



Research paper

Laboratory experiment and numerical simulation on authigenic mineral formation induced by seabed methane seeps

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ABSTRACT

In natural gas hydrate marine settings, cold seepage of methane fluid is a widely observed phenomenon, where authigenic minerals serve as an indication of potential gas hydrate-bearing reservoirs at depth. In this study, based on the data from the site HD196 near Dongsha Island, northern continental slope of South China Sea, laboratory experiments and numerical simulation studies were conducted to investigate the biogeochemical processes of authigenic mineral formation induced by methane seepage. The bioreactor experimental results show that in response to methane flux, pH increased to 8.5 after 20 days of reaction, and Eh declined rapidly first and then remained unchanged at about 100 mV. The decrease in SO_4^{2-} , HS^- and HCO_3^- concentrations indicated the occurrence of anaerobic oxidation of methane coupled with sulfate reduction (AOM-SR). The depletion of Fe^{2+} implied the formation of iron-bearing minerals, e.g., iron sulfides. Carbonate minerals were also identified in the experimental phase of this study. Most iron sulfides existed as massive pieces, and in some cases as spherical or rod-shape pieces. The calcium carbonates were observed as blocky pieces. Numerical simulations were also performed to reproduce the biogeochemical reactions that occurred in the reactor experiments. Based on experimental data, kinetic parameters associated with the observed reactions were calibrated. The model simulated results are general consistent with those obtained by the experiments conducted in this study. The combination of simulation and experimental studies provided a powerful tool to investigate the biogeochemical processes in the methane leakage environment at different temporal and spatial scales. This study gave a new perspective to understand the formation of cold seep authigenic minerals in marine sediments, and was significant for future investigations on the effects of hydrate decomposition.

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1. Introduction

Natural gas hydrates are receiving increasing attention around the world since its finding in marine sediments (Luff and Wallmann, 2003). Gas hydrates are typically composed of natural gas, main methane (CH_4), within a rigid lattice of water molecules, and form an important reservoir in the global carbon cycle (Dickens, 2001; Chuang et al., 2013). At the same time gas hydrates are unstable in shallow sediments below sea bed, the change of temperature and pressure may lead to the decomposition of gas hydrate and release a large amount of methane (Maslin et al., 2004).

There are two important biogeochemical processes induced by the leakage of methane: (1) Microbially-mediated redox reactions such as anaerobic oxidation of methane coupled to sulfate

reduction (AOM-SR) (Joye et al., 2004; Segarra et al., 2013). (2) The formation of associated authigenic minerals in sediment pore space, which 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 part of South China Sea (Himmeler et al., 2013; Tong and Chen, 2012; Treude et al., 2003; Tsunogai et al., 2002). These associated minerals have great significances for studying ocean gas hydrate and global carbon and sulfur cycle (Liu et al., 2016b).

Most existing studies of cold seep biogeochemistry focus on the field investigations (Knittel and Boetius, 2009; Reeburgh, 2007). Due to the complexity and diversity of the seabed environments, it is necessary to use laboratory incubation and numerical simulation methods to investigate these biogeochemical processes. Some studies have shown the feasibility to do vitro experiments about biogeochemical processes in marine sediments (Steeb et al., 2014; Zhang et al., 2010, 2014b). Wei et al. (2015) did some experiments on the effect of sulfate-reducing reaction on water chemical

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compositions in methane seeping environment. Liu et al. (2016a) studied anaerobic oxidation of methane and precipitation of carbonate mediated by microbes. Meanwhile several numerical models have been developed, which concentrated mainly on the characterization of AOM-SR rate (Regnier et al., 2011), the effective leakage rate of methane emission from sediment to seawater (Luff and Wallmann, 2003; Liu and Wu, 2014). However, there are few experimental and simulation studies on the formation of authigenic minerals (Han et al., 2016; Liu et al., 2016b).

In this study, based on the data taken from the site HD196 in Dongsha Island, northern continental slope of South China Sea, we focus our efforts on investigating the microorganism-mediated reaction and mineral alteration through experiments and numerical simulations. A bioreactor system was used to create a real, methane-rich seabed environment, under which the formation of authigenic mineral is expected to occur. Numerical simulations were also carried out to reproduce the biogeochemical processes taking place in the bioreactor. Comparing the data obtained by modellings and experiments, some kinetic parameters used to calculate the mineral reaction rate were calibrated.

2. Materials and methods

2.1. Preparation of materials

2.1.1. Cultivation of microorganisms

The microorganisms were cultivated from the seabed sludge samples collected from the seabed of Northern continental slope of South China Sea. Some sludge was taken into the appropriate amount of sterile water, after 5 min' shaking and 2 h' standing; the supernatant was obtained and then placed it into the culture medium and kept in an anaerobic and thermostatic container. The container was filled with excess methane at a temperature of 35 °C. Each liter of the culture medium consisted of: K₂HPO₄ 0.5 g, NH₄Cl 1.0 g, Na₂SO₄ 0.5 g, MgSO₄·7H₂O 2.0 g, NaCl 2.0 g, CaCl₂·2H₂O 1.0 g, (NH₄)₂Fe(SO₄)₂·6H₂O 0.1 g, and bicarbonate solution 10 ml. The bicarbonate solution was configured according to the work of Widdel and Bak (1992). After a period of time (usually 15 days), the process was repeated several times more, the cultivation was finished until that the microbial concentration and purity met the requirements, bacterial solution for later experiments was obtained.

2.1.2. Configuration of simulated seawater

The simulated seawater contained inorganic salts at concentrations found in sediment pore water at site HD196 in Dongsha Island, northern continental slope of South China Sea (Table 1).

Superfluous nitrogen and phosphorus were provided to promote the microbial growth. The concentration of sulfate (SO₄²⁻) here was higher than the value at sulfate-methane transition zone (SMTZ) to investigate the initial process of sulfate reduction.

2.1.3. Preparation of solid phases

The mineral composition of superficial sediments in the northern South China Sea is very complex, and is also difficult to define the original mineral composition prior to formation of authigenic minerals. In this study, we chose the specially processed quartz sands (99.9% purity) as the media, which were only used to simulate the sedimentary framework and not participated in the reactions. The possible effects of the dissolution of primary minerals in the actual environments were reflected by the simulated seawater. The quartz sands were selected and grinded first to ensure that their particle sizes were within the range of 1.5–14.0φ (Zhang et al., 2003). Limited by the laboratory experiment conditions, the amount of minerals formed during the experiment was small, which leads to the difficulty in collecting and testing the authigenic minerals. We used processed granite slices (collected from the Songliao Basin, China) as intermediate carriers to collect the minerals during the experiment. These quartz sands and granite slices were washed in dilute hydrochloric acid, and then rinsed by distilled water and dried at 35 °C. Before the experiment, X-ray Diffraction (XRD, DX-2700, Kemait NDT Co., Ltd, China) analysis of granite slices was carried out. The results of XRD show that the granite slices consist of (in wt. %) n 22 quartz, 33 plagioclase, 37 alkali feldspar, 5 biotite and 3 Kaolinite on average. According to Acker and Bricker (1992), the dissolution rate of biotite in the near-neutral and low temperature environment is very slow. Under our experimental conditions and the short experimental time, biotite can be regarded as not involved in the reaction.

2.2. Experiment

2.2.1. Experimental set-up and procedures

The experiments were conducted in a low-temperature and high-pressure bioreactor with a volume of 1.0 L, which allowed for external control and monitoring of pressure and temperature (Fig. 1). The minimum operating temperature and maximum pressure were 0 °C and 30 MPa, respectively.

At first, the simulated seawater and solids were put into the sterilized bioreactor, granite slices were placed in the middle of sands layer, and then bacteria solutions were injected into the bioreactor through an injection system. After evacuation, methane was added into the bioreactor to raise the pressure. During the entire experiment, temperature (10 °C) and pressure (5 MPa) in the

Table 1

Geochemistry of pore waters from sediments at site HD196 and corresponding composition of the simulated seawater.

Sediment pore water ^a		Simulated seawater			
Chemical species	Concentration (g/L)	Chemical	Concentration (g/L)	Chemical species	Concentration (mol/L)
Ca ²⁺	0.402	CaCl ₂ ·2H ₂ O	1.485	Ca ²⁺	0.0101
Mg ²⁺	1.232	MgCl ₂ ·6H ₂ O	10.414	Mg ²⁺	0.0513
Na ⁺	11.868	NaCl	24.125	Na ⁺	0.516
K ⁺	0.475	KCl	0.703	K ⁺	0.0122
Fe ²⁺	—	(NH ₄) ₂ FeSO ₄ ·6H ₂ O	0.075	Fe ²⁺	0.000254 ^b
SO ₄ ²⁻	2.732	Na ₂ SO ₄	4.011	SO ₄ ²⁻	0.02856
HCO ₃ ⁻	—	NaHCO ₃	0.168	HCO ₃ ⁻	0.002 ^b
Cl ⁻	19.879	—	—	Cl ⁻	0.5667
NH ₄ ⁺	—	NH ₄ Cl	1.150	NH ₄ ⁺	0.0220 ^c
PO ₄ ³⁻	—	K ₂ HPO ₄	0.240	PO ₄ ³⁻	0.00138 ^c

^a (Wu et al., 2010).

^b (Liu et al., 2016b).

^c The concentration has been adjusted to achieve a C:N:P ratio of 106:16:1.

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