

## Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.**

Once completed, the report should be submitted electronically to the User Office via the User Portal:

<https://www.esrf.fr/misapps/SMISWebClient/protected/welcome.do>

### ***Reports supporting requests for additional beam time***

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

### ***Reports on experiments relating to long term projects***

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

### ***Published papers***

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.


### **Deadlines for submission of Experimental Reports**

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

### **Instructions for preparing your Report**

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.



	<b>Experiment title:</b> Thermal recovery of self-irradiation induced defects	<b>Experiment number:</b>
<b>Beamline:</b> BM20	<b>Date of experiment:</b> from: 09.06.2015 to: 14.06.2015	<b>Date of report:</b> 24.01.2017  <i>Received at ESRF:</i>
<b>Shifts:</b> 12	<b>Local contact(s):</b> A. Scheinost	
<b>Names and affiliations of applicants</b> (* indicates experimentalists): D. Prieur <sup>a,*</sup> , R. C. Belin <sup>b</sup> , J.-F. Vigier <sup>a</sup> , P. Martin <sup>c</sup> <sup>a</sup> European Commission, Joint Research Centre (JRC), Postfach 2340, 76125 Karlsruhe, Germany <sup>b</sup> CEA, DEN, DEC, SPUA, LMPC, F-13108 Saint-Paul-lez Durance, France <sup>c</sup> CEA, DEN, DEC/SESC/LLCC – 13108 Saint-Paul-Lez-Durance cedex, France.		

## Report:

The use of Minor Actinides bearing MOX as a future nuclear fuel cycle requires a full and accurate assessment of their thermal properties. This is indeed mandatory for fuel pin design to meet safety criteria in the reactor core. Furthermore, these fundamental data are essential for the prediction of the in-pile fuel behaviour with fuel performance codes. The charge distribution of these materials must be then properly assessed as it affects the thermal properties and hence the nuclear safety.

XANES measurements have been performed on approximately 1 mg of powdered sample mixed with 20 mg of boron nitride (BN). The data have been collected at the Rossendorf beamline (ROBL) of the European Synchrotron Radiation Facility (ESRF) situated in Grenoble (France) <sup>1</sup>. To determine the oxidation states of the cations, XANES spectra at L<sub>III</sub> edge were compared to data collected on reference compounds on the same beamline using the same experimental setup. For uranium, the reference compounds were UO<sub>2.00</sub> and U<sub>4</sub>O<sub>9-δ</sub>. Their structures were confirmed using both XRD and neutron diffraction <sup>2</sup>. For americium, Am<sup>+IV</sup>O<sub>2</sub> <sup>3</sup> and a mixed oxalate U<sup>+IV/V</sup><sub>0.85</sub>Am<sup>+III</sup><sub>0.15</sub>O<sub>2</sub> <sup>4</sup> were used. For plutonium, the reference materials were Pu<sup>+IV</sup>O<sub>2.00</sub> <sup>5</sup> and Pu<sup>+III</sup> oxalate. Np<sup>+IV</sup>O<sub>2.00</sub> <sup>6</sup> was used as a reference for Np L<sub>III</sub>. The molar fractions of the cations were assessed using a linear combination of reference compounds from the normalized μ(E) spectra. Experimental data were fitted between -20 eV and +30 eV of the white line energy maximum.

XANES spectra have been recorded for U, Pu, Np and Am. From the analysis of XANES spectra collected at  $L_{III}$  edge, one can conclude that U, Np and Pu are strictly tetravalent (the data are not presented) while Am is clearly trivalent (cf. Figure 1). Due to the very low Am content in the Np-MOX, the XANES could not be collected. For this reason, it was assumed, by analogy with the Am-MOX compound, that Am is also trivalent in Np-MOX as the same annealing conditions had been used to remove the influence of damage from the stored samples. One can then determine that the global O/M ratios of Np-MOX and Am-MOX are equal to 1.99 (1) and 1.98 (1), respectively.

The thermal properties of these Np- and Am-MOX solid solution materials were investigated<sup>7</sup>. Their linear thermal expansion, determined using high temperature X-ray diffraction from room temperature to 1973 K showed no significant difference between the Np and the Am doped MOX. The thermal conductivity of the Am-MOX is about 10% higher than that of Np-MOX. The melting temperatures of Np-MOX and Am-MOX, measured using a laser heating self crucible arrangement were  $3020 \pm 30$  K and  $3005 \pm 30$  K, respectively.

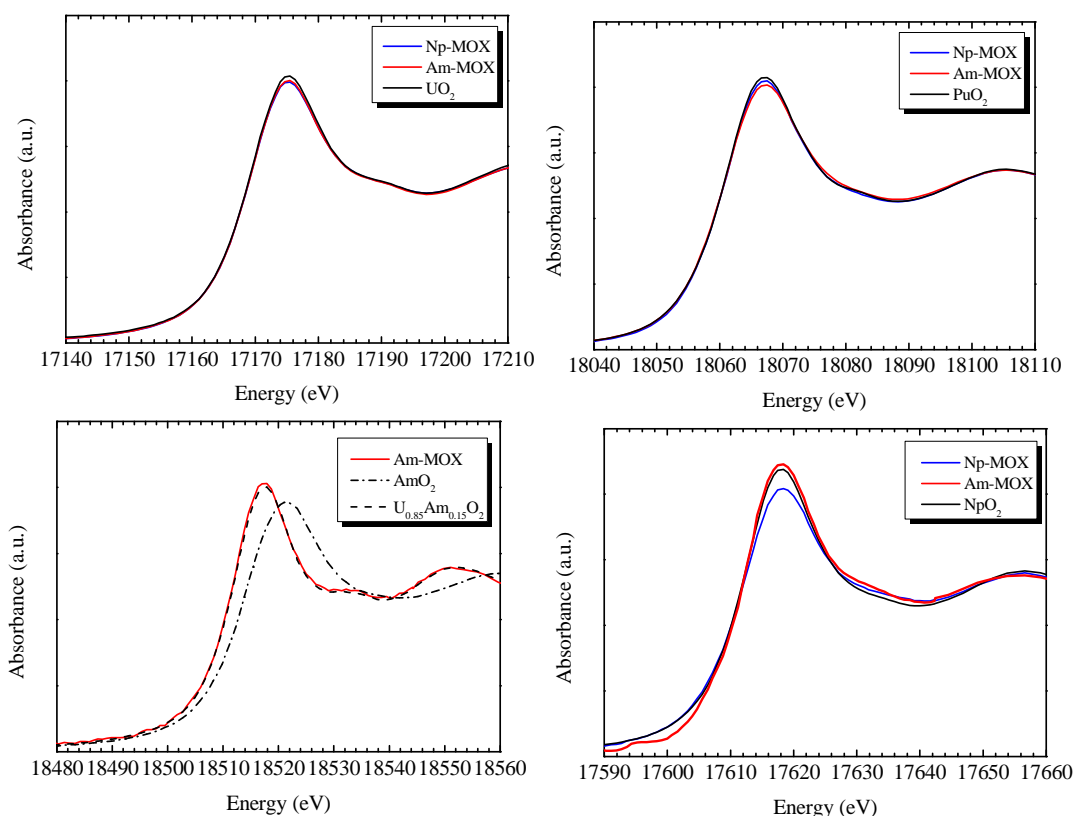


Figure 1:  $L_{III}$  XANES spectra of Np- and Am-MOX in comparison with the reference materials.

## References:

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2. Desgranges, L.; Baldinozzi, G.; Rousseau, G.; Niepce, J.; Calvarin, G., *Inorganic Chemistry* 2009, 48, 7585-7592.
3. Belin, R. C.; Martin, P. M.; Valenza, P. J.; Scheinost, A. C., *Inorganic Chemistry* 2009, 48, 5376-5381.

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5. Prieur, D.; Carvajal-Nunez, U.; Vitova, T.; Somers, J., *European Journal of Inorganic Chemistry* 2013, 2013, 1518–1524.
6. Prieur, D.; Vigier, J.-F.; Wiss, T.; Janssen, A.; Rothe, J.; Cambriani, A.; Somers, J., *Journal of Solid State Chemistry* 2014, 212, 7-12.
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