コマチアイトと二酸化炭素に富んだ海水との反応による水素発生の実験的研究 Experimental study on H_2 generation through reactions between komatiite and CO_2 -rich seawater

*上田 修裕^{1,2}、渋谷 岳造²、澤木 佑介¹、斎藤 誠史²、高井 研²、丸山 茂徳³ *Hisahiro Ueda^{1,2}, Takazo Shibuya², Yusuke Sawaki¹, Masafumi Saitoh², Ken Takai², Shigenori Maruyama³

1.東京工業大学大学院理工学研究科地球惑星科学専攻、2.海洋開発研究機構、3.地球生命研究所
1.Tokyo institute of technology, 2.JAMSTEC, 3.ELSI

To understand the chemical nature of hydrothermal fluids in the komatiite-hosted seafloor hydrothermal system in the Hadean, we conducted two hydrothermal serpentinization experiments involving synthetic komatiite and a CO_2 -rich acidic NaCl fluid (pH = 4.9 at 25 °C) at 250 °C and 350 °C, 500 bars. During the experiments, the total carbonic acid concentration (ΣCO_2) in fluids at 250 °C and 350 °C decreased from approximately values from 400 to near 30 and 170 mmol/kg, respectively, which is consistent with the greater amount of alteration carbonate mineral at 250 °C than at 350 °C in the serpentinized/carbonated komatiites (Shibuya et al., 2013). Furthermore, the precipitated carbonate species strongly influenced Mg concentration in the hydrothermal fluid: Mg concentration at 250 °C (carbonate as Fe-bearing dolomite) was 36-40 mmol/kg, which was 30-40 times higher than that at 350 °C (carbonate as calcite). Therefore, in contrast to modern seafloor hydrothermal systems, the reactions between komatiite and CO_2 -rich seawater at temperatures where dolomite was stable could have been the source of Mg for the Hadean ocean (e.g., Alt, 1995). More importantly, the carbonation of komatiites potentially suppressed H_2 generation in the fluids. The Fe content in dolomite at 250 °C (3-8 wt%) was clearly higher than that of calcite at 350 °C (< 0.8 wt%), while the steady-state H₂ concentration in the fluid was approximately 0.024 and 2.9 mmol/kg at 250 °C and 350 °C, respectively. This correlation between the Fe content in carbonate mineral and the H₂ concentration in the fluid suggests that the incorporation of ferrous iron into the carbonate mineral probably limited the magnetite formation and consequent generation of hydrogen in the fluid during the serpentinization of komatiites. In comparison with modern H_2 -rich seafloor hydrothermal systems, the H₂ concentration of the fluid in the experiment at 350 °C corresponds to that of Kairei hydrothermal field (Central Indian Ridge) (Takai et al., 2004; Gallant and Von Damm, 2006; Kumagai et al., 2008; Nakamura et al., 2009), where hydrogenotrophic methanogens dominate in the prosperous microbial ecosystem. Accordingly, the high-temperature serpentinization of komatiite would provide the H₂-rich hydrothermal environments that were necessary for the emergence and early evolution of life in the Hadean ocean. In contrast, considering that carbonate minerals become more stable with decreasing temperature in the komatiite-H₂O-CO₂ system, H₂-rich fluids may not have been generated by serpentinization at temperatures below 250 °C, even in the komatiite-hosted hydrothermal systems of the Hadean Earth.

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