Research paper

Methane hydrate formation in thick sand reservoirs: 1. Short-range methane diffusion

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ABSTRACT

We show with a comprehensive hydrate formation model that diffusion of dissolved methane along the local concentration gradient alone cannot transport enough methane to form the concentrated methane hydrates in thick ($>=$ several m) marine sand reservoirs. In this model, methane is generated locally by biodegradation of organic matter in mudstones and diffused into the bounding sandstone where the methane hydrate solubility decreases abruptly and hydrate precipitates from the extra methane in water. Hydrate formed by local diffusion has peak concentrations at the top and base of the sandstone, and it decreases sharply toward the center; salinity is slightly elevated relative to seawater, and its gradient is negligible across the sandstone and adjacent mudstones; dissolved methane concentration decreases from the mudstones to the sandstone. Overpressure is generated in the sediment due to reduction of sediment permeability and volume expansion when hydrate replaces pore water. There is a shallow zone where pore pressure converges toward the lithostatic pressure and fracturing is possible. This study provides further insights into hydrate formation by local diffusion of dissolved methane. In addition, the numerical model we developed in this study fully couples the hydrate-liquid capillary effect, dynamic pore size change, and transient pore pressure-dependent sediment compaction with the multiphase (gas, liquid and hydrate) flow and multicomponent (methane, water and salt) transport. Our model not only predicts pore pressure, but also incorporates the effect of hydrate formation on overpressure generation and subsequent dissipation. These modeling approaches are now at a level of sophistication that we can start to distinguish what physical processes, of the several proposed, are viable explanations for observed hydrate deposits.

1. Introduction

Methane hydrate is crystalline, ice-like and nonstoichiometric substance consisting of methane encased in open water lattices (Sloan and Koh, 2007). Methane hydrate has great energy potential (Boswell and Collett, 2011; Milkov, 2004). The amount of gas estimated to be stored in the world's hydrate accumulations is enormous, ranging from 2.8×10^{15} to 8×10^{18} m^3 (Boswell and Collett, 2011; Collett et al., 2009; Milkov, 2004). However, the largest volume of gas hydrate is in sub-economic stratigraphic accumulations dissipating through marine mud with low energy concentration and high production challenges (Boswell and Collett, 2011; Collett et al., 2009; Milkov, 2004). The most promising potential targets for successful resource extraction are those local, anomalous and high concentration methane hydrate accumulations in thick sand reservoirs in Arctic and marine environments, such as those in Northern Gulf of Mexico, in the Nankai trough, on the Cascadia margin, and at the Northern Slope of Alaska (Boswell and Collett, 2011; Uchida et al., 2009).

One critical question is how methane is transported to the thick sandstones to form the concentrated methane hydrates as observed. There are three major ways of methane transport in geological systems: (1) advection in liquid phase; (2) advection in gas phase; (3) diffusion in liquid phase.

Hydrate can form by the process of aqueous transport or by methane advection in a saturated liquid phase, where water with dissolved methane flows upward through the hydrate stability zone (HSZ), and hydrate is precipitated as the solubility of methane in water decreases upward (Xu and Ruppel, 1999). This mechanism has been invoked to explain concentrated hydrate formation in thin (on the scale of cm) sandstones embedded in thick mudstones (Daigle and Dugan, 2011). More recently, Nole et al. (2016) proposed that where a large overpressure is present (several MPa), methane saturated water is focused into permeable layers; this focused flow of dissolved methane could generate concentrated hydrate in thick (on the scale of m), coarse-grained sandstones. However, there is very little field evidence for the existence of such large overpressures in the hydrate locations studied by

Nole et al. (2016).

The second model for methane transport and hydrate formation is advection in a separate gas phase proposed by Liu and Flemings (2006) and quantified in Liu and Flemings (2007); You et al. (2015, 2016). In this model, methane is sourced below the BHSZ and transported into the HSZ by upward gas flow along a permeable layer under the force of buoyancy. The advantage of this model is vast amount of mass transport within a short period of time.

The third mechanism is short-range methane diffusion proposed by Malinverno (2010). In this model, methane is generated locally by microbial conversion of organic matter within fine-grained mudstones, and is transported by diffusion as a saturated liquid phase into the adjacent coarse-grained sandstones (Daigle and Dugan, 2011; Malinverno, 2010; Rempel, 2011). This diffusion is driven by the contrast in solubility between sandstones and their encasing mudstones. The methane solubility to form hydrate is increased in confined pores because of increase in hydrate-liquid capillary pressure (Clennell et al., 1999; Liu and Flemings, 2011). Hence, solubility increases with decreasing size of pores (Clennell et al., 1999; Liu and Flemings, 2011). This is why methane hydrate solubility is notably increased in the pores of mudstones where the largest size is on the scale of nm. In contrast, solubility in sandstones is much smaller, because pores are larger, in the order of μm. Therefore, methane is sourced within the mudstone where its solubility is much greater. Methane is then transported by diffusion from its source in the mudstone to the adjacent sandstone. As methane enters the sandstone, the abrupt decrease in solubility between the mudstone and the sandstone results in the extra methane being fixed into hydrate in the sandstone. This model has been used to explain occurrences of hydrate in thin sandstones (on the scale of cm) sandwiched with hydrate-free mudstones at Cascadia Margin, Andaman Sea of Indian Ocean, and Nankai Trough (Cook and Malinverno, 2013; Daigle and Dugan, 2011; Malinverno, 2010; Malinverno and Goldberg, 2015). It has also been used to explain the average hydrate saturation across a 2.5-m thick sandstone in a shallow depth of Northern Gulf of Mexico (Cook and Malinverno, 2013). However, it is not clear whether the diffusion model can explain concentrated hydrate across several m-thick sandstones, such as those observed at Walker Ridge (Boswell et al., 2012), Nankai Trough (Fujii et al., 2009), North Slope of Alaska (Boswell et al., 2011) and Mallik site (Dallimore and Collett, 2005). This is because diffusive transport is a slow process, and it is debatable whether it can transport sufficient volumes of methane to explain the high levels of hydrate saturation observed (e.g., 90% near the base of the hydrate stability zone (BHSZ) at Walker Ridge 313, northern Gulf of Mexico (Boswell et al., 2012)).

The goal of the current paper is to quantitatively investigate whether diffusion of dissolved methane along the local concentration gradient can transport enough methane to form the concentrated hydrate that are commonly observed in thick sandstones. We quantify hydrate formation in thick sandstones from short-range methane diffusion by developing a new two-dimensional (2D) numerical model. We apply this model to simulate hydrate formation in a 12 m-thick dipping sandstone at Walk Ridge 313, Northern Gulf of Mexico, and illustrate the process of hydrate formation through this mechanism. We conduct simulations with different methane generation rate and diffusion rate and find that high hydrate saturations are only formed at the top and base of the 12 m-thick sandstone with little or no hydrate in the center 8 m when the sandstone is buried to ~810 m below seafloor (mbsf) at Walker Ridge 313. This simulated amount of hydrate is much less than that interpreted from the resistivity data. Therefore, short-range methane diffusion alone cannot form the concentrated methane hydrate in thick sandstones as observed in many field locations.

2. Mathematical model

In the short-range methane diffusion model, methane is generated by biodegradation of organic matter in the mudstones and diffused into the bounding sandstone as a saturated liquid phase. This brings the dissolved methane concentration to the solubility limit and drives hydrate to gradually precipitate in the sandstone as the sediments and hydrate are buried with time (Malinverno, 2010). We describe the mass conservations involved, the effect of liquid-hydrate capillary pressure, the dynamic change of effective pore size during hydrate formation, the sedimentation and compaction, the microbial methane generation, and the initial and boundary conditions for this model in the following.

2.1. Mass conservation equations

In our model, we include three components: water (w), methane (m) and salt (s). These components can form three phases: liquid (l), gas (g) and hydrate (h). We assume:

(1) Local thermodynamic equilibrium (Liu and Flemings, 2007; Rehder et al., 2002; Sun and Duan, 2005; Zatsepina and Buffett, 2003);
(2) Methane is the only component in the gas phase because the solubility of water in the gas phase is small (Duan et al., 1992; Liu and Flemings, 2007);
(3) Salt is limited to the liquid phase;
(4) Temperature is at steady state. It increases linearly with depth with a constant geothermal gradient and fixed seafloor temperature.

We use the Lagrangian reference frame. The reference frame is fixed on, and moves with the solid grains. The mass conservation equation for water, methane and salt can be described as

$$\text{Water:} \frac{D\ln(M^w)}{Dt} - \frac{D\ln[(1-n)\rho_R]}{Dt} + \frac{\nabla(q_l\rho_l X_l^w)}{M^w} - \frac{q^w}{M^w} = 0, \quad (1)$$

$$\text{Methane:} \frac{D\ln(M^m)}{Dt} - \frac{D\ln[(1-n)\rho_R]}{Dt} + \frac{\nabla(q_l\rho_l X_l^m + q_g\rho_g)}{M^m}$$
$$- \frac{\nabla[nS_lD_l^m\nabla(\rho_l X_l^m)]}{M^m} - \frac{q^m}{M^m} = 0, \quad (2)$$

$$\text{Salt:} \frac{D\ln(M^s)}{Dt} - \frac{D\ln[(1-n)\rho_R]}{Dt} + \frac{\nabla(q_l\rho_l X_l^s)}{M^s} - \frac{\nabla[nS_lD_l^s\nabla(\rho_l X_l^s)]}{M^s}$$
$$- \frac{q^s}{M^s} = 0, \quad (3)$$

where $\frac{D}{Dt}$ is the substantial time derivative; t is time (s); the superscripts m, w and s denote methane, water and salt, respectively; the subscripts l, g, h and R denote the liquid, gas and hydrate phases and sediment grains, respectively; and M^w, M^m and M^s are the mass of water, methane and salt per volume of sediment (kg m^{-3}), respectively. These masses are described as

$$\text{Water:} \ M^w = nS_l\rho_l X_l^w + nS_h\rho_h X_h^w, \quad (4)$$

$$\text{Methane:} \ M^m = nS_l\rho_l X_l^m + nS_g\rho_g + nS_h\rho_h X_h^m, \quad (5)$$

$$\text{Salt:} \ M^s = nS_l\rho_l X_l^s; \quad (6)$$

where n is porosity (dimensionless); ρ_β and S_β are the density (kg m^{-3}) and saturation (dimensionless) of β phase, respectively; X_β^κ is the mass fraction of component κ in phase β (dimensionless); D_l^m and D_l^s are the molecular diffusion coefficients (m^2 s^{-1}) of methane and salt in sediments, respectively, and $D_l^m = nD_{l0}^m$, and $D_l^s = nD_{l0}^s$ (Liu and Flemings, 2007). D_{l0}^m and D_{l0}^s are the molecular diffusion coefficients (m^2 s^{-1}) of methane and salt in free water, respectively. q^m, q^w and q^s are the local generation or consumption rate (kg m^{-3} s^{-1}) of methane, water and salt, respectively; q_l and q_g are the volumetric flux of liquid and gas relative to sediment grains (m^3 m^{-2} s^{-1}), respectively. These fluxes are described by Darcy's law

$$\text{Liquid:} \ q_l = -\frac{kk_{rl}}{\mu_l}(\nabla P_l - \rho_l g), \quad (7)$$