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# Effects of Al(III) on the ferrihydrite – ordered ferrimagnetic ferrihydrite – hematite transformation

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Ferrihydrite, a ferric oxyhydroxide nanomineral with high chemical reactivity, is ubiquitous in various Earth surface environments and especially in the edaphosphere. Aluminium is also very common in soils and therefore is tipically associated with iron oxides.

Here, we study the influence of Al on the hydrothermal transformation of Al-ferrihydrites to better understand both the structure (much debated in the last years; Michel et al., 2010, Manceau, 2011) and properties of this poorly crystalline phase. Suspensions of synthetic aluminium 2-line ferrihydrite [with Al/(Fe+Al) from 0 to 19 mole%] doped with citrate (citrate/Fe molar ratio = 3%) in order to retard rapid transformation into hematite was aged at 175 °C for periods ranging from 0 to 18 h. X-ray diffraction in the real- and reciprocal-space analyses derived from synchrotron high-energy x-ray total scattering, Mössbauer spectroscopy, magnetic measurements and transmission electron microscopy were used to characterize the dry final products.

The presence of Al delays the transformation of ferrihydrite into hematite. In the low-Al samples there is a clear sharpening of the ferrihydrite peaks during aging that is consistent with the formation and growth of the intermediate ordered ferrihydrite. Samples with higher Al contents show minor changes in intensity and peak width that suggest these samples produce less and/or smaller sized intermediate ordered ferrihydrite during aging prior to the formation of hematite. The a- and c-dimensions of the ferrihydrite unit cell [according to the Michel et al. (2010) structure] both decrease with increasing Al content. Uniform changes in the unit cell provide evidence for substitution in all three cation sites in the ferrihydrite structure. The pair distribution functions (from the real XRD pattern) suggest virtually no change in particle size with increasing Al.

Increasing the Al concentration results in a decrease in the magnetic enhancement that occurs during aging. The presence of Al also causes such enhancement to persist for longer times. A saturation magnetization maximum of about 90 Am2kg-1 (comparable to that of magnetite) was obtained for the sample with 2 mole% Al after 6 hours of aging. Mössbauer spectra in the paramagnetic state indicate a considerable variation of the local coordination of Fe. Low-temperature spectra indicate that the magnetic properties are strongly influenced by interparticle effects.

In summary, the structural and magnetic properties of the ordered ferrimagnetic ferrihydrites resulting from the transformation of the 2-line Al-ferrihydrites are consistent with the model proposed by Michel et al. (2007, 2010).

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