Uranium speciation in two Freital mine-tailing samples: SSE, μ-XRD, and μ-XRF results

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Introduction

• During the uranium ore extraction, uranium is converted from relatively insoluble U(IV) minerals to highly mobile U(VI) species.
• Contaminated mine waste piles and tailings and surrounding soils are usually covered by soil materials to reduce the Rn emission and radionuclide spreading by wind and water erosion.
• Little is known on the further fate of uranium at these sites. Only by a combination of micro-spectroscopic and wet chemical approaches, which are fundamental to achieve reliable results.

Elemental Mapping (μ-XRF)

Distribution of U, K, S (left) and μ-XRF of selected microspots (right)

Sample F1 (pH 8): U predominates homogeneously distributed at relatively low concentrations (mg/kg) in aggregates with a diameter of tens to hundreds of µm, where it is associated with K and 3d metals

Distribution of U, Cu, V (left) and μ-XRF of selected microspots (right)

Sample F3 (pH 4): U is highly enriched in local spots (g/kg) with distinct elemental compositions suggesting a variety of U-containing minerals

Identification of soluble phases (SSE)


Sample F1: U is predominantly water soluble and/or ion exchangeable

Sample F3: U is predominantly hosted by recalcitrant solid phases, together with Th, Fe, Pb, and V

Identification of crystalline phases (μ-XRD)

Sample F1 (pH 8): U is associated with the layer silicates muscovite, illite and kaolinite.

Sample F3 (pH 4): U is hosted by a variety of U minerals, including coffinite, uraninite, uranyl hydroxide, and vanadinite, but also associated with layer silicates

Materials & Methods

Samples:

<table>
<thead>
<tr>
<th>Sample</th>
<th>Ph</th>
<th>E [keV]</th>
<th>µXRF</th>
<th>µXRD</th>
</tr>
</thead>
<tbody>
<tr>
<td>F1</td>
<td>8</td>
<td>18</td>
<td></td>
<td></td>
</tr>
<tr>
<td>F3</td>
<td>4</td>
<td>18</td>
<td></td>
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</tr>
</tbody>
</table>

-10^10 ph/s flux at E=18 keV

XR data extraction with AXIL (Univ. of Antwerp)

Pixel conversion with Fit2D (Hammersley, ESRF)

Conclusions

Sample F1 (pH 8): The diffuse distribution of low U concentrations among aggregates of layer silicates, and the high extractability suggest that U is sorbed to layer silicate surfaces. The type of minerals (muscovite, illite and kaolinite) and the high pH are in line with inner-sphere sorption on edge sites of the minerals, while the extraction during SSE 1 suggest outer-sphere sorption (ion exchange).

Sample F3 (pH 4): The combination of μ-XRF and μ-XRD enabled us to identify several U(VI) and U(IV) containing minerals like coffinite, uraninite, uranyl hydroxide, and vanadinite. Only a minor part of U is sorbed to layer silicates.

Geochemical implications:
At smaller depth (F1, 5 m), acidity from the ore extraction procedure was conserved. U(IV) minerals may precipitate as secondary minerals. Uranium remains highly mobile.

At greater depth (F3, 12 m), the low pH was conserved. U(VI) minerals may precipitate as secondary minerals. Uranium remains highly mobile.