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Stability of Al-substituted jarosite in the presence of Fe(II)

Andrew Grigg[1], Luiza Notini[1], Ralf Kaegi[2], Laurel ThomasArrigo[1], and Ruben Kretzschmar[1]
[1]Soil Chemistry Group, Institute of Biogeochemistry and Pollutant Dynamics, Department of Environmental Systems Science, ETH Zurich, Universitätstrasse 16, CHN, CH-8092 Zurich, Switzerland
[2]Eawag, Swiss Federal Institute of Aquatic Science and Technology, Überlandstrasse 133, CH-8600 Dübendorf, Switzerland

Jarosite is a ferric iron sulfate mineral [$(KFe_3(SO_4)_2(OH)_6$] that is commonly formed in acidic environments that are rich in iron and sulfate, such as acid-sulfate soils or acid mine drainage. The stability of jarosite is important because the mineral contains embodied acidity and may scavenge trace elements by sorption and co-precipitation. Although stable under high Eh and low pH conditions, previous studies have shown that jarosite is prone to transformation by hydrolysis at circumneutral pH, or may undergo Fe(II)-catalysed transformation where ferrous ions are present [1-3]. Jarosite may be exposed to Fe(II) at circumneutral pH in reducing environments, such as in flooded acid-sulfate soils [2]. Jarosite is a member of the alunite supergroup and forms a solid solution series with alunite by substitution of Al for Fe. However, the effect of Al substitution on the stability of jarosite in the presence of Fe(II) has not previously been investigated. Here, we performed batch experiments using samples of a synthetic jarosite without aluminium substitution, and synthetic jarosite containing 7.3% Al-for-Fe substitution. Mineral samples were reacted with 0.5 mM and 5 mM Fe(II) at pH 7.1 (50 mM MOPS buffer) for up to 24 hours. Rietveld analysis of X-ray diffraction patterns was used to quantify mineral transformations and to determine the crystallinity of, and Al substitution in, product phases. Complete transformation of jarosite to mixtures of ferrihydrite, goethite and lepidocrocite occurred within several hours for all jarosite samples and Fe(II) treatments. The 10-fold increase in Fe(II) concentration resulted in a 50% increase in jarosite transformation rate, and pure jarosite transformed 110% to 280% faster than Al-substituted jarosite. The transformation products of Al-substituted jarosite contained a smaller proportion of lepidocrocite than the products of pure jarosite transformation, and the unit cell size of the lepidocrocite that initially formed from Al-substituted jarosite indicates that Al was substituted into the structure. These results demonstrate that structural Al can stabilise jarosite against transformation, which has implications for understanding the longevity of jarosite, and its importance to trace element cycling, in reducing environments.

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