

Pyritization and natural hydrogen genesis

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We aim to explain the H₂ genesis in Earth's crust in a sulfur environment. While most of the known reactions are usually associated with minerals reacting with water under oxidizing conditions, such as the serpentinization and siderite decomposition, the role of pyritization has been significantly underestimated. We use thermodynamic simulations (SUPCRTBL) to predict the direction of a range of possible metamorphic reactions regarding Gibbs free energies as a function of depth in the crust, temperatures up to 1500 °C, pressure up to 10 kbar and including the relative partial pressures of H₂ and H₂S gases. We show that iron-based oxide precursors under hydrogen sulfide lead to H₂ formation and pyrite FeS₂ in the upper part of the continental crust (down to 12km using the thermal gradient of 30°C at Qr=1). As the crust can be considered as a semi-open system for gas mobility (lower H₂ partial pressure with H₂ leaking), its genesis may occur at deeper levels. The slow cooling of continental crust through time in cratonic area is also compatible with the enhancement of exothermic pyritization reactions. The pyritization induces a reduction of ferric into ferrous iron forming hydrogen, completing the iron cycle to the inverse oxidation process represented by the serpentinization. Those complementary reactions can indeed co-exist in acidic black smokers (pyritization) and basic white smokers (serpentinization). The complete cycle of iron Redox states may occur either in continental crust or in oceanic crust when including the chemical reactions associated with subduction of an oceanic slab. Magnetic changes (through magnetite formation/dissolution), thermal gradient changes, pH of fluids and pyritization are then indicators of the localization of the formation of H₂. It implies that natural hydrogen is a renewable raw matter on Earth which will revalorize the interest in exploring and investing on such clean energy for fuel cells and other industrial applications.

Mots-Clés : Thermodynamics, FeS₂, iron oxides, sulfur, black smokers

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