



## Research paper

# Numerical modeling study of mineralization induced by methane cold seep at the sea bottom



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## ABSTRACT

In order to investigate the response of authigenic minerals to gas hydrate geo-systems, the biogeochemical processes and its induced mineralization were predicted by employing the comprehensive reactive transport modeling approach. Based on the available data extracted from the northern continental slope area of the South China Sea, a 1-D vertical column model was developed. Three cases with different upward methane flux rates and three cases with different mineral compositions, i.e., a total of six cases were designed to investigate the effects of variations in the depth of sulfate methane transition zone (SMTZ) and in the mineral composition on the formation of authigenic minerals. The simulation results indicate that the SMTZ depth influenced by both the upward methane flux rate and the initial composition played an important role in the formation of authigenic minerals. The AOM reaction is intensive at the interface, and the precipitation amount of calcite is large, which is mainly controlled by AOM. When the methane leakage rate is 20 times higher than the base case, aragonite starts to precipitate. During the simulation, oligoclase, k-feldspar, smectite-Na, smectite-Ca, chlorite dissolved. Our study specific to this area as a starting point may provide a quantitative approach for investigating carbonate and pyrite formation in hydrate-bearing sediments accounting for methane oxidation and sulfate reduction. The method presented here and the model built in this study can be used for other sites with similar conditions. In addition, this study may serve as an indication for the potential natural gas hydrate reservoir in depth, and is also significant for marine carbon and sulfur cycle.

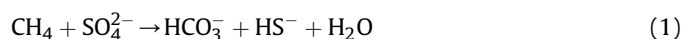
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## 1. Introduction

Natural gas hydrate found in marine sediments has attracted much attention in the world, because it could become a major future energy resource. Authigenic rocks or minerals associated with methane seeps from gas hydrate are widespread in hydrate occurrence regions, so-called cold-spring deposits. For example, a large amount of carbonate rocks and authigenic pyrite have been observed in the black ridge in the United States, the Northeast Pacific hydrate ridge of the Gulf hydrate zone, Nankai trough in eastern Japan, the Northern South China Sea (Torres et al., 2002; Treude et al., 2003; Himmler et al., 2013; Sassen et al., 2004; Tsunogai et al., 2002; Tong and Chen, 2012; Ge et al., 2010). The associated authigenic minerals are common phenomenon in natural gas hydrate sedimentary environments. Carbonate minerals mainly include calcite, aragonite, and siderite, which are related to

the anaerobic methane (CH<sub>4</sub>) oxidation reaction. These associated minerals have great significances for studying ocean gas hydrate, and global carbon and sulfur cycle.

Anoxic marine sediments are the largest methane reservoirs on Earth, either dissolved in the formation water or as condensed gas hydrates (Dale et al., 2008a). At Hydrate Ridge (HR), Cascadia convergent margin of offshore Oregon of the United States, marine sediments contain massive gas hydrates formed due to upward-moving methane together with fluids from deeper reservoirs (Treude et al., 2003). When methane encounters sulfate diffused downward from seawater, it could be oxidized anaerobically by a microbial consortium, releasing bicarbonate and sulfide into the pore water (Hinrichs et al., 1999; Boetius et al., 2000; Luff et al., 2005). This process is called anaerobic oxidation of methane (AOM). During the AOM, methane is oxidized using sulfate as an electron acceptor via the following net equation:

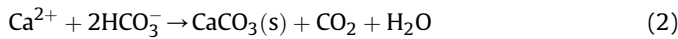


The AOM consortium predominant at Hydrate Ridge consists of

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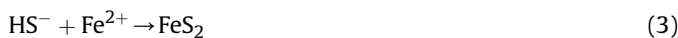
sulfate-reducing bacteria of the branch *Desulfosarcina/Desulfococcus* and archaea of the ANME-2 group (Boetius et al., 2000; Treude et al., 2003). The anaerobic oxidation of methane increases the alkalinity and triggers carbonate precipitation:



Carbonate formation in sediment pore space takes place when pore-fluids are highly supersaturated with respect to the carbonate phase (e.g., calcite or aragonite).

As upward methane delivery increases, the depth of the AOM layer moves upward in order to supply a balancing amount of sulfate derived from overlying seawater (Borowski et al., 1996). The depth and thickness of the AOM horizon mainly depends on the strength of the upward fluid flow and the concentration of methane in the ascending fluids (Luff et al., 2004). Carbonate precipitation in these sediments is regarded as a major sink of methane derived carbon and seawater Ca (Han and Suess, 1989), but precipitation mechanisms and rates are still poorly understood.

Sulfur is a representative of the geochemical reduction phase, it has significant meaning for our understanding of the environment of the early diagenetic stage. Pyrite is one of the most widely distributed sulfides in marine sediments. The formation of pyrite needs stronger reductive environment than that of siderite. The presences of organic matter and hydrogen sulfide ( $\text{H}_2\text{S}$ ) and the associated anaerobic environment are important conditions for the precipitation of pyrite. The  $\text{HS}^-$  generated from Equation (1) reacts with  $\text{Fe}^{2+}$  provided by dissolution of some Fe-bearing minerals, resulting in pyrite precipitation (expressed by Eq. (3)). The pyrite in cold-spring carbonates often occurs in the shape of strawberry.



Studies on distribution of sea-bed cold-spring deposits and gas hydrates occurrence conditions are carried out mostly through the drilling and sediment sampling method, which are very costly and cannot be widely used. Here we attempt to use numerical simulation methods to investigate processes of mineralization induced by the cold seeps of  $\text{CH}_4$  at the sea bottom and associate conditions.

Several numerical models have been developed, which concentrated mainly on the characterization of AOM rate (Regnier et al., 2011), the effective leakage rate of methane emission from sediment to seawater (Luff and Wallmann, 2003; Liu and Wu, 2014), and the relationship between the rate of methane leakage and the type of self generated minerals (Luff and Wallmann, 2003). The theoretical model of AOM reaction rate is divided into three categories (Regnier et al., 2011): biogeochemical models, metabolic models and biomass-explicit models. In addition, several researchers have studied the control effect of AOM process on methane emission (e.g., Luff et al., 2004; Luff and Wallmann, 2003; Dale et al., 2008b; Liu and Wu, 2014). The results of these studies show that methane flux would be consumed in the sediment via the dissolution, sorption, and AOM reaction. Therefore, a portion of methane is able to enter the water column and possibly escape to the atmosphere only when the flux flows at a higher rate. The currently existing works are mainly focused on the control of AOM by biological and bioenergy and the relationship between the rate of methane leakage and the type of self generated minerals. Much less attention was paid to the relationship between AOM-SR reaction and diagenesis. Here a model coupling AOM, solute transport and diagenesis was used to explore the AOM biogeochemical processes and its inducing mineralization. The data used in this study were extracted from methane-bearing sediments located in the northern continental slope area of the South China Sea. One-dimensional (1D) column model was built to investigate the

response of AOM and authigenic minerals to the gas hydrate geosystems. Three scenarios were designed to identify variations in the depth of sulfate methane transition zone (SMTZ) and the mineral composition.

## 2. Geologic setting

A great deal of specific and detailed information is required to model the AOM biogeochemical processes in methane-bearing sediments at any particular site. Before moving into site-specific investigations, general features and issues should be explored. This can be done by investigating a methane-bearing sediment system below sea bottom that abstracts site-specific features and thereby attempt to represent characteristics that are common to many such systems. Here, geological characteristics and physico-chemical conditions are mainly the northern continental slope area of the South China Sea (Fig. 1).

The South China Sea (SCS) is one of the largest marginal seas in the West Pacific and is located at the junction of the Eurasian, Pacific, and Indian-Australian tectonic plates (Zhang et al., 2014). The marine sediments on the continental slope of the NE South China Sea are considered a favorable place to host gas hydrates, because the region has a high sedimentation rate, thick sediments and suitable pressure–temperature regime (Yang et al., 2010; Zhang et al., 2014).

Our study area is located in the NE South China Sea, near the bottom of the continental slope of Taiwan Island (Fig. 1). The area is near the border of the Luzon subduction system transforming into an incipient arc-continent collision with the Philippine Sea Plate moving at a rate of ~8 cm/yr toward the Eurasia Plate (Lim et al., 2011). Fluid seepages and gassy sediments as well as mud diapirs were identified (Chiu et al., 2006; Lim et al., 2011). Normal faults, thrust faults, mud volcanoes, and mud diapirs could provide good conduits for methane-charged fluids migrating from deeper sediments (Lim et al., 2011). According to the study of Liu et al. (2006), the bottom simulating reflectors (BSRs) covered most of the study area, indicating abundant gas hydrate at depth in the region. Chuang et al. (2006) showed that the area is characterized by high concentrations of methane in sediments. In addition to high concentrations of methane in the pore water, high content of authigenic carbonates and pyrite were observed in this area (Yang et al., 2010; Tong et al., 2013; Han et al., 2014; Zhang et al., 2014; Ge et al., 2015).

## 3. Methodologies

### 3.1. Numerical simulator

All simulations were performed using TOUGHREACT, which is a numerical simulation program for chemically reactive non-isothermal flows of multiphase fluids in porous and fractured media. The program was written in Fortran 77 and developed by introducing reactive chemistry into the multiphase fluid and heat flow simulator TOUGH2 (Xu et al., 2011). The numerical method for fluid flow and chemical transport simulations is based on the integral finite difference (IFD) method for space discretization. TOUGHREACT allows comprehensive modeling of biogeochemical interactions between fluid and solid phases that are coupled to solute transport. The program is applicable to one-, two-, or three-dimensional geologic domains with physical and biogeochemical heterogeneity. The further descriptions about this code are provided in Appendix A.

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