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Beyond the Water-Rich Paradigm: Experimental Evidence for H_2 Generation and Abiotic Hydrocarbon Formation in Vapor-Dominated Serpentinizing Systems

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Hydrogen (H_2) generation during serpentinization is commonly investigated in water-rich laboratory systems. However, in many natural ultramafic settings, water availability can be limited, and the system may evolve from liquid-water-dominated to water-vapor-only conditions as reactions proceed. This raises a key question: as a system shifts toward nearly dry conditions, can serpentinization and H_2 production continue, and what carbon-bearing gases form (if any)? We conducted controlled hydrothermal experiments at 200°C with three objectives: (1) assess water availability and phase state as first-order controls on serpentinization and H_2 generation; (2) clarify the role of CO_2 and test whether a "H_2 promotion window" exists; and (3) constrain the conditions for abiotic hydrocarbon formation in serpentinized systems.

Our results show that H_2 is produced under both liquid- and vapor-phase conditions. The phase state strongly affects kinetics: H_2 generation persists after the system becomes water-vapor-only, but the production rate decreases by about a factor of 10 compared with liquid-dominated conditions. This provides direct experimental evidence that vapor-phase serpentinization can sustain H_2 generation, offering a process-based explanation for the spatial heterogeneity of H_2 production in natural systems where fluid supply varies.

Contrary to the expectation that CO_2 uniformly inhibits H_2 production, we find its effects are variable depending on conditions and reaction stage. In water-rich experiments where serpentinization had already progressed for one month, CO_2 injection increased the H_2 generation rate, pointing to a probable "promotion window". In water-limited experiments, CO_2 had little effect on the H_2 generation rate but consistently shortened the production time window of H_2, resulting in earlier plateaus and lower cumulative yields.

Natural hydrogen seeps worldwide are often accompanied by CH_4 whose compositional variability remains poorly understood. By systematically simulating abiotic hydrocarbon generation under controlled temperature and water-availability conditions, we find that CH_4 production exceeding background levels is only observed at 250°C with no detectable signal at lower temperatures. These results provide an experimental baseline for interpreting co-emitted gases at hydrogen seeps and assessing natural hydrogen resource potential.