バイカル湖南湖盆Kedr泥火山の天然ガスハイドレートのエタン濃縮過程 Processes of ethane enrichment in hydrate-bound gas at the Kedr mud volcano, Southern Baikal Basin

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In this report we propose an ethane-enrichment process in double structure gas hydrates discovered at Lake Baikal. The structure II gas hydrates, those hydrate-bound gas contains 14-15% of ethane, have been retrieved from eight places (Kukuy K-2, K-3, K-4, K-10, K-P, PosolBank, Kedr, and Kedr2), widely distributed in central and southern Baikal Basins. In the framework of Multi-phase Gas Hydrate Project, our international collaboration between Japan, Russia, and Belgium, has revealed distribution of near-surface gas hydrates at the Kedr and Kedr2 mud volcano (southern Baikal basin) from 2015 to 2017. We focus on relation between crystallographic structure of gas hydrates and their molecular and isotopic compositions of hydrate-bound gas.

Kedr and Kedr2 mud volcanoes locate 25km south of Listvyanka, and the distance between them is about 1.5km. They consist of many small mounds and pockmarks related to near-surface gas hydrates. We retrieved 28 sediment cores (including 14 hydrate-bound cores) and 16 cores (including 2 hydrate cores) from Kedr and Kedr2, respectively. Gas hydrate crystals were collected onboard and stored in liquid nitrogen. Samples of hydrate-bound and sediment gases were also obtained and stored in 5mL vials. We obtained the powder X-ray diffraction (PXRD) patterns of the crystals and measured molecular and stable isotope compositions of the gas samples.

PXRD results showed double-structure gas hydrates, composed of the cubic structure I and II, while several sediment cores showed only the structure I or II. We found that  $\delta D$  of ethane in the structure II was around 13‰ smaller than that in the structure I. Methane  $\delta D$  of hydrate-bound gas was around 5‰ smaller than that of sediment gas, suggesting that the gas hydrate is in an equilibrium condition. However, only three cores retrieved from the central pockmark of Kedr mud volcano showed no difference in methane  $\delta D$  between them. These results suggest the idea that gas supply at the central pockmark of Kedr stops and gas hydrate dissociates now. The structure I gas hydrate (composition of ethane: 2-3%) dissociates and forms the structure II, that enriches light ethane according to hydrogen isotope fractionation.

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