HZDR HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF ROBL-CRG	**Experiment title:** ***Investigation of the complex stoichiometry of [Am(CyMe$_4$BTPhen)$_x$] complexes in ionic liquid***	**Experiment number**: ***37063 20-01-751***
Beamline: BM 20	**Date of experiment**: from: 2015-05-18 to: 2015-05-19	**Date of report**: 2017-02-02
Shifts: 3	**Local contact(s)**: Andreas Scheinost, André Rossberg	*Received at ROBL:*

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Report:

Ionic liquids have very interesting properties for a potential application in solvent extraction for the separation of actinides from used nuclear fuel, e.g. the low volatility and high flash point. However, the extraction mechanism in ionic liquids differs from the extraction mechanism in conventional diluents and furthermore is even depending on the initial nitric acid concentration. Therefore, investigations towards a clarification of the extraction mechanism are extremely important. One important aspect is the complex stoichiometry of the extracted complexes in the ionic liquid.
The extractant CyMe$_4$BTPhen is currently under investigation in European collaborative projects and it was shown to be highly selective for trivalent actinides over lanthanides in conventional diluents. Therefore, the extraction of Am(III) was also studied in ionic liquids. A "boomerang" shaped extraction behaviour was found as a function of the nitric acid concentration, implying a competition of different extraction mechanisms at the different nitric acid concentrations. Further solvent extraction studies have not been able to clarify the complex stoichiometry at the different nitric acid concentrations. Therefore, EXAFS measurements were conducted to answer these questions.

Three samples of Am(III) extracted from different nitric acid concentrations into an ionic liquid phase containing $CyMe_4BTPhen$ were prepared. The ionic liquid phases were separated and used to produce the EXAFS samples. These were shipped to ESRF, Grenoble, and measured at the ROBL beamline.

Sample preparation using higher quantities of $^{243}Am(III)$ worked as foreseen and the EXAFS samples were prepared without difficulties.
The EXAFS measurements themselves were successful and nearly the whole allocated measurement time could be used. In total 32 scans were collected from the samples as shown in Figure 1. The noise to signal ratio (normalized to the edge jump) was acceptable for all samples.

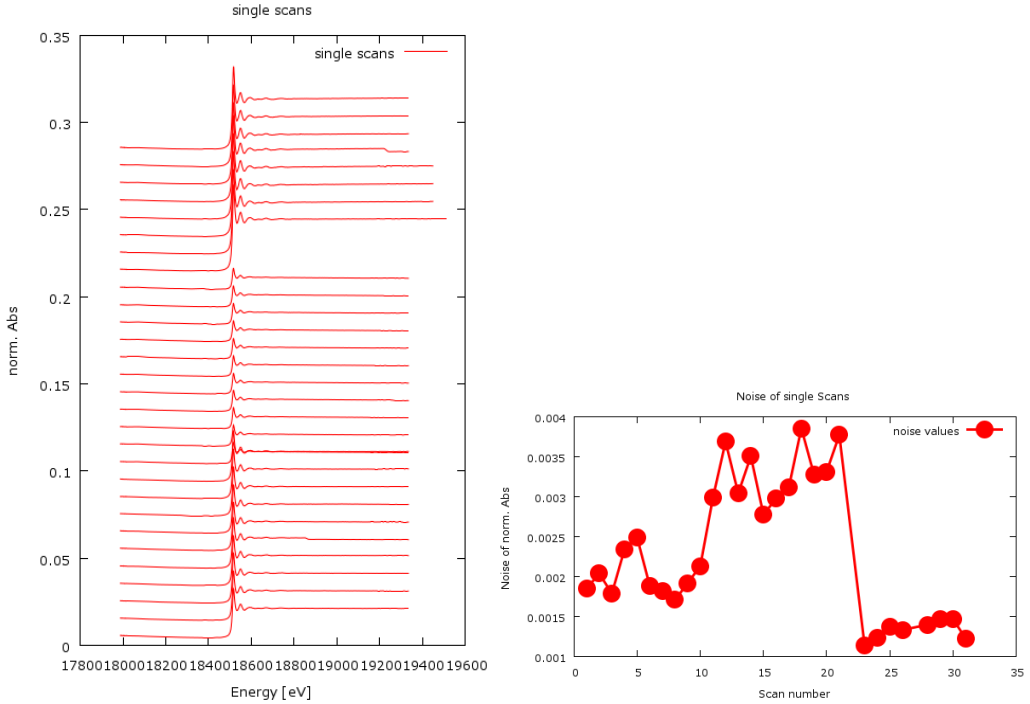

Figure 1. Overview of the EXAFS scans (left) and normalized noise (right).

The data reduction and interpretation is still going on, as different structural models have to be evaluated.

In a first attempt of fitting the EXAFS signal, a structural model as indicated by Figure 2 was used with a first coordination shell of 2 nitrogen atoms, a second shell with another 2 nitrogen atoms, and a third shell with 2 carbon atoms.

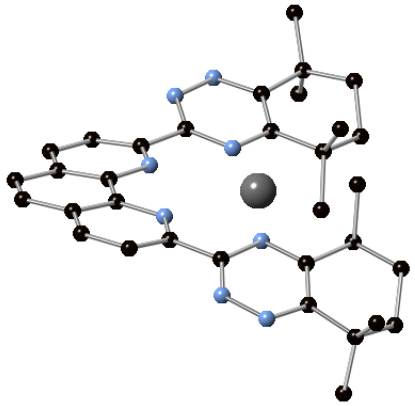

Figure 2. Structural model. Black = C, Blue = N, Grey = Am.

The best fit obtained so far with this model is shown in Figure 3. The data is well fitted for radial distances up to ca. 3 Å, but at larger distances deviations are observed.

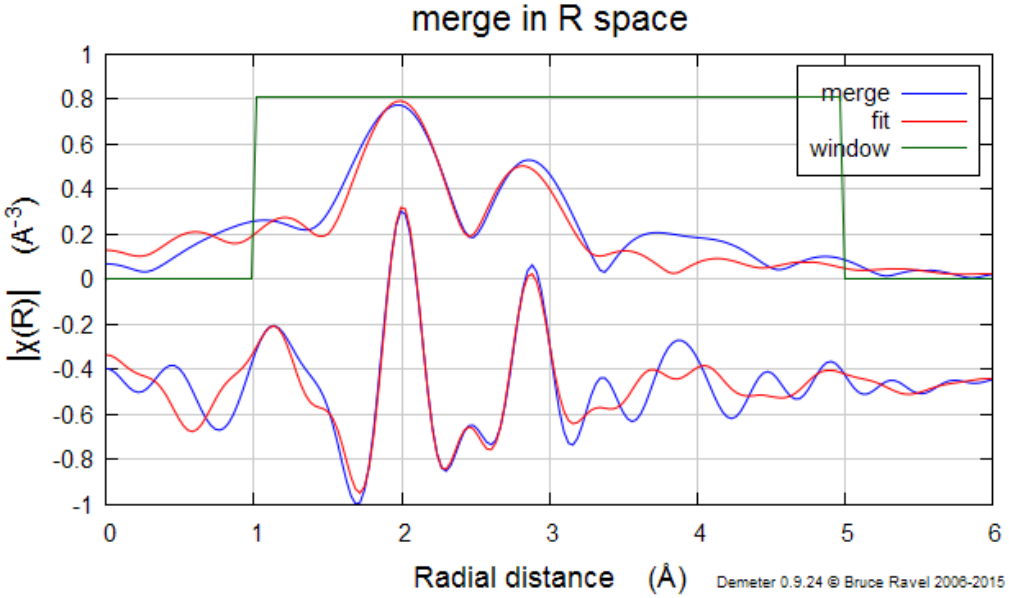

Figure 3. Data fitting.

A collaborative publication is foreseen after the data has been interpreted and a structural model has been implemented. The EXAFS data will be compared to results from solvent extraction as well as theoretical considerations.