



# Experimental study on the effective thermal conductivity of methane hydrate-bearing sand



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## ABSTRACT

The thermal conductivities of methane hydrate-bearing sand samples, formed from moist sand with different initial water saturation levels, were measured by Gustafsson's transient plane source (TPS) technique. The thermal conductivity values show weak negative dependence on temperature similar to a crystal-like material, which agrees well with most of the published results on sedimentary and pure methane hydrates. Similar dependence has also been observed in the wave speed studies. The effective thermal conductivity of hydrate-bearing sediment is strongly dependent on the morphology of the sediment. In partially water-saturated, gas-rich environments, the hydrates tend to cement sediment grains together, and even a small amount of hydrates presence can significantly increase the effective thermal conductivity of the sediments. In sand samples with high water concentrations and water-saturated sand, the effective thermal conductivity does not show an obvious increase with the hydrate saturation level.

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## 1. Introduction

Sediments containing natural gas hydrates are discovered in permafrost areas and marine environments within the hydrate stability zone, where the pressure and temperature conditions support the stable existence of these compounds [1]. While natural gas hydrates are a promising potential future energy source [2], they may also play an important role in exacerbating global warming and may act as geo-hazards [3–7].

An understanding of the thermal properties of hydrate-bearing sediments is important to design the drilling and gas extraction processes that utilize gas hydrate resources. However, little data is available in the literature on the effect of various parameters on the thermal conductivity of hydrate-bearing sediments. Several studies have been conducted on the thermal conductivity of synthetic hydrate-bearing sediments [8–10] and various models describing the effective thermal conductivity of multiphase hydrate systems have been proposed [11,12]. Muraoka et al. [13] measured and compared the thermal properties of synthetic hydrate-bearing sediment samples with sand, water, methane, and methane hydrate using the hot-disk transient plane

source technique. The results show that thermal conductivity and diffusivity of the samples slightly increased as hydrate saturation increased from 0 to 0.3. They also performed thermal conductivity calculations using simple thermophysical models and pointed out that better prediction models are required to accurately determine the thermal properties of four-component systems. Most of these models describe the effective thermal conductivity as a function of the thermal conductivities of individual phases weighted by their volume fractions. Although predictions based on volume fractions are convenient to use, they are likely to be inaccurate because individual phases are unlikely to be distributed uniformly within the sediments. Moreover, the thermal conductivity of hydrate-bearing sediments may be influenced by complex interplay among various material characteristics such as particle size, effective stress, porosity, and pore fluids for some grained materials [14].

Based on the types of hydrate inclusions within sediments, two mechanically extreme cases of hydrate morphologies (hydrates that are cemented around grains or formed within pores) have been considered in the effective medium theory, which predict acoustic wave velocities [15,16]. However, Revil [17] modeled the effective thermal conductivity by scaling up the local equations using a differential effective medium approach and accounted for the pore space distribution and complexity of the thermal flow lines through the porous material by introducing a thermal formation factor. The main assumption of the model was that the continuity between the grains is small. By comparing the modeling

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results with the available experimental data, they found that the assumption was valid for unconsolidated sand-shale mixture sediments. However, microscale processes were not considered in this model. In fact, the variations in gas hydrate concentrations at different geological locations show the dependency on sediment mineralogy [18]. In hydrate-bearing sediments, gas hydrates can be (a) free-floating in the sediment matrix, (b) contacting and existing sediment grains, but not cementing or (c) cementing and stiffening the bulk sediments [19,20]. Moreover, for the exploration of gas hydrates, many physical properties depend primarily on the spatial distribution of hydrates in the pore space of sediments [21]. Kleinberg et al. [22] suggested that hydrates tend to replace the water in the pore spaces and form in the centres of the pores rather than as a coating on the grains. However, Waite et al. [23] used the rock physics models to differentiate the potential pore-space hydrates distributions and suggested the methane hydrates cementing the unconsolidated sediments. Moreover, hydrate saturation is another important concern. Hydrate formation may be influenced by various factors, such as gas pressure, temperature, particle size, porosity, pore fluids, etc. [24–29]. Zhao et al. [21] observed the microstructure and growth of gas hydrates in porous media by magnetic resonance imaging and confirmed the random nucleation of gas hydrates in porous media. Mekala et al. [30] found that the gas consumption of CO<sub>2</sub> in hydrate is more for smaller silica sand particle and decreases as the size of the sand increases. Moreover, the presence of salts significantly affects the kinetics of hydrate formation in porous media [31].

In this study, we applied a single-sided transient plane source (TPS) technique [11] to determine the thermal conductivity of hydrate-bearing sediments and investigate the effects of pore water composition changes on the microscale processes of methane hydrate formation in marine sediments. In this technique, the thermal conductivity can be measured by contact rather than by inserting a probe into the sample. Moreover, a new Sierpinski carpet model was also applied to discuss the effective thermal conductivities of hydrate-bearing sediment.

## 2. Experimental

### 2.1. Apparatus

The measurement apparatus consisted of a hydrate formation system and a thermal conductivity test system, as shown in Fig. 1. The device allowed hydrate formation and subsequent direct compaction of the sample on a TPS element attached to an insulating support.

The hydrate formation system consisted of a cylindrical stainless steel reaction vessel, an air-cooling bath, a gas support unit, a sample compaction unit, and a data-recording unit. Details of the pressure vessel are shown in Fig. 2. The inner diameter of the pressure vessel was 5.0 cm and the sample was compacted using a hydraulic piston driven by a hand pump. A linear motion sensor with an accuracy of  $\pm 0.025$  mm, fixed on the shell of the vessel, was used to measure the sample thickness.

The entire vessel was placed inside an air bath (Angelantoni Challenge 250 climatic chamber) that was capable of controlling temperature within  $\pm 0.1$  K. The temperature ramp cycles and soak intervals. The temperature of the air bath was measured using a four-line PT100 RTD (Resistance Temperature Detector) with an accuracy of  $\pm 0.1$  K and the pressure was measured using a Senex DG1300-BZ pressure sensor with an accuracy of  $\pm 20$  kPa. The gas pressure, temperature, and piston motion data were collected using an Agilent 34970 Data Acquisition/Switch Unit and logged with an Agilent BenchLink Data Logger program. A one-sided TPS technique was applied to compact the sample in situ. The TPS was adhered to polytetrafluoroethylene (PTFE), which served as a backing support and a partial insulator and the thermal conductivities of the

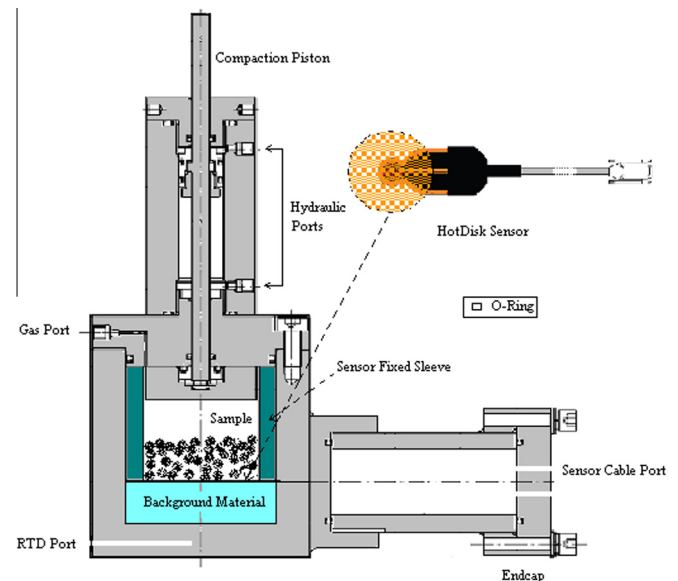


Fig. 2. Reactor for in situ measurements of the thermal conductivity of gas hydrates.

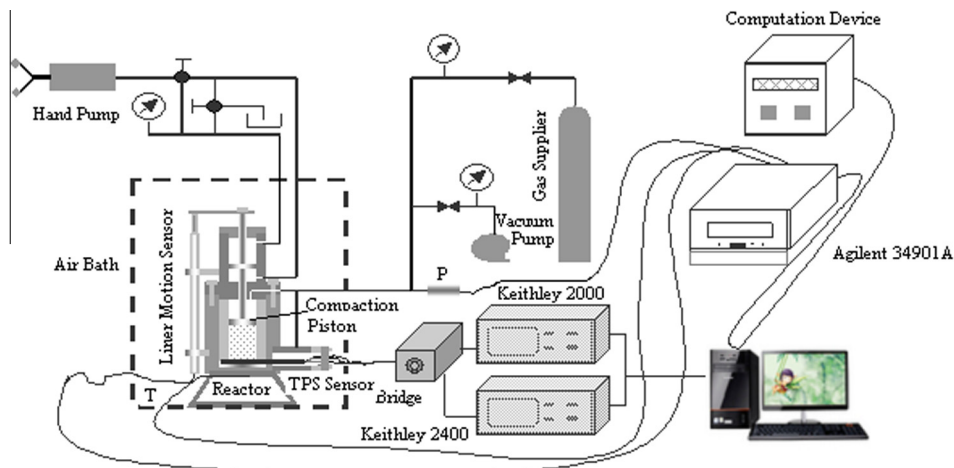


Fig. 1. Schematic illustration of the methane hydrate formation and thermal conductivity measurement system.

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