

## H<sub>2</sub>-driven pyrite/pyrrhotite redox cycle: a sustained electron generator for the autotrophic origin of life

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Electrochemical survey of Okinawa trough hydrothermal fields and the subsequent laboratory simulations have suggested that the electricity generation in deep-sea hydrothermal vent environments has served as a crucial source of autotrophic energy through the origin and evolution of life. However, although thermodynamic calculation has predicted that H<sub>2</sub> oxidation in alkaline hydrothermal fluids ( $\text{H}_2 \Rightarrow 2\text{H}^+ + 2\text{e}^-$ ) is a powerful process for generating strongly negative electric potentials, the factors controlling the potentials and current available in realistic geochemical systems remain largely unknown: there should be a gap between the thermodynamic potentials and the potentials provided to the mineral–seawater interface. Here I show preliminary results of potential measurement and linear sweep voltammetry of iron sulfide under HS<sup>−</sup> and H<sub>2</sub>-containing alkaline hydrothermal fluids. It was found that, despite of the presence of H<sub>2</sub>, iron sulfide controls the electric potential through the mackinawite-pyrite equilibrium ( $\text{FeS} + \text{HS}^- \Rightarrow \text{FeS}_2 + \text{H}^+ + 2\text{e}^-$ ). H<sub>2</sub> chemically reduces the resultant pyrite, realizing a sustained electricity generation. With increasing temperature, the monitored electric potential approached to the thermodynamic potential for the mackinawite/pyrite equilibrium, reaching less than −650 mV versus the standard hydrogen electrode (at 25°C) under naturally realistic alkaline hydrothermal conditions. Additionally, higher temperatures led to greater current generation under oxidative potentials. These results indicate that in deep-sea systems, iron sulfide flexibly changes its oxidation state in response to the local electric potentials, serving as a natural battery between the reductive hydrothermal fluids and oxidative seawater.

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