The INE-Beamline for Actinide Research at ANKA

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Abstract. The INE-Beamline for actinide research at the synchrotron source ANKA is now fully operational. This beamline was designed, built, and commissioned by the Institut für Nukleare Entsorgung (INE) at the Forschungszentrum Karlsruhe (FZK), Germany. It is dedicated to actinide speciation investigations related to nuclear waste disposal as well as applied and basic actinide research. Experiments on radioactive samples with activities up to 10^6 times the limit of exemption inside a safe and flexible double containment concept are possible. The close proximity of the beamline to INE's active laboratories is unique in Europe. Currently, experiments can be performed in an X-ray energy range from around 2.15 keV (P K edge) to 24.35 keV (Pd K edge). The INE-Beamline design is optimized for spectroscopy with emphasis on surface sensitive techniques. A microfocus option is presently being installed at the INE-Beamline. Access to the INE-Beamline is possible through cooperation with INE, through the ANKA proposal system and via the European Network of Excellence for Actinide Sciences (ACTINET).

Keywords: Actinide, Sulfur, Phosphorous, XAFS, EXAFS, XANES, GI-XAFS, XRF, TXRF, XRD
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INTRODUCTION

The Institut für Nukleare Entsorgung (INE) at the Forschungszentrum Karlsruhe (FZK) has constructed and commissioned a beamline dedicated to actinide research at the synchrotron source Ångstrømquelle Karlsruhe (ANKA). Official operation of the INE-Beamline commenced on October 1st, 2005. There is presently a growing demand for synchrotron facilities, where the infrastructure, safety equipment, and expertise are available for performing research on radioactive samples. One great advantage of the INE-Beamline is that the ANKA accelerator and INE's active laboratories are both located within the same FZK site. This symbiosis has numerous advantages including profiting from the existing infrastructure, backed by decades of know-how. Samples can be prepared, characterized, and analyzed using the spectroscopic, analytical, microscopic, and structural methods available at INE's active laboratories before being transported to ANKA and investigated using synchrotron based methods at the INE-Beamline. The time-span between sample preparation and experiment is relatively short, which allows investigations of dynamic systems within a time-frame of hours. The samples are also retrievable, so that sample characterization in INE laboratories following the experiments at the INE-Beamline is possible. In addition, hazards associated with transporting radioactive samples to and from INE and nearby ANKA are minimized and the administrative requirements associated with such transports simplified.

BEAMLINE DESIGN

Because actinide speciation associated with nuclear disposal concerns is manifold, the major aim in the INE-Beamline design was to ensure variability, i.e., provide a multi-purpose station, where a number of methods are possible, including standard, surface sensitive, and spatially resolved methods.

For details of the optics section of the INE-Beamline design see ref. [1].

Experimental Station And Exemplary Measurements

The INE-Beamline is optimized for spectroscopic X-ray methods, X-ray absorption fine structure (XAFS), X-ray fluorescence (XRF), and combined XAFS and X-ray diffraction (XRD). Because many reactions of actinides in the hydro- and geo-sphere are at interfaces and junctions, emphasis at the INE-Beamline is being placed on surface sensitive

techniques based on grazing incidence (GI) geometry such as GI-XAFS, X-ray reflectivity and total external reflection X-ray fluorescence analysis (TXRF).

Our GI-XAFS investigation of the corrosion layer of a high level waste glass is an example of an grazing incidence experiment at the INE-Beamline. The uranium-containing glasses were kept for two years in a solution containing opalinus clay at two different temperatures, 50°C and 90°C. Figure 1 depicts the Fourier transforms of the U L3 EXAFS spectra of the two glasses (GI 50°C and GI 90°C) measured in grazing incidence geometry and two bulk measurements of an leached and a freshly prepared glass.

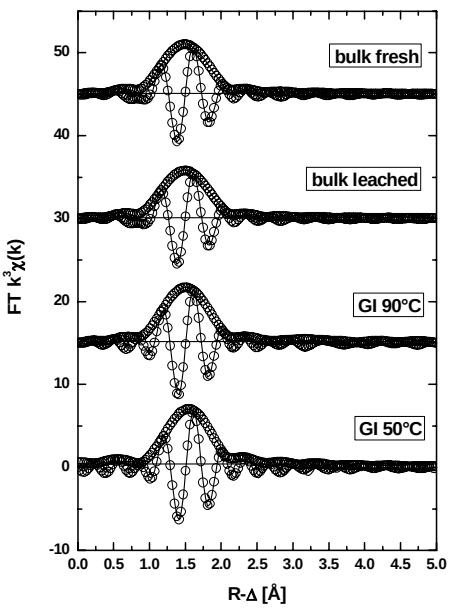

FIGURE 1. Fourier transformed spectra of the investigated samples. Experimental data are shown as lines and fits curved as symbols (see text for detail).

Two oxygen shells comparable to known uranyl containing compounds are revealed in the bulk samples: two short axial oxygen atoms (O1) at 1.77(2) Å and four further distant oxygen atoms (O2) bound to U(VI) in the equatorial plane at 2.21(3) Å. The U coordination structure in the glass bulk is not affected by leaching, as both bulk fresh and the bulk leached results are the same. Analysis of the GI-XAFS shows that U retains two shorter O1 distances within the corrosion layers. The number of O2 atoms increases to approximately five in the corrosion layer. This finding is supported by the near edge (XANES) spectra (Figure 2). The white line increases for samples GI 50° and GI 90°, which may indicate a reduced U coordination symmetry in the corrosion layer. Similar trends have been reported for Hf(IV) sorbed onto mineral surfaces [2]. Both the symmetry decrease and O2 number increase in the corrosion layer can be explained by additional coordinated water molecules within this layer [3]. The FT spectra exhibit no contributions at R-Δ values beyond the U-O peak, indicating that neither U agglomerates are present in the bulk glass nor do secondary U-phases form on the surface of the glass after leaching. This is in contrast to previous GI-XAFS investigations of the leaching behavior of other borosilicate glasses [4], where the agglomeration of uranium oxides was reported.

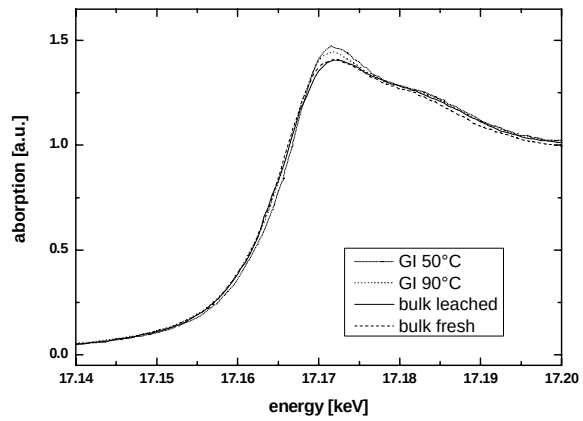

FIGURE 2. U L3 XANES spectra of the four samples.

We have designed, constructed and tested an inexpensive position sensitive XRD setup for performing combined XAFS/XRD investigations using flexible image plates made of radiation-sensitive, europium-doped barium fluoride films. The films are read out using a scanner with a laser beam and can be re-used after exposure to the day UV-light. The experimental setup is depicted in Figure 3.

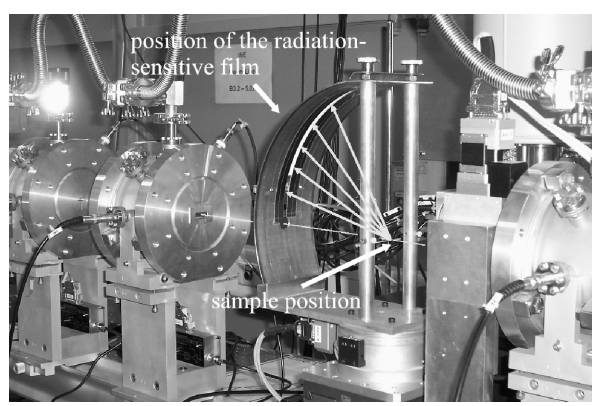

FIGURE 3. Setup for combined XAFS/XRD measurements.

As proof of feasibility, the Debye-Scherrer diffractogram of a powdered quartz sample has been

measured with an excitation energy corresponding to the Cu $K_{\alpha 1}$ energy of approximately 8.04 keV. The diffraction pattern shown in Figure 4 matches well the diagrams recorded with a conventional setup and a calculated one without splitting of the of the reflexes from the $K_{\alpha 1}$ and $K_{\alpha 2}$ in the conventional source.

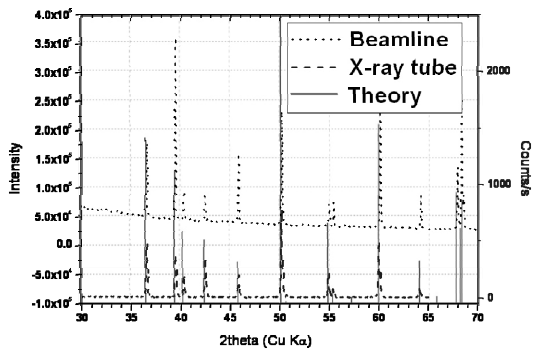

FIGURE 4. Comparison of a diffractogram of a powdered quartz sample extracted from the read-out of the films with a conventionally measured and a calculated diffractogram.

In cooperation with the Physikalische Institut, Bonn University, Germany, a setup for measurements down to the phosphorous K edge has been constructed and tested at the INE-Beamline (see Figure 5).

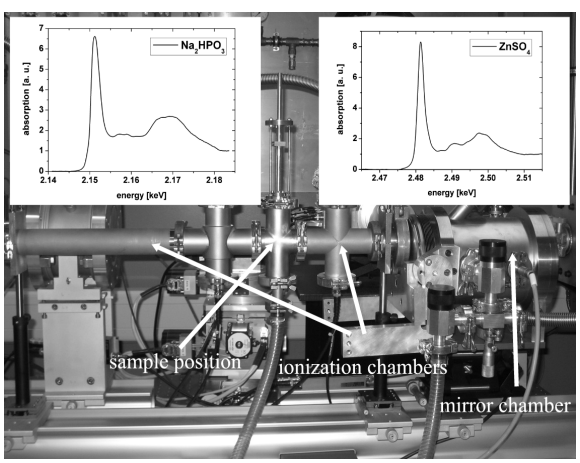

FIGURE 5. Setup for low energy measurements: First measurements of $ZnSO_4$ and Na_2HPO_3 at the sulfur K and phosphorous K edge, respectively, are shown as insets.

The experimental setup is connected to the end of the beamline separated by a 14 μm Kapton® window. It is pumped to low pressure to reduce the loss of incoming photons by absorption in air. An additional mirror chamber is installed prior to the first ionization chamber. It contains a fused silica crystal, which can be position into the beam close to the angle of total external reflection to suppress contributions from higher harmonics.

A micro-focus option is being currently designed for an upgrade to offer spatially resolved measurements. This will allow chemical state imaging (μ-XAFS), elemental mapping (μ-XRF), and identification of phases (μ-XRD). The combination of X-ray methods with other techniques, e.g., laser based methods, will be possible at the beamline. First test using a planar compound refractive lens as one dimensional focusing optic (i.e. virtual slit) are planned for the end of 2006.

BEAMTIME REQUEST

The INE-Beamline and INE active laboratories are one of the pooled facilities of the EU European Network of Excellence for Actinide Science (ACTINET) and access to the beamline is possible via this avenue. A portion of annual beamtime at the INE-Beamline (30%) is available via the standard ANKA facility proposal procedure (for detailed information see [5]). ACTINET users are prompted to also submit a proposal to the ANKA facility for their beamtime. Experiments are also possible through cooperation with INE. INE-Beamline scientists are to be contacted prior to proposal submittal to ensure feasibility of any experiment involving radioactivity (contact persons can be found in [6]). INE provides a radiation protection officer during active measurements, who is responsible for all radioactive substances.

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