Experimental determination of permeability of porous media in the presence of gas hydrates

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ABSTRACT

Permeability variation, particularly in the presence of gas hydrates, greatly influences production of natural gas from hydrate reservoirs. A series of experiments were performed to investigate the variation of permeability in gas hydrates-bearing sediments. Carbon dioxide hydrate was formed in Ottawa 20/30 sand samples contained within a rigid cell using a partial water saturation formation methodology. Different initial water saturations were used to achieve hydrate saturations up to 45% and the corresponding water permeability was measured during steady-state flow. The experimental permeability results were compared with several theoretical models using both the quantitative and graphical analyses. A hybrid modeling approach based on the weighted combination of grain coating and the pore filling models was used to fit the measured experimental data. The experimental results were also compared to relevant experimental studies that used similar methods to form hydrates. Our analysis indicates a gradual reduction in permeability with increasing hydrate saturation, which is consistent with earlier studies. Further analysis using hybrid modeling suggests a progressive change in the hydrate formation morphology from cementing to that of the pore filling with increasing hydrate saturation.

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1. Introduction

Gas hydrates are ice-like crystalline compounds composed of water and guest molecules such as carbon dioxide or methane. Gas hydrates form under high pressure and low temperature conditions wherein water molecules form a cage-like structure that are stabilized by guest gas molecules. Natural gas hydrates are commonly found in permafrost regions such as arctic subsurface and outer continental (and insular) margins such as deep-sea sediments (Booth et al., 1998; Ruppel and Noserale, 2012). While gases such as the ethane, carbon dioxide, and propane can form gas hydrates, methane is the most common gas that is abundantly found in naturally occurring gas hydrate sediments (Kvenvolden, 1993). Both methane and carbon dioxide hydrates form Type I hydrate structure (Lee et al., 2003). Gas hydrates are important for their role in future energy resource (Moridis and Collett, 2004), submarine geohazard (Maslin et al., 2010; Grozic, 2010) and global climate change (Lashof and Ahuja, 1990; Goel, 2006). Gas hydrate also has practical implications on flow assurance and safety of oil and natural gas pipelines (Hammerschmidt, 1934; Sloan and Koh, 2008). Therefore, gas hydrates have witnessed tremendous research interest both in fundamental aspects as well as development efforts in commercialization.

Production of natural gas from hydrates can be achieved by inducing dissociation through depressurization (Ji et al., 2001; Sloan and Koh, 2008), thermal stimulation (McGuire, 1982), inhibitor injection (Kamath and Godbole, 1987), or their combinations (Moridis, 2003). Numerical (or mathematical) models are commonly used to evaluate the production potential of dissociation techniques and to assess the factors and conditions affecting the production (Moridis, 2003; Wilder et al., 2008). Numerous predictive models of gas hydrate dissociation ranging from simple energy balance models to complex three-dimensional, three-phase numerical simulators have been developed (Sawyer et al., 2000). These numerical models require the knowledge of several geophysical properties of the porous media in the presence of gas hydrates (Waite et al., 2009).

Permeability variation of porous media in the presence of gas hydrates is an important geomechanical parameter used in numerical modeling of gas hydrate production. The effectiveness of destabilizing pressure and temperature pulses from wellbores is critically influenced by the relative permeability of the hydrate bearing sediment (Moridis et al., 2011). Secondly, the relative permeability determines the gas transportation rate to the production wells (Hong, 2003; Sakamoto et al., 2008). Reduction of

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permeability occurs due to decreased porosity when hydrate forms within pore spaces. Permeability variation of porous media in the presence of hydrates is critically affected by where and how hydrate forms within pore spaces. If hydrate forms at the grain surfaces or at grain contacts as cement (grain coating or cementing), a more gradual reduction in permeability is expected. Permeability reduction can be larger if the hydrates were to form in the middle of the pores (pore filling). Drastic reduction in permeability occurs if the hydrates formation plugs the pore throats besides pore bodies.

Permeability measurement from natural core samples is difficult due to core disturbance in the retrieval and processing. For instance, hydrate redistribution after repressurization in natural core samples has been observed during the experimental analysis (Waite et al., 2008; Kneafsey et al., 2011a). Natural samples differ in mineralogy, host sediment, confinement, and the hydrate formation mode to name a few. Thus, it is imperative to study the hydrate formation mode and its impact on permeability using various media, sample preparation method under controlled environment. Laboratory synthesis provides a viable alternative and flexibility to form samples with desired characteristics such as the hydrate saturation and specific preparation method. Several methods such as the ice seeding (hydrate from ice), partial water saturation method using excess gas or water, dissolved gas method, and the hydrate premixing have been reported in literature (Moridis et al., 2011). The advantages and limitations of the above preparation methods are discussed elsewhere (Waite et al., 2009). Earlier experimental studies have inferred a strong relationship between the sample preparation method and the way in which hydrate forms within the pore spaces (Waite et al., 2009; Moridis et al., 2011). For example, hydrates formed with excess water in the pore space and limited gas tends to be frame supporting (Priest et al., 2009). On the other hand, hydrate formed with the partial water saturation method using excess gas will likely cement grain contacts forming a stiff sample (Waite et al., 2004; Kneafsey et al., 2005; Priest et al., 2009). However, other experimental studies have reported pore filling behavior for hydrates formed using excess gas (Kleinberg et al., 2003; Jaiswal, 2004). Thus, there is a definite need to measure the relative permeability of the porous media in the presence of hydrates and compare them with previous results.

Permeability measurements covering the entire range of hydrate saturations is often difficult to achieve using a particular hydrate formation method. Secondly, individual laboratory studies on permeability variation in the presence of hydrates differ in the hydrate formation method, mineralogy, host sediment, confinement, and the measurement techniques (Moridis et al., 2011). In the absence of reliable experimental data and field measurements, numerical reservoir simulators have largely depended on the theoretical models for permeability prediction (Kumar et al., 2010). Comparing experimental permeability data with the theoretical models provides valuable insight into the hydrate growth within porous media as they are derived using specific assumptions. As a consequence, they also show large variation in predicted permeabilities for a given hydrate saturation. Therefore, the theoretical models can be used within numerical reservoir simulators only after validating the underlying hydrate growth through experimental investigation. Besides, the suitability of a specific theoretical model to experimental results is largely qualitative through graphic analysis.

In this work, we performed a series of experiments in which carbon dioxide hydrate was formed in partially-water saturated Ottawa 20/30 sand samples. Hydrate formation was inferred by gas consumption (pressure drop) and concurrent temperature rise during the experiment. Water permeability was calculated based on Darcy's law after achieving steady-state flow. The experimental results were then compared with a number of theoretical models to understand the underlying hydrate formation behavior. Two quantitative performance measures were used to determine the fitness of the theoretical models to the experimental results. Secondly, a hybrid modeling approach was used to better predict the experimental permeability values. Finally, conclusions were drawn based on the comparison between the results obtained here and similar experimental studies.

2. Experimental method

2.1. Experimental set up

The schematic diagram of the developed apparatus adopted for hydrate formation and permeability measurement is shown in Fig. 1. Ottawa 20/30 silica sand (ASTM) was used as the porous media, which is medium sized and uniformly graded. The particle size distribution is between 600 µm and 850 µm with an average size of 720 µm. While the coefficient of uniformity is similar to that of glass beads at around 1.18, the particles are more sub-rounded (roundness around 1.10). The specific gravity is around 2.65. The maximum and minimum void ratio is around 0.78 and 0.48 respectively (Salgado et al., 2000). A stainless steel rigid cell with 6.5 cm inner diameter and 8.2 cm outer diameter was used as a pressure vessel in the experimental set up. The rigid cell is designed to withstand pressures up to 8 MPa. The rigid cell was placed between the top and the bottom platens of a triaxial set up, which set the axial length of the sample to be 12.9 cm. A minimal load was applied to the rigid cell with the axial actuator of the triaxial set up. Fig. 2 shows the experimental apparatus including the rigid cell and glycol bath. The cooling coil from the temperature controller covered the entire rigid cell. The sample can thus be maintained at a desired temperature by circulating glycol through the coil.

The temperature of the sample was monitored with Type-T thermocouples (Class 1) that have an accuracy of $\pm 0.5\,°C$ for temperatures between -40 and $250\,°C$. The thermocouples T1 and T2 were attached to the outer surface of the rigid cell at a distance of 4 cm and 8 cm from the bottom of the sample. Therefore, an independent experiment was performed to determine the temperature inside the rigid cell. A DST micro-T implantable temperature recorder ($8.3\,mm \times 25.4\,mm$) with a range of -2 and $40\,°C$ and $\pm 0.2\,°C$ accuracy was placed within the sample to measure the steady-state temperature. After retrieving and processing the recorded data, it was observed that the temperatures within the rigid cell were between 1 and 2 °C cooler than the outer surface. The system pressure was monitored using a Honeywell FP2000 gauge transducer that is capable of measuring pressures up to 5 MPa with an accuracy of 0.25%. The pressure transducers were mounted on the lines near the top (P1) and the bottom (P2) of the rigid cell. The pressure transducers were calibrated against a known pressure source prior to testing. The pressure transducers and thermocouples were connected to a data acquisition system, which recorded the readings at 1 s interval.

The rigid cell restricted any lateral movement; vertical movements were restricted with an axial actuator and any additional induced vertical load due to potential volumetric expansion or contraction was monitored. Because the specimen was constructed in a dense state, very little change in the axial loading was observed. The experimental set up was designed to facilitate gas injection from both the top and the bottom of the rigid cell during hydrate formation. Carbon dioxide (99.9% pure) was used as the hydrate forming gas. A back pressure regulator was installed downstream to maintain a constant back pressure at the bottom of the cell during permeability measurement. A cylindrical cell was