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Analytic modeling and large-scale experimental study of mass and heat transfer during hydrate dissociation in sediment with different dissociation methods

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ABSTRACT

An analytic model of the mass and heat transfers during hydrate dissociation in porous media by depressurization, thermal stimulation, and depressurization in conjunction with thermal stimulation without any empirical correlation is established in this work. Meanwhile, the PHS (Pilot-Scale Hydrate Simulator), a three-dimensional 117.8 L pressure vessel, is used for the investigation into the characteristics of the heat transfers and gas production behaviors during hydrate dissociation with the above dissociation methods. The model is solved analytically, and the predicted results are in good agreement with the experimental data. The results indicate that the sensible heat in porous media is firstly consumed for the hydrate dissociation, and then the heat transferred from the boundaries is employed for dissociating the hydrate in the reservoir by depressurization. The maximum deviations of the predicted gas production and the predicted temperatures with the model are 7.4% and 0.36%, respectively. With the thermal stimulation method, the maximum deviation of the predicted moles of the dissociated hydrate is 7.6%. There are two moving boundaries of the hydrate dissociation in the hydrate reservoir by depressurization in conjunction with heat stimulation. A synergistic effect of depressurization and heat stimulation on hydrate dissociation enhances the hydrate dissociation rates.

1. Introduction

Gas hydrates are ice-like, naturally occurring compounds that are composed of gas, typically methane (CH_4), physically trapped inside a solid lattice of water molecules. Gas hydrates form naturally under the conditions of the high pressures, the relatively low temperatures and the sufficient sources of methane and water. Under these conditions, methane molecules are compressed into very tightly packed ice-like cages. As a result, methane hydrates have high energy density. For example, the solid methane hydrate of 1 m^3 will release methane of approximately 160 m^3 when dissociated at normal surface temperatures and pressures [1,2]. In addition, the hydrate technology can be applied in the process of the hydrogen storage [3], sea water desalination [4], and the capture of carbon dioxide [5].

Scientists have known the existences of “clathrate” substances since their creation in the laboratory in the early 1800s. However, in the 1970s, the scientists began to suspect that gas hydrates could be a significant part of the natural environment. The first indications of naturally-occurring gas hydrates came from the Siberia and the Black Sea. By the mid 1990s, it was widely accepted that the volume of energy stored in gas hydrates could exceed that of all the world's coal, oil, and conventional natural gas combined. Today, it is known that gas hydrates is a potential energy resource, which occur naturally in Arctic regions and in shallow marine sediments along continental margins [6,7].

So far, the technologies for the commercial production of natural gas from the hydrate are still being developed. Recently, the known methods for the hydrate decomposition consist of (1) the depressurization method, with which the hydrate reservoir pressure is reduced below the equilibrium decomposition pressure to decompose the hydrates [8,9], (2) the thermal stimulation method,

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with which the hydrate reservoirs are heated above the equilibrium decomposition temperature to decompose the hydrates [10,11], (3) the chemical injection method, with which the chemicals (such as methanol or ethylene glycol) are injected into the hydrate reservoirs to change the equilibrium hydrate decomposition conditions, and thus decompose the hydrates [12,13], and (4) the CO_2 replacement method, with which the CO_2 is injected into the hydrate reservoirs to replace the methane gas [14,15]. A series of field production testes in hydrate reservoirs has confirmed the utility of these methods [16]. The researchers indicate that the depressurization method and the thermal stimulation method are the most practical methods. Meanwhile, the modeling and experimental studies are underway to advance our understanding of this potential energy resource [17,18].

Over the past two decades, some mathematical models and numerical codes have been applied for methane hydrate decomposition and gas production from the hydrate, and the laboratory experiments from the centimeter scales to the large scales for simulating field conditions have also been carried out [2,6]. Generally, the researches indicate that the heat transfer in the decomposing zone, the intrinsic kinetics of hydrate decomposition, and the multiphase flow during gas production are the three primary mechanisms during hydrate decomposition process in porous media [19]. The determining factor of gas production depends on the reservoir scale. For example, the determining factor in the core-scales is the intrinsic kinetics of hydrate decomposition, and the heat transfer and mass transfer are not the dominant factors in such small scales [20]. Conversely, in the field scales, the heat transfer and mass transfer become the predominant factors in the production stage. Thus, the phase change kinetics is negligible [20]. The hydrate dissociation is an endothermic process, which means the hydrate decomposition occurs with heat absorption. Thus, the heat transfer characteristics directly influence the efficiency and economy for gas production from the hydrate reservoir.

In last decades, a series of theoretical studies on heat transfer during hydrate dissociation had been reported. In 1985, Selim and Sloan reported heat transfer during hydrate dissociation as a moving-boundary ablation process [21]. However, this result was only useful in modeling of pure hydrate dissociation. In 1989, they presented a physical model, which describes hydrate dissociation under thermal stimulation in porous media. The model also views the dissociation as a moving boundary process [22]. The moving boundary separates the hydrate reservoir into the dissociated zone and the undissociated zone. In 1997, Makogon used the classical Stefan's melting model to describe the process of hydrate decomposition. The energy equation was used to describe the thermal condition of natural gas in the porous layer. The conductive and convective heat transfers as well as the effects of the throttling process were included [23]. However, most of the modeling studies were not verified from the large-scale experimental data. Recently, more and more experimental studies on the hydrate dissociation were presented. Li et al. [24] investigated the hydrate decomposition in a three-dimensional reactor. The results indicated that the hydrate dissociation in porous media by thermal stimulation is a moving-boundary ablation process. Zhao et al. [25] studied the hydrate dissociation by the thermal method, and he indicated that the heat is mainly transferred by conduction from the dissociated zone to the dissociating zone. The experimental investigation of Li et al. [9] indicated that hydrate dissociation by depressurization absorbs the sensible heat from the reservoir and the boundary. However, the dissociation rate is limited in the later stage when the heat is insufficient. Wang et al. [26] found that the thermal stimulation in conjunction with depressurization as a better choice for gas production from the hydrate. Generally, these experimental investigations focus on the gas production behaviors. However, the theoretical analysis for the heat transfer characteristics during methane hydrate dissociation is lack. It is significant to establish a theoretical model of the heat transfer during methane hydrate dissociation in the sediment.

In this work, a PHS (pilot-scale hydrate simulator), a three-dimensional 117.8 L pressure vessel, is used for the investigation into gas production behaviors from hydrate dissociation in porous media by depressurization, thermal stimulation, and depressurization in conjunction with thermal stimulation. The size of the vessel is big enough to distinguish heat-driven dissociation from the kinetics-driven dissociation. Thus, the experimental results are close to those for the field-scale gas production from hydrate reservoir. The analytic model of the mass and heat transfers in porous media without any empirical correlation is established to describe hydrate dissociation by depressurization, thermal stimulation, and depressurization in conjunction with thermal stimulation. The analytical solutions of the model are obtained and the predictions of the model are compared with the experimental data. In addition, the heat and mass transfer characteristics during each stage of these methods are analyzed.

2. Experiments

2.1. Experimental apparatus

Fig. 1 shows the schematic of the apparatus. This apparatus has been employed to investigate into the gas production from methane hydrate in porous media by the huff and puff method [27], the depressurization method [8], and the SAGD method [28]. The experimental apparatus involves a high-pressure reactor, a control system of ambient temperature, a gas and liquid injection system, a production control system, a data acquisition system, and some measurement units. The details of the experimental apparatuses have also been reported in the previous work [27]. The core component of the apparatus is the high-pressure reactor, which is named as the PHS. The PHS is a cylindrical pressure vessel, which is made of 316 stainless steel. The inner diameter is 0.50 m, and the length is 0.60 m (inner volume is 117.8 L). The external diameter is 0.80 m (wall thickness is 0.15 m). The apparatus is encircled by a water jacket (−15 to 30 °C, ±0.1 °C). The apparatus and the water jacket are all placed in a walk-in cold room (−8 °C to 30 °C, ±2 °C). The water jacket and the cold room constitute the ambient temperature control system. Fig. 2 gives the schematic of the inner PHS and the well design in the PHS. From this figure, we can find that the inner room of the apparatus is divided into 3 layers (Layer A–A, Layer B–B, and Layer C–C), and the distance between each layer is 0.15 m. There are two central vertical wells (W_{inj} and W_{pro}) along the centerline of the reactor, and four vertical wells (W_1–W_4) in the four corners of the reactor. The production wells (W_{pro} and W_1–W_4) are distributed on the top layer (Layer A–A), and the injection well (W_{inj}) is distributed on the bottom layer (Layer C–C). The W_{pro} is used in the depressurization method as the production well. The W_1–W_4 are used in the thermal injection method and the conjunction method as the production wells, and the W_{inj} is the injection well. In our previous work, this well design has been employed to investigate the gas production behaviors from methane hydrate in the CHS (cubic hydrate simulator) [11]. In the PHS, a total of 147 ($7 \times 7 \times 3$) thermal couples distributes on three layers. As seen in Fig. 2, the name of thermal couples can be illustrated as follows: as an example, the 49th thermal couples on Layer A–A to Layer C–C are named as T49A, T49B, and T49C, respectively.

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