulator includes a tubular reactor, which consists of three sapphire tubes and four stainless steel tubes assembled together with associated instrumentation. A schematic diagram of the ODVHS is shown in Fig. 1(a). Fig. 1(b) and (c) show a schematic diagram and a photograph of the tubular reactor, respectively. The length of the reactor is 1.0 m and the volume is about 0.51 L. There are three sapphire tubes (2.54 cm inner diameter, 6.0 cm out diameter, 10.0 cm height) and four stainless steel tubes (316L, 2.54 cm inner diameter, 15.0 cm or 20.0 cm height). The seven tubes (sapphire and stainless steel) are assembled to make a vertical tubular simulator, which is fixed on a steel frame. A heater is installed on the bottom stainless steel tube. Two porous plates (porous sintered stainless steel, one at the top of the simulator and the other at the bottom) are used to retain the sediments.

Four resistance temperature detectors (RTD, type PT100, T1–T4) are installed on the tubular reactor. The RTD sensors are installed in the middle of each stainless steel tube, with the temperature detecting position at the center of the tube. Two pressure transducers (Trafag 8251 model, one is installed at the top of the reactor and the other one is at the bottom) are used to measure the pressure in the system. Combining the data acquisition modules (KLM-4542S and KLM-4514 model from Beijing Kunluntongtai Automatic, Inc.), the temperature measurement accuracy is ± 0.1 K and the pressure measurement accuracy is ± 0.05 MPa.

There are 12 electrode tips in the reactor. The electrode tips are comprised of a wire and Teflon rod. One end of the wire is peeled and twined on the Teflon rod. The Teflon rod is fixed in the center of the tubular reactor so that the measurement point is fixed. Three Teflon rods are installed in the simulator in total. Each rod has six measurement points numbered 1–6 (Fig. 1(b)). The electrode tips are wired out through a thread guide in the middle of the stainless steel tube. An LCR meter (DF-2819 model, Ningbo Zhongce Electronics, Ltd.) is used to measure the electrical resistivity between the two electrode tips. Two measurement points (such as 1–2 = R1, 2–3 = R2, 4–5 = R3, and 5–6 = R4) are connected and then the electrical resistivity is measured by the LCR meter. Each Teflon rod can be used to measure four electrical resistivity datasets. Twelve electrical resistivity datasets are measured in total (four datasets/rod by three rods). Two electromagnetic relays controlled by a square wave generator are used to switch the pair of measurement points.

Temperature and pressure are recorded by a data acquisition software package (MCGS, Beijing Kunluntongtai Automatic, Inc.) in real time. Sediment morphology in the sapphire tube is photographed by a digital camera. The ODVHS is housed in a large thermostated chamber (3.5 m in length, 2.2 m in width, and 2.8 m in height). The chamber temperature can be maintained at 258–293 K with an accuracy of ± 0.5 K.

2.2. Materials and experimental procedure

Three types of sediments (coarse sands, silts, and clay marine sediments) were examined in the experiments. Natural yellow silica coarse sands (20–40 meshes, porosity 38%) and silts (220–280 meshes, porosity 42%) are from Taihang Mountain (Hebei province, north of China). The clay marine sediments (mud, porosity 45%) were drilled from the Bohai seafloor (innermost gulf of north China) by China Oilfield Services Limited (COSL). All sediments were washed with deionized water and then thoroughly dried. Brine with a salinity of 3.35% (NaCl aqueous solution, weight

Download English Version:

<https://daneshyari.com/en/article/6475222>

Download Persian Version:

<https://daneshyari.com/article/6475222>

[Daneshyari.com](https://daneshyari.com)