



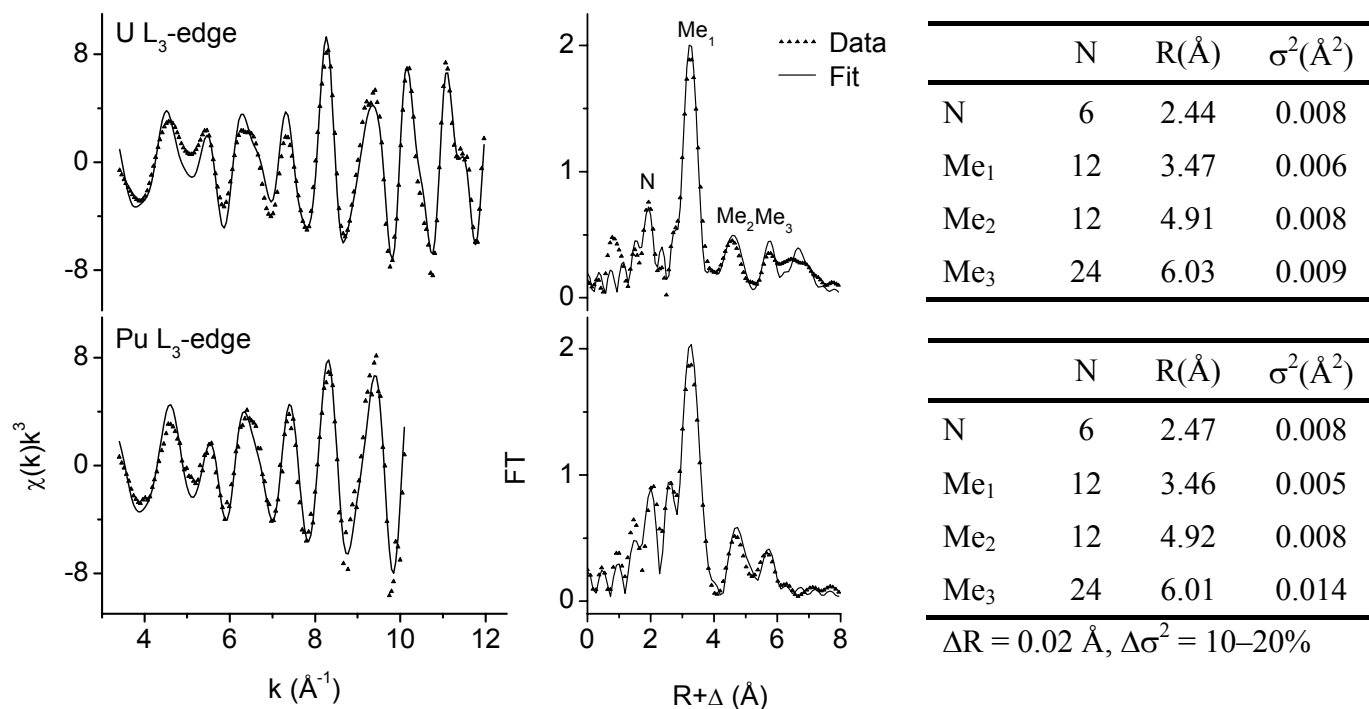
	Experiment title: Local atomic structure in (Zr,Pu,Am)N	Experiment number: CH-2192
Beamline: BM20	Date of experiment: from: 15.11.2006 to: 18.11.2006	Date of report: 07.07.2007
Shifts: 9	Local contact(s): Christoph Hennig	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): M. Walter*, J. Somers, C. Nästren*, D. Bregiroux*, S. Stohr*, A. Rossberg*, Harald Funke*, C. Hennig*, A. Scheinost* European Commission, Joint Reserch Centre, Institute for Transuranium Elements P.O. Box 2340, 76125 Karlsruhe, Germany		

Report:

The long-term radiotoxicity of minor actinides (MA: Am, Cm, Np) can be reduced by their transmutation in fast flux reactors or accelerator driven systems. For this application nitride fuels are of interest due to their thermal properties. High MA transmutation efficiency can be reached by MA incorporation in ZrN, forming (Zr,MA)N. Alternatively, MA can be incorporated in UN or (U,Pu)N to give (U,Pu,MA)N, a fuel foreseen for gas-cooled fast reactors within the Generation IV initiative. ZrN and the actinide mononitrides form solid solutions of the rock salt structure. For the (Zr,U)N solid solution it was shown that the metal-nitrogen bond distance can differ from the bulk crystallographic structure [1], but for (Zr,MA)N and (U,MA)N no such information are available yet. As the Zr K-edge (17998 eV) and the Pu L₃-edge (18057 eV) are close to each other, the investigation of the local structure by EXAFS analysis has been limited to the (U,Pu)N based fuel.

(U_{0.82}Pu_{0.18})N and (U_{0.8}Pu_{0.18}Am_{0.02})N fuels were produced by gel-supported precipitation, Am infiltration and carbothermic reduction. Solutions of U and Pu were mixed and polymers were added to increase the viscosity. In addition, carbon black was added in a slight excess (C/(U+Pu)=2.4) to ensure the presence of sufficient carbon for the carbothermic reduction. This solution was then atomized and the droplets were collected in an ammonia bath. After washing and drying, the resulting beads were calcined under Ar/H₂. The batch was then divided, and half of the material was infiltrated with ²⁴¹Am nitrate solution. The carbothermic reduction was performed at 1400°C (12 h in N₂) and the remaining excess carbon was subsequently removed under N₂/H₂ at 1400°C (18h). The nitride beads were then pressed into pellets and sintered at 1650 °C in N₂/H₂ atmosphere (30 h). X-ray diffraction analysis indicates the presence of single nitride phases of (U_{0.82}Pu_{0.18})N (4.8906 Å) and (U_{0.80}Pu_{0.18}Am_{0.02})N (4.8919 Å).

For XAS measurements, the nitrides were milled, mixed with carbon and pressed in disks. The Am containing sample was placed between plexiglass windows of a screwed stainless steel/brass cuvette, developed for highly radioactive samples. The XAS measurements on the U L₃-edge was performed in transmission mode, whereas the Pu and Am L₃-edge XAS were recorded in fluorescence mode using a 13 element HPGe detector. The evaluation of the XAS data was performed according to standard procedures using EXAFSPAK. A detailed descriptions of the fitting procedure is given in Ref. [1].



The figure shows the k^3 -weighted U and Pu L₃-edge EXAFS spectra and FT's of (U_{0.80}Pu_{0.18}Am_{0.02})N together with the derived structural parameters. The corresponding spectra of (U_{0.82}Pu_{0.18})N are similar. The measurement of the Am L₃-edge XAS was difficult, since the energy of the Pu and Am fluorescence lines are too close to be discriminated by the multiple channel analyser during measurement. The recorded Am L₃-edge XAS data are not of sufficient quality and furthermore contain information of the Pu L₃-edge as the counting rate of selected fluorescence channels was Pu:Am 6:1.

The interatomic distances obtained by EXAFS analysis are in general agreement with the distances calculated using the bulk crystal structure. There are no significant differences for (U_{0.82}Pu_{0.18})N and (U_{0.80}Pu_{0.18}Am_{0.02})N. For both compounds, the derived Pu–N distance is higher than the U–N distance and the values calculated from the lattice parameters.

[1] Walter M., Somers J., Fernández-Carretero A., Rothe J., Local atomic structure in (Zr_{1-x}U_x)N. J. Nucl. Mater., in press doi:10.1016/j.jnucmat.2007.05.027