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Radiation-formed radicals and their reaction products in carbon dioxide hydrate

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Carbon dioxide hydrate (CO_2 hydrate) is a clathrate compound by water cages including a guest molecule, CO_2 and may form naturally on the Earth (ex. Konno et al., 2006) and Mars (Pellenbarg et al., 2003). Natural CO_2 hydrate should be irradiated by natural radiation from radioisotopes in sediments and/or cosmic rays. What reaction is induced by such radiation in CO_2 hydrate? We have investigated radiation-formed radicals and their reaction products in CO_2 hydrate.

After gamma-ray irradiation to synthetic CO₂ hydrate at 77 K, HCO₂ radicals are observed by electron spin resonance (ESR). ESR signal intensity of the HCO₂ radicals decreased around 150 K, although CO₂ hydrate is stable even at 200 K under 0.1 MPa (Falabella, 1975). Ion chromatography analysis showed that formic acid (HCOOH) was formed in the gamma-irradiated sample. Hydrogen atom detached from water molecule may be captured by CO₂ and change to HCO₂ radical. One more hydrogen atom may be captured by the radical and form formic acid. Therefore, natural radiation may induce hydrogen addition reaction on guest molecule in CO₂ hydrate less than 200 K without CO₂ hydrate dissociation.