ROBL-CRG	**Experiment title:** **Interaction of actinides with predominant bacteria isolated at nuclear waste repositories using X-ray absorption spectroscopy**	**Experiment number**: **20-01-611**
Beamline: BM 20	**Date of experiment**: From: 15.06.2005 to: 17.06.2005	**Date of report**: 09.03.2006
Shifts: 6	**Local contact(s):** Ch. Hennig	*Received at ROBL:*
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Report:

X-ray absorption spectroscopy (XAS) is a powerful tool to characterize the speciation of heavy metals in a broad range of systems. The two parts, XANES and EXAFS, are giving in combination element specific information concerning the oxidation state and local structure of an absorbing metal. In this study, XAS was used for a detailed characterization of the microbial processes taking place in the system Pu – *D. äspöensis*. This strain predominate the indigenous SRB population at the Äspö aquifer system in Sweden.

Experimental: The spectrum from the Pu(VI) reference sample (0.05 M in 1 M HNO_3) was taken from earlier measurements. The Pu(IV) reference sample (0.08 M in 1 M $HClO_4$) was prepared as described in /1/. 1 mL of the bacterial suspension in 0.9% NaCl was incubated with 5 mL of ^{242}Pu, 127 mg/L, at pH 5. After shaking the samples for 96 h under N_2 atmosphere, the biomass was separated by centrifugation, washed with 0.9% NaCl, and sealed in a polyethylene cuvette. The bacterial sample was measured as wet past.

Results: The low intensity of the white line (WL) and the feature near 18080 eV are indicating dominating plutonyl species in the blank, the supernatant and in the Pu(VI) reference (data not shown). This shoulder above the WL, results from multiple scattering processes of the photo electron wave between Pu and the two axial oxygens. The Pu accumulated by *D. äspöensis* occurs in the tetravalent oxidation state. The characteristic changes in the absorption spectra were used to determine the relative concentrations of the Pu species by applying the iterative transformation factor analysis (FA).

The Pu in the blank is surrounded by two close axial oxygens at 1.77 Å and appr. 5 equatorial oxygens at 2.42 Å (see Fig. 1).

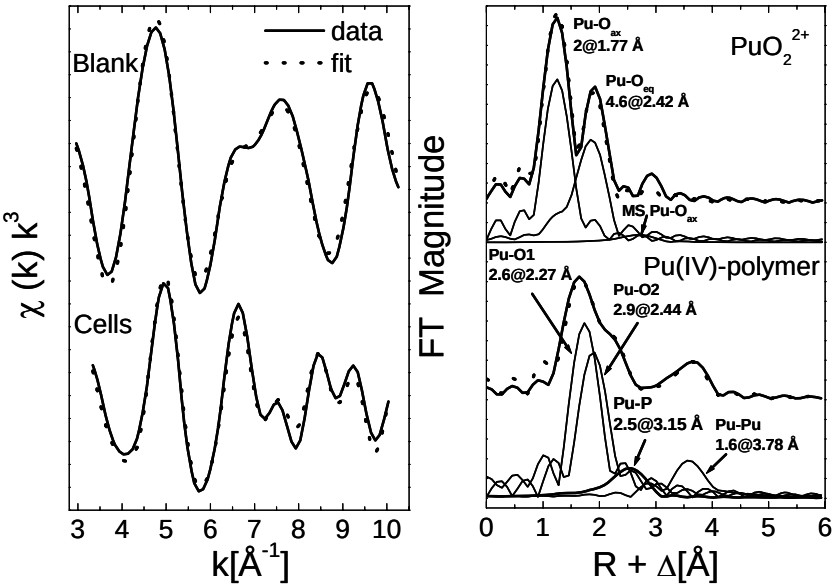

Fig. 1: Pu L_{III}-edge k^3-weighted EXAFS spectra (left) and the corresponding Fourier transforms (right) and the theoretical fits (dotted line).

Unfortunately, we cannot present structural parameter of Pu in the supernatant solution after separating the cells. Changes in the XANES from scan to scan showed that the sample was not stable in the synchrotron beam. The EXAFS oscillation of the cell-bound Pu show close similarities to the spectra of colloidal Pu(IV) species published in /2/. The presence of an additional symmetric EXAFS oscillation giving a FT peak at R+Δ of app. 2.8 Å gives evidence for an interaction of the Pu(IV)-polymers with light atoms of the biomass. The best fit result could be obtained using P which points to an interaction with organic phosphate groups of the cell membrane structure as postulated for Cm(III) in /3/. In conclusion, the XAS investigation of the Pu - *D. äspöensis* system confirmed the results of the solvent extraction experiments /4/. The cell-bound Pu exists as Pu(IV)-polymers which are interacting with the biomass.

Acknowledgements. This work is supported by the BMWi (no. 02E9491). We thank Ch. Den Auwer (CEA Marcoule, France) for providing us with the Pu(IV) reference sample.

Reference

/1/ Charrin, N. et al. (2000) Radiochim. Acta 88, 25-31.
/2/ Rothe, J. et al. (2004) Inorg. Chem. 43, 4708-4718.
/3/ Moll, H. et al. (2004) Environ. Sci. Technol. 38, 1455-1459.
/4/ Moll, H. et al. (2006) submitted to Radiochimica Acta.