

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 determining the chemical forms of uranium, one will know if uranium remains mobile for extended time periods, or if it precipitates as secondary minerals.
- We investigated the uranium speciation in covered mine tailings by a combination of micro-spectroscopic and wet chemical approaches, which are fundamental to achieve reliable results.

Materials & Methods

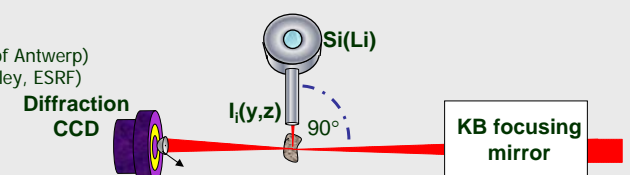
Samples:

Uranium mine tailings Freital (former Wismut site, Saxony, Germany)
Acidic ore extraction from 1949 - 1960
After 1968 covered with construction waste and tin-ore mining waste
Samples F1 and F3 collected from 5 and 12 m depth
Large range in pH (4-8)
Oxic pore water conditions at sampling (1994)

	F1	F3	F1	F3
pH/	8.0	4.0		
Eh/mV	450	500	Co	45
U	444	425	Ni	193
P	651	597	Cu	682
S	25900	5433	Zn	876
Cl	52	127	As	254
K	27950	33500	Se	15
Ca	12390	1261	Hg	3
Mn	71	135	Pb	1410
Fe	21950	26800	Th	16

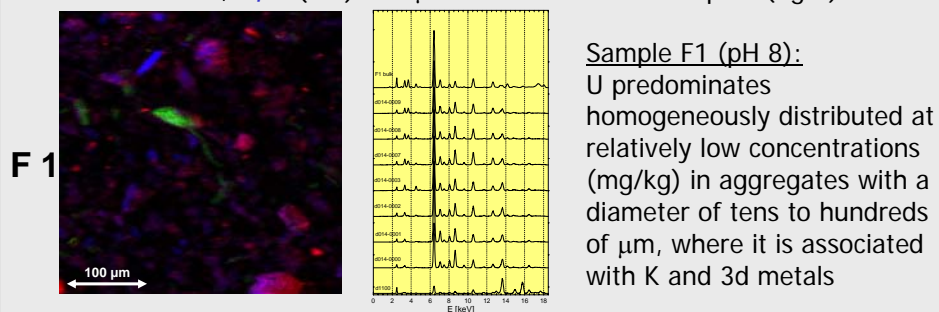
μ -XRF and μ -XRD at ID-22 (ESRF):

μ spot: $4 \times 1.5 \mu\text{m}^2$
 $\sim 10^{11}$ ph/s flux at $E=18$ keV
XRF data extraction with AXIL (Univ. of Antwerp)
Pixel conversion with Fit2D (Hammerley, ESRF)

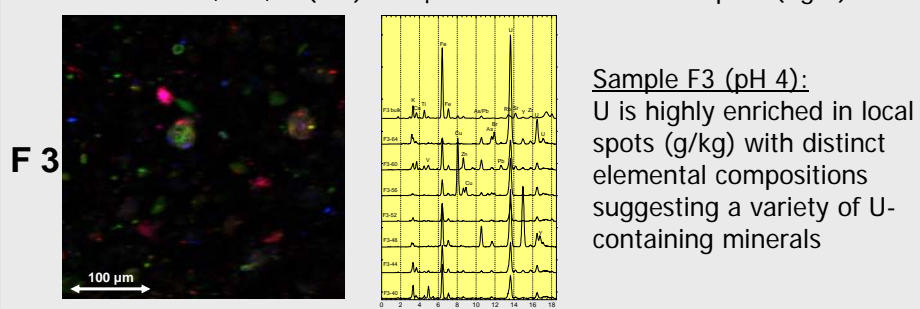


Elemental Mapping (μ -XRF)

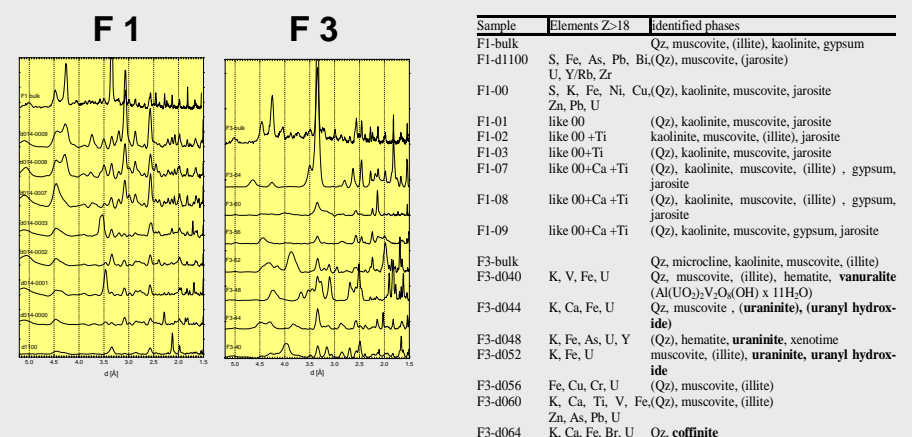
Distribution of U, K, S (left) and μ -XRF of selected microspots (right)



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



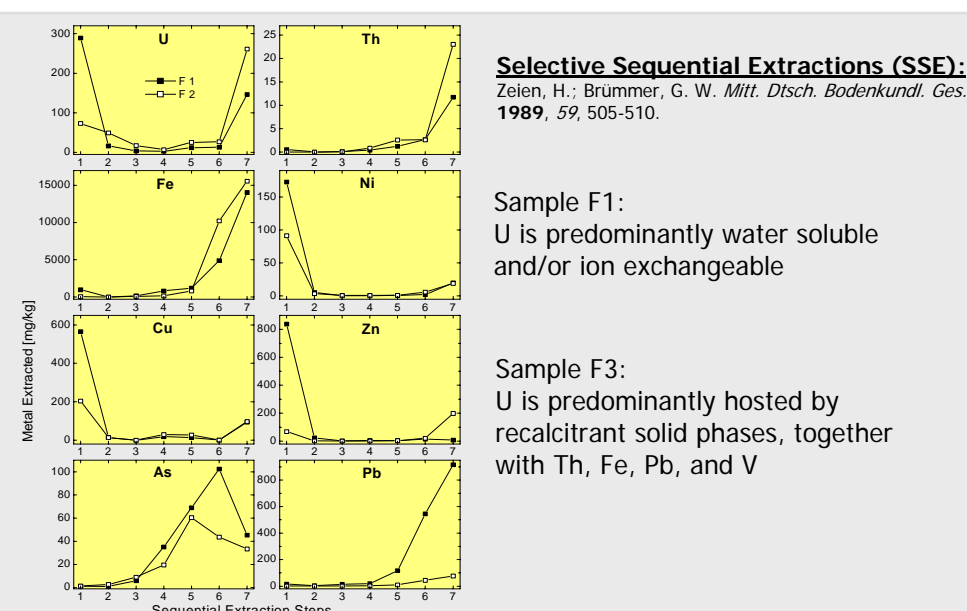
Identification of crystalline phases (μ -XRD)



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 vanuralite, but also associated with layer silicates

Identification of soluble phases (SSE)



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(IV) and U(VI) containing minerals like coffinite, uraninite, uranyl hydroxide, and vanuralite. 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 completely neutralized by the construction waste (jarosite, gypsum precipitation). Uranium remains highly mobile. At greater depth (F3, 12 m), the low pH was conserved. U(IV) minerals may have precipitated due to lower redox, and/or indicate a smaller ore processing efficiency for these older sediments. Uranium mobility is low.

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