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New developments at 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 operated by the Institut für Nukleare Entsorgung (INE) at the Forschungszentrum Karlsruhe. 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. One great advantage of the beamline is its close proximity to INE's active laboratories with its equipment for manipulation of actinide materials and state-of-the-art spectroscopic, analytical, and microscopic instrumentation. This constellation is unique in Europe.

The INE-Beamline is built primarily to serve INE in-house research associated with safe disposal of high level nuclear waste such as actinide speciation or coordination-, redox-, and geo-chemistry of actinides. A wide energy range from around 2.1 keV to 25 keV covering the K-edges from P to Pd and the L3, L2, and L1 edges for actinides from Th to Cm can be used. The INE-Beamline is optimized for X-ray absorption spectroscopy techniques (XANES/EXAFS), but x-ray fluorescence (XRF) analysis and powder diffraction (XRD) are also possible, as well as surface sensitive measurements in grazing incidence geometry (GI-XAFS). Upgrades of instrumentation and extension of experimental capabilities at the INE-Beamline are driven by user needs. Two of the recent upgrades are presented: 1) installation of a microfocus option for spatially resolved studies (μ -XRF, μ -XANES, μ -XRD) and investigations of small volumes (e.g., heterogeneous natural samples and diamond anvil high pressure cells); 2) construction, and commissioning of a high resolution x-ray emission spectrometer (HRXES); 3) availability of an electrochemical cell for investigation of redox sensitive systems.

1. Introduction

The Institut für Nukleare Entsorgung (INE) at the Forschungszentrum Karlsruhe (FZK) operates a beamline dedicated to actinide research at the synchrotron source Ångströmquelle Karlsruhe (ANKA). Official operation of the INE-Beamline began on October 1st, 2005. The symbiosis of the INE-Beamline at the ANKA accelerator and INE radiochemical laboratories, which are both located within the same FZK site, is unique in Europe. Samples can be prepared and then characterized and analyzed before and following synchrotron-based investigations at the INE-Beamline using the state-of-the-art spectroscopic, analytical, microscopic, and structural methods available at INE laboratories.

The major research and development activities at INE concern high level nuclear waste disposal safety. Speciation investigations of the actinide elements are a major topic, as they comprise the largest contribution to the long-term radiotoxicity of high level nuclear waste. The beamline is also

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available for external users for a wide variety of physics, chemistry and biology experiments on radioactive material, not possible at any facility in Germany and only few world-wide. Therefore, the major goal in INE-Beamline design and development is to provide a multi-purpose station, where a number of methods are possible, including standard, surface sensitive, and spatially resolved techniques. 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). Scientific investigations related to nuclear waste disposal safety often involve reactions of actinides in the hydro- and geosphere at interfaces and junctions. For this reason emphasis at the INE-Beamline is placed on surface sensitive techniques based on grazing incidence (GI) geometry such as GI-XAFS. In addition, spatial resolution in the micrometer range is desirable to investigate heterogeneous natural systems.

2. Beamline design and recent developments

2.1. Characteristics overview

The schematic drawing of the INE-Beamline, as well as details of the optics design, are published elsewhere [1,2]. A wide energy range from around 2.1 keV to 25 keV covering the K-edges from P to Pd and the L3, L2, and L1 edges for actinides from Th to Cm can be used. Experiments on radioactive samples with activities up to 10^6 times the limit of exemption (i.e., 1GBq Np-237, 10GBq Pu-242) inside a safe and flexible double containment concept are possible.

The complete list of methods, sample environments and detectors available is given on the ANKA website [3].

2.2. Recent developments

2.2.1. Microfocus optics. A microfocus beam delivered by a polycapillary half-lens can be used at the INE-Beamline in scanning mode to perform spatially resolved μ -XRF and μ -XANES investigations. An application example is presented elsewhere in these proceedings [4]. Here we report on first results using a microfocus beam, produced by a single-bounce capillary, specially designed to adapt to the high horizontal divergence of the beam (IFG Berlin, Germany), for performing combined μ -XRD and μ -XAFS. In this experiment 243-Am phase transitions from the AmI phase (dhcp), to AmII (fcc), to AmIII (face centered orthorhombic *Fddd*) to the highest known pressure AmIV phase (*Pnma*) at nominal pressure up to 25 GPa [5] in a high pressure diamond anvil cell (DAC) is studied. The pressure inside the DAC is measured by in situ monitoring of wavelength changes in the fluorescence emission line of a ruby crystal. At discrete pressures an XRD pattern is recorded to validate the Am phase and a Am L3 edge XAFS spectrum recorded in transmission geometry. The experimental setup is shown in figure 1. The capillary is mounted on a remote controlled hexapod positioner for alignment. The beam size obtained with the capillary at 18.5 keV was 35 μ m diameter at a focal length of 3mm. This allows easy beam

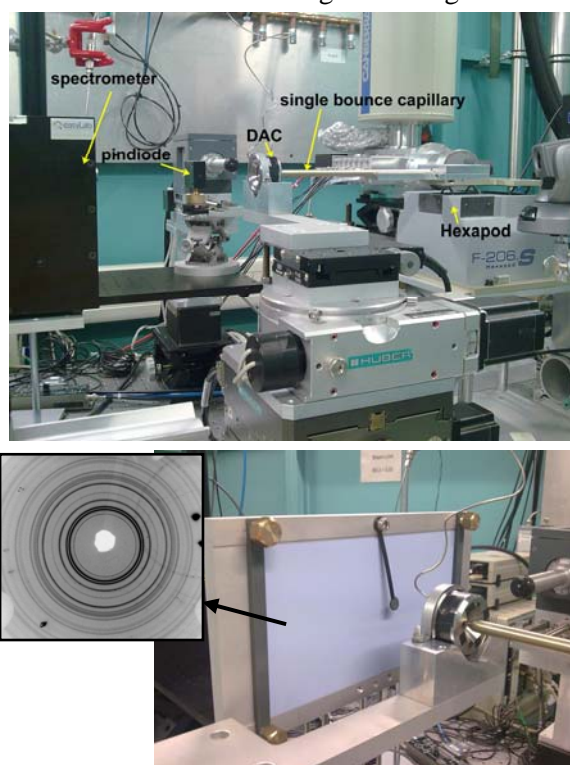


Figure 1 Photographs of the setup for μ -XAS (top) and μ -XRD (bottom) measurements in the DAC experiment. The inset depicts an example of XRD ring pattern obtained for the AmIII phase.

entrance into the diamond aperture (200 μ m). The XRD patterns are recorded using radiation sensitive image plates with a high dynamic range. An indium foil cut-out mounted on a plastic rod is used as beamstop. The image plates are read out using a scanner (Packard BioScience, Dreieich, Germany) and re-used after exposure to UV-light. The transmitted intensity (I_1) through the DAC is recorded using a pin diode. The incident intensity (I_0) is measured by moving the Am in the diamond out of the beam path using the motorized positioners and recording counts with the pin diode. The absorption spectrum is then according to the Beer-Lambert law $\ln(I_0/I_1)$.

2.2.2. HRXES. INE has built a Johann spectrometer for performing high resolution x-ray emission spectroscopy and resonant inelastic scattering investigations (HRXES and RIXS) [6]. It will be available for use at the INE-Beamline for the investigation of radioactive materials. The spectrometer is of a compact, modular design, optimized for attaining a wide range of energies with a dynamically bent analyzer crystal (figure 2). The principle of the spectrometer is fluorescence emitted from the sample is energy dispersed by a secondary dynamically bent monochromator crystal and focused onto a position sensitive detector. A positioning device is used to change the Bragg angle of the crystal mounted in the crystal bender.

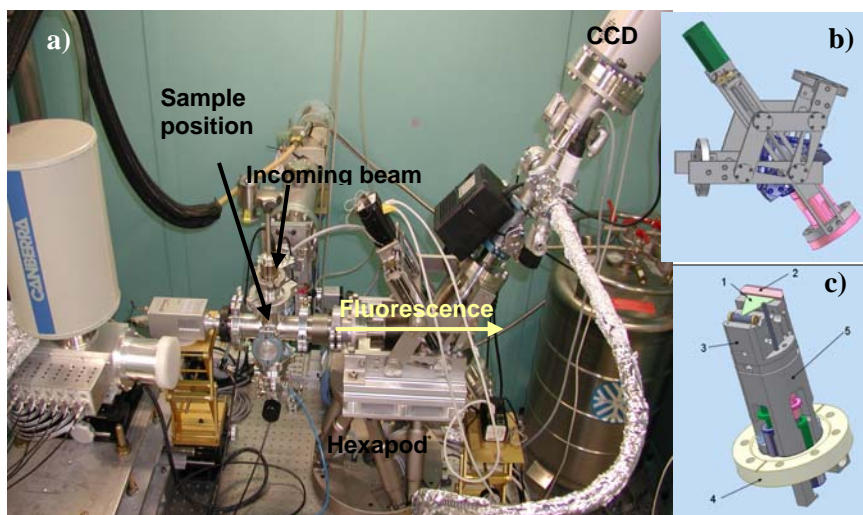


Figure 2. (a) Photograph of the spectrometer in use at the INE-Beamline. (b) 3 dimensional drawing of the Bragg arm. The crystal bender is indicated in pink (c) Drawing of the crystal bender showing the crystal (1) held by a clamp (2). (3) indicates the plate covering the shaft used to apply force onto the crystal tip, thereby bending it, (5) the corpus and (4) the flange attached to the main plate of the Bragg device.

2.2.3. Spectro-electrochemistry. An electrochemical cell for in situ XAFS investigation of redox sensitive systems was developed and following successful test with inactive (Se), used for investigations of the redox chemistry of radioactive ^{237}Np . Figure 3a shows a photograph of the cell installed at the beamline during the first active measurements. The Np L3-edge XANES of pure ^{237}Np species prepared electrochemically in the same solution are shown in figure 3b. Technical design and further details are given in [7].

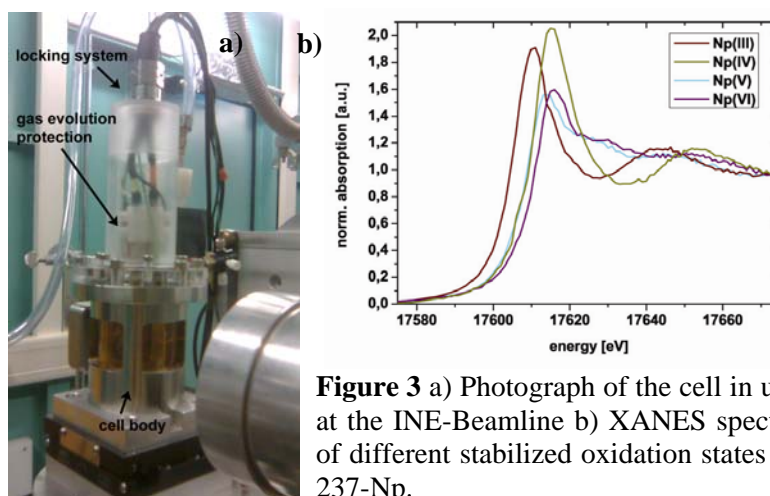


Figure 3 a) Photograph of the cell in use at the INE-Beamline b) XANES spectra of different stabilized oxidation states of ^{237}Np .

3. Future Upgrades

Instrumentation development and upgrades planned for the near future at the INE-Beamline are related to optimization of the μ -focus options at the beamline, improvements of the HRXES spectrometer, and designing new sample cells for special user requirements. Specifically are planned:

- Eliminate horizontal beam drift during DCM crystal rotation (i.e., large energy scans) with high precision actuators tuning the second DCM crystal roll position.
- Extend the microfocus capabilities to confocal irradiation-detection geometry for additional resolution in the third (depth) dimension.
- First active measurements with the HRXES spectrometer, extension of attainable emission energy range and upgrade to a new position sensitive detector.
- Combine the electrochemical cell with UV-Vis spectroscopy signal detection for a secondary in-situ verification of oxidation states.
- Adapt and test a liquid cell for high temperature, high pressure investigations.

4. Beamtime request

The INE-Beamline and INE active laboratories are among the pooled facilities of the European Network of Excellence for Actinide Science (ACTINET). Access to the beamline is possible via this avenue. 30% of annual beamtime at the INE-Beamline is available via the standard ANKA facility proposal procedure (for details see <http://ankaweb.fzk.de>). 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 [8] prior to proposal submission to ensure feasibility of any experiment involving radioactivity. INE provides a radiation protection officer during active measurements, who is responsible for safety of all radioactive substances.

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