Dry abiotic oxidation of H₂ by hematite, pyrite, and other redoxactive minerals at subsurface conditions (120°C, 200 bar)

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Despite widespread H2-generating processes in the subsurface like serpentinization or radiolysis of water, molecular hydrogen in the subsurface rarely occurs in higher concentrations [1]. Hence, significant amounts of H₂ are lost in a variety of processes that encompass microbial activity as well as abiotic oxidation. The knowledge of reaction rates of abiotic oxidation of H₂ at reactive mineral surfaces or by dissolved oxidizing species are not only useful for a quantitative description of natural H2 flows but also for underground hydrogen storage (UHS). UHS is a promising technology for buffering fluctuations in electricity production by renewable sources [2]. The extent of abiotic oxidation of H₂ under UHS conditions is one crucial parameter for the assessment of a potential storage formation. However, there is a lack of kinetic data for low-temperature abiotic H₂ oxidation processes.

To fill the gap we conducted a series of initially dry experiments with H₂ at potential UHS conditions. H₂ was reacted with several redox-active minerals (hematite, pyrite, magnetite, goethite, anhydrite and chlorite) typically occuring in porous sandstone formations or shale cap rocks to (1) identify H₂-oxidizing mineral surfaces and (2) to derive kinetic data of abiotic H₂ oxidation for minerals with significant reactivity. The experiments used sealed gold capsules as reaction cells in high-pressure reactors at 120°C, 200 bar for 2-14 days. The reaction progress was monitored by quantifying the amount of consumed H₂ and, in addition, the amount of generated reaction products as H₂O or H₂S by means of gas chromatography and mass spectrometry.

In experiments with initially dry conditions, only hematite and pyrite along with the less reactive goethite and smectite exhibited a detectable H₂-consumption. Thus, kinetic data of H₂ oxidation by hematite and pyrite in the dry system were obtained in follow-up experiments at different temperatures. Current investigations focus on hydrothermal experiments of redox-active minerals with an initial liquid water phase and H₂.

 Sherwood-Lollar *et al.* (2014) Nature **516**, 379-382.
Liebscher *et al.* (2016) in Hydrogen Science and Engineered Materials, 629-658.