U-Th radioactive disequilibrium analyses of carbonate nodules from methane seeps at Naoetsu-oki, the Sea of Japan

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Methane seep sites are often associated with authigenic carbonate such as crusts, chimney and concretion. These carbonate precipitation is triggered by the increase in alkalinity during anaerobic methane oxidation via sulfur reduction. The reaction that precipitates carbonate is controlled by upward flux of methane. Therefore, authigenic carbonate is thought to be a useful chronometer that records when methane seep is active. This study aims at testing the feasibility if a carbonate can record the activity of methane seep, using the samples of carbonate nodules from Naoetsu-oki, the Sea of Japan.

Naoetsu-oki is the boundary between Eurasia Plate and North American Plate. In the summer of 2004, R&T/V Umitaka-maru of Tokyo University of Marine Science and Technology investigated small ridge at Naoetsu-oki sedimentary basin in the eastern margin, the Sea of Japan. At the ridge, methane seeps were detected high concentration of methane in seawater (Ishida et al., 2005) and by a high-resolution echo-sounder survey (Aoyama et al., 2005). Piston core recovered gas hydrate, which is dominated by methane with minor ethane, whereas the delta13C of gas hydrate methane is -40 to -42 permil PDB (Matsumoto et al., 2005; Ishida et al., 2005). According to the deep-exploratory drilling near this study area, deep-seated gases are dominated by thermogenic methane with -40 permil PDB of delta13C. Therefore, methane seep and ocean floor gas hydrate are likely to be connected with deep-thermogenic gas reservoirs at Naoetsu-oki.

Samples for U-Th dating were recovered by piston cores and grabs during the cruise of 2004 summer. Carbonate nodules are grey to blown color, are composed of calcite or aragonite. Carbonate samples also contains detritus such as silicate and organics. Since pure carbonate could not be separated from detritus physically, carbonate nodules were analyzed by total sample digestion.

Carbonate nodules from Naoetsu-oki have U concentrations of 2.9 to 11.5 ppm, and Th concentrations of 1.1 to 2.8 ppm. Therefore, the amount of initial 230Th cannot be neglected to acquire accurate U-Th ages.

Isochron method was tested by triple measurements of a carbonate nodule. As the result, an isochron was drawn. Points of analyzed sediment are on the isochron, showing that carbonate nodules contain the sediment as impurity. Accordingly, to correct for initial 230Th, this study assumes that the carbonate nodules contain Th derived from impurity with the same (230Th/232Th) activity ratio of the local sediment, 1.07. 230Th derived from impurity was subtracted using 232Th in the samples and (230Th/232Th) activity ratio of the local sediment. As the result, U-Th correction ages showed 12 to 35 ka, and many of them concentrate about 20 ka.

We also compared U-Th corrected ages with 14C ages, which were older than U-Th ages. At methane seep sites, it is undeniable that carbonates reflect dead carbon, which makes 14C ages older, derived from methane. Therefore, we can conclude that U-Th disequilibrium dating is a more reliable geo-chronometer than 14C dating for evaluating activities of methane seeps.

During time interval of U-Th ages, from 12 ka to 35 ka, environmental condition must have been favorable for enhanced methane fluxes through sediment. Furthermore, extensive methane flow at 20 ka consistent with lowest-stand sealevel during the last glacial. Therefore, we speculate that the decrease of hydraulic pressure due to low-stand sealevel triggered the decomposition of gas hydrate and/or ascent of gas. This study also suggests that U-Th ages of carbonate could be used as a reliable chronometer with regard to the activation of methane seep.