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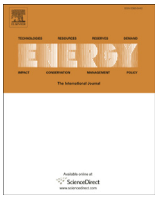

Experimental study on the effective thermal conductivity of hydrate-bearing sediments

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ABSTRACT

Gas hydrates are considered as a potential strategic energy source for sustainable development. The thermal properties of hydrate-bearing sediments govern the hydrate dissociation behavior and gas production process that accompany phase transformation and multiphase flow. This paper presents a thermistor-based measuring method to obtain the effective thermal conductivity of tetrahydrofuran hydrate-bearing sediments. The effects of different porosities, hydrate saturations and porous materials on the effective thermal conductivity were investigated. The porosity and the hydrate saturation were obtained using an X-ray CT (computed tomography) apparatus. The findings indicated that the effective thermal conductivity of hydrate-bearing sediments increased from 0.6468 W/(m K) to 0.7318 W/(m K) with porosity decreasing from 42.5% to 37.2%. Increasing hydrate saturations from 0% to 100% decreased the effective thermal conductivity from 0.7876 W/(m K) to 0.7318 W/(m K). Additionally, existing effective medium correlations were examined using the experimental data. The results showed that none of the existing correlations can suitably predict the measured data. Therefore, a hybrid correlation was proposed by optimizing the weighting parameters of the Parallel correlation and the Series correlation using the PIKAIA genetic algorithm. The agreement of the fitting correlation with the experiments is given, and the effective prediction of other researchers' work confirms the feasibility of our correlation.

1. Introduction

Natural gas hydrates, which exhibit high energy density, are clean and for which a large amount of resources are available, are generally accepted as a potential strategic energy form for sustainable development [1,2]. Small gas molecules, such as methane, can form ice-like solid crystalline gas hydrates by interacting with water framework cages under high pressure and low temperature [3]. Two different geographic settings favor gas hydrate formation: the permafrost and the continental slope, where low temperature and high pressure conditions naturally coexist [4].

Thermal properties, which represent the physical reality, lay the foundation for the theoretical research and engineering applications [5–8]. Therefore, studies of the thermal physical properties of gas hydrates are of significance not only for the future production of natural gas from gas hydrates (especially for the thermal stimulation method) but also for the mechanical stability of seafloor gas hydrate-bearing sediments, plugging problems of natural gas transport pipelines, global climate change and other issues [9,10]. As a vital thermodynamic parameter, the thermal conductivity is broadly employed to assess the heat transfer process [11–15]. However, measuring the thermal conductivity of gas hydrates requires the use of pressurized equipment and maintaining a low temperature. It is also difficult to synthesize a uniform gas hydrate sample [16].

Consequently, a THF (tetrahydrofuran) aqueous solution with a specific concentration can form THF hydrates at a temperature above the freezing point and atmospheric pressure. The physical and thermal properties of THF hydrates are similar to those of natural gas hydrates, making THF hydrates a convenient surrogate to study gas hydrates [17–19]. Krivchikov et al. [20] measured the thermal conductivity of tetrahydrofuran hydrates at a temperature range from 2 to 220 K using the steady-state potentiometric method, and the temperature dependence was analyzed. Andersson et al. measured the thermal conductivity of normal and deuterated tetrahydrofuran clathrate hydrates at temperatures ranging from 55 to 250 K using the transient hot-wire method [21]. The thermal conductivity was revealed to slightly increase with the temperature, which indicated a glass-like behavior. Moreover,

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researchers have also conducted abundant experiments and simulations to obtain the thermal conductivity of methane hydrates in porous media. The composite thermal conductivity of the sample was determined and compared, and the agreement of the calculated results with the experimental data was estimated [11,16,22,23]. However, the effects of the porous system properties, including the porosity, content saturation and physical characteristics of the porous materials, on the effective thermal conductivity of the hydrate-bearing sample were not systematically discussed. Furthermore, studies on these aspects are significant for realizing the actual thermal properties of the natural repository and the heat transfer process during gas production. In addition, the thermal conductivity is most commonly measured with the plate heat source method or the transient hot-wire method. Few studies have attempted to determine the thermal conductivity of the hydrate-bearing sediments using the point heat source method, which better reflects the actual local properties due to the smaller heating unit and thermally affected region.

Numerous effective medium correlations have been proposed to determine the effective properties of porous media [24–26]. The mixing law correlation is a type of classic and representative prediction correlation that assumes that the composite properties of a multicomponent system can be represented by an equation of the components properties and their saturation [5]. Huang et al. compared the measured thermal conductivity of THF hydrate-bearing sand porous media with the six classic correlations and found that the Woodside correlation agreed well with the experimental data [16]. Braun, Katika and Garahan et al. studied the effective optical and thermal properties of nanocomposite thin films using effective medium correlations [27–29]. Hamdami et al. measured the effective thermal conductivity of a sponge using a heat probe system, and the agreement of two predictive correlations of porous food (Krischer and Maxwell correlations) with the measured data was determined [30]. Additionally, Ould-Lahoucine et al. developed a thermistor probe method to measure the thermal conductivity of bentonite and mixtures of bentonite and silica-sand, and numerous prediction correlations were validated with the experimental data [31]. Gong et al. proposed a novel effective medium theory that unified the five basic structural correlations to predict the thermal conductivity of porous materials [32]. However, few studies of the effective thermal conductivity prediction correlation of multicomponent porous systems containing hydrates have been reported. These correlations are vital for researchers to understand the factors that influence the effective thermal conductivity of porous media that contains hydrates and to obtain these parameters without complicated measurements. Moreover, most existing effective medium correlations were revised or modified to suit a specific porous system; therefore, their applicability is limited. Furthermore, the volume fraction or saturation of each component is an important factor in the effective medium correlations. The saturation is traditionally obtained via calculations based on the experimental condition [33,34]. However, the visualization technique, which uses X-CT (X-ray computed tomography), can directly provide the actual distribution. Jin et al. investigated the structure of natural gas hydrates sediments using microfocus X-ray CT [35,36]. The spatial distribution of different components was obtained and the corresponding saturation was also determined. The calculation method proposed by Akin and Perrin et al. can also be adopted to analyze the properties of porous media [37,38]. However, few studies of the thermal properties of hydrate-bearing sediments using effective medium theory combined with the visualization method have been published.

In this study, a thermistor-based point heat source method was developed and employed to investigate the effective thermal conductivity of THF hydrate-bearing samples. Various sizes of silica sand and grains with different thermal conductivities were used to simulate porous media of different porosities and thermal properties for the formation of THF hydrates of varying saturation. An X-ray CT was used to acquire the porosity and THF hydrate saturation. The measured effective thermal conductivity was compared with the results calculated using the typical prediction correlations, and the agreement was evaluated. Moreover, a fitting correlation was developed by linearly combining the Parallel correlation and the Series correlation using the PIKAIA genetic algorithm. Lastly, the work of other researchers was predicted using our fitting correlation.

2. Material and methods

2.1. Experimental apparatus

The schematic of the experimental apparatus and circuit used in this study is shown in Fig. 1. A 50-ml vessel with a diameter of 25 mm was used to pack the grains for THF hydrate formation. This vessel was sufficiently large for the thermally affected region; therefore, it was considered infinite [39]. The operation temperature was controlled by a thermostatic water bath (FP51, Julabo Co., Germany) with a precision of 0.01 °C. The measuring method developed by Ould-Lahoucine et al. using a thermistor probe was improved to suit our experimental conditions [31,40]. The precision of the method was evaluated by measuring the thermal conductivity of pure materials as shown in Table 1. We embedded a calibrated thermistor probe (Shibaura Electronics Co., Japan) in the center of the packed grains, with the probe unit fully contacting the sample. The thermistor was then connected to the control circuit with a protecting variable resistance and a stabilized voltage supply. A data acquisition and switch unit (34972A, Agilent Co., United States) transformed the analog signals of the voltage drop of the thermistor and the current rise to digital signals and sent these signals to the computer.

A microfocus X-ray CT system (SMX-225CTX-SV, Shimadzu Co., Japan) was used in this study to determine the porosity and THF hydrate saturation of the sample. The maximum resolution of this apparatus can reach 4 μm/pixel. While scanning, we placed the samples on the precision rotation stage and the X-ray was emitted. A three-dimensional image was then obtained after rotating the stage 360° in 60 s. Liquid nitrogen was blown on the sample to control the surrounding temperature during scanning. The CT raw data were collected at a source voltage of 100 kV and a current of 40 μA. The CT images were acquired in this study with a 16-bit gray

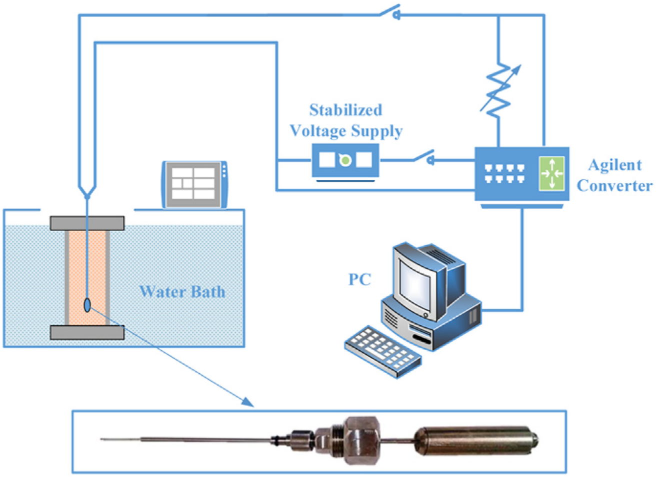

Fig. 1. Schematic of the experimental apparatus and circuit used in this study.

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