



# Methane bubble growth in fine-grained muddy aquatic sediment: Insight from modeling



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## ABSTRACT

Methane (CH<sub>4</sub>) is the most abundant hydrocarbon and one of the most important greenhouse gases in the atmosphere. CH<sub>4</sub> bubble growth and migration within muddy aquatic sediments are closely associated with sediment fracturing. In this paper we present the modeling of buoyancy-driven CH<sub>4</sub> bubble growth in fine-grained muddy aquatic sediment prior to the beginning of its rise. We designed a coupled mechanical/reaction-transport numerical model that enables a differential fracturing over the bubble front (as it occurs in nature), when the fracturing increment stays constant at the bubble head and subsides towards bubble tail during bubble growth. We show that this differential fracturing over the bubble front controls the bubble shape and size temporal evolution, and is significantly affected by the critical stress intensity factor of the muddy sediment. The intercalated stages of elastic expansion and fracturing during the bubble growth shorten with time as the bubble approaches its terminal size (prior to its ascent). Our simulations reveal a high asymmetry in the bubble shape growing with time, with respect to its initial symmetric penny-shaped configuration. It was found that the bubble grows allometrically, while the importance of the bubble surface area growth with time. We also confirmed the earlier predictions about the "inverted tear-drop" bubble cross-section just prior to the beginning of its rise. Modeling of the terminal bubble characteristics will permit prediction of the delivery of gaseous methane from the sediment to the atmosphere via the water column.

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## 1. Introduction

Methane (CH<sub>4</sub>) is the most abundant hydrocarbon and one of the most important greenhouse gases in the atmosphere. Over the last century the CH<sub>4</sub> concentration has risen by 1% per year (Rowland, 1985). Despite their importance, CH<sub>4</sub> fluxes from the aquatic sediments recently reported in the literature ranged over an order of magnitude (Soumis et al., 2005; St. Louis et al., 2000) indicating that they have not been properly quantified (Del Sontro et al., 2010). CH<sub>4</sub> emission from aquatic systems to the atmosphere is usually dominated by gas ebullition (Ostrovsky et al., 2008; Del Sontro et al., 2010): In shallow lakes up to 98% of CH<sub>4</sub> release originated from bubbles while only 2% came from dissolved CH<sub>4</sub> (Keller and Stallard, 1994; Del Sontro et al., 2010). Formation of methane bubbles and their transport within the sediments is a subject of recent investigations.

### 1.1. The growth and migration of bubbles within muddy sediments

The perceived pliability of soft muddy sediments and the observed fluidization patterns (e.g. gravity flow), erroneously suggest that such sediments could act fluidly or plastically in response to stress induced by growing bubble (Wheeler, 1988). However, recent laboratory simulations have shown that these sediments respond mechanically as fracturing elastic solid (Best et al., 2004; Boudreau et al., 2005; Barry et al., 2010). The importance of grain size in determining the behavior of gassy sediments has recently been demonstrated by Jain and Juanes (2009), and Choi et al. (2011). They suggested that gas migration in fine-grained (muddy) sediments is governed by a fracture-dominated regime due to the large capillary-entry pressure precluding gas from entering pore throats without breaking the inter-granular bonds, while in coarse-grained (sandy) sediments it occurs by capillary invasion through the sediment framework.

Exploring the bubble shape, Anderson et al. (1998) observed that bubbles in shallow muddy sediment were often non-spherical, with eccentricity increasing with their volume. Imaging bubble in gassy sediments, Best et al. (2004) demonstrated that in contrast to the small (2 mm in diameter) sub-spherical bubbles in silty sands, most bubbles observed in the clayey silts appeared as low aspect

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ratio cavities, up to  $\sim 40$  mm long, with their longest axes aligned in the sub-vertical plane.

Van Kessel and van Kesteren (2002), Winterwerp and van Kesteren (2004), Johnson et al. (2002) demonstrated experimentally that a bubble initially grows quickly to entirely fill the pore space it occupies, and then starts deforming the surrounding sediment skeleton. After numerous observations of crack initiation and propagation in soft sediments, it was suggested that these cracks can be described by linear elastic fracture mechanics (LEFM), the theory applicable to cracks in which the region of plastic strain at their tips is small relative to the crack size (Lawn and Wilshaw, 1975; Broek, 1986).

## 1.2. Recent modeling approaches

Recent modeling efforts addressed the issue of methane dynamics in sediments through several alternative perspectives.

### 1.2.1. Diagenetic reaction-transport models

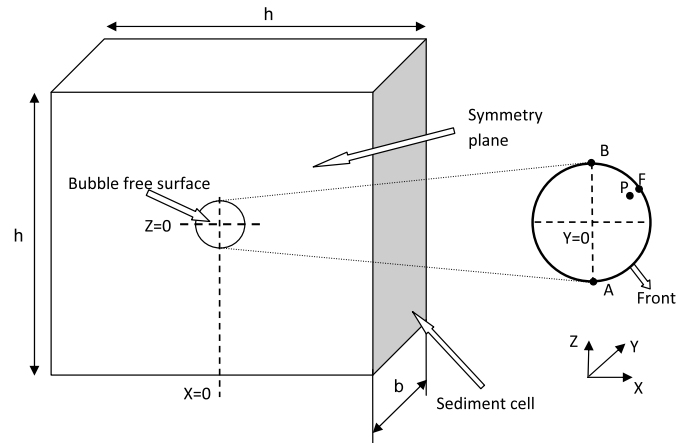
Despite the importance of exploring gaseous methane dynamics, models incorporating an explicit gas phase representation usually use a multiphase fluid dynamics approach that is better justified for coarse-grained soils (Oldenburg et al., 2010) or fractured aquifers (Rubin et al., 2008).

Martens et al. (1998) incorporated a gas phase into a diagenetic model, balancing in a steady-state sedimentation and methane production from organic matter against ebullition of gaseous methane,  $\text{CH}_4(\text{g})$ . Haeckel et al. (2004) represented  $\text{CH}_4(\text{g})$  as a source term for dissolved methane, implying that  $\text{CH}_4(\text{g})$  is not transported explicitly through the sediment, while Davie and Buffett (2001) assumed that gas transport follows burial. Alternatively, Haeckel et al. (2007) considered gas phase transport explicitly through tube structures. Further, Dale et al. (2008) added a mass-conservation equation for  $\text{CH}_4(\text{g})$ , assuming that exchange of methane between the dissolved and gaseous phases is proportional to the departure from the local solubility (Duan et al., 1992). Mogollon et al. (2009, 2011) derived a separate 1D mass and momentum conservation equations for the solid, aqueous, and gas phases coexisting within a common control volume, in addition to conservation of the individual species.

### 1.2.2. Mechanical models

Microscopic Discrete-Element Model coupling two-phase (gas-brine) flow with sediment mechanics was implemented in Jain and Juanes (2009) allowing sediment fracture by an advancing gas phase, caused by localized breaking of cohesion between adjacent sediment grains.

A steady state (Gardiner et al., 2003a) and transient (Algar and Boudreau, 2009, 2010) reaction-diffusion model describing supply of dissolved methane,  $\text{CH}_4(\text{aq})$ , to the growing bubble was combined with principles of LEFM to simulate sediment elastic expansion, followed by uniform fracturing at the bubble front during its growth. A key assumption involved in the model of Gardiner et al. (2003a) is that bubbles grow slow enough to enable the solute concentration field next to the bubble be readily adjusted. This assumption allowed Gardiner et al. (2003b) to find an analytical solution to the diffusion equation in oblate-spheroidal coordinates. However, fracture event results in sudden increase in bubble volume and drop in internal gas pressure (Johnson et al., 2002), implying that transient reaction-diffusion equation must be solved in combination with LEFM, as implemented by Algar and Boudreau (2009, 2010). When buoyancy was added to the model, an initial rise (propagation) of a large (mature) bubble was simulated by Algar et al. (2011b), neglecting mass transfer between the bubble and sediment. Fracturing in the model was permitted to occur at the bubble head only.



**Fig. 1.** Sediment cell is presented as a block cut by a symmetry plane (only half a block is modeled). The initial bubble seed is presented as a small penny-shaped crack placed on the symmetry plane. Only one of the two bubble free surfaces is modeled. Points B and A are the head and tail points on the bubble front respectively. Point P prescribed on the bubble surface is used for the calculations of Stress Intensity Factor at the adjacent point F at the bubble front (see Section 2.1.3).

Coupling mechanical and reaction-transport processes is the issue addressed by a structural diagenesis (Laubach et al., 2010). This coupling is greatly important for general understanding of bubble growth and migration in fine-grained muddy sediments. In general, such a feedback can help to obtain a scientific knowledge about the low-temperature realm of sedimentary basins that is of great intrinsic and practical interest. However, despite the undoubted progress in development of such models, the accurate reproduction of the processes governing bubble dynamics is still scarce.

In this paper we present a model describing the process of the buoyancy-driven bubble growth in muddy sediments prior to beginning of its ascent toward the sediment-water interface. For the first time we simulate the differential fracturing over the bubble front, as it occurs in nature, and show that the fracturing controls the bubble shape and size evolution. The model has a large potential for simulating buoyancy-driven upward migration of bubble, and predicting the delivery of gaseous methane from sediments to the atmosphere.

## 2. Methods

### 2.1. The model

To model bubble growth within soft sediment we used approach of bubble growth within an elementary cell that was applied by Prousevitch et al. (1993), Prousevitch and Sahagian (1996), Navon et al. (1998), Favelukis (2004) for fluids, and by Algar and Boudreau (2009, 2010) for muddy sediments. Our model extends the above approaches by including a fully-coupled numerical treatment of transport and mechanical components. It especially concentrates on the precise modeling of the differential fracturing of muddy sediments at the growing bubble front, which is carried out accordingly to the principles of fracture mechanics. The modeling setup is depicted in Fig. 1. The sediment cell is presented as a block cut by a symmetry plane (only half of the block is modeled). The initial bubble seed is presented as a small penny-shaped crack with one of its free surfaces located on the symmetry plane (Algar et al., 2011b).

#### 2.1.1. Modeling of the region outside the bubble

Transport of dissolved methane and solid mechanics are modeled in the bulk of the sediment cell outside the bubble (Fig. 1).

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