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Local structure and crystallization-transformation of hydrous ferric arsenate in acidic systems: Implications for acid mine drainage and arsenic geochemical cycling

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Hydrous ferric arsenate (HFA) is a common thermodynamically metastable phase in acid mine drainage. However, little is known regarding the structural forms and transformation mechanism of HFA. We investigated the local atomic structures and the crystallization-transformation of HFA at various Fe(III)/As(V) ratios in acidic solutions. Results show that the Fe(III)/As(V) in HFA decreases with decreasing initial Fe(III)/As(V) at acidic pHs. The protonation degree of As(V) in HFA increases with increasing As(V) concentrations. The Fe *K*-edge EXAFS and XANES results reveal that each FeO_6 is linked to more than two AsO_4 in HFA precipitated at Fe(III)/As(V)<1. Furthermore, the formation of scorodite ($FeAsO_4 \cdot 2H_2O$) is greatly accelerated by decreasing the initial Fe(III)/As(V). The release of As(V) from HFA is observed during its crystallization-transformation process to scorodite at Fe(III)/As(V)<1, which is different from that at Fe(III)/As(V)≥1. SEM results show that Oswald ripening is responsible for the coarsening of scorodite regardless of the initial Fe(III)/As(V) or pH. Moreover, the formation of crystalline ferric dihydrogen arsenate as an intermediate phase at Fe(III)/As(V)<1 is responsible for the enhanced transformation rate from HFA to scorodite. This work provides new insight into the local atomic structure of HFA and its crystallization-transformation that may occur in acid mine drainage and has important implications for arsenic geochemical cycling.

