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Experimental investigation on dissociation driving force of methane hydrate in porous media

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ABSTRACT

As one kind of clean and widely distributed unconventional energy resource, natural gas hydrate (NGH) is attracting increasing attention these years. In this study, the methods of depressurization and thermal stimulation are adopted to dissociate the NGH experimentally, when the initial temperature is 1.0 °C, the initial pressure is 3.1 MPa, the porosity is 0.35 and initial NGH saturation is 32%. The results reveal that during the depressurization process, there exists a pressure difference threshold (ΔP = 0.19 MPa), and similarly a temperature difference threshold (ΔT = 1.4 °C) exists during the process of thermal stimulation. NGH will not dissociate until pressure difference or temperature difference outweighs the corresponding threshold value. In addition, the influencing factors of driving force on the gas production performance have been studied. When thermal stimulation is adopted, the driving force of the temperature difference will grow with the increasing hot-brine injection rate and the brine temperature, in which case the gas production rate will drive up. When depressurization is used, the pressure difference driving force will go up with the increasing depressurizing range and depressurizing rate. Driving force is a major factor during NGH dissociation by depressurization and thermal stimulation.

1. Introduction

The NGH is crystalline solid composed of water and gas molecules, distributed mainly in marine sediments beside continental slopes and continental permafrost zones [1]. It is estimated that the amount of natural gas trapped in hydrates around the world is approximately two times larger than the recoverable gas and oil in conventional reservoirs [2,3]. Therefore, it is attracting increasing attention all over the world nowadays.

NGH could be dissociated through depressurizing or heating, which could happen when the equilibrium condition is broken. Dissociation driving force is defined as a kind of parameter that can promote the dissociation of hydrate, like the differential between system temperature and the equilibrium value or the differential between system pressure and the equilibrium value correspondingly. In 1987, Kim et al. established a kinetics model of hydrate dissociation and proposed that the hydrate dissociation rate is proportional to the total area of remaining hydrate and the differential fugacity of methane [4]. This theory is experimentally verified later by Sun et al. [5,6]. According to Kim's kinetics model, Yousif et al. developed a simulator (1 dimension, 3 phases), which takes depressurization-driving force into consideration, and fitted well with the experiments [7]. Goel et al. developed an improved model and showed that there is an exponential relationship between the hydrate dissociation rate and the differential driving force [8]. Kamath et al. conducted a research on thermal dissociation of methane and propane hydrate, and showed that differential temperature was considered to be the driving force of hydrate dissociation [9]. In 1989, Jamaluddin et al. proposed a new model coupled the mass transfer equations and the heat transfer equations, which showed that the fugacity difference and the activation energy of hydrate affected the driving force a lot [10]. In 1991, Clarke et al. investigated how porosity and hydrate saturation influenced the hydrate decomposition driving force. And the results showed that hydrate dissociation rate was proportional to the porosity and hydrate saturation [11]. In 2002, Kashchiev et al. studied the driving force of gas hydrate dissociation. They defined the difference between the chemical potentials of the old and the new phases as the driving force. And this difference was called supersaturation [12]. In 2005, He et al. established a particle shrinking dynamic model for methane hydrate dissociation, and the gas concentration difference between inside and outside of

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the phase interface was considered as the dissociation driving force, which indicated that interface chemical reaction was the control of the process [13]. In 2006, Zhao et al. found that the dissociation driving force had a linear relationship with apparent dissociation rate through experiments, and calculated the decomposition rate of constant [14]. In 2007, Tang et al. conducted experiments on gas production from hydrate-bearing core by depressurization with three different depressurization ranges, and got the intrinsic hydrate dissociation constant of 10^4 mol m^{-2} Pa^{-1} s^{-1}, by fitting the experimental data with TOUGH-Fx/Hydrate [15]. In 2009, Pang et al. researched the hydrate dissociation in a middle-sized reactor with thermal method, and concluded that the heat transfer rate and the thermodynamic driving force were the major controlling factors [16]. In 2012, Zhao et al. studied hydrate dissociation experimentally and concluded that there existed temperature difference in the radial direction during NGH dissociation [17]. In 2014, Zhao et al. established a two-dimensional axisymmetric simulator to study the sensible heat as well as heat flow transfer in the cap and base of sediment, and the effects of conductive and convective heat transfer on gas production [18]. In 2015, Li et al. conducted experimental study on hydrate dissociation and gas production behavior by thermal stimulation in a 2D experimental system and revealed that the heat injection pattern of continuous heat injection is superior to that of intermittent heat injection [19]. In 2015, Cheng et al. experimentally investigated the effect of sensible heat of the hydrate sediments and heat flow rate from over-underburden layers on hydrate dissociation by depressurization [20].

Apart from being a source of energy, NGH could be a disaster to the environment, since CH_4 is a greenhouse gas which has a large impact on global climate [21,22]. It is estimated that the effect of CH_4 on global warming is 10–20 times larger than that of CO_2 [23,24]. Dickens established a numerical simulator and investigated the effect of gas hydrate on global carbon cycling in 2003 [25]. Kvenvolden argued that compared with global climate change, a more potential effect of gas hydrate is on the submarine geohazard [26]. Boswell et al. indicated that the most promising global hydrate resource occurrences are also the most dangerous areas that were more likely to respond to climate change [27]. Rutqvist et al. developed a numerical model to analyze on the geomechanical stability of hydrate-bearing sediments, and revealed that the stability of hydrate-bearing sediments would be affected by gas production and some other factors [28,29]. Li et al. designed a triaxial system to investigate the mechanical properties of synthetic hydrate-bearing sediments and indicated that temperature had an important impact on the strength of hydrate-bearing sediments [30]. The dissociation of NGH could weaken the stability of wellbore and reservoir.

Therefore, it is very important to figure out the mechanisms of NGH dissociation, especially the driving force of NGH dissociation. Earlier researches mainly focused on establishing and describing the dynamic model of hydrate dissociation, but neglected the effects of driving force during the process, as well as gas production performance. In this paper, a series of experiments are conducted to study the decomposition driving force and its effects on hydrate dissociation rate and gas production.

2. Experimental apparatus and procedure

2.1. Experimental apparatus

Experiments are carried on a self-designed NGH production simulation system. Fig. 1 shows the schematic diagram of the experimental system. The 1D stainless steel tube is $\Phi 80 \times 800$ mm, and its working pressure is up to 20 MPa. 11 suits of thermocouples, pressure transducers and electrode probes are distributed along the axis equidistantly. The inlet of the tube is connected with gas cylinder and liquid supply equipment. The outlet is connected with back-pressure regulator and gas–liquid separator. The experimental data will be automatically and timely collected by the data collection and processing program.

2.2. Material and procedure

In this experiment, brine with a mass concentration of 2% has been used, the purity quotient of NaCl is 99.5%. The purity of methane is 99.9%.The diameter of the sand packed in the tube is ranged from 300 μm to 450 μm. After filling sand, the porosity is 33.4% and the water permeability is 1.2 μm^2.

The procedure is as follows:

(1) Hydrate isovolumetric formation. ① Inject the brine into the sand packed tube with the same injection rate to ensure full saturation. ② Displace a certain amount of brine by injecting methane. ③ Inject methane with a constant rate to the scheduled pressure, i.e. 8.6 MPa. ④ Close the inlet and outlet valves and then keep the temperature of the thermostat at

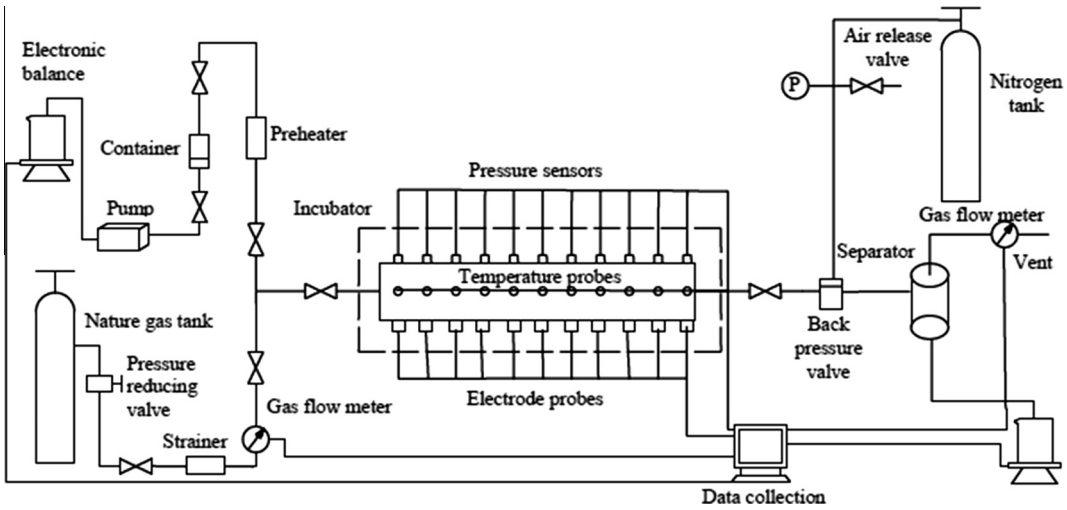

Fig. 1. NGH formation and dissociation of 1D experimental simulation system.