Microstructural controls on pyrrhotite weathering and secondary mineral formation: A TEM study

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Weathering processes are strongly associated with the hydrosphere, atmosphere, and biosphere and are controlled by reactions at mineral surfaces. The kinetics of mineral dissolution and the precipitation of secondary phases play a fundamental role for the local and global geochemical cycles. Moreover, the oxidative alteration of sulfidic ores due to natural events and mining activities leads to the production of acid rock drainage (ARD) and acid mine drainage (AMD), respectively, releasing acidity (low pH) as well as high heavy metal and sulfate concentration to the environment and is a concern to the water quality of ground and surface waters worldwide.

Pyrrhotite is found in a wide range of magmatic and metamorphic rocks as well as in massive deposits and is, after pyrite, the most common iron sulfide in nature. The variable Fe/S ratio ($Fe_{1-x}S$, with x = 0 – 0.125) allows for various stacking sequences and ordered superstructures. Furthermore, twinning, stacking faults, and dislocations are common in natural samples (e.g., Posfai et al., 2000). The oxidation rates of pyrrhotite are on the order of 20 to 100 times higher compared with pyrite (Nicholson and Scharer, 1994).

This study focuses on the influence of the microstructure on the reacting front and the spatial relationship between the primary mineral and the secondary phases at nanometer scale. Pyrrhotite (from Dalnegorsk, Russia) is associated with pyrite and secondary goethite. Transmission electron microscopy (TEM) reveals the 4C modification as dominant superstructure as well as numerous stacking faults. Weathered rims have a thickness of about 20 μm and are compositionally heterogeneous. A first shell (about 15 μm) consists of fine-grained Fe-oxyhydroxide which is loosely bound to pyrrhotite. A second shell (less than 5 μm) is formed by massive secondary FeS_2 (probably marcasite). Where pore space is available, grains of goethite (5 to 20 μm in diameter) have been formed.

The observed sequence of secondary phases is in contrast to a XPS/AES study of Pratt et al. (1994) on pyrrhotite oxidation in air who reported a sequence Fe_7S_8-Fe_2S_3-FeS_2-Fe(III) oxide. A discussion of the results on the background of thermodynamic and kinetic considerations will be presented.

References

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