Contents lists available at ScienceDirect



Perspective

Journal of Energy Chemistry



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

Energy-efficient methods for production methane from natural gas hydrates

Jun Chen, Yan-Hong Wang, Xue-Mei Lang, Shuan-Shi Fan*

Key Lab of Enhanced Heat Transfer and Energy Conservation, Ministry of Education, South China University of Technology, Guangzhou 510640, Guangdong, China

ARTICLE INFO

Article history: Received 21 July 2015 Revised 21 August 2015 Accepted 22 August 2015 Available online 4 September 2015

Keywords: Gas hydrates Exploitation Energy efficiency EROI Strategy

ABSTRACT

Gas hydrates now are expected to be one of the most important future unconventional energy resources. In this paper, researches on gas hydrate exploitation in laboratory and field were reviewed and discussed from the aspects of energy efficiency. Different exploiting methods and different types of hydrate reservoir were selected to study their effects on energy efficiencies. Both laboratory studies and field tests have shown that the improved technologies can help to increase efficiency for gas hydrate exploitation. And it also showed the trend that gas hydrate exploitation started to change from permafrost to marine. Energy efficiency ratio (EER) and energy return on energy invested (EROI) were introduced as an indicator of efficiency for natural gas hydrate exploitation. An energy-efficient hydrate production process, called "Hydrate Chain Energy System (HCES)", including treatment of flue gas, replacement of CH₄ with CO₂, separation of CO₂ from CH₄, and storage and transportation of CH₄ in hydrate form, was proposed for future natural gas hydrate exploitation. In the meanwhile, some problems, such as mechanism of CO₂ replacement, mechanism of CO₂ separation, CH₄ storage and transportation are also needed to be solved for increasing the energy efficiency of gas hydrate exploitation.

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All natural gas hydrate reservoirs can be divided into four categories which can be attributed to Class I. Class II. Class III. and Class

1. Introduction

Energy plays an important role in the history of human development. Firewood, coal, oil, natural gas and so on are considered as the energy resource for warmth, cooking, automobile and other human activities until now. However, environmental problems associated with the use of the energy also began to appear, and tended to be more serious. One of the ways to solve the contradiction between environmental problems and energy demand is searching for new and clean energy resource.

Natural gas hydrates now are expected to be one of the most important clean energy resources among unconventional resources of coal bed gas, shale gas, biogas, and hydrated gas. Natural gas hydrate is a non-stoichiometric solid, which often forms at lower temperature and elevated pressure. It is known to occur in both terrestrial and marine environments where natural gas and water are present, and where pressure and temperature conditions are in favor of hydrate formation. The amount of methane in the form of hydrate below the ocean floor was estimated to be about 20,000 trillion m³ in the world [1]. The amount of estimated natural gas hydrates is more than that of all the current fossil fuel energy combined as shown in Fig. 1 [2].

IV [4–10]. Class I is the flowable gas-water plus the natural gas hydrate layer, while Class II is the flowable water plus natural gas hydrate layer. Class III was considered as the natural hydrate layer sandwiched between impermeable layers, while Class IV is the low saturated natural gas hydrates distributed in homogeneous strata. A small part of hydrates in arctic can be classified to Class I hydrate as shown at the top of Fig. 2, which can be easily developed. However, large amounts of hydrate were distributed in marine area, which can be classified to Class IV hydrate reservoir with difficulty in exploiting as shown at the bottom of Fig. 2. The recoverable gas hydrate is less than 1/1000 with current technologies and economic feasibility. So, over the long term, to improve energy efficiency plays a major role in ensuring adequate future supplies of natural gas and moderating future energy prices. One of the methods to improve energy efficiency can focus

One of the methods to improve energy efficiency can focus on the exploited method. Thermal stimulation [11–16], depressurization [17–20], and inhibitor injection [21], are three common methods to exploit natural gas hydrate. New methods, such as replacement of hydrate by CO_2 , [22,23], may help improve energy efficiency in natural gas hydrate exploitation. In the meanwhile, new idea is also important for improving energy efficiency during natural gas hydrate exploitation. Energy efficiency ratio (EER) and energy return on energy invested (EROI) can be used as the

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^{*} Corresponding author. Tel: +86 2022236581; Fax: +86 2022236581. *E-mail address:* ssfan@scut.edu.cn (S.-S. Fan).



Fig. 1. Modified proportion of organic carbon in various forms from Kvenvolden [2].



Fig. 2. Gas hydrate resource pyramid from Schoderbek and Boswell [3].

reference to explain the energy efficiency during natural gas hydrate exploitation.

In this paper, progress of gas hydrate exploitation which includes experiments in laboratory and field test was reviewed and discussed from an economic point of view. Energy efficiency ratio and energy return on energy invested were introduced for natural gas hydrate exploitation. Finally, an energy-efficient hydrate production process, called "Hydrate Chain Energy System (HCES)", including treatment of flue gas, replacement of CH_4 with CO_2 , separation of CO_2 and CH_4 mixtures, and storage and transportation of CH_4 in hydrate form, was proposed for future natural gas hydrate exploitation.

2. Progress of gas hydrate exploitation

2.1. Research in laboratory

Many countries, including United States of America, Japan, China, Korea, France, Germany, Russia, India and so on are interested in natural gas hydrate exploitation.

In order to simulate gas hydrate reservoir in real environment, different experimental apparatuses have been designed. Yousif and Abass [24] designed an apparatus for gas hydrate dissociation in sediment core. The apparatus which collected a lot of informations about gas hydrate dissociation could be the earliest apparatus to simulate gas hydrate exploitation. Eaton et al. [25] adopted a high pressure reactor to simulate gas hydrate formation and dissociation in the seabed. The volume and rated pressure of the reactor were 72 L and 20 MPa, respectively. Visual window and ultrasonic were used to confirm and analyze gas hydrate formation and dissociation. A 117.8 L

pilot scale reactor was set up to conduct three-dimensional synthesis and simulation [26]. Konno et al. [27] have set the world's largest reservoir simulating vessel with the internal volume of 1606 L, which can be more close to gas hydrate reservoir in nature. Except for apparatuses, Rempel and Buffett [28] simulated gas hydrate formation in marine environmental condition by using their vimineous apparatus without stirrer. They first found that gas hydrate can form in natural porous medium without the existence of free gas.

In addition, a lot of properties, such as thermal conductivity [29], phase equilibrium [30], permeability [31], wave velocity [32] have been studied which may help improve energy efficiency for natural gas hydrate exploitation. Other properties are also studied. For example, the mechanical properties of the hydrate structure were tested and the replacement of gas hydrate by carbon dioxide was simulated at Dalian University of Technology [33,34]. Li et al. [35,36] also have conducted a series of experiments to measure physical parameter of gas hydrate.

Gas hydrate formation and dissociation process also plays an important role in gas hydrate exploitation. Yusuke et al. [37] observed CH₄ and Krypton (Kr) hydrate dissociation at the surface of ice using an optical scanning microscopy. They found that ice formation behavior is different with different dissociation methods, which means different methods can change energy efficiency in natural gas hydrate development. A high pressure reactor was used to study gas hydrate formation and dissociation for gas hydrate exploitation in China University of Petroleum (Beijing) [38,39]. The results showed that ice are likely to form at the initial gas hydrate dissociation, and gas hydrate dissociation was controlled by heat transfer. From the results reported by Yoshihiro et al. [40], depressurization-induced gas production can be accelerated in the ice-formation regime. Du et al. [41] have in situ studied gas hydrate formation and dissociation for gas hydrate exploitation in a two-dimensional reactor. Qindao Marine Geological Survey also conducted experiments of hydrate dissociation. They concluded that gas hydrate was easily affected by heat transfer. The heat transfer efficiency of the porous medium is decreased by the center direction. Hydrate dissociation rate and the distance from the heat source is two function attenuation relationship [42-44]. The national energy laboratory of national geological survey of America, University of Columbia also conducted gas hydrate formation and dissociation research [45-47].

Methods to exploit gas hydrate are also very important. Li et al. [48] conducted gas hydrate exploitation by injection of NaCl solution. The temperature and injected rate of NaCl solution were 60 °C, 80 °C, 100 °C and 12 mL/min, 15 mL/min, 18 mL/min, respectively. The results have shown that higher injected rate and temperature made gas production rate higher. However, increased injected rate and temperature would reduce energy efficiency. They thought that depressurization or injection of saline water may be more effective to exploit gas hydrate [49,50]. Mikami et al. [51] have analyzed replacement of CH₄ hydrate by CO₂ and found that formation of CO₂ hydrate would result in replacement process occurring only at the surface, which can prevent further CO₂ replacement process. Masuda et al. [52] have studied exchange ratio by injection of N_2/CO_2 mixtures into hydrate-bearing sediments. Injection rate and pressure are kept in 80 L/min and 7 MPa, respectively. The results have shown that higher exchange ratio existed in lower gas hydrate saturation. Exchange ratios were about 30% and 5% for gas hydrate saturation of 41% and 60%, respectively. They also thought that formation of N₂/CO₂ hydrate can prevent further replacement process. Qi et al. [53] studied the effect of pressure and temperature on the replacement rate by using a self-made gas hydrate device. The experimental results showed that temperature had a great influence on the replacement process. Because carbon dioxide formation condition is milder than that of methane hydrate formation, replacement efficiency can reach 48% at the pressure of 2.5 MPa in their experiments.

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