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# 科学研究費助成事業　　研究成果報告書

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研究課題名（和文）Do microcrystalline dolomite inclusions provide fossil evidence for the onset of gas hydrate formation?: Isotopic and biogeochemical investigations of methane hydrate in Joetsu Basin, Sea of Japan.

研究課題名（英文）Do microcrystalline dolomite inclusions provide fossil evidence for the onset of gas hydrate formation?: Isotopic and biogeochemical investigations of methane hydrate in Joetsu Basin, Sea of Japan.

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研究成果の概要（和文）：微生物活動は高温の温泉水、深海堆積物など様々な場所で見つかっているが、ガスハイドレート結晶中に高塩分微小空間が存在し、その中で微生物が代謝を繰り返していたとは研究チーム。私たちは新潟県沖上越堆積盆で、長い連続的なガスハイドレートのコアを採取・分解し、分解後に残ったハイドレート融解水中に、幸運にも、中心部分に暗黒有機物質と生命活動の痕跡を包含する微小なマイクロドロマイトが残されているのを発見した。その後の分析と研究により、ハイドレートの微小空隙中では微生物がオイルのような複雑な有機物質を代謝し、その活動の副産物として微小のマイクロドロマイトが形成され続けたことが明らかになった。

研究成果の学術的意義や社会的意義
Meters-thick gas hydrate was recovered from marine sediments and the role of mantle gases in methane migration and gas hydrate growth in the sea of Japan was documented. Microbial life was discovered inside of the gas hydrate, living within spheroidal aggregates of microdolomite.

研究成果の概要（英文）：Microbial life has been found in a variety of extreme environments such as hot springs, glaciers, and in deep marine sediments. Researchers at Meiji's GHRL recovered the first long cores of massive gas hydrate from the seafloor in Joetsu Basin, just southwest of the Japan's port city of Niigata in order to study this unconventional energy resource. While melting the hydrate to study methane gas, a fine microdolomite powder was discovered which consists of microscopic spheroids with dark cores. Further research revealed a unique, microenvironment where microbial life is able to metabolize complex macromolecules to produce extracellular polymeric substance that promote the formation of spheroidal microdolomites. In addition, the noble gases helium and neon were also measured, permitting the discovery that gases from the mantle are involved in the migration of methane in the region which eventually accumulates as gas hydrate in shallow marine sediments.

研究分野：Isotope geochemistry, marine chemistry

キーワード：gas hydrate　Sea of Japan　microdolomite　Bacteriodetes　helium　stable isotopes

１．研究開始当初の背景（Background)

The widespread distribution of gas hydrate along the world's continental margins first captured the interest of geoscientists in the 1990s as a possible future energy source. Initial studies suggested that the amount of organic carbon found in these deposits of solid methane and water ice in marine sediments could potentially supply the world's energy needs for hundreds of years. On the other hand, the dissociation of gas hydrate to water and gas bubbles could be a potential geohazard, provoking underwater landslides along continental slopes in areas where conventional offshore hydrocarbon exploration is being carried out. Climate change far into the Earth's geologic past is also thought to have been influenced by the release of greenhouse gases to the atmosphere as a result of widespread gas hydrate dissociation. All of these factors have led to an increase in scientific interest in gas hydrates in recent years.

The first shallow massive gas hydrate was recovered from the Sea of Japan in 2004, about 100 km to the southwest of the port of Niigata, at a submarine mound site now known as Umitaka Spur. Several participants in this most recent study were present on the cruise, including Ryo Matsumoto the current director of Meiji University's Gas Hydrate Research Laboratory (GHRL), and researchers Glen Snyder and Hitoshi Tomaru. In 2011, the Tohoku earthquake and nuclear disaster led the Japanese government to reassess the country's dependency on nuclear power, and to evaluate the potential of non-conventional energy sources, including gas hydrate. Research to date has focused on a number of sites along Japan's continental margins, yet Umitaka Spur still remains a prime location for the study of gas chimneys, methane seeps, and massive gas hydrate. The site also provides an important natural laboratory in which to investigate the unusual underwater ecology associated with natural submarine gas seeps.

２．研究の目的（Purpose)

In the course exploratory drilling of gas chimneys in Joetsu Basin during the 2014 and 2015 research seasons, a number of unusual features were found, including areas with massive hydrate several meters thick, and the presence of mineral inclusions within the hydrate itself. As these were both novel features, the initial purpose of our research was to characterize the gas chemistry in relation to the history of gas hydrate accumulations, and to determine the composition of the mineral inclusions and their significance. The mineral inclusions turned out to be almost pure spheroidal microdolomite with dark inner cores. As the size of the microdolomites was sufficient to potentially host microbial life within them, our focus shifted to the internal content of the microdolomites as well.

３．研究の方法（Method)

The work leading to the discovery of gas hydrate microdolomites and microbial communities represents an interdisciplinary effort by researchers from a variety of scientific specialties. Shipboard collection of microdolomite-bearing gas hydrate was carried out by the Meiji University Gas Hydrate Research Lab (GHRL) team including Ryo Matsumoto, Glen Snyder and Yoshihiro Kakizaki, as well as by Hitoshi Tomaru of Chiba University and Takumi Imajo of the Tokyo University of Marine Science and Technology. The research was carried out in conjunction with Japan's Ministry of Economy, Trade and Industry (METI) and the National Institute of Advanced Industrial Science and Technology (AIST)， Once on land, the samples were analyzed by X-ray diffraction, scanning electron microscopy (SEM) and stable isotopic mass spectrometry by Glen Snyder, Naizhong Zhang, and Yoshihiro Kakizaki at the GHRL.

Oils released by melting the gas hydrate were analyzed by Stephen Bowden of the University of Aberdeen in Scotland. Epifluorescence of microbial DNA and phylogenetic identification was carried out by Yohey Suzuki and Mariko Kouduka at the University of Tokyo Hongo-Sanchome Campus. Internal composition and elemental mapping of the microdolomite grains, as well as additional noble gas and stable isotope analyses of hydrate gases were carried out at the University of Tokyo Atmosphere and Ocean Research Institute (AORI) by Kentaro Tanaka, Naoto Takahata, and Yuji Sano.

４．研究成果（Results)

The research carried out by this project is groundbreaking in a number of regards. It is the first study using noble gas geochemistry to show that fluids from the Earth's mantle can, in the case of the western margin of the Japan Sea, be involved in the migration of deep hydrocarbons that result in the accumulation of shallow gas hydrate. It is also the first study to show that saline pockets found within massive gas hydrate can host microbial life which is encapsulated within spheroidal aggregates of microdolomite. General findings can be summarized as follows:

- Fluid overpressures generated by hot mantle circulation has resulted in both the production of thermogenic gas in Miocene sediments at approximately 5 km sediment depth and in subsequent rupturing and fluid migration to the seafloor.
- Gas seepage in methane chimney mounds in the Sea of Japan has led to ongoing growth of solid gas hydrate, consisting primarily of ice-like accumulations of methane and water. The rapid growth of massive gas hydrate in shallow sediments has led to the entrapment of oil and saline inclusions within the hydrate.
- Although previous studies have shown that a consortia of microbial life grows on the outer surfaces of gas hydrate where it oxidizes methane, the microbial life hosted within the hydrate primarily consists of the phylum *Bacteriodetes,* which metabolizes complex molecules such as oils.
- The growth of these microorganisms in saline pockets within the hydrate has resulted in the production of extracellular polymeric substances which provide a substrate for the precipitation of spheroidal microdolomite, effectively encasing the microorganisms as well as a small amount of the surrounding fluids.
- During long-term burial of the gas hydrate, oils which were also expelled in the gas chimneys and incorporated in the gas hydrate become increasingly biodegraded.
- While the presence of this newly discovered microenvironment is unusual, the widespread occurrence of gas hydrate along continental margins and in permafrost suggest possibly a much wider distribution. Given that methane has been shown to exist on other cold planets, microbial microdolomites could be an analogue for extraterrestrial life as well.

５．主な発表論文等

〔雑誌論文〕　計5件（うち査読付論文　5件／うち国際共著　4件／うちオープンアクセス　4件）

| | |
|---|---|
| １．著者名<br>Snyder, G. T. ,　Sano, Y.,　Takahata, N., Matsumoto, R., Kakizaki, Y. and Tomaru, H., | ４．巻<br>535 |
| ２．論文標題<br>Magmatic fluids play a role in the development of active gas chimneys and massive gas hydrates in the Japan Sea. | ５．発行年<br>2020年 |
| ３．雑誌名<br>Chemical Geology | ６．最初と最後の頁<br>119462 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>https://doi.org/10.1016/j.chemgeo.2020.119462 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

| | |
|---|---|
| １．著者名<br>Snyder, G.T., Matsumoto, R., Suzuki, Y., Kouduka, M., Kakizaki, Y., Zhang, N., Tomaru,H., Sano, Y.,, Takahata, N., Tanaka, K., Bowden, S.A., Imajo, T. | ４．巻<br>10 |
| ２．論文標題<br>Evidence in the Japan Sea of microdolomite mineralization within gas hydrate microbiomes | ５．発行年<br>2020年 |
| ３．雑誌名<br>Scientific Reports | ６．最初と最後の頁<br>1-13 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>https://doi.org/10.1038/s41598-020-58723-y | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

| | |
|---|---|
| １．著者名<br>Zhang, N., Lin, M., Snyder, G.T., Kakizaki, Y., Yamada, K., Yoshida, N., Matsumoto, R. | ４．巻<br>512 |
| ２．論文標題<br>Clumped isotope signatures of methane-derived authigenic carbonate presenting equilibrium values of their formation temperatures | ５．発行年<br>2019年 |
| ３．雑誌名<br>Earth and Planetary Science Letters | ６．最初と最後の頁<br>207-213 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>https://doi.org/10.1016/j.epsl.2019.02.005 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスではない、又はオープンアクセスが困難 | 国際共著<br>該当する |

| | |
|---|---|
| １．著者名<br>Tanikawa, W., Ohtomo, Y., Snyder, G., Morono, Y., Kubo, Y., Iijima, Y., Noguchi, T., Hinrichs, K.-U., and Inagaki, F. | ４．巻<br>337 |
| ２．論文標題<br>Publication dateData report: water activity of the deep coal-bearing basin off Shimokita from IODP Expedition 337. | ５．発行年<br>2018年 |
| ３．雑誌名<br>Proceedings of the Integrated Ocean Drilling Program, 337: Tokyo (Integrated Ocean Drilling Program Management International, Inc.). | ６．最初と最後の頁<br>1-12 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>doi:10.2204/iodp.proc.337.204.2018 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

| | |
|---|---|
| 1．著者名<br>Kakizaki Y, Snyder G, Tanahashi Y, Ishida N, Matsumoto R | 4．巻<br>124 |
| 2．論文標題<br>Concentration, carbon-isotopic change of dissolved gas from Murono mud volcano in Tokamachi City, Niigata Prefecture, central Japan), just before, after the 2014 Kamishiro Fault Earthquake | 5．発行年<br>2018年 |
| 3．雑誌名<br>Journal of the Geological Society of Japan | 6．最初と最後の頁<br>127-140 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>http://dx.doi.org/10.5575/geosoc.2017.0065 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>－ |

〔学会発表〕　計11件（うち招待講演　1件／うち国際学会　9件）

| |
|---|
| 1．発表者名<br>G.Snyder, R.Matsumoto, Y. Kakizaki, Y. Suzuki, N.Zhang, Y.Sano, N. Takahata, K. Tanaka, T. Imajo, S. Bowden, H. Tomaru |
| 2．発表標題<br>Microdolmite grains in Japan Sea Gas Hydrate: What do they tell us about present and past hydrate growth? |
| 3．学会等名<br>Pacific Conference of Marine Science and Technology (PACON), Vladivostok, Russia（国際学会） |
| 4．発表年<br>2019年 |

| |
|---|
| 1．発表者名<br>G.Snyder, R.Matsumoto, Y. Kakizaki, Y. Suzuki, N.Zhang, Y.Sano, N. Takahata, K. Tanaka, T. Imajo, S. Bowden, H. Tomaru |
| 2．発表標題<br>Microdolomites are not MDACs: microbially-derived carbon source for microcrystalline dolomite found within massive gas hydrates and implications for the shallow marine carbon cycle |
| 3．学会等名<br>Japan Geoscience Union Meeting（招待講演）（国際学会） |
| 4．発表年<br>2019年 |

| |
|---|
| 1．発表者名<br>Glen Tritch Snyder, Yohey Suzuki,Takumi Imajo, Ryo Matsumoto, Yoshihiro Kakizaki, Hitoshi Tomaru, Aya Iguchi, Yuji Sano, Naoto Takahata, Naizhong Zhang |
| 2．発表標題<br>Microbial microdolomite aggregates found within massive gas hydrate: Implications for the Earth's deep biosphere and for extraterrestrial life? |
| 3．学会等名<br>American Geophysical Union Fall Meeting（国際学会） |
| 4．発表年<br>2018年 |

１．発表者名
Naizhong Zhang, Glen Tritch Snyder, Keita Yamada, Yoshihiro Kakizaki, Akihiro Kano, Naohiro Yoshida, Ryo Matsumoto

２．発表標題
Clumped isotopic signatures of methane-dervied authigenic carbonates and gas hydrate-derived microcrystalline dolomite in the Sea of Japan

３．学会等名
American Geophysical Union Fall Meeting（国際学会）

４．発表年
2018年

１．発表者名
Glen Snyder, Yoshihiro Kakizaki, Ryo Matsumoto, Naizhong Zhang, Yohey Suzuki, Yuji Sano, Naoto Takahata, Kentaro Tanaka, Takumi Imajo, Hitoshi Tomaru, Aya Iguchi

２．発表標題
Stable isotopic composition of microdolomitic aggregates associated with saline inclusions within Japan Sea massive gas hydrate G

３．学会等名
International Sedimentological Congress

４．発表年
2018年

１．発表者名
Glen Snyder, Yohey Suzuki,Takumi Imajo, Ryo Matsumoto, Yoshihiro Kakizaki, Hitoshi Tomaru, Aya Iguchi, Yuji Sano, Naoto Takahata, Naizhong Zhang

２．発表標題
Microdolomitic evidence for deep biosphere contained inside of the saline inclusions within Japan Sea massive gas hydrate

３．学会等名
Japan Geoscience Union (JPGU) Annual Meeting

４．発表年
2018年

１．発表者名
Glen Snyder, Ryo Matsumoto, Yoshihiro Kakizaki, Hitoshi Tomaru, Yohey Suzuki

２．発表標題
Compositional and textural trends of microcrystalline dolomite found within massive gas hydrate in Joetsu Basin

３．学会等名
JpGU-AGU Joint Annual Meeting（国際学会）

４．発表年
2017年

１．発表者名
G. Snyder, Y. Kakizaki, R. Matsumoto, Y. Suzuki, Y. Sano, N. Takahata, K. Tanaka, T. Imajo

２．発表標題
Mineralization of Dolomite Associated with Gas Hydrate Formation in Sea of Japan Sediments.

３．学会等名
International Conference on Gas Hydrates (ICGH9) (国際学会)

４．発表年
2017年

１．発表者名
G. Snyder, Y. Kakizaki, R. Matsumoto, Y. Suzuki, Y. Sano, N. Takahata, K. Tanaka, T. Imajo

２．発表標題
Noble Gas and Stable Isotopic Composition of Methane Hydrates in Umitaka Spur: Three End-Member Mixing and Elemental Fractionation

３．学会等名
International Conference on Gas Hydrates (ICGH9) Denver, Colorado (国際学会)

４．発表年
2017年

１．発表者名
G. Snyder, Y. Kakizaki, R. Matsumoto, Y. Suzuki, Y. Sano, N. Takahata, K. Tanaka, T. Imajo

２．発表標題
Stable isotopes and noble gases in marine methane hydrate and associated dolomite: Investigations in the Sea of Japan.

３．学会等名
International Conference on Gas Hydrate in Continental Margins, Taipei, Taiwan (国際学会)

４．発表年
2017年

１．発表者名
G. Snyder, Y. Kakizaki, R. Matsumoto, Y. Suzuki, Y. Sano, N. Takahata, K. Tanaka, H. Tomaru, T. Imajo, A. Iguchi

２．発表標題
Microcrystalline dolomite within massive Japan Sea methane hydrate: origin and development ascertained by inclusions within inclusions.

３．学会等名
American Geophysical Union (AGU) Fall Meeting, New Orleans, U.S.A. (国際学会)

４．発表年
2017年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

Extreme Microbes Found in Sea of Japan crystals
https://www.livescience.com/64532-microbes-inside-gas-hydrate-crystals.html
ガスハイドレート研究所
http://www.kisc.meiji.ac.jp/~hydrate/index.html
'Flammable ice' discovery offers up clue to life on other planets
https://www.abdn.ac.uk/news/13715/

6．研究組織

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