

Experimental investigation of different factors influencing the replacement efficiency of CO₂ for methane hydrate

Ye Chen^{a,b}, Yonghai Gao^{a,b,*}, Yipeng Zhao^a, Litao Chen^{a,b}, Changyin Dong^a, Baojiang Sun^{a,b,*}

^a China University of Petroleum (East China), Qingdao, Shandong 266580, China

^b National Engineering Laboratory for Subsea Equipment Testing and Detection Technology, Qingdao, Shandong 266580, China

HIGHLIGHTS

- Simulate the CO₂ replacement in methane hydrate-bearing deposits with an injection well and a production well.
- Evaluate the replacement efficiency at the methane hydrate saturation of 0–32%.
- Analyze the influencing rules of different sensitive factors on CO₂ replacement.
- Propose the corresponding suggests to improve CO₂ replacement efficiency with less cost.

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ABSTRACT

The objective of this research is to study the replacement efficiency of methane hydrate with CO₂, which is an important index to describe the use of CO₂ for this application and to evaluate its economic benefits. An experimental study was designed to simulate the replacement of methane hydrate with CO₂ in a low permeability, porous medium, and analyze the influence of different factors such as injection rate (0.5–1.5 ml/min), total amount (1.8–5.4 L), temperature (275–279 K) and pressure (3–5 MPa) by controlling these variables. The results indicated that the total amount at injection and the temperature had relatively significant impacts on the replacement efficiency. The CO₂ injection rate and pressure primarily affected the CH₄ production rate, rather than the total amount produced. The injection temperature and pressure should be comprehensively optimized for improved economic benefits. The ultimate replacement efficiency ranged from 22.9% to 44.6%, and the ultimate production ratio ranged from 3.35% to 13%, confirming the necessity of a comprehensive optimization of the influencing factors and implying that most of the large cavities had not yet been occupied by CO₂.

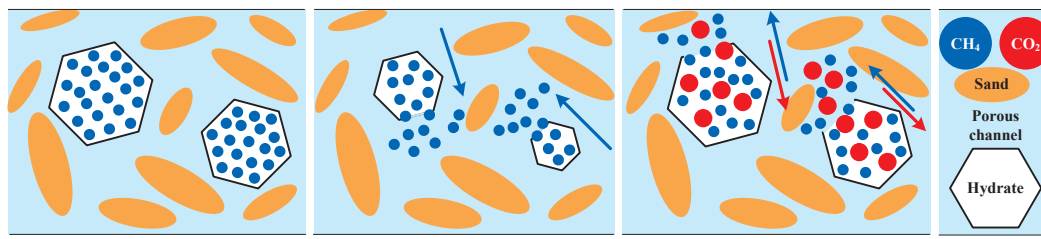
1. Introduction

Methane hydrate is a potential source of alternative energy, as it has abundant resources, a wide distribution and low pollution [1]. During the hydrate exploration process, dissociation and reformation phase changes will occur, which is different from conventional fossil fuels [2]. A series of developmental methods have been proposed based on the special characteristics of the hydrate [3]. Thermal stimulation, depressurization and inhibitor injection are widely used methodologies that are employed to make the natural gas hydrate dissociate into methane and water [4–6]. Through a series of experiments and theoretical analysis, depressurization is believed to be the most promising method because it is low cost and has been already applied in the USA, Japan and Canada [7,8]. However, when the solid hydrate dissolves, it gradually discharges a large amount of gas into the limited reaction

space. As a result, the mechanical structure of the entire layer will be dramatically changed or even destroyed, and geological disasters such as land collapse and landslides are easily induced by this artificial disturbance [9].

Injecting CO₂ into a hydrate reservoir for fuel production was first proposed in the 1990s and was further improved by other methods [10]. The injection of CO₂ not only allows CH₄ to be obtained from the hydrate but also buries large quantities of CO₂ under the ocean floor and slows down the greenhouse effect [11]. Research has found that the hydrophilicity of CO₂ is higher than that of CH₄ [12]. Under the same pressure and temperature, CO₂ becomes a hydrate more easily with a more stable chemical structure than CH₄. The newly formed CO₂ hydrate can maintain the mechanical structure of the pores, uphold the reservoir stability and reduce the possibility of geological disasters, as shown in Fig. 1. In addition, by simulating the hydrate layers with a

* Corresponding authors at: School of Petroleum Engineering, China University of Petroleum, Qingdao, China.
E-mail addresses: upcgaoyh@163.com (Y. Gao), sunbj@upc.edu.cn (B. Sun).

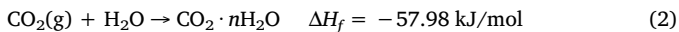
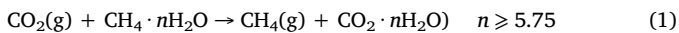


(a) CH₄ hydrate reservoir (b) CH₄ hydrate dissociation (c) Replacement process illustrations

Fig. 1. CO₂ hydrate replaces CH₄ hydrate in the reservoir.

remodeling core, the strength at failure of CO₂ hydrate-bearing sediment was found to be generally higher than that of CH₄ hydrate-bearing sediment, verifying the improved safety of CO₂ as a CH₄ replacement [13].

In the past few decades, CH₄-CO₂-H₂O phase equilibria curves have been plotted using experimental data [14]. It was believed that the transformation of CH₄ hydrate to CO₂ hydrate would slightly distort the host lattice and decrease the binding strength of the guest molecules, but it was dominated by an entropic contribution [15]. Accurate space-averaged, time-resolved, in situ data were measured by cryo-SEM, Raman spectroscopy, and neutron diffraction measurements to scatter the fluid components during CH₄-CO₂ exchange at conditions relevant to sedimentary matrixes of continental margins. The results reflected that the main aspect of the replacement took place during the second stage with slow, permeation-limited gas swapping [16]. The CH₄-CO₂ replacement that occurred via style II with natural gas hydrates for CH₄ recovery and CO₂ sequestration was also investigated with a primary focus on the thermodynamic, microscopic, and kinetic aspects [17]. The energy released during CO₂ hydrate formation can provide enough of a decomposition force for CH₄ hydrate once the replacement reaction starts, and can be described by the following [18,19]:



To date, related investigations have mainly focused on theoretical analysis, such as the feasibility of the kinetics and thermodynamics of this reaction [20,21]. CO₂ hydrate kinetics in porous media with and without salt have been studied [22], and the distribution of excess water during CO₂ hydrate formation and dissociation has been monitored [23]. The results indicated that an initial water-saturated state can effectively control hydrate saturation, but it is not the main controlling factor, compared to temperature and pressure. A two-dimensional CFD model of CO₂ hydrate mixture flow based on the Eulerian multiphase flow modeling approach was developed. This model used COMSOL Multiphysics built-in application modes and coupled the mass, momentum and energy equations [24]. In situ Raman and X-ray diffraction were also employed to record the dynamic process of CO₂/N₂ replacement [25,26]. Since efficiency and utilization are key factors for large-scale development, and other methods have already been evaluated [27,28], it is necessary to study the efficiency of CO₂ replacement while considering these main influencing factors in a porous medium. The efficiency is believed to depend on certain conditions such as temperature, pressure, and hydrate saturation [29,30].

The objective of this study is to carry out a series of experiments to simulate CO₂ replacement in a porous medium with an injection well and a production well. The natural gas hydrate-bearing samples with specific saturations are prepared by injecting methane and water into packed sand at low temperature and under high pressure. The horizontal permeability of the original porous medium can be calculated by Darcy's law. The feasibility of CO₂ replacement for a natural gas hydrate is verified, and both the corresponding ultimate replacement efficiency

and ultimate production ratio are obtained. In addition, four key parameters, including injection rate, total injected amount, injection temperature and injection pressure, have been simulated separately within a reasonable range [31,32]. Related influencing factors have been analyzed and compared, with the hope of providing theoretical support for optimization of the CO₂ replacement process.

2. Experimental apparatus and procedures

2.1. Experimental apparatus

The experimental system mainly contains a core holding unit, thermostat, injecting unit, back-pressure valve, buffer tank, constant pressure/flow pump, pressure gauge, and recovery and metering unit, as shown in Fig. 2.

The core holding unit, in which the replacement reactions take place, is the main part of the experimental set-up and has a working pressure of up to 20 MPa. It is a steel cylinder, 280–320 mm in length and 13 mm in diameter, with two lids blocking the sides and a sensor recording the temperature in the center. The removable lids can change the internal volume and compaction of the porous medium by adjusting the internal effective length. Since the sample prepared in these experiments is unconsolidated, the lids are equipped with a special-sized mesh, allowing the gas and water to flow through while preventing solid particles from migrating.

The injecting unit mainly contains air bottles, a multiport valve, buffer tank and constant pressure/flow pump. It is used to drive fluid, including methane, CO₂ and water, into the core holding unit. The multiported valve can make each fluid flow individually. The constant pressure/flow pump not only can provide the water necessary for hydrate formation but also serves as a driving force for gas injection. The real-time pressure of the inlet and outlet can be recorded by pressure gauges.

The thermostat uses a circulating liquid bath (using the ethanol solution as a cryogen) with a working temperature range from 263 to 373 K. The buffer tank is divided into two parts by a rubber sleeve and is filled with gas in the upper compartment and water in the lower compartment. The pressure and volume of the upper compartment can be controlled by injecting water into the lower compartment.

The system pressure can be maintained by a back-pressure valve. The recovery and metering unit contains a certain concentration of an alkaline solution and collects, separates and measures the drained gas.

2.2. Experimental procedures

First, the core holding unit should be cleaned by aqua distillate and dried with N₂ gas. To simulate the porous medium, a certain size of sand (270–550 μm) was packed into the core holding unit until the required volume was achieved. Then, the exposed ports were covered with the lids.

Aqua distillate was injected into the porous medium to drive out all of the original gas. The volume of the escaped original gas was recorded as V_p . Then, the aqua distillate was partially displaced by methane gas

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