

Pyrite (FeS₂) oxidation as a function of pH: a multitechnique approach

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This work intends to identify the FeS₂ oxidation mechanism and to evaluate the effects of carbonates during its dissolution. Oxidative dissolution experiments are carried out at several pH associating solution speciation to surface solid analysis (XPS, FTIR, SEM and Nuclear Microprobe Analysis). The influence of carbonates is investigated at several contents in equilibrium with air.

We show that fast FeS₂ air oxidation explains the disparity of both results and oxidation mechanisms published.

Previous works indicate S at oxidation number 0 (FeSSO) and 2 (FeS₂O₃) on localized points of FeS₂ surface (Descostes *et al.*, 2001). SO₄²⁻ and S₂O₃²⁻ are the main S aqueous species whatever the pH. Under acidic conditions (pH<3) a S deficit leads to ratios $R=[S]_{tot}/[Fe]_{tot}<2$ (Descostes *et al.*, 2004). Given a kinetic model FeS₂ dissolution may be ruled by a localized first solid-state oxidation step followed by S₂O₃²⁻ dissolution. In acidic media S₂O₃²⁻ dismutation into S⁰ and S₄O₆²⁻ explains the observed uncongruence with a predicted S/Fe ratio: R=1.6. This work offers a new sight of the well accepted iron preferential dissolution models.

In buffering conditions $\Sigma CO_2 \geq 10^{-2}$ mol/L 90% of aqueous S is under S₂O₃²⁻ form. The dissolution rate raises with ΣCO_2 . The observed FeS₂ → siderite FeCO₃ → goëthite α -FeOOH → lepidocrocite γ -FeOOH → ferric sulfate transformation can't be predicted by an equilibrium thermodynamic model. The competition between Fe(II) oxidation and complexation - precipitation explains the raising of the dissolution rate vs ΣCO_2 . Concurrently a component GR-SO₃ from the green rust compounds family incorporates SO₃²⁻.

References

- Descostes M., Mercier F., Beaucaire C., Zuddas P. and Trocellier P. (2001), *Nucl. Instr. and Meth. B*, **181**, 603-609.
- Descostes M., Beaucaire C. and Vitorge P. (2004), *Geochimica et Cosmochimica Acta*, **68**, 4559-4569