

Isotope systematics of CO₂-H₂-H₂O-CH₄ in hydrogenotrophic methanogenesis examined by laboratory incubation

S. KAWAGUCCI¹, T. OKUMURA¹, Y. SAITO¹, Y. MATSUI¹, K. TAKAI¹ AND H. IMACHI¹

¹ JAMSTEC, 2-15 Natsushima-cho, Yokosuka, Kanagawa 237-0061, Japan (kawagucci@jamstec.go.jp)

The stable isotope ratios of carbon (¹³C/¹²C) and hydrogen (D/H) in methane have been used as geochemical tracers to deduce its origin. The stable isotope diagnosis is justified by the current understanding of stable isotope systematics with respect to each of the methanogenic processes and factors. It has been well confirmed that the magnitude of the carbon isotope fractionation depends on the substrate availability of the methanogenic environment: a greater ¹³C-depletion in the produced CH₄ occurs under a lower H₂ concentration [1]. In contrast to the carbon isotopic characteristics, the hydrogen isotope characteristics of methanogenesis remain less understood in terms of the factors affecting fractionation such as the H₂ availability. While possible relationship between the CH₄-H₂O hydrogen isotope fractionation and the H₂ availability has been reported, as well as the case in carbon, we recently demonstrated that the hydrogen isotope ratio of substrate H₂ also affects the hydrogen isotope ratio of the produced CH₄ [2], called “δD_{H2} effect.”

In this study, we examined patterns of both carbon and hydrogen isotope fractionations among CO₂-H₂-H₂O-CH₄ with respect to H₂ availability for hydrogenotrophic methanogens by laboratory incubations of pure culture (high p_{H2} condition) and coculture with heterotrophic H₂-producing bacteria (low p_{H2} condition) [3]. The CH₄-H₂O isotope fractionation at high p_{H2} varied due to the δD_{H2} effect while that at low p_{H2} ranged narrow and agreed with that examined previously. Noteworthy the CH₄-H₂O isotope fractionation at low p_{H2} exhibited in laboratory cultures is lower than that found in the corresponding geological samples. The observations including δD_{H2} effect can be interpreted consistently by kinetics of methanogenic pathway and molecules transport across the cell membrane.

References

- [1] Valentine, D. et al. (2004) *Geochim. Cosmochim. Acta* **68**, 1571-1590.
- [2] Kawagucci, S. et al. (2014) *Geochim. Cosmochim. Acta* **142**, 601-614
- [3] Okumura, T. et al. (under review)