

## Study on Gas Hydrate Dissociation in Small Bodies of Hydrate-bearing Sediments under Water-heating Condition

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### ABSTRACT

High efficiency, economic, and safe exploitation of natural gas hydrate is an important researching topic. Mechanical-Thermal exploitation is a new presented potential efficient method for shallow marine hydrate exploitation, and contains the following procedures: In-situ mining of hydrate-bearing sediments, cutting the sediments into small bodies, mixing the sediments with surface injected seawater, transporting the multiphase fluid with hydrate dissociation in the exploitation well, and backfilling the sediments, etc. The physical processes of small bodies of hydrate-bearing sediments and water flow accompanying hydrate dissociation are described in the main controlling parameters. Then some trial observational tests are conducted to obtain information on the dissociation process of gas hydrate in small bodies under water-heating condition.

**KEY WORDS:** Mechanical-Thermal exploitation; multiphase flow; hydrate.

### INTRODUCTION

Gas hydrate (GH) is a solid compound of hydrocarbon gas and water molecules. Gas hydrate-bearing sediments (GHBS) consist of hydrate, water or/and gas, sand/clay etc. GH exists in cementing or filling status with soil skeleton, and widely distributes in the sea, permafrost and deep lakes (Kvenvolden and Lorenson, 2001; Koh, 2002; Song et al., 2014).

Countries such as Russia, Canada, America and Japan have carried out trial productions of GH in the permafrost (Makagon et al., 2005, 2013; ConocoPhillips, 2012a, b; Collett et al., 2012; ) and deep marine (Fujii et al., 2013; Chee et al., 2014; Terao et al., 2015). The methods of GH exploitation include thermal injection, depressurization, and CO<sub>2</sub> displacement. The trial production of GH gives the confidence that increasing temperature or/and decreasing pressure could release methane from GHBS in a short period, but the efficiencies of these productions are hard to satisfy a commercial-viable application.

The physical processes in the production contain heat conduction, phase transformation, multiphase seepage and soil deformation (Moridis et al., 2009). The ratio of the characteristic times is 10<sup>9</sup>:10<sup>7</sup>:10<sup>6</sup>:1. The heat conduction is the slowest physical effect, and controls the coupling processes. Lack lasting supply of heat into the GHBS leads to the low efficiencies of the in-situ trial production, and constrains the utilization of the methods such as depressurization and thermal injection (Hong et al., 2003; Zhang et al., 2014a).

The formation and geological characteristics of GHBS in South China Sea are more complex and inhomogeneous. The reserve is large but in a scattered spatial distribution. The sediment is soft and the permeability is low. Through a preliminary estimation of thermal injection, the expansion length of hydrate dissociation zone gets to merely about 30 m after 20 years, leading to a serious situation of high investment and low profit, especially at the period of low price of international oil and natural gas (Zhang et al., 2014a, b).

In view of the concern in the production efficiency, new technologies are required to increase the rate of heat transfer. Improving the heat area of GHBS is an essential solution, and transferring GHBS into small blocks and heating in warm water is a potential method. Mechanical-thermal exploitation is a new presented method for shallow marine hydrate exploitation considering the utilization of surface warm seawater and convective heat transfer (Zhang et al., 2014b).

The aim of this paper is introducing the new method of mechanical-thermal exploitation, obtaining the dimensionless controlling parameters during hydrate dissociation, and presenting some trial tests to illustrate the dissociation characteristics of small bodies of GHBS in warm water.

### MECHANICAL-THERMAL HYDRATE EXPLOITATION

Mechanical-thermal hydrate exploitation method is presented considering the hydrate formations of large reserve, low permeability, soft soils, and low connectivity. The basic idea of mechanical-thermal

exploitation is that utilizing the huge heat of surface seawater, and convective heat transfer between water and small bodies of GHBS. The new technology includes following procedures as in Fig. 1.

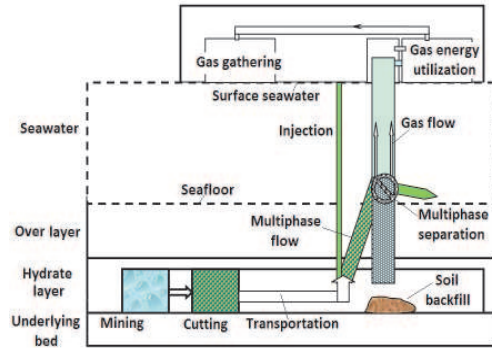


Fig. 1 Sketch of mechanical-thermal hydrate exploitation

Underground in-situ mining similar to coal mining, cutting the sediments into small bodies, and transporting them to the well: The GHBS is excavated from the ground, crushed into a certain size (mm-cm) of small bodies, forced into the well, and mixed with injected warm surface seawater.

Multiphase flow with hydrate dissociation: Surface seawater is injected into the well in certain velocities to force the small bodies of GHBS flowing with water to a certain height (The fluidization status is appreciated due to the energy optimization). GH dissociates in the well during the upward flow, and the dissociation rate is determined by the water temperature, flow velocity, and contact area, etc.

Gas separation and soil backfilling: On the seafloor, a multiphase separation system is required to trap the soil bodies, backfill the soils into the excavated zones to keep the stratum stable, discharge water into the sea, and let the gas flow upwards to the gathering installation.

The main problems in the mechanical-thermal hydrate exploitation method include:

(i) Deep water mining under high pressure and low temperature. This is a researching project in many countries. It can be referred to some mechanical installations in coal mining, deep water mining on the seabed, and the plan of Sweden deep formation mining of environmental viable, safe, automatic, continuous operation (Hoagland et al., 2010; Lu and Zhang, 2012; Boschen et al., 2013). The mechanical mining technologies are beyond our research domain and not discussed further here.

(ii) Multiphase flow containing GH dissociation. Small bodies of GHBS will dissociate rapidly in the warm seawater, releasing a large number of bubbles. Then different flow patterns (solid-liquid flow, solid-liquid-gas bubble flow, liquid-gas flow, etc.) occur in the well. These physical processes involve non-equilibrium dynamics, and important to the optimization design of the new technology. The kinetic model of small bodies of GHBS in water should be established considering convective heat transfer and migration of gas bubbles in the small bodies.

(iii) The separation of the mixture of gas, water, soil and the backfilling of the sediments. In this problem, the aim is to decrease the elevation of the sediments. The centrifugal separation and the variable cross-section separation are two potential methods.

(iv) The instability of stratum with a slope. How to optimize to excavate the stratum and keep it stable is to be studied. Local settlement, failure and large slide should be concerned in a deep water condition.

In view of energy and economy, the method is viable through preliminary estimation. If the expansion energy of released gas is utilized fully, the energy consumption in the mechanical work, hydraulic transportation can be self supplied. In the present value method of economic estimation, assuming 10000 m<sup>3</sup> gas produced from one well, the income of the produced gas will exceed the investment after about 4 years (Lu and Zhang, 2012).

## PHYSICAL PROCESSES AND CONTROLLING PARAMETERS

Here, we concern the multiphase flow containing gas hydrate dissociation in the new technologies, i.e. small bodies of GHBS and seawater flow upwards in the well, the small bodies are heated in the water, and hydrate dissociates fast. First the hydrate soil bodies (temperature  $T_{w0}$ , supply rate  $v_{hs0}$ ) and water (temperature  $T_{w0}$ , pressure  $P_{w0}$ , supply rate  $v_{hs0}$ ) form a fluidized bed in well 1, the hydrate in small soil bodies dissociates into water and gas, and the gas separates from the soil and flows with the water. Then the hydrate is anticipated to complete dissociation when the soil reaches the seafloor, where centrifugal or other separation method will be used to let the gas flow upwards, backfill the soil, and discharge the water.

In view of the above processes, the gas occurs in the form of dissolved state, small bubbles, and gas plug. So the multiphase flow can be described into four categories in the well: solid-liquid two-phase flow, solid-liquid-gas bubble flow with hydrate dissociation, solid-liquid-gas flow, liquid-gas plug flow, as in Fig. 2. These physical processes involve in non-equilibrium dynamics problems, and are essential to the optimization design of well size, flow rate and solid-liquid concentration.

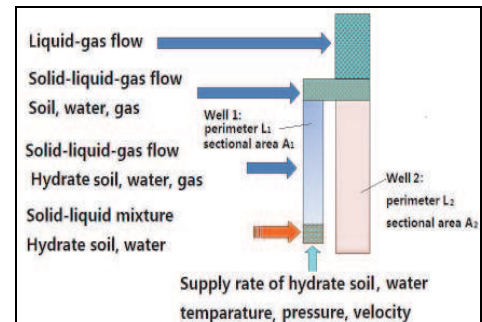


Fig. 2 Multiphase flow containing hydrate dissociation

The physical processes contain different geometrical scales and time scales. The choice of the characteristic time  $\bar{t}$ , length  $\bar{x}$ , temperature  $\bar{T}$  is important (Tan, 2011). The main dimensionless controlling parameters are obtained through dimension analysis and expressed in table 1. Here,  $\bar{x} = d, \bar{u} = v_{w0}, \bar{t} = d / v_{w0}, \bar{P} = P_{w0}, \bar{T} = T_{w0}$ ,  $\xi(T)$  is hydrate dissociation constant,  $H_{w-hs}$  is interaction force between water and hydrate-soil bodies,  $q_{w-s}$  is heat flow of well side.

Three characteristic times are included in this problem, i.e. fluid

flow  $\bar{u} \cdot \bar{t} / \bar{x}$ , convective heat transfer  $6\psi(\text{Re}, \text{Pr})\bar{t} / \rho_w C_w \bar{x}$ , phase change  $\xi(T)\bar{t} / \rho_w$ , and heat conduction  $\lambda_w \bar{t} / \rho_w C_w \bar{x}^2$ .

We assume that the water velocity is 1 m/s, the diameter of the soil bodies is 1cm, the diameter of the well is 10cm. Other parameters are referred to Waite et al. (2009) and Zhang et al. (2014a). The ratios of the times are as follows:

$$\frac{\bar{x}}{\bar{u}} \frac{\rho_w}{\xi(T)} = 10^{-7}, \quad \frac{\bar{x}}{\bar{u}} \frac{\rho_w C_w \bar{x}^2}{\lambda_w} = 4 \times 10^{-5}, \quad \frac{\bar{x}}{\bar{u}} \frac{\rho_w C_w \bar{x}}{6\psi(\text{Re}, \text{Pr})} = 4 \times 10^{-4}.$$

Table 1. Dimensionless parameters

Dimensionless parameters	Physical meanings	Dimensionless parameters	Physical meanings
$\bar{u} \cdot \bar{t} / \bar{x}$	Fluid flow	$\bar{u} \cdot \bar{t} \cdot H_{w-hs} / \rho_w$	Interaction force
$\xi(T) / (\rho_w \cdot \bar{x})$	Dissociation	$\bar{u}^2 / (C_w \cdot \bar{T})$	Ratio of kinetic to internal energy
$\lambda_w \cdot \bar{t} / (\rho_w C_w \bar{x}^2)$	Heat conduction	$q_{w-s} \cdot l \cdot \bar{t} / (\rho_w \cdot C_w \cdot \bar{T} \cdot A)$	Side heat conduction
$\bar{u}^2 / (g \cdot \bar{x})$	<i>Fr</i> number	$\bar{P} / (\rho_w \cdot \bar{u}^2)$	<i>Eu</i> number
$l \cdot \bar{t} / (\rho_w \cdot \bar{u} \cdot A)$	Side friction	$d / D$	Ratio of grain size to well diameter
$\psi \cdot \bar{t} / (\rho_w \cdot C_w \cdot \bar{x})$	Convective heat transfer	$\bar{u} \cdot \rho_w \cdot \bar{x} / \mu_w$	<i>Re</i> number
$\Delta H / (C_w \cdot \bar{T})$	Latent heat	$\beta_w \cdot \beta_{hs}$	Volumetric fraction

## TRIAL TESTS AND TESTS

Kim model and Stefan model (Moridis et al., 2009) are used to describe the dissociation of GH in GHBS, but research should be conducted to make clear that whether these models are applicable to hydrate dissociation in small bodies in a convective heat transfer condition. After the solution of the dissociation rate of the hydrate-bearing small bodies, how to optimize the size of the small bodies, the solid concentration, the size of the well, and the velocity of the injected seawater will be determined.

### Test preparation

The test material was prepared in the hydrate-bearing sediment formation apparatus. The soil skeleton was silty-sand, with a dry density of 1.6 g/cm<sup>3</sup> and porosity of 0.4. The hydrate saturation was 2%-40%, and the test samples were cut into small cubic bodies of diameter 2 cm -3 cm in a cold room to prevent hydrate dissociation. The test apparatus of hydrate dissociation and flow was a cylindrical organic glass tube of 5 cm diameter and 2 m length, a temperature sensor was arranged in the apparatus to measure the evolution of the water temperature during hydrate dissociation. The gas was gathered at the top of tube and measured in displacement method.

### Testing phenomena

In the static water with a temperature of 30°C, hydrate in the small bodies dissociated rapid, as in Fig. 3. The testing phenomena can be expressed as follows: (i) Rapid dissociation of hydrate. Gas released from the soil bodies similar to an explosive case, and small bubbles flowed in the water upwards. In this process, the enough heat was transferred into the surface hydrate of the small soil bodies due to direct contacts between water and hydrate, and the rapid react occurred, leading to explosive gas release. A group of gas bubbles moved due to the buoyant force. (ii) Surface soil particles were separated from the small bodies and elevated to a certain height. The reason was that after

From the three ratios, a solution can be concluded that the characteristic time of the hydrate dissociation is the slowest, followed by the heat conduction and the convection heat transfer, and the fluid flow is the fastest. The time of hydrate dissociation is two orders longer than others, so the physical process can be solved in a decoupling method, i.e. the hydrate dissociation of small hydrate soil bodies in water can be analyzed first.

the dissociation of hydrate, the cohesion between soil particles decreased greatly. Then the surface soil particles were separated due to gravity, but the gas expansion led to a phase-interaction force and forced small particles upwards, then the dispersed or settled particles were observed in the test apparatus. Soil particles (several microns to tens of microns) moved up to a certain height with the bubbles. The present models are invalid to describe these processes.

In the following tests, the affect of temperature was investigated. The temperature arrangements, the sample length, and the size of small bodies were given in table 2. For the former five groups of tests, the samples were cut into small cubic bodies of 2 cm, and dissociated in 30 °C water in a static status. For the later two groups of tests, the small bodies were decomposed in 11 °C and 0 °C water in a stirring status.

Table 2. The basic test conditions

Hydrate saturation (%)	Total sample length (cm)	Small body length (cm)	Temperature(°C)
2.0	10	2	30
2.4	10	2	30
2.5	12	2	30
11	14	2	30
18	12	2	30
23	2.8	2.8	11
23	2.5	2.5	0



(a) dissociation phenomenon



(b) fluid flow with soil grains during dissociation

Fig. 3 Test phenomena during hydrate dissociation of small GHBS bodies in warm water

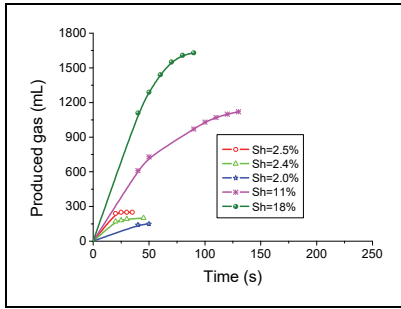


Fig. 4 Test results in static water

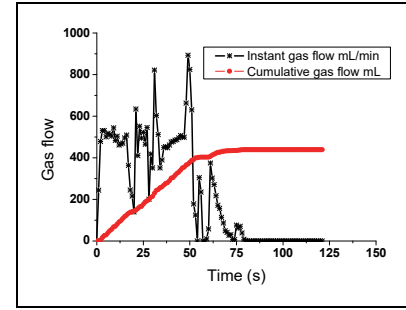
Hydrate-soil bodies dissociated fast in the warm water as in Fig. 4. At the temperature of 30 °C, the dissociation completed in 100 seconds. In the convective heat transfer condition, the rate would increase. If the estimation of characteristic length ( $x = \sqrt{\lambda t / \rho C}$ ) of solid heat conduction is used, the influence length of hydrate dissociation is about 1 cm in 100 seconds, and the length will be less referred to Kamath et al. (1984; 1987) which considers phase transition in heat conduction.

In Fig. 5, the instant and cumulative gas production was recorded in the tests of water stirring condition. In the case of 11 °C, the dissociation rate was nonlinear, changed with the stirring processes, and the final production gas volume was consistent with the measured total gas volume in the hydrate-soil bodies. While in the case of 0 °C, the dissociation rate decreased rapidly after 10 seconds, and the final produced gas volume was much less than the measured total gas volume. The change of the dissociation rate could be explained that during the stirring process, the contact area between the water and hydrate and local water velocity changed accompanying hydrate dissociation. In the case of 0 °C, the initial hydrate dissociation decreased the temperature of the local water, ice could form around the soil bodies, and prevented the continuous dissociation of hydrate. So the heat energy should be estimated considering the requirement of hydrate dissociation and ambient heat.

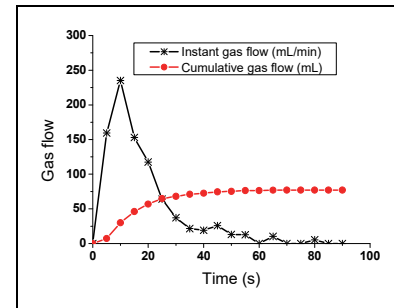
According to above test results, high temperature and large heat supply can lead to the rapid GH dissociation and unstable multiphase flow, while low temperature and small heat supply can lead to GH reformation or freezing. Provided the temperature of the surface seawater is about 25 °C, it will reduce about 10 °C via the vertical well to the hydrate stratum (Lu and Zhang, 2012), and the volumetric ratio between injected water and small bodies of GHBS should be higher than about 0.8 to satisfy the requirement of the complete GH dissociation.

GH dissociation rate and the hydraulic elevation velocity should be

controlled, and the aim is that when the small bodies of GHBS reach the seabed, GH has dissociated completely. It is noted that the mechanism of GH dissociation rate in small bodies of GHBS will be studied in further experimental and theoretical work, especially the physical effect of convective heat transfer and the heat distribution in each phase (gas, water, soil) during hydrate dissociation.



(a) temperature 11 °C



(b) temperature 0 °C

Fig. 5 Test results in stirring water

## CONCLUSIONS

In this paper, a new hydrate exploitation method, mechanical-thermal hydrate exploitation, is presented for the shallow marine hydrate deposits, the physical processes of multiphase flow containing gas hydrate dissociation are described in the main controlling parameters. Then observational tests are conducted to obtain information on the dissociation process of gas hydrate in small bodies under water-heating condition.

The procedures of the new method include in-situ mining of hydrate-bearing sediments, cutting the sediments into small bodies, mixing the sediments with injected surface seawater, transporting the multiphase fluid accompanying hydrate dissociation in the exploitation well, and backfilling the sediments, etc.

The small bodies of GHBS and water flow in the well contains four characteristic times, as heat conduction, convective heat transfer, hydrate dissociation, fluid flow. Through the three ratios of these times, hydrate dissociation is two orders longer than others, and the physical processes can be analyzed in a decoupled method i.e. hydrate dissociation in water can be analyzed first.

Small bodies of GHBS dissociate much faster in the warm static water at the temperature of 30 °C than solid heat conduction. The dissociation rate increases after stirring of the water. When the sum of heat is lack, the dissociation slows even stops due to ice formation or hydrate reformation.

This work presents a preliminary research on the mechanical-thermal hydrate exploitation method, and several complex non-equilibrium dynamics problems will be solved in future in order to optimize the efficient, economic, and stratum safe utilization of the technologies, especially to determine the water flow velocity, the size of hydrate soil small bodies and well, solid concentration, etc.

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## REFERENCES

- Boschen, R E, Rowden, A A, Clark, M R, et al. (2013). "Mining of deep-sea seafloor massive sulfides: A review of the deposits, their benthic communities, impacts from mining, regulatory frameworks and management strategies," *Ocean & Coastal Management*, 84: 54-67.
- Collett, T S, Boswell, R, Lee, M W, et al (2012). "Evaluation of long-term gas-hydrate-production testing locations on the Alaska North Slope," *SPE Reservoir Evaluation & Engineering*, 243-264.
- ConocoPhillips (2012a). "ConocoPhillips gas hydrate production test," *Oil & Natural Gas Technology*, Progress report first half 2012, Office of Fossil Energy, Anchorage, USA, July 31.
- ConocoPhillips (2012b). "ConocoPhillips gas hydrate production test," *Oil & Natural Gas Technology*, Progress report second half 2011, Office of Fossil Energy, Anchorage, USA, January 31
- Chee, S, Leokprasirtkul, T, Kanno, T, et al (2014). "A deepwater sandface monitoring system for offshore gas hydrate production," *Offshore Technology Conference*, Houston, Texas, USA, 2014
- Fujii, T, Noguchi, S, Takayama, T, et al. (2013). "Site selection and formation evaluation at the 1st offshore methane hydrate production test site in the eastern Nankai Through," *75th EAGE Conference & Exhibition incorporating SPE EUROPEC 2013*, London
- Hong, H, Pooladi-darvish, M, Bishnoi, PR (2003). "Analytical modeling of gas production from hydrates in porous media," *Journal of Canadian Petroleum Technology*, 42(11): 45-56.
- Hoagland, P, Beaulieu, S, Tivey, M A, et al. (2010). "Deep-sea mining of seafloor massive sulfides," *Marine Policy*, 34: 728-732.
- Kamath, VA, Holder, GD, Angert, PF (1984). "Three phase interfacial heat transfer during the dissociation of propane hydrates," *Chemical Engineering Science*, 39(10): 1435-1442.
- Kamath, VA, Holder, GD (1987). "Dissociation heat transfer characteristics of methane hydrate," *AIChJ*, 33(2): 347-350.
- Kvenvolden, K A, Lorenson, T D (2001). "The global occurrence of natural gas hydrate," *Geophysical Monograph*, 124:3-18.
- Koh, C A (2002). "Towards a fundamental understanding of natural gas hydrates," *Chem. Soc. Rev.*, 31:157.
- Lu, X B, Zhang, X H (2012). "Feasibility study on mechanical-thermal hydrate recovery method," *Scientific Report in Institute of Mechanics*, Chinese Academy of Sciences (in Chinese)
- Makogon, Y F, Holditch, S A, Makogon, T Y (2005). "Russian field illustrates gas-hydrate production," *Oil and Gas Journal*, 103: 43-47.
- Makogon, Y.F., Omelchenko, R.Y. (2013). "Commercial gas production from Messoyakha deposit in hydrate conditions," *Journal of Natural Gas Science and Engineering*, 11: 1-6.
- Moridis, G J, Collett, T S, Boswell, R, et al. (2009). "Toward production from gas hydrates: Current status, assessment of resources, and simulation-based evaluation of technology and potential," *SPE Reservoir Evaluation & Engineering*, 12(5): 745-751.
- Song, YC, Yang, L, Zhao, JF, et al. (2014). "The status of natural gas hydrate research in China: A review," *Renewable and Sustainable Energy Review*, 31: 778-791.
- Tan, QM (2011). *Dimensional Analysis: With Case Studies In Mechanics*. Berlin, Heidelberg: Springer-Verlag Berlin Heidelberg.
- Terao, Y, Lay, K, Yamamoto, K (2015). "Design of the surface flow test system for 1st offshore production test of methane hydrate," *Offshore Technology Conference Asia*, Kuala Lumpur Malaysia.
- Waite, WF, et al. (2009). "Physical properties of hydrate-bearing sediments," *Rev. Geophys.*, RG4003/2009, 47: 1-38.
- Zhang, XH, Lu, XB, Zheng, ZM, et al. (2014a). "Heat-induced evolution of phase transformations in tetrahydrofuran hydrate-bearing sediment," *Journal of Heat transfer-Transaction of the ASME*, 136: 052002.
- Zhang, XH, Lu, XB, Liu, LL (2014b). "Advances in natural gas hydrate recovery methods," *Progress in Geophysics (in Chinese)*, 29(2): 0858-0869.