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Investigation of formation efficiency of volatile organic carbons by gamma-rays in methane hydrate

Takuya Higuchi^{1*}, Atsushi Tani¹

¹Sci., Osaka Univ.

Clathrate hydrate is a crystal compound of water molecules encaging guest molecules. Natural gas hydrate, whose guest molecule is mainly methane, is found under deep sea and in permafrost region. Estimation of formation age in natural gas hydrate has been attempted using the ¹²⁹I method (e.g. Fehn et al., 2003), which is an indirect age determination method. In contrast, we have investigated to establish a direct age determination method from hydrate crystal itself. Since natural gas hydrate is formed in sediment and irradiated by natural radiation due to natural radioisotopes like ⁴⁰K, U-series and Th-series, chemical reaction via radicals may occur in natural samples. In methane hydrate, methyl radicals are induced by gamma-rays (Takeya et al., 2004). However, they are unstable at the temperature and pressure of natural gas hydrate occurrence, and dimerize to ethane (Ishikawa et al., 2007). In another reaction, methanol is also formed after gamma-ray irradiation to methane hydrate (Tani et al., 2008). If the amount of radiation-formed compounds in hydrate increases after hydrate formation, it may suggest a formation age of natural gas hydrate. The formation efficiency of radiation-induced compounds is necessary for estimation of the formation age. Therefore, we have investigated the dissociated water of gamma-irradiated methane hydrate and analyzed quantitatively radiation-formed compounds in methane hydrate by gas chromatography-mass spectrometry (GC-MS).

Methane hydrate was synthesized and irradiated by gamma-rays of about 6 kGy at 273 K under high pressure to avoid dissociation of the hydrate. Headspace gas of the dissociated water was analyzed by GC-MS. Methanol and formaldehyde were formed and the other small peaks were observed. The amounts of methanol and formaldehyde by gamma-rays in methane hydrate were almost the same. We will discuss the formation efficiencies of those compounds in the presentation.

Keywords: clathrate hydrate, methane, gamma rays, GC-MS, radical, volatile organic compounds