Abiotic methane formation not from H_2 but from H_2O in the serpentinite-hosted Hakuba Happo hot spring

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Serpentinite-hosted hydrothermal system is considered to be important for prebiotic synthesis as well as habitat for the earliest life. Fluids derived from serpentinites are characterized by high concentrations of H_2 and CH_4 [e.g. 1]. It is generally assumed that the methane and hydrocarbons are produced abiotically from the H_2 and CO_2 via Fischer-Tropsch Type (FTT) synthesis [e.g. 1,2]. However, the production mechanism of the methane is not adequately understood yet. We report systematic isotopic study of a new serpentinite-hosted hydrothermal system: Hakuba Happo hot spring in the Shiroumadake area, Japan. The hot spring water was directly collected from 500-1000 m deep two drilling wells that show high pH over 10 and rich in H_2 and CH_4. The δD values of H_2 and H_2O from both wells are almost the same, whereas the δD-CH_4 values are different by approximately 80‰. The CH_4-H_2-H_2O hydrogen isotope systematics indicates at least two different mechanisms are required for the methane formation. The higher δD-CH_4 with respect to equilibrium relation is similar to other serpentinite-hosted system reported earlier and demonstrates that the source of hydrogen of CH_4 cannot be H_2 but directly derived from H_2O. This implies that the CH_4 is not produced via the FTT synthesis, but instead, likely produced by hydration reaction of olivine. On the other hand, lower δD-CH_4 with respect to equilibrium relation suggests incorporation of biological methane. Based on the comparison of δD systematics between our result and other serpentinite-hosted hydrothermal system, we suggest that dominant methane formation mechanism in a general serpentinite-hosted system is not FTT reaction. Hydration of olivine may play more significant role for abiotic methane production than previously thought.

[1] Holm & Charlou (2001) *EPSL* **191**, 1-8. [2] Proskurowski *et al*. (2008) *Science* **319**(5863), 604-607.

Organic and inorganic contaminant remediation by biogenic nanopalladium

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Microbial synthesis of nanoparticles is an innovative process for precious metal recovery and preparation of nanocatalysts for transformation of environmental contaminants. Here, we report preparation of microbiologically synthesized palladium (Pd) nanoparticles (Bio-Pd) using *Clostridium* sp. BC1 [1] and microbial granules (MG) under fermentative growth conditions [Fig. 1]. Microbial granules were cultivated in bubble column sequencing batch reactors [2]. Reduction of dissolved Pd (II) to insoluble, black colored Pd (0) by BC1 or MG was instantaneous and complete. Removal of Pd (II) via sorption by BC1 or MG was negligible. Reduction of Pd (II) was mediated by the microbially generated hydrogen and the Pd (0) nanoparticles were predominantly associated with the bacterial cells or MG. The Bio-Pd catalysed the reduction of Cr (VI) to Cr (III) and the transformation of *p*-nitrophenol to *p*-aminophenol [Fig. 2]. Fermentatively produced hydrogen continued to act as the reducing agent for contaminant transformation. Our results demonstrate the potential use of MG for synthesis of Bio-Pd (0), Pd (II) recovery and efficient treatment of toxic contaminants in acidic environment. We also investigated the potential application of Bio-Pd in nitrate reduction.

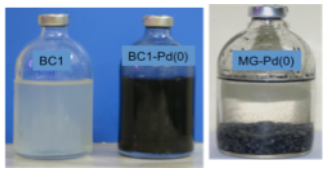

Fig 1. *Clostridium* sp. BC1 culture, Pd (0) nanoparticles associated with BC and microbial granules (MG).

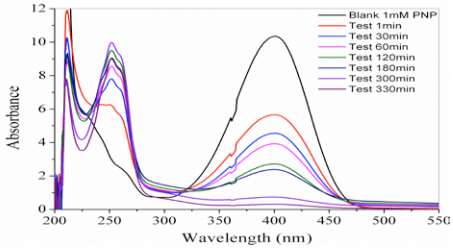

Fig 2. Bio-Pd mediated transformation of *p*-nitrophenol to *p*-aminophenol.

[1] Nancharaiah and Francis (2011) *Bioresour Technol*. 102(11)6573–8. [2] Suja *et al*. (2012) *Appl Biochem Biotechnol*. 167:1569–1577.

10.1180/minmag.2013.077.5.19