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Formation of amorphous Fe arsenates and their roles in the crystallization of scorodite

DR. XIANGYU ZHU[1], PEI CHANG[2] AND HUI HENRY TENG[3]

[1]School of Earth System Science, Institute of Surface-Earth System Science, Tianjin University
[2]Chengdu University of Technology
[3]School of Earth System Science, Institute of Surface-Earth System Science, Tianjin University, Tianjin

Amorphous ferric arsenate (AFe^{III}) and amorphous ferrous arsenate (AFe^{II}) serve as crucial arsenic sinks in acidic, arsenic-rich environments, playing key roles in the geochemical cycling and mineralization of iron-arsenic-bearing minerals such as scorodite ($FeAsO_4 \cdot 2H_2O$). Despite their significance, the precipitation kinetics and structural transformations of these amorphous phases remain poorly understood. This study examines the formation and crystallization pathways of AFe^{III} and AFe^{II} under varying pH, As/Fe ratios, and temperature conditions using a combination of in situ and ex situ techniques, including small- and wide-angle X-ray scattering SAXS and X-ray absorption spectroscopy XAFS .Our results reveal a two-stage precipitation process for AFe^{III}: (i) the initial formation of subnanometer- to nanometer-sized particles (8.87–10.11 Å radius), likely consisting of arsenate-FeO_6 octahedral complexes, followed by (ii) particle growth via a non-classical attachment mechanism involving hydrogen bonding. Elevated temperatures enhance particle growth in Stage I by promoting FeO_6 incorporation, while higher pH and As/Fe ratios accelerate precipitation by reducing electrostatic repulsion. In contrast, AFe^{II} exhibits longer Fe–O bond lengths (2.02 ± 0.01 Å) relative to AFe^{III} (1.99 ± 0.01 Å) and scorodite, leading to slower transformation kinetics due to structural misfit and a reduced capacity to supply aqueous Fe^{3+}. Consequently, AFe^{II} crystallization results in the formation of larger, euhedral scorodite crystals. These findings improve our understanding of Fe-As mineralization mechanisms and provide insights into arsenic remediation strategies applicable to both natural and industrial settings.