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Mössbauer evidence of rapid pyrite formation during the interaction of dissolved sulfide and lepidocrocite

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Mössbauer spectroscopy has been applied to study mineral transformation during the interaction between lepidocrocite and dissolved sulphide with two different initial concentration ratios. Synthetic lepidocrocite enriched with the Mössbauer sensitive isotope Fe-57 was reacted with dissolved sulphide at neutral pH in an anoxic glove box, the initial molar ratios of iron and sulphide being 3.125 and 0.5, respectively. Solid samples for Mössbauer analysis were collected by filtration at different time steps after 15 min, 2hrs, 48hrs, 72hrs and 1 week for the 3.125 iron-sulphide ratio; and after 72hrs and 1 week for the 0.5 iron-sulphide ratio. Dissolved and solid phase ferrous iron, dissolved sulphide and elemental sulphur were measured with wet chemistry analysis techniques in parallel runs.

Mössbauer spectra provide evidence for rapid pyrite formation within 48hrs during the reaction at an initial molar ratio of 3.125. Pyrite formation was accompanied by a decrease of elemental sulphur and mackinawite, which is the main iron intermediate product with minor pyrrhotite and magnetite. At the ratio of 3.125, excess lepidocrocite existed and dissolved sulphide was completely consumed after 15min. Acid extractable Fe(II) concentration was in excess of that of Fe(II) bonded as FeS, which has been reported before[1,2] and the difference of the two iron species was defined as excess Fe(II)[2]. In the experiment with an initial ratio of 0.5, lepidocrocite was completely transferred to mackinawite. In this system dissolved sulphide was not completely consumed even after 1 week. No pyrite could be detected. Species including elemental sulphur and mackinawite were stable in 1 week without any further transformation. The long term preservation of unstable iron sulphide minerals at a low concentration of dissolved sulphide is consistent with the observation in marine sediment [3].

These experiments show an important linkage between rapid pyrite formation and surplus lepidocrocite and/or the excess Fe(II) formed during reaction. Together with wet chemistry analysis results we propose that some of the electrons donated by sulphide oxidation are preserved temporally in the lepidocrocite bulk and lead to formation of the excess Fe(II) fraction and ultimately rapid pyrite formation.

[1] Poulton et al.(2004) Geochim. Cosmochim. Acta.
[2] Hellige et al. (2011) Geochim. Cosmochim. Acta.
[3] Gognon et al. (1995) Geochim. Cosmochim. Acta.