

Abiotic mineral-fluid-gas reactivity in underground H₂ storage: Preliminary results on reductive pyrite dissolution kinetics at 60-150 °C and elevated P_{H2}

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H₂ is a future energy storage medium, as excess energy from renewable sources can be used to produce green H₂. Large-scale H₂ storage in geologic subsurface environments (e.g. salt caverns, depleted natural gas fields, porous rock aquifers) is, therefore, anticipated as part of the energy transition. However, very few studies have investigated fluid-rock-H₂ interactions at conditions relevant for underground storage (40-120 °C, P_{H2} up to 250 bar), including the production of H₂S as a gaseous by-product phase [1, 2].

To address these issues, the reaction kinetics of reductive pyrite dissolution at conditions relevant for underground H₂ storage are currently being investigated at T = 60-150 °C and P_{H2} = 7 bar (70 bar Ar/10%-H₂ gas mixture) in Ti-batch reactors, whereas other experiments at P_{H2} >> 7 bar (pure H₂) are being performed in stainless steel batch reactors at 90 °C. The experiments are based on natural pyrite (50-100 μm) altered in 30 mmol/L NaCl solutions buffered to pH ≈ 8 by calcite. The temporal evolution of dissolved sulfide concentrations is determined using electrochemistry, photometry, and an SO₄²⁻-derivative approach coupled to ICP-OES. After periods of 3-4 weeks approximate steady-state dissolved sulfide concentrations of 0.1-1.5 mmol/L at 90-150 °C have been measured, in addition to gaseous H₂S production. SEM of post-mortem pyrite grains reveals the formation of authigenic pyrrhotite (Fig. 1), in agreement with [1, 2]. Experiments using pyrite bearing trace amounts of native sulfur (< 1 wt.%) correlate positively both with significantly faster initial sulfide release kinetics and higher total dissolved sulfide concentrations at the same T and P_{H2} conditions, suggesting that the reduction of native sulfur is another source for H₂S.

[1] Truche, Berger, Destrigneville, Guillaume & Giffaut (2010), *Geochimica et Cosmochimica Acta* 74, 2894-2914.

[2] Truche, Jodin-Caumon, Lerouge, Berger, Mosser-Ruck, Giffaut & Michau (2013), *Chemical Geology* 351, 217-228.

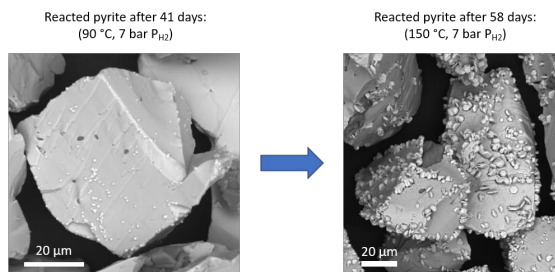


Fig. 1: Reacted pyrite (FeS₂) showing increasing degrees of secondary pyrrhotite (FeS) formation.