

SIGNIFICANCE AND FATE OF JAROSITES IN BROWN COAL MINING SITES: A FIELD AND LABORATORY STUDY

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In disused brown coal mining pits ascending groundwater is expected to change chemical conditions and to mobilize hazardous elements. With it, the stability relations of minerals containing iron and sulfur like jarosite $AFe_3(SO_4)_2(OH)_6$ ($A = Na, K, H_3O, NH_4$), schwertmannite $Fe_{16}O_{16}(SO_4)_2(OH)_{12}$ and goethite $FeOOH$ influence the distribution of metals, mainly iron and sulfur between solid phases, surface and ground water.

The present study focuses on jarosite because it has been detected in large amounts in brown coal sites of the Central German and the Lusatian District (Germany). From its iron to sulfur ratio of 1.5 in comparison with 0.5 in the original iron disulfide, jarosite enriches iron and high amounts of sulfate result in the related surface waters. Formation of mainly K-rich jarosite is due to the release of K^+ from the most common K-bearing silicate minerals (muscovite, K-feldspar) of the Tertiary sediments through reaction with sulfuric acid. But also Na-rich jarosites and NH_4 -bearing jarosites have been detected. NH_4 might result from nitrate stimulated oxidation of FeS_2 . No end member oxonium jarosites have been found in the dump sites until now. But from lattice constants it is likely that part of the alkalis in the jarosite structure is replaced by H_3O .

Investigations of site materials often give only partly information about location, duration and mechanisms of mineral formation. Just these data are needed to reconstruct acidification processes. Therefore mineral precipitation on test substances placed in an acidic mining lake (pH 2.6, Lusatian District) show the most intensive coatings close to the lake surface which have been identified by Raman spectroscopy as jarosite.

Jarosite precipitated in the lake water takes part in the sediment formation: up to 40 wt% jarosite have been measured. Assemblage of goethite, jarosite and rarely schwertmannite in the sediments (pH ≥ 3) of the lake under investigation indicate instability of jarosite under aerobic conditions. An aim of remediation measures could be to initialize neutralization processes by microbial sulfate reduction. Thus, the behaviour of jarosite under both aerobic and anaerobic conditions has to be known. Laboratory experiments show that by hydrolysis under aerobic conditions sulfate release decreases in the order of H_3O -, Na-, K-, K(Fe,Cr)-jarosite. Cr(III) in the structure seems to stabilize K-rich jarosites. Red-brown coloured surface coatings indicate iron oxyhydroxides. Increasing run duration confirms goethite as a product of hydrolysis for K-rich jarosite. Under anaerobic conditions in the presence of sulfate reducing bacteria, K-rich jarosite decomposes and iron sulfides precipitate as black coloured covers. Jarosite surfaces during and after hydrolysis are characterized using atomic force microscopy (AFM).

Structure refinement from X-ray powder and single crystal investigations as well as IR and Raman spectroscopic measurements supply crystallographic and crystallochemical data which are necessary to calculate the stability of jarosite crystal faces to get to know its surface properties.