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Replacement of pyrrhotite by pyrite and marcasite under hydrothermal conditions up to 220 °C: An experimental study of reaction textures and mechanisms

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Abstract

The transformation of pyrrhotite to Fe disulfide (pyrite and/or marcasite) under hydrothermal conditions was studied experimentally by probing the effects of temperature (up to 220 °C, vapor-saturated pressures), ΣS(-II) concentrations, pH, and availability of oxygen on reaction progress and on the resulting textures.

The pyrrhotite to Fe disulfide reaction proceeded by a dissolution-reprecipitation mechanism under all conditions. Marcasite and pyrite formed under both oxic and anaerobic conditions, which is inconsistent with the traditionally assumed polysulfide route for FeS_2 formation (oxidants required for polysulfide formation). The nature of the products was controlled by the level of supersaturation of the solution with respect to Fe disulfide minerals. Marcasite formed preferentially at low pH or S(-II)-deficient solutions (saturation index << 1000), while pyrite was the main product at saturation indices >1000.

Different textures were obtained for the replacement of pyrrhotite by either pyrite or marcasite. Pyrite formation proceeded by direct replacement of pyrrhotite and, simultaneously, by overgrowth from solution. The pyrite crystals were >10 μm in size and randomly oriented. In comparison, marcasite crystals were <1 μm in size, and no significant overgrowth was observed. At $pH_{21°C}$ <3, the marcasite nanocrystals showed the well-known crystallographic relationship with pyrrhotite, but at $pH_{21°C}$ 3.96, the marcasite crystallites were randomly oriented. These experimental results confirm that the preservation of the crystallographic orientation is not a distinguishing feature between dissolution-reprecipitation and solid-state reactions. The different textures among pyrite and marcasite reflect the dominance of crystal growth (pyrite) vs. nucleation (marcasite) as a precipitation mechanism.

Keywords: Hydrothermal replacement, pyrrhotite alteration, pyrite, and marcasite, dissolution-reprecipitation reaction, crystallographic relationship, textures