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Cm(III)/Eu(III)- Coprecipitation with trioctahedral smectite (hectorite)

H. PIEPER, D. BOSBACH, P.J. PANAK, K. DARDENNE,
J. ROTHE, M.A. DENECKE AND TH. FANGHÄNEL

Institut für Nukleare Entsorgung, Forschungszentrum
Karlsruhe (pieper@ine.fzk.de)

Smectites are common secondary phases forming upon alteration/dissolution of natural Si-rich systems. The trioctahedral smectite, hectorite $\text{Na}_{0.7}[\text{Li}_{0.7}\text{Mg}_{5.3}\text{Si}_8\text{O}_{20}(\text{OH})_4]$, forms also within the alteration layer of corroded nuclear HLW (high level waste) glass [1]. Smectites represent a significant retention potential for the migration of solved pollutants including toxic and radioactive metal ions like actinides.

Due to their structural complexity and chemical variability, sorption of trivalent actinides involves several unique sorption mechanisms: (1) adsorption via inner-sphere and outer-sphere complexation, (2) cation exchange in the interlayer and (3) incorporation into the octahedral sheet of the smectite structure.

We have used a new method to synthesize organo-hectorite nano particles [2]. The synthesis was modulated by exerting Cm(III) and homologues Eu(III) coprecipitated with $\text{Mg}(\text{OH})_2$ as a precursor. The Eu- respectively Cm-containing hectorite was used as a model for the corrosion product hectorite.

Extended X-Ray Absorption Fine Structure Spectroscopy (EXAFS) allowed to probe the local environment around the coprecipitated Eu-cations and to identify the sorption mechanisms. Fit results indicate the presence of a dominant Eu-O interaction at 2.34 Å (5-6 O) and a more distant oxygen shell at 2.42 Å (3 O). The dominant Eu-O interaction can be interpreted as an octahedral O-coordination of the Eu-cation and leads to the hypothesis of trivalent actinides incorporation into the octahedral layer. Time Resolved Laser Fluorescence Spectroscopy (TRLFS) allowed to identify the sorption species. Measurements clearly show, that an unhydrated Eu(III) species is closely associated with the solid phase. Our observations are compatible with the assumption that trace amounts of Eu(III)/Cm(III) occupy an octahedral lattice site. The incorporation of Eu(III)/Cm(III) into an amorphous silica phase, remaining during hectorite synthesis cannot be excluded. Finally, only TRLFS measurements of the Cm-containing hectorite affirmed the incorporation into the octahedral sheet of the smectite structure and allowed to differentiate unambiguously between the Cm-Silica complex and the Cm-species incorporated into the hectorite.

References

- [1] Zwicky et al. (1989) *Mat. Res. Soc. Symp. Proc.* **127**, 129-136
[2] Carrado et al. (1997) *Langmuir* **13**, 2895-2902

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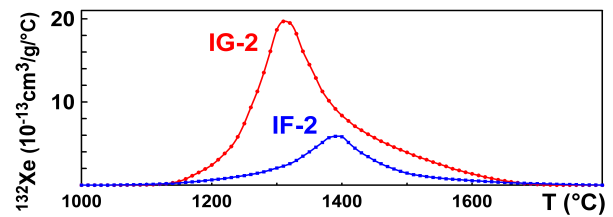
Fissiogenic Xenon in ground zero of Trinity nuclear test

A. MESHK, O. PRAVDIVTSEVA AND C.M. HOHENBERG

McDonnell Center for Space Sciences and Physics
Department of Washington University, 1 Brookings
Drive, Saint Louis, Missouri, USA (am@wustl.edu)

Earlier analyses of the soil from nuclear test site revealed Xe isotopic anomalies which were not completely understood [1]. The site of first nuclear test (at Trinity in the Jornada del Muerto, southeast of Socorro) is now open to the public, except at the very epicenter. Here we report new measurements of these large Xe isotopic anomalies in two trinites, minerals formed at Trinity ground zero, and present a quantitative model for the mechanism by which these anomalies were formed. We demonstrate that sand at the Trinity ground zero experienced differential flash heating > 1300–1400°C for about 1 minute.

Observed anomalous xenon was evidently produced by two processes: (1) neutron induced fission of ^{235}U in the sand and (2) rapid temperature-activated diffusion of immediate fission products (mainly β -active Sb). When the molten sand cools down and solidifies (forming trinitite), effectively terminating Sb diffusion, the accumulation of Xe isotopes in the trinitite is determined by free decay of Sb isotopes retained at that moment. According to our calculations, ~ 1 minute after the neutron burst, the isotopic composition of Sb expected is very close to that observed in Xe from subsequent β -decay. Comparison of the peaks in the Xe temperature release profiles from IG-2 and IF-2 suggests a maximum temperature gradient, subsequent to the event, of about 1°C/m.



Isotopes	^{136}Xe	^{134}Xe	^{132}Xe	^{131}Xe
$^{235}\text{U} + n_{th}$	$\equiv 1$	1.238	0.680	0.456
IF-2	$\equiv 1$	7.6 ± 0.1	59.2 ± 0.8	955 ± 13
IG-2	$\equiv 1$	22.7 ± 0.4	157 ± 3	497 ± 9
[1]	$\equiv 1$	16.2 ± 0.2	86 ± 1	97 ± 2

Xe release profiles and isotopic data from samples IF-2 and IG-2 (~10 m and ~100m from ground zero, respectively).

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References

- [1] Pravdivtseva O. V. et al (1986) Proc. IX All-Union Symposium on Isotopes in Geochemistry, 289, Moscow.