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Experimental evaluation of formation kinetics for abiotic methane in hydrothermal systems

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To better understand the kinetics of methane formation through abiotic reaction pathways (i.e., Fischer-Tropsch type synthesis) in hydrothermal systems, a series of laboratory experiments are performed. Using a flow-through reactor, dissolved CO_2 and H_2 are introduced with the presence of mineral catalyst. Reactions proceed in a thermodynamically open system, in which the steady state is maintained. The key reaction variables, including dissolved CO_2 and H_2 concentrations, temperature, and fluid residence time, are adjusted independently or in combination during the experiments, facilitating the systematic assessment of the potential controlling factors.

The experiments are conducted at temperature ranging from 150 to 300 °C. The residence time of the fluids varies between 3 and 36 hours. Different minerals, magnetite (Fe_3O_4) or bunsenite (NiO), are used in the experiments as the reaction catalyst. The concentration of dissolved CO_2 and H_2 varies, with the ratio of $H_{2(aq)}/CO_{2(aq)}$ ranging from 1 to 10, representing distinct redox conditions.

Experimental results have shown the effect of variables on methane formation to different extents. Bunsenite is more effective on methane generation than magnetite. The formation rate of methane is also temperature-dependent. The highest rate is observed at about 200 °C with bunsenite, and at 170 °C with magnetite. The overall formation rate (R in mmol/kg/hour) of methane can be expressed as:

$$R = \frac{a}{b} e^{-0.5\left(\frac{T-c}{b}\right)^2} \frac{{H_{2(aq)}}^2 + dCO_{2(aq)}}{H_{2(aq)} * CO_{2(aq)}}$$

where T is temperature in K, and $H_{2(aq)}$ and $CO_{2(aq)}$ are the concentration of dissolved H_2 and CO_2, respectively. The coefficients (a, b, c, and d) vary with minerals, with a = 1272.4, b = 24.7, c = 479.5, d = -2.8 for bunsenite, and a = 66.1, b = 24.3, c = 442.1, d = 2581.7 for magnetite.

Heavier light alkanes (C_2 to C_4) are also generated, but with slower rates than methane. Identification of other organic compounds in solution and on mineral surfaces are currently in progress. Altogether, those results may help shed light on the reaction mechanism of abiotic methane formation in hydrothermal systems, and evaluate the contribution of abiotic hydrocarbons to the carbon cycle on Earth.