



Burial-driven methane recycling in marine gas hydrate systems

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ABSTRACT

Natural gas hydrate may be buried with sediments until it is no longer stable at a given pressure and temperature, resulting in conversion of hydrate into free gas. This gas may migrate upward and recycle back into the hydrate stability zone to form hydrate. As of yet, however, no quantitative description of the methane recycling process has been developed using multiphase flow simulations to model burial-driven gas hydrate recycling. In this study, we present a series of 1D multiphase transport simulations to investigate the methane recycling process in detail. By invoking the effects of capillary phenomena on hydrate and gas formation in pores of varying size, we find that a free gas phase can migrate a significant distance above the bulk base of hydrate stability. Since the top of the free gas occurrence is often identified as the base of the hydrate stability zone from seismic data, our results demonstrate that not only could this assumption mischaracterize a hydrate system, but that under recycling conditions the highest hydrate saturations can occur beneath the top of the free gas occurrence. We show that the presence of pore size distributions requires a replacement zone through which hydrate saturations progressively decrease with depth and are replaced with free gas. This replacement zone works to buffer against significant gas buildup that could lead to fracturing of overlying sediments. This work provides a framework for simulating flow and transport of methane within the 3-phase stability zone from a mass conservation perspective.

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1. Introduction

Natural gas hydrate is known to exist all over the world in environments characterized by high pressure and low temperature (Sloan and Koh, 2007) where both water and the guest gas are both in relative abundance (Kvenvolden, 1998; Buffett and Zatsepina, 2000). Gas hydrate is typically found within the pore space of sediments either beneath the seafloor along continental margin environments (Kvenvolden and Barnard, 1982) or beneath arctic permafrost (Collett and Dalimore, 2000). Understanding the distribution of gas hydrate in sediment pore space can yield better insight into the potential role of gas hydrate in regional slope failure events (Mienert et al., 2005), as a future natural gas energy resource (Boswell and Collett, 2011), as an exploratory drilling hazard (McConnell et al., 2012), and as a significant component of mobile carbon in the global carbon cycle (Dickens, 2003; Archer et al., 2009).

Characterizing gas hydrate occurrences in natural sediments often first involves constraining the thickness of the gas hydrate stability zone (GHSZ). The GHSZ is defined as the depth interval over which hydrate is the thermodynamically stable phase in equilibrium with dissolved methane in water and is typically formulated as a function of *in situ* pressure and temperature (Xu and Ruppel, 1999). The base of the hydrate stability zone (BHSZ) is traditionally defined as the depth where pressure and temperature of the pore fluids reach the phase boundary between hydrate and free gas (Fig. 1; Moridis, 2003). This description typically neglects capillary phenomena and is referred to here as the bulk three-phase equilibrium condition. Deeper than the 3-phase boundary, hydrate is no longer stable and only free gas can form when dissolved gas concentrations exceed aqueous solubility. The BHSZ is typically estimated by analyzing regional seismic data, seafloor temperature measurements, and water depth measurements. However, discrepancies between these data sets at many sites worldwide illustrate how these methods can mischaracterize a hydrate system when assuming bulk thermodynamic equilibrium (Pecher et al., 2010; Crutchley et al., 2011; Majumdar et al., 2016).

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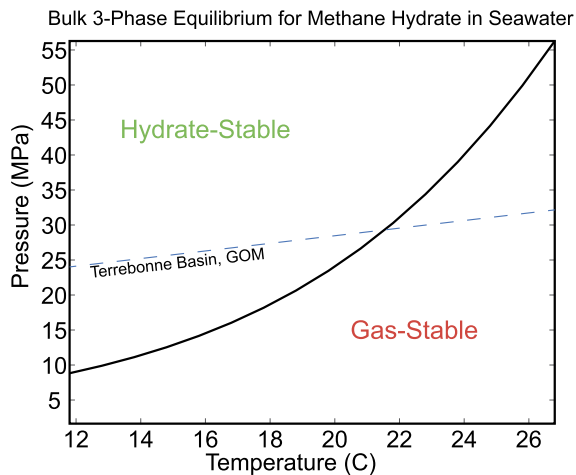


Fig. 1. The phase boundary between a hydrate–water and a gas–water system at bulk thermodynamic equilibrium. Along the phase boundary, all 3 phases can coexist. For reference, the hydrostatic pressure and temperature profile of a system like the Terrebonne Basin in the Gulf of Mexico is also depicted.

Instead of the single phase boundary line that is predicted from thermodynamic equilibrium between a single gas guest molecule and bulk water, a region of three-phase stability is possible by invoking the presence of either a pore size distribution in hydrate- and gas-bearing sediments (Liu and Flemings, 2011), the presence of different types of hydrate-forming gases besides just methane (Paganoni et al., 2016), or significant local gradients in salinity owing to salt exclusion during hydrate formation (though the latter is a transient process; i.e., Liu and Flemings, 2006). These observations are important to understand because poorly constraining the thickness of the GHSZ can lead to inaccurate predictions regarding total amount of hydrate in place, the depth of the BHSZ, and quantities like heat flow that may be derived from seismic data (e.g., Townend, 1997; Phrampus et al., 2017).

This work focuses on the role that capillary phenomena can play in altering the equilibrium behavior of a system comprised of methane hydrate and aqueous dissolved methane (Clennell et al., 1999; Rempel, 2011). If gas hydrate is to form within the pore space of sediments, and if gas hydrate is considered a nonwetting phase, then the interface between gas hydrate and pore water must have a nonzero curvature. Known as the Gibbs–Thomson effect, precipitating a solid crystal with progressively higher curvature from the dissolved phase requires a correspondingly stronger driving force. For hydrate growing in porous media, this translates into a progressive depression of the three-phase equilibrium temperature for hydrate growing into pores with larger and larger surface curvature (Anderson et al., 2009). Similarly, since the free gas phase does not wet the grain surfaces in a water wet medium, forming a gas phase in pores requires a capillary pressure (the difference between gas pressure and water pressure) to maintain a curved gas–water interface in those pores; this pressure increases with increasing interface curvature, which is constrained by the size of the pore containing the nonwetting phase.

Maintaining a curved interface between gas hydrate and water inside of a pore with finite curvature requires a stronger driving force than in bulk water and progressively shifts the phase boundary to higher pressures and lower temperatures at higher-curvature interfaces. If a sediment pore network were composed entirely of one pore size, then this phase boundary would correspond to a precise pressure and temperature; therefore, it would be crossed at one particular depth just as in the bulk equilibrium scenario. Natural sediments contain a distribution of pore sizes, and therefore a range of pressures and temperatures can exist for which 3-phase equilibrium is possible (Liu and Flemings, 2011).

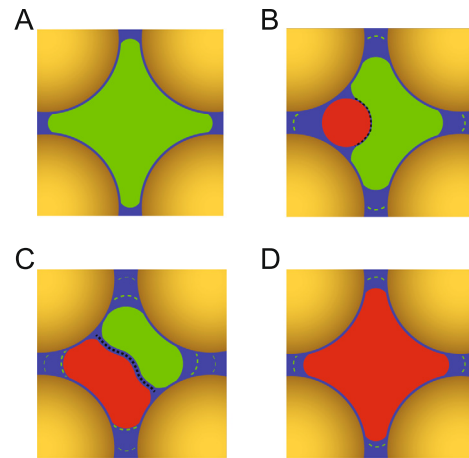


Fig. 2. The three-phase equilibrium model used in these simulations when transitioning from a 2-phase hydrate–water system to a gas–water system (A to D) or the other way (D to A). The black dashed line denotes the hydrate-free gas interface, which in this work is assumed to always be separated by a thin water film. A) Hydrate (green) initially exists in sediment (brown) pore space, and its interface with water (blue) has a finite curvature. B) Shifting past the phase boundary requires methane to form a free gas phase (red), which consumes some hydrate. C) The curvature of the free gas phase (and capillary pressure) increases progressively while the curvature of the hydrate phase progressively decreases, until eventually D) only a free gas phase remains.

The work presented here develops a simulation framework for modeling flow and transport of methane through the 3-phase hydrate- aqueous-free gas stability zone defined by a distribution of sediment pore sizes in marine geologic environments, from a mass conservation perspective. Simulations presented here in particular examine the methane recycling process, or the process by which methane hydrate buried past the BHSZ converts to buoyant free gas, which then migrates back up into the GHSZ and forms gas hydrate. This phenomenon is often suggested as a hydrate-forming mechanism (Haacke et al., 2007; Davies and Clarke, 2010), but the work presented here describes the mechanics of this process when methane mass is conserved.

2. Methods

This work expands upon the formulation of Liu and Flemings (2011) by simulating the coupled flow mechanics of phase transitioning in a methane-constrained system. Additionally, we describe three-phase coexistence in pore space as generally being controlled by the effective curvatures of hydrate and free gas phases at the interfaces between these nonwetting phases and the aqueous phase.

Take for example a system with only free gas and water, where water is the wetting phase. At low saturations, free gas exists in the largest pores of a distribution because they have the lowest capillary entry pressures. At a given gas saturation the capillary pressure is defined by the highest-curvature interface between free gas and the aqueous phase. In this study we associate this interface with a pore size by assuming the nonwetting free gas phase must exist within the pore of a given size. The same treatment is applied to the hydrate phase: a higher-curvature hydrate–water interface is associated with smaller pores and a more significant shift in the phase boundary. We assume the only interfaces within the pore space are hydrate–water and free gas–water.

In a system at 3-phase equilibrium, the link between phase curvature and pore curvature becomes more nuanced. Consider a single pore system containing gas hydrate (Fig. 2A). The curvature of the interface between hydrate and water is constrained by the size of the smallest pore occupied by hydrate. If the system is brought just past the phase boundary, then hydrate is no longer

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