	Experiment title: Speciation of americium(III) by illite under Boom Clay conditions: Influence of dissolved organic carbon	**Experiment number**: EV-125
Beamline:	**Date of experiment**: from:18-Jul-15 to: 21-Jul-15	**Date of report**: 10-09-2015
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Report:

The speciation of Americium on Illite (www.catclay.org) has been investigated under Boom Clay conditions. Influence of DOC at pH 6 has been investigated in different background electrolytes (0.1 M $NaClO_4$, 15 mM $NaHCO_3$ and in situ pore water of Boom Clay) under anoxic conditions (see table 1). The most important parameters of the investigated in situ pore water can be found in De Craen et al. [1]

Table 1. Summary of the EXAFS samples

Sample	Preparation
1	[Am]: 4. 10^{-5} M, in 0.1 M $NaClO_4$ on illite
2	[Am]: 4. 10^{-5} M in 15 mM $NaHCO_3$ on illite
3	[Am]: 8. 10^{-5} M in EGBS (HA-FA:120 mg /L) on illite
4	[Am]: 8. 10^{-5} M in F8 (HA-FA :132 mg /L) on illite
5	[Am]: 8. 10^{-5} in SPRING (FA: 60 mg /L) on illite
6	[Am]: 8. 10^{-5} in 15 mM $NaHCO_3$ + BC-HA :120 mg /L

The EXAFS analysis was performed with the software packages EXAFSPAK [2] and FEFF8.20 [3]. The scattering phases and amplitudes were calculated using the crystal structures of $La_2(CO_3)_3(H_2O)_8$ [4] and $Eu_{0.83}Fe^{+2}_{0.5}Fe^{+3}_{1.5}Al_{10}O_{19}$ [5], where La and Eu were replaced by Am to model possible Am-O, Am-C, Am-Si/Al , Am-Fe and Am-Am interactions at the illite surface.

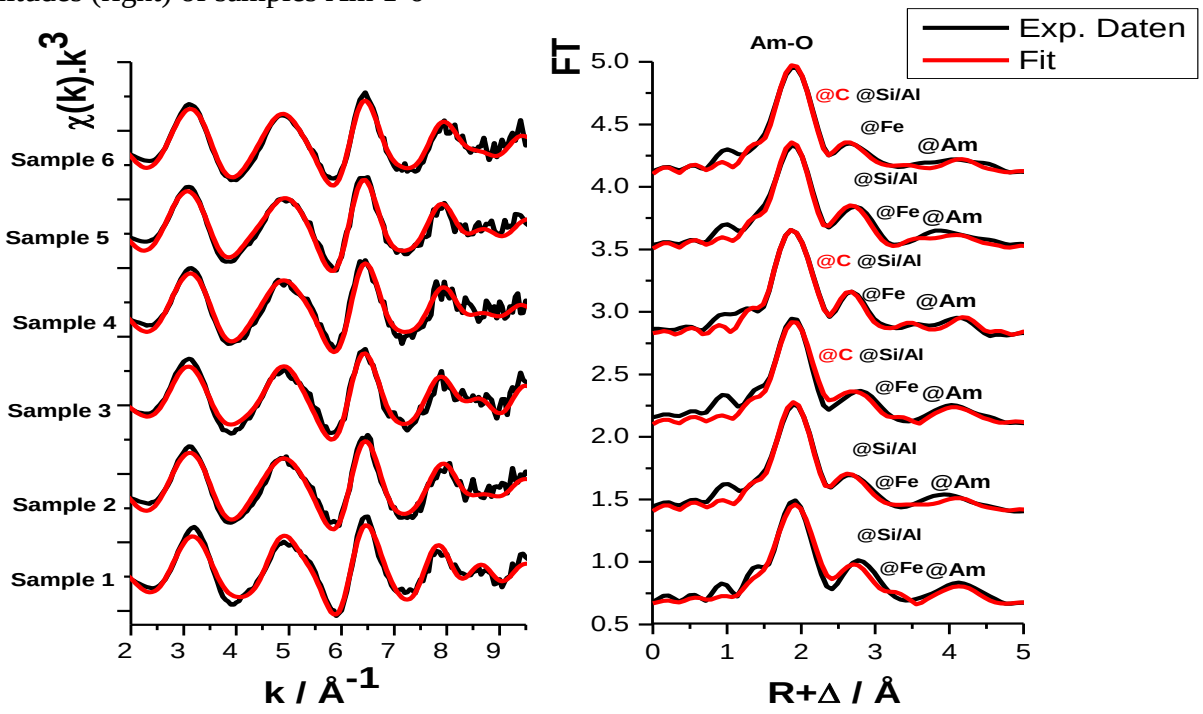
Table 1 shows a summary of the samples which were measured in this EXAFS study. The second sample was prepared in order to improve the understanding of the influence of high carbonate concentrations as expected in Boom Clay pore water on the Am speciation on illite. The other samples (3-5) were prepared with in situ pore water as background electrolyte in order to better understand the influence of DOC from Boom Clay pore water on the Am speciation on illite that originates from the MORPHEUS piezometer (Mobile Reganic matter and Pore water extraction in the Hades Experimental Underground Site). The FT shows peaks associated with Am–O, Am-C, Am–Si/Al, Am-Fe and

Am–Am interactions in these samples. The first shell is related to 8–9 oxygen atoms at a distance of 2.45 Å which is in good agreement with literature data for the Am(III) aquo ion and other trivalent actinides [6]. All samples have show a Al/Si shell at 3.17 – 3.19 Å. The detection of Am-Al/Si interactions in all samples is indicative for inner-sphere sorption of Am on the illite surface. The FT magnitude of sample 3-4-6 shows an additional peak due to Am-C coordination (2.7 Å). Runde at al. reported that the FT peaks located at about 4.0 Å in the spectra of $Eu(OH)_3$ and $EuOHCO_3$ are due to Eu–Eu interactions [7]. Also in the present work Am-Am interaction is found at a distance of 4.1 Å indicating a precipitation of Am(III) at the surface or the formation of polynuclear species (e.g. colloids).The structural parameters derived from the EXAFS fits are summarized in Tab. 2

Table 2: Fit parameters for the raw k^3-weighted Am L_{III}-edge EXAFS spectra shown in Figure 1

	Am-O			**Am-C**			**Am-Si/Al**			**Am-Fe**			**Am-Am**			**ΔE_0 / eV**	**red. Error**
	N	R / Å	σ^2 / Å^2	N	R / Å	σ^2 / Å^2	N	R / Å	σ^2 / Å^2	N	R / Å	σ^2 / Å^2	N	R / Å	σ^2 / Å^2		
1	9.5	2.46	0.014	-	-	-	4.4	3.19	0.010	1.7	3.55	0.008*	1.9	4.12	0.003	1.94	0.07
2	9.4	2.46	0.013	-	-	-	3.3	3.17	0.007	1.8	3.56	0.008*	2.5	4.10	0.010	0.19	0.14
3	8.9	2.46	0.012	1.9	2.74	0.006	3.3	3.18	0.007	1.6	3.57	0.008*	1.3	4.10	0.002	0.49	0.15
4	8.3	2.46	0.010	2.2	2.75	0.004	2.8	3.18	0.004	1.9	3.58	0.008*	2.5	4.12	0.010	1.64	0.18
5	10.1	2.46	0.015	-	-	-	4.5	3.18	0.009	1.8	3.57	0.008*	1.5	4.10	0.003	-0.74	0.11
6	7.4	2.45	0.009	2.8	2.70	0.005	2.3	3.16	0.004	2	3.57	0.008*	1.7	4.11	0.006	1.27	0.11

Figure 1. Am L_{III}-edge k^3-weighted EXAFS spectra (left) and the corresponding Fourier transform magnitudes (right) of samples Am 1-6



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