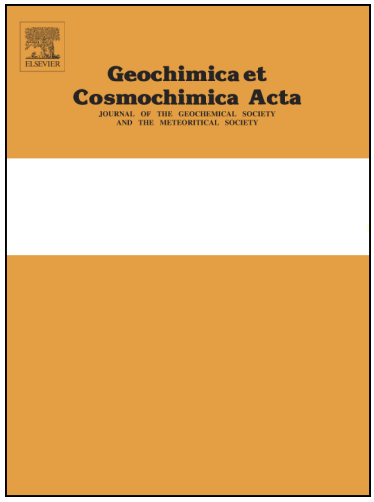

Cerium sequestration and accumulation in fractured crystalline bedrock: the role of Mn-Fe (hydr-)oxides and clay minerals

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Cerium sequestration and accumulation in fractured crystalline bedrock: the role of Mn-Fe (hydr-)oxides and clay minerals

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Abstract

This study focuses on the mechanisms of Ce sequestration and accumulation in the fracture network of the upper kilometer of the granitoid bedrock of the Baltic Shield in southeast Sweden (Laxemar area, Sweden). The material includes 81 specimens of bulk secondary mineral precipitates ("fracture coatings") collected on fracture walls identified in 17 drill cores, and 66 groundwater samples collected from 21 deep boreholes with equipment designed for retrieval of representative groundwater at controlled depths. The concentrations of Ce in the fracture coatings, although varying considerably (10^{th}-90^{th} percentiles: 67-438 $mg \cdot kg^{-1}$), were frequently higher than those of the wall rock (10^{th}-90^{th} percentiles: 70-118 $mg \cdot kg^{-1}$). Linear combination fitting analysis of Ce L_{III}-edge X-ray absorption near-edge structure (XANES) spectra, obtained for 19 fracture coatings with relatively high Ce concentrations ($\geq$ 145 $mg \cdot kg^{-1}$) and a wide range of Ce-anomaly values, revealed that Ce(IV) occurs frequently in the upper 10 m of the fracture network ($Ce(IV)/Ce_{total}$ = 0.06-1.00 in 8 out of 11 specimens) and is mainly associated with Mn oxides (modeled as Ce oxidatively scavenged by birnessite). These features are in line