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Large scale reactor details and results for the formation and decomposition of methane hydrates via thermal stimulation dissociation

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A B S T R A C T

An in-situ heating method to efficiently produce methane gas from hydrates was recently developed. It is a modification of the thermal stimulation method using a down-hole combustion technique, to more efficiently bring necessary heat to the desired location within the reservoir. To test the concept of methane production via in-situ heating and characterize other alternative production methods, a 70 l reactor vessel has been designed, manufactured and assembled to simulate relevant field environments. Tests show that the reactor satisfies the conditions for both methane and carbon dioxide hydrate formation. During the tests hydrate was formed at 2.2 °C and 4.2 MPa using 99.97% pure methane gas and deionized water in unconsolidated porous quartz sand pack. Heating tests at 100 W resulted in a peak production rate of 1.5 standard liters per minute (slpm) of methane gas, resulting in the dissociation of 35% of total hydrate in the system. A thermal energy efficiency of 72% was calculated based on net thermal heat input and the total heating value of the methane produced during a 10 h test period. Based on mass balance and calculations using the Peng–Robinson EOS it was calculated that hydrate saturations of 4.5, 10.2, 21 and 33% were produced prior to heating tests. Phase equilibrium data is presented for mixed gas hydrates formed from a 50:50 mol fraction mixture of CO_2:CH_4. Measurements and simulations suggest that hydrate began forming at the top section of the reactor initially, and continued to form downwards within the sediment in a non-homogeneous manner.

1. Introduction

Current estimates forecast that world oil production will peak between 2010 and 2030 (Aleklett et al., 2010), which translates into continued oil use for the foreseeable future. During this time new domestic and distributed sources of energy can be developed to ensure that society's demands are met while transitioning away from oil. The best carbon-based energy source is natural gas, because of its high hydrogen to carbon ratio, thus yielding the greatest energy per unit carbon. Worldwide efforts increasing compressed natural gas shipments provide further evidence to a shift to methane based fuel.

The largest sources of methane worldwide are trapped in the permafrost and marine sediments in the form of gas hydrates and are estimated to be as large as all oil, coal and conventional gas *combined* (Moridis et al., 2009). Methane hydrates have the potential to provide the needed energy in a sustainable manner because their extraction can possibly be coupled to a system that allows for the permanent and stable sequestration of carbon dioxide. Interest in methane hydrates as a potential source for clean hydrocarbon energy has been increasing. Natural gas, or methane hydrates are solid, non-stoichiometric compounds of small guest molecules and water (Sloan, 2003). Hydrates form when methane originating from biogenic or thermogenic sources combines with water at sufficiently low temperatures and high pressures where the guest molecule becomes trapped within atomic scale crystalline cages of H_2O. Natural forming hydrates have been found as lenses, nodules, and pore infillings on and beneath the sea floor at shallow depths within the ocean sediment around the world, particularly along continental shelves (Alexi and Sassen, 2002). The most promising regions containing natural gas hydrates are located in polar continental sedimentary rock and in the permafrost areas of the Arctic. In comparison with other important carbon deposits, gas hydrates store an extremely large quantity of organic carbon. There are some uncertainties with regard to the global budget, yet it is believed that gas hydrate formations contain approximately from 0.2×10^{15} to 120×10^{15} m^3 of methane at STP (Sloan and Koh, 2008). USGS surveys have produced estimates of hydrate deposits off the coast of North and South Carolina in the range of 3.6×10^{13} m^3, a deposit of this size could supply the US with gas for 55 years at 2010 consumption rates (Demirbas, 2010). Gas hydrates could therefore represent a potentially significant alternative source of energy in the future, replacing or supplementing conventional fossil fuel sources.

There has been a tremendous amount of research done on locating and quantifying gas hydrate resources (Trofimuk and

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