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Reviews

Introduction to exploration research on gas hydrates in Japan

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Abstract : Marine gas hydrates around Japanese islands are regarded as one of the prospective unconventional hydrocarbon resources in Japan. The Geological Survey of Japan (GSJ) is vigorously conducting the basic research on exploration techniques and feasibility studies for the development of gas hydrate. In relation to the GSJ research, the Ministry of International Trade and Industry (MITI) and the Japan National Oil Corporation (JNOC) have a plan for the first offshore drilling for hydrate exploration on the continental slope inside the Nankai Trough in 1999.

1. Introduction

Natural gas hydrates are a kind of clathrates, which contain natural gas such as methane in a framework of water molecules. 1 cubic meter of pure methane hydrates theoretically can contain up to 172 cubic meters of methane gas at standard condition. However, in natural gas hydrates, methane gas volume can be expected approximately 40 to 165 times greater. Therefore, natural gas hydrates are regarded as one of the unconventional potential resources in future, distributed in the permafrost regions of Arctic and Antarctic and deep-sea areas of continental margins including those around the Japanese Islands.

The phase equilibrium indicates that the maximum temperature for the occurrences of gas hydrates is higher than water freezing point under high pressures. The pressure higher than 26 atmospheres at 0℃ and 76 atmospheres at 10℃ are necessary for the methane hydrate formation (Fig. 1).

If 1 atmosphere is assumed for 10 meters of the water depth, the pressure of 26 atmosphere at 0℃ and that of 76 atmospheres at 10℃ correspond to the water depths of 260 and 760 meters in the ocean respectively, for the occurrences of methane hydrates. Therefore, it is considered that the occurrences of methane hydrates in nature are controlled by the factors such as temperature, pressure (depth) and compositions of gas and water. It is generally considered that exploration targets of marine methane hydrates in the middle latitudes are confined in the areas deeper than 500 meters.

Strong negative phase of anomalous reflections are often observed on seismic reflection profiles (Fig. 2) at the depth of more than 500 meters around the continental margins of the world, which is corresponding to the base of hydrate stability zone estimated by the geothermal gradient and the depth. These reflectors are called Bottom Simulating Reflectors (BSRs), which can be regarded as a significant indicator at the initial stage of natural exploration.

The Geological Survey of Japan (GSJ) has identified the potential of natural gas hydrates in the offshore area around Japan based on the studies of the BSRs on the seismic profiles and the geothermal gradient corresponding to the results of methane hydrate synthesis experiments. The BSRs are correlated to the methane hydrate phase equilibrium obtained from the experiments in our laboratory, which revealed interrelationship among the factors such as temperature, pressure (depth) and compositions of gas and water.

This paper gives an outline of the methane hydrate research in GSJ as an introduction to this special issue on hydrates, and special emphasis is given on some experimental results of the hydrate formation and dissociation using a unique equipment for methane hydrate synthesis. The distribution of methane hydrates around Japan obtained by correlation of the BSRs and geothermal gradient distributions to the methane hydrate phase diagram are also presented. In addition, a five year program of MITI for the exploration of natural gas hydrate including a drilling in 1999, which was induced by the results of above mentioned explorations and experiments, are also introduced.

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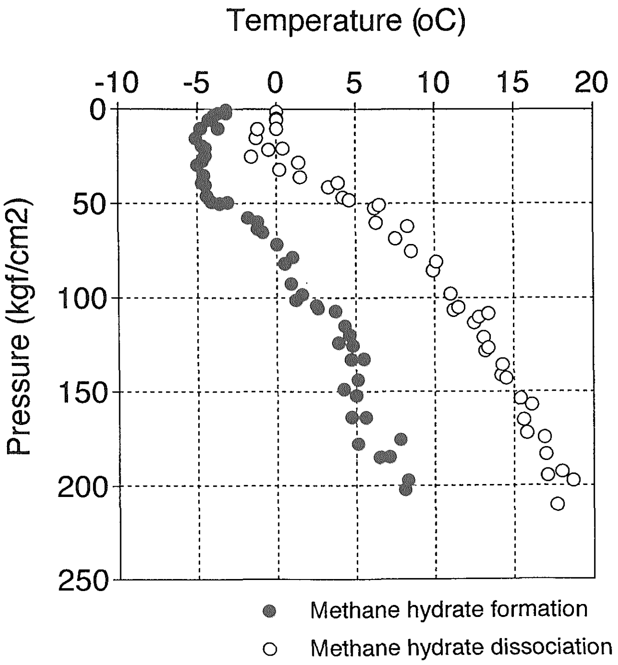
Keywords : Methane hydrate, Bottom simulating reflector, Japan



Data from Itoh (1993)

Fig. 1 Phase diagram of methane and pure water (modified from Itoh, 1993).

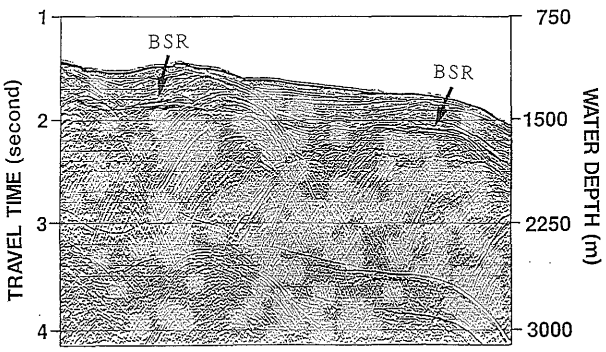


Fig. 2 An example of BSRs in the Tosa basin at the north of the Nankai Trough (Okuda, 1993).

2. Basic Concept of Experiment on Methane Hydrate Phase Diagram

2.1 Equipment and method of methane hydrate synthesis

The first synthesis equipment for gas hydrates in GSJ (Fig. 3) was originally designed by Itoh (1993) which consists of a high-pressure hydrate chamber immersed in an anti-freezing water bath, a gas compressor, a temperature control unit, and optical devices to detect methane hydrate formation and dissociation.

To form methane hydrates, the chamber containing highly pressurized methane supplied from a gas compressor and required amount of water is immersed in a temperature controlled bath filled with cold anti-freezing water for sufficient time. Thermocouples are set just beneath the water level inside the chamber in order to measure the water temperature. The detection device of methane hydrate formation and dissociation consists of a light source from a high intensity halogen lamp, an optical sensor and an optical power meter. Thus, changes of temperature, pressure and intensity of light penetrating through water in the hydrate cell are monitored and recorded during the synthesis experiment.

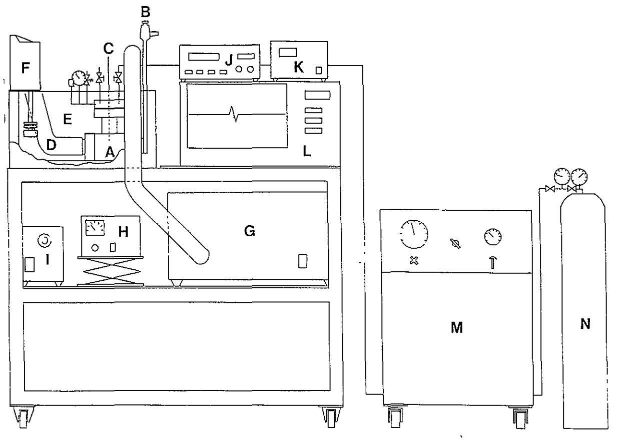


Fig. 3 System configuration of the methane hydrate synthesis equipment (modified from Itoh, 1993).
A : Chamber B : Monitor fiber scope
C : Thermocouple D : Optical sensor
E : Temparture controlled water bath
F : Temparture controller G : Cooler
H : Hagnetic stirrer I : Halogen lamp
J : Optical power metter K : Digital pressure gauge
L : Recorder M : Gas compressor N : Gas cylinder

As this equipment in GSJ has been greatly improved until now, the similar concept is employed principally for the latest equipment (Maekawa *et al.*, 1995).

2.2 Results of methane hydrate synthesis from pure water and pure methane

The formation of methane hydrate was clearly detected from the sharp reduction in intensity of light through water recorded on the charts, whereas the hydrate dissociation was detected from the sharp increase in intensity of light penetrating through methane hydrates (Itoh, 1993; Fig. 4). Phase equilibrium of methane-water can precisely be given by plotting the temperature and pressure at the starting point of change in the intensity of light on the chart.

The results of methane hydrate synthesis from pure water and methane (Fig. 1) show that the lower temperature and higher pressure are necessary for methane hydrate formation compared to methane hydrate dissociation. It suggests that water in the intermediate zone between the boundaries of formation and dissociation can be regarded as meta-stable zone. The water and the methane are converted to methane

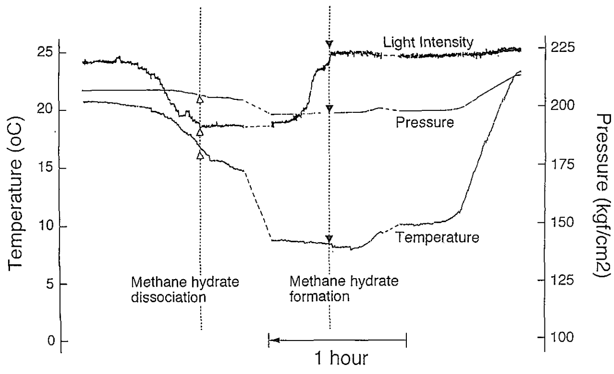


Fig. 4 An example of the recording chart during the experiment of the methane hydrate synthesis (Itoh, 1993).

hydrate crystals at lower temperature and higher pressure relative to the conditions of phase equilibrium curve of methane hydrate formation. The phase equilibrium curve of formation is normally varies depending on the experimental conditions, such as stirring and cooling speed. The formation curve of natural gas hydrates is useful for the exploration, which is presumably close to the dissociation curve due to long time of the hydrate formation in the natural condition.

2.3 Results of methane hydrate synthesis from brine water and pure methane

Since the natural formation water beneath the sea bottom generally contains some amount of sodium chloride, the conditions for methane hydrate dissociation in brine water were also determined by experiments in the laboratory, and the phase equilibrium is plotted in Fig. 5. The results show that the pressure of methane hydrate dissociation is raised up and the temperature is depressed when the concentrations of sodium chloride in water are increased in the solutions (Maekawa *et al.*, 1995).

For example, in 3.5 wt.% sodium chloride water, which is nearly equivalent to the sea water, approximately 1℃. less temperature is necessary for hydrate formation at any given pressure compared to the methane hydrate dissociation for the pure water-pure methane system. Then, more than five degrees in centigrade are necessary for hydrate dissociation in 10 wt.% sodium chloride solution, and more than twelve degree in centigrade in case of 20 wt.%.

The additional pressure between 0.5 and 2 Ma are necessary for hydrate formation at temperature below 15℃ in 5-wt.% Sodium Chloride solution compared to pure water. Similarly additional pressure between 2 and 3 Ma is essential for hydrate formation in 10-wt.% Sodium Chloride solution and more than 7 Ma in case of 20-wt.% Sodium chloride solution.

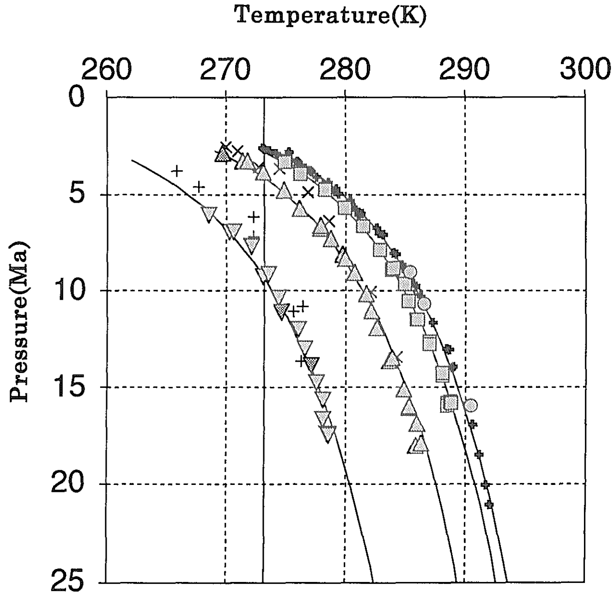


Fig. 5 Phase diagram of methane and brine water (Maekawa et al., 1995).

○ 0 wt% NaCl
□ 3.5 wt% NaCl
△ 10 wt% NaCl
▽ 20 wt% NaCl
—— 0 wt% NaCl (calc.)
—— 3.5 wt% NaCl (calc.)
—— 10 wt% NaCl (calc.)
—— 20 wt% NaCl (calc.)
✚ Methane hydrate (Sloan, 1991)
× 10 wt% NaCl (Kobayashi et al., 1951)
+ 20 wt% NaCl (Kobayashi et al., 1951)

2.4 Implication of phase diagram obtained from the methane hydrate synthesis

The experimental results indicate that the hydrates theoretically occur in nature at the water depth of more than 280 meters where the sea water temperature is 0℃. But, more than 500 meters depth may be necessary for economical development of the hydrate in the offshore area where the normal thermal gradient is measured in the middle latitudes.

If free gas reserve exists just beneath the hydrate layers, exploitation of the free gas causes the pressure reduction in the gas reservoir and consequent hydrate dissociation may result in the production of dissociated gases. But, in the case of absence of free gas beneath the hydrate layers, some kinds of solvent may be necessary to exploit gases from methane hydrates to control the gas dissociation rate. In this situation, the seawater can be used which might be easily condensed as a good solvent for hydrates as indicated by the phase equilibrium curve for the brine water and the methane hydrate.

These facts suggest that injecting condensed sea water into the underground hydrated strata for exploitation of gas from gas hydrates may be a possible method to produce gases from the hydrated layers in

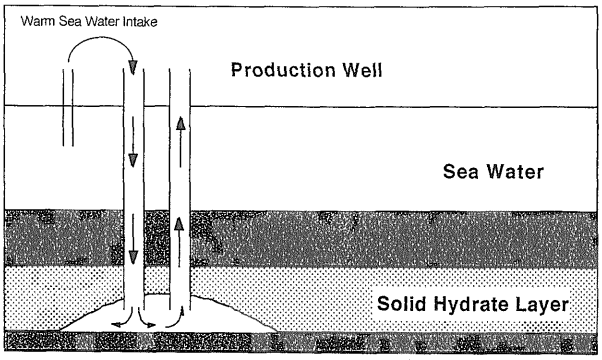


Fig. 6 A model of offshore gas production from a gas hydrate layer.

future. In other words, the seawater is possible to be a fairly good solvent at the bottom of the hydrate stability zone, which corresponds to the phase boundary. Further, as the seawater dissolves methane hydrates slowly, this can be used with control in hydrate exploitation (Fig. 6).

Otherwise, economical chemical solvents should be developed for the hydrate gas production. For this, the Tokyo Gas Co. in co-operation with the GSJ research program has tested various types of chemical solvents for gas hydrates.

3. Distribution of Natural Gas Hydrate around Japan

3.1 Outline of natural gas hydrate data and distribution around Japan

Since 1974, the Geological Survey of Japan has been carrying out marine geological and geophysical surveys around Japan using the R/V Hakurei-maru to produce marine geological maps. Meanwhile, the Ministry of International Trade and Industry has been conducting reconnaissance geophysical investigations for oil and gas exploration in the offshore area mainly shallower than 2000 meters in depth around Japan. These seismic reflection data indicate that BSRs are distributed mainly in the area of inner slope of Nankai Trough and offshore areas around Hokkaido (Fig. 7).

3.2 Geologic setting and BSRs in Nankai Trough area

The Nankai Trough is regarded as a kind of young trench, where the Philippine Sea plate has subducted beneath the Eurasian Plate since Pliocene. In the inner continental slope, several deep sea sedimentary basins filled with Neogene sediments are distributed as forearc basins approximately between 800 and 2000 meter depths, and accretionary prisms are well developed in the deeper slope outside of the forearc basins (Okuda, 1977; Okuda 1984).

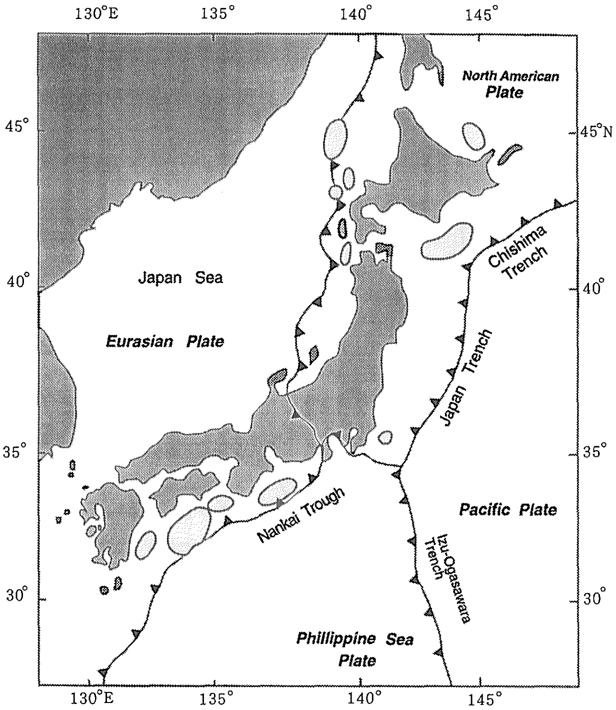


Fig. 7 Distribution of BSRs around the Japanese Islands.

Generally, the BSRs are distributed in the forearc basins north of Nankai Trough. Units of the BSRs often form closures independently, which correspond to several topographic highs and are presumably conformed by geological tectonic structures of lower sedimentary strata and their basements (Fig. 8).

BSRs are regionally distributed in the lower continental slope of the Nankai trough, where accretionary prisms are well developed. They are often faulted by thrusts and disturbed by slumps, so that the distribution patterns of the BSRs are fairly complicated.

Seismic reflectivity in the deep sea sedimentary basins in the Nankai Trough region is generally the strongest in the shallowest part within a BSRs unit, which indicates high probable existence of free gas beneath the methane hydrate layer.

3.3 Geologic settings and BSRs around Hokkaido

Cretaceous to Quaternary sedimentary basins are developed both sides of collision belt oriented almost in NS direction in the central part of the Hokkaido Island. In the western offshore area of Hokkaido, a plate boundary between North American and Japan Sea plates presumably runs, where compressive feature is indicated by several historical big earthquakes in the past. Chishima trench, where the Pacific Plate subducts beneath the North American Plate, exists in the offshore areas of south-east Hokkaido in the offshore area. The Okhotsk Sea, a marginal sea, is stretched at the northwestern extension of Tertiary sedimentary basin in the northwestern Hokkaido.

As the seawater temperature around Hokkaido is relatively lower than other offshore areas around

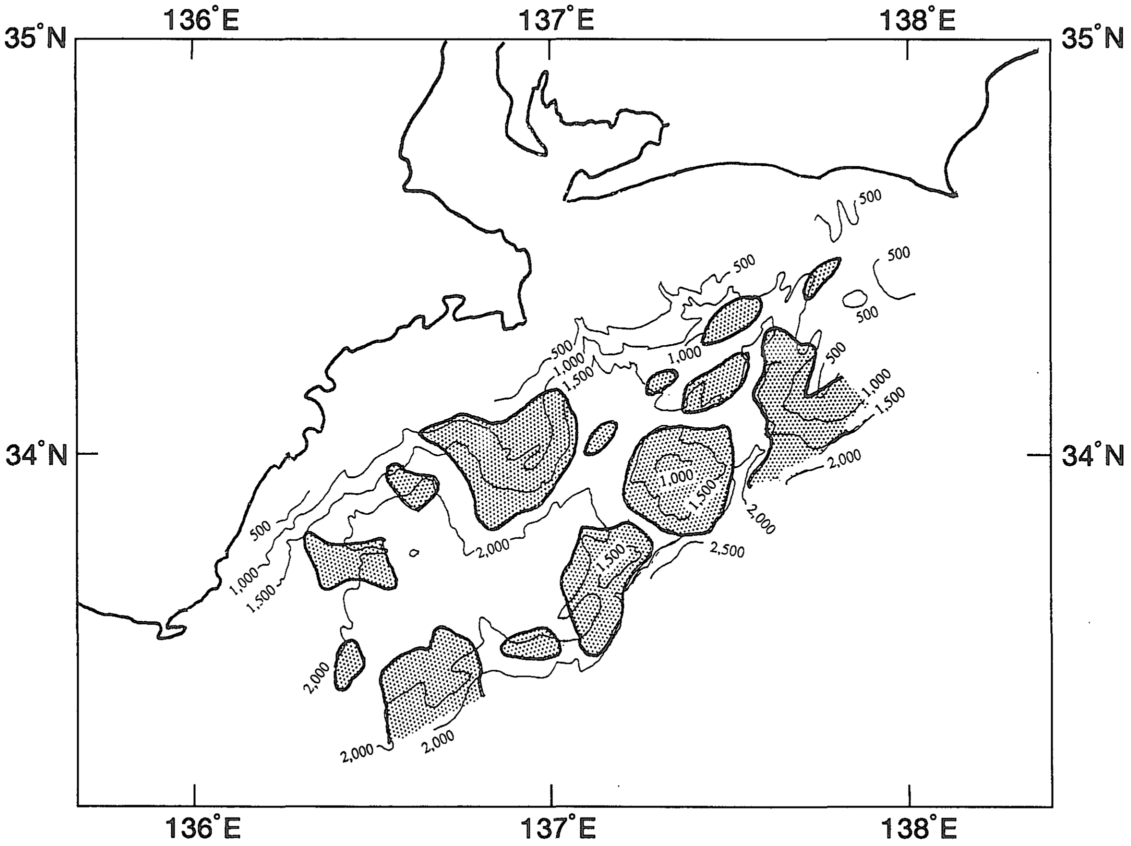


Fig. 8 Distributions of BSRs the north of the eastern Nankai Trough.

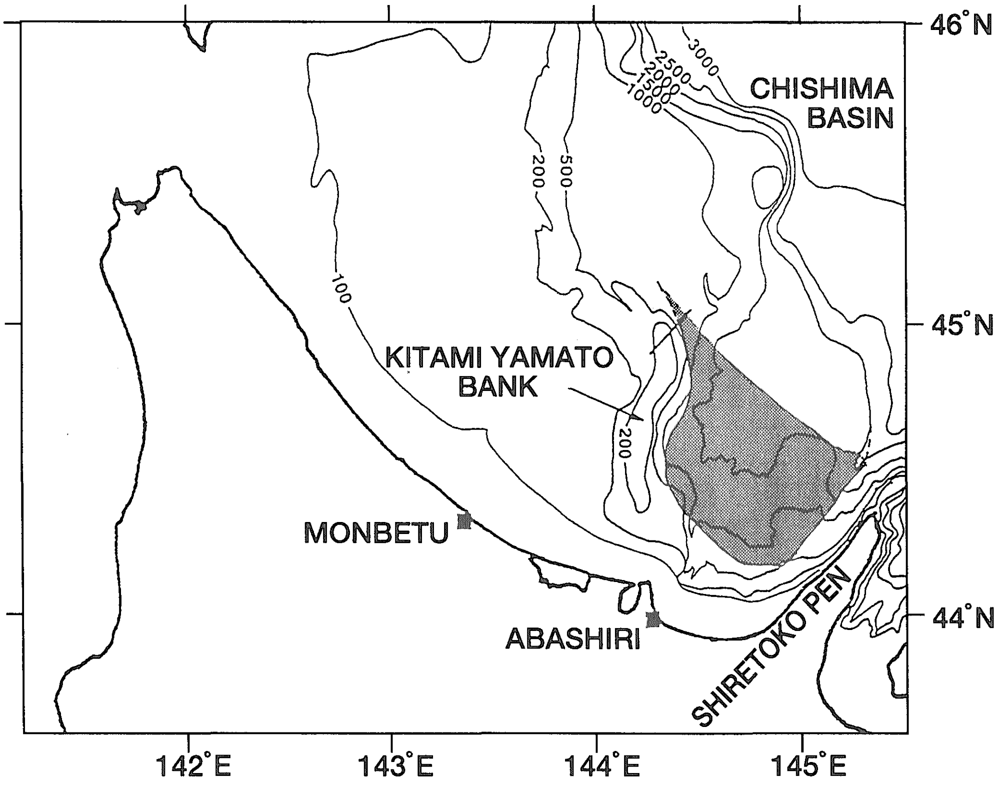


Fig. 9 Distribution of confirmed BSRs north of Abashiri, Hokkaido.

Japan, the BSRs are reasonably recognized at many places around the Hokkaido. Especially, several layers of strong BSRs are regionally distributed in the Okhotsk Sea at the depths of more than 500 meters (Fig. 9).

3.4 BSRs Distribution Patterns

There are two types of BSRs distribution observed around Japan. One is a closure type distribution, which roughly coincides with a topographic high related to the geological structure of lower strata. The other is tentatively named a plain type distribution, which shows homogeneously distribution of the BSRs and coincides with the distribution of sedimentary plains.

The former one is typically recognized in the eastern end of the deep-sea terraces of the Nankai Trough. A cost well was drilled in 1983 near the deep-sea terraces at a water depth of 469 meters though the depth is slightly shallower than that of BSRs distribution. The drilling data in the deeper part show fair hydrocarbon source potential and maturity in the lower Miocene strata, accompanied by gas showings detected by the drill stem test during our old drilling exploration. Therefore, thermogenic gas supply from the deeper part to the hydrate stability zone may be considered in this area.

The latter one is typically recognized in the Okhotsk Sea region, where thick sediments possibly younger than lower Miocene are distributed.

4. Japanese Research Programs on Gas Hydrates

Based on the above case studies in 1993, the Geological Survey of Japan estimated the resources of the methane hydrates in place around Japan as approximately 6 trillion cubic meters (Okuda, 1993), which is about 100 times of the total domestic consumption of natural gas in Japan in 1992.

Following the above estimation, the GSJ has been doing the basic research program on gas hydrates for four years in cooperation with Tokyo Gas Co., Osaka Gas Co. and Japan Petroleum Exploration Co. since 1994. Some of the results are presented in this special issue of the GSJ Bulletin on gas hydrates.

The above estimation made an impact to the Petroleum Council in Japan, and they presented an advisory report to the Minister of the MITI in 1994 related to gas hydrates. The report recommended geophysical and drilling explorations for five years between 1995 and 1999, and a drilling exploration in 1999 for gas hydrates during the 8th phase of a national program was planned. Consequent to the advisory report, the MITI constituted "Methane Hydrate Promotion Committee" in the Japan National Oil Corporation in 1995. For a successful deep-sea drilling in the Nankai Trough in 1999, the committee conducted a comprehensive study on technical issues mainly for drilling and geophysical survey in 1996 as a part of five years project. Therefore, most of the scientists and technicians related to the hydrate exploration in Japan were concentrated on the drilling program. The program partly includes the preliminary studies of the drilling at the Leg 164 of the Ocean Drilling Program (ODP) in 1995 and permafrost drilling programs under international research cooperation with Canada, USA and Russia.

Recently, GSJ and other 7 domestic research organizations started feasibility study for gas hydrate exploration and development in the 21st century to plan a new realistic technical research program after 1999. Some of the interim results are also presented in this special issue.

5. Summary

The potential of natural gas hydrates in the offshore area around Japan was investigated using methane hydrate synthesis experiment to understand the interrelations among factors such as temperature, pressure (depth) and compositions of gas and water. In addition, the BSRs on seismic reflection profiles are correlated to the methane hydrate phase diagram resulted from the experiments.

The seismic reflection data around Japan show that BSRs are regionally distributed mainly in the area on the inner slope of Nankai Trough and offshore areas around Hokkaido.

There are two types of BSRs distribution around Japan. One is the closure type which roughly coincides with topographic high and the geological structure of lower strata. The other is tentatively named as a plain type, which coincides with sedimentary plain and homogeneously distributed.

The pressure rise and temperature lowering of methane hydrate dissociation due to the concentrations of sodium chloride in the solution indicate that the method of injecting seawater as a solvent into underground for exploitation of gases with control from gas hydrates will be the methods in future.

Based on the above findings, the MITI has a plan to carry out a drilling in the Nankai Trough area in 1999.

References

Itoh, S. (1993) Synthetic experiment for methane hydrate. *Abs. 100 th meeting of Jap. Geol. Assoc.* **100**, 176 (in Japanese).

Maekawa, T., Itoh, S., Sakata. S., Igari, S. and Imai N. (1995) Presssure and temperature conditions for methane hydrate dissociation in sodium chloride solutions. *Geochm. Jour.* **29**. 325–329.

Okuda, Y. (1977) Geological Map off outer zone of Southwest Japan. Marine *Geol. Map. Ser.,* **8**, 1: 1,000,000, Geol. Surv. Japan.

Okuda, Y. (1984) Tectonic development of the continental margin off Southwest Japan during the late Cenozoic. *Rep. TRC. Jap. Nat. Oil. Corp.,* **19**, 33-93.

Okuda, Y. (1993) Natural Gas Hydrate as Future Resources. *Jour. Jap. Inst. Ener.* **6**, 425-435 (in Japanese with English Abstract).

Okuda, Y.(1994) Natural Gas Hydrate. *Jour. Jap. Soc. Energy and Resources.* **15**, 1, 40-46 (in Japanese).

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日本におけるガスハイドレート探査の基礎研究

奥田義久

要　旨

近年，海洋ガスハイドレートは，日本周辺海域に分布する非在来型炭化水素資源として有望視されつつある．通産省では，南海トラフ内側大陸斜面（御前崎沖）海域において，ガスハイドレート探査を目的とした国の基礎試錐を1999年に実施することを予定している．これらの動きに対応して，地質調査所では，国内の他の研究機関と共同でガスハイドレートの探査，開発にかかわる地球科学的な基礎的研究，とりわけ基礎的な海洋地質調査と地球化学的な基礎実験を実施してきている．本特集号は現在までに得られたこれらの研究の成果をとりまとめたものである．