PLUTONIUM SPATIAL DISTRIBUTION IN SOILS STUDIED BY TRACK ANALYSIS AND SEM-EDS

Vlasova I.E. [1]*, Kalmykov S.N. [1, 2], Kashkarov L.L. [2], Clark S. B. [3], Aliev R.A. [1]

[1] - Moscow State University, Department of Chemistry, Radiochemistry division

[2] –Vernadsky Institute of Geochemistry and Analitycal Chemistry RAS

[3] – Washington State University, Department of Chemistry

* - *ivlas@radio.chem.msu.ru;* Phone (095) 939-32-20

Key words: ATA, FTA, plutonium spatial distribution, SSNTD, alpha-emitting radionuclides, SEM-EDS

Introduction

Radionuclides in soils could be distributed irregularly as a result of radionuclide sorption by different mineral particles or due to the presence of so-called "hot" particles. Alpha Track Analysis (ATA) and Fission Track Analysis (FTA) could be used to determine spatial distribution of alpha-emitting and fissionable radionuclides, measure their size and activity of "hot" particles [1, 2]. However mineralogical and chemical composition of the studied particles could not be determined using radio graphical methods. Kurosaki with co-authors [3] propose to use the SEM finder grid, coated with ^{7}B to obtain grid image on the solid state nuclear track detector (SSNTD) using (n, α) reaction.

The aim of this research was to develop the track analysis methods to locate microdistribution of ^{239}Pu and ^{235}U combined with the analysis of contaminated microparticles by scanning electron microscopy with energy-dispersed spectrometry (SEM-EDS).

Experiment

Anion exchange beads "DOWEX-1" to which natural uranium at 0.7 mBq per bead were used in model experiments. Two types of SSNTD, CZ and CR-39, were used for ATA and were exposed for 21 days. CZ detectors were etched using 6M NaOH solution (65^0C) for 5 hours and CR-39 detectors using 6,25 M NaOH solution (75^0C) for 5 hours.

Fission tracks of uranium were generated either by photofission using (γ, f) reaction or by neutron irradiation using (n, f) reaction. Two types of plastic detectors were used: lavsan (polyterephtalate), Germany, and "LEXAN GE". Irradiation of samples by gamma photons was performed at microtone at Skobeltsyn Institute of Nuclear Physics MSU with energy of electron beam of 35 MeV and intensity of 5 mA. The impulse duration was 4 μs and frequency of 10 Hz. Duration of irradiation was about 6 hours. Neutron irradiation was performed using thermal neutron beam in MEPhI Research Reactor ($1.6x10^{11} n \cdot cm^{-2} s^{-1}$ for 106 min). SSNTD were etched with 6M NaOH solution during 60 min (60^0C) and 10 min (70^0C) for Lavsan and "LEXAN GE" correspondingly.

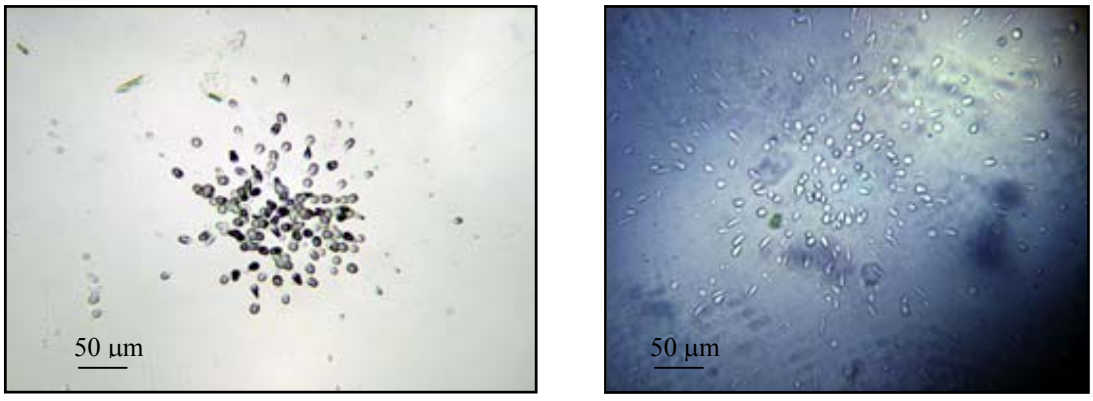

Fig. 1, a. Spatial distribution of alpha-tracks from U-coated anion exchange beads (left –CR-39 SSNTD, right –CZ SSNTD)

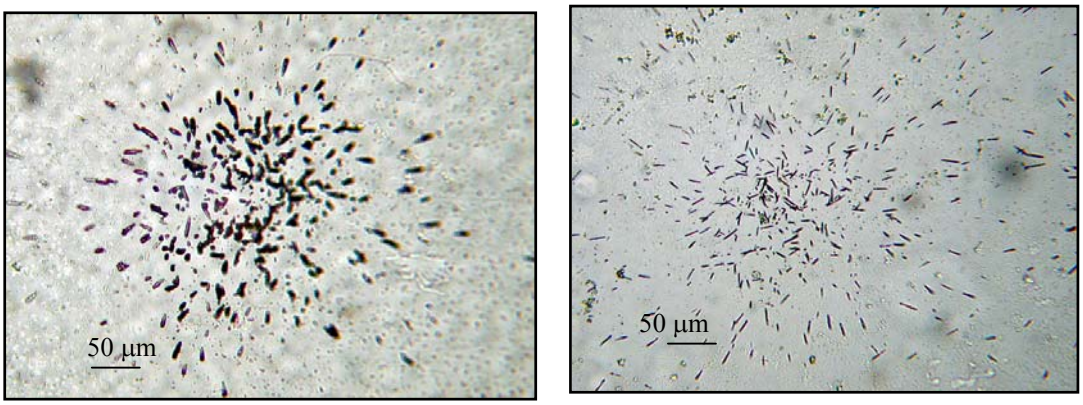

Fig. 1, b. Spatial distribution of fission tracks from U-coated anion exchange beads (left –lavsan SSNTD, right – "LEXAN GE" SSNTD)

Soil samples were collected in May 1986 around Chernobyl NPP. Spatial distribution of alpha-emitting radionuclides was obtained by ATA using CZ SSNTD. Exposition depends on activity of the sample and varied from 15 hours to 25 days.

To fix the exact location of "hot" particles SEM finder grid to which ^{239}Pu has been electrodeposited was used. This grid was located between SSNTD and sample that lead to the formation of the image of grid on the SSNTDs. The SEM-EDS (JEOL-840 (JSM-840A)) was used for microanalysis of single particles. For several particles alpha-spectrometry with ion-implanted detector was used that confirm the presence of ^{238}Pu, ^{239,240}Pu.

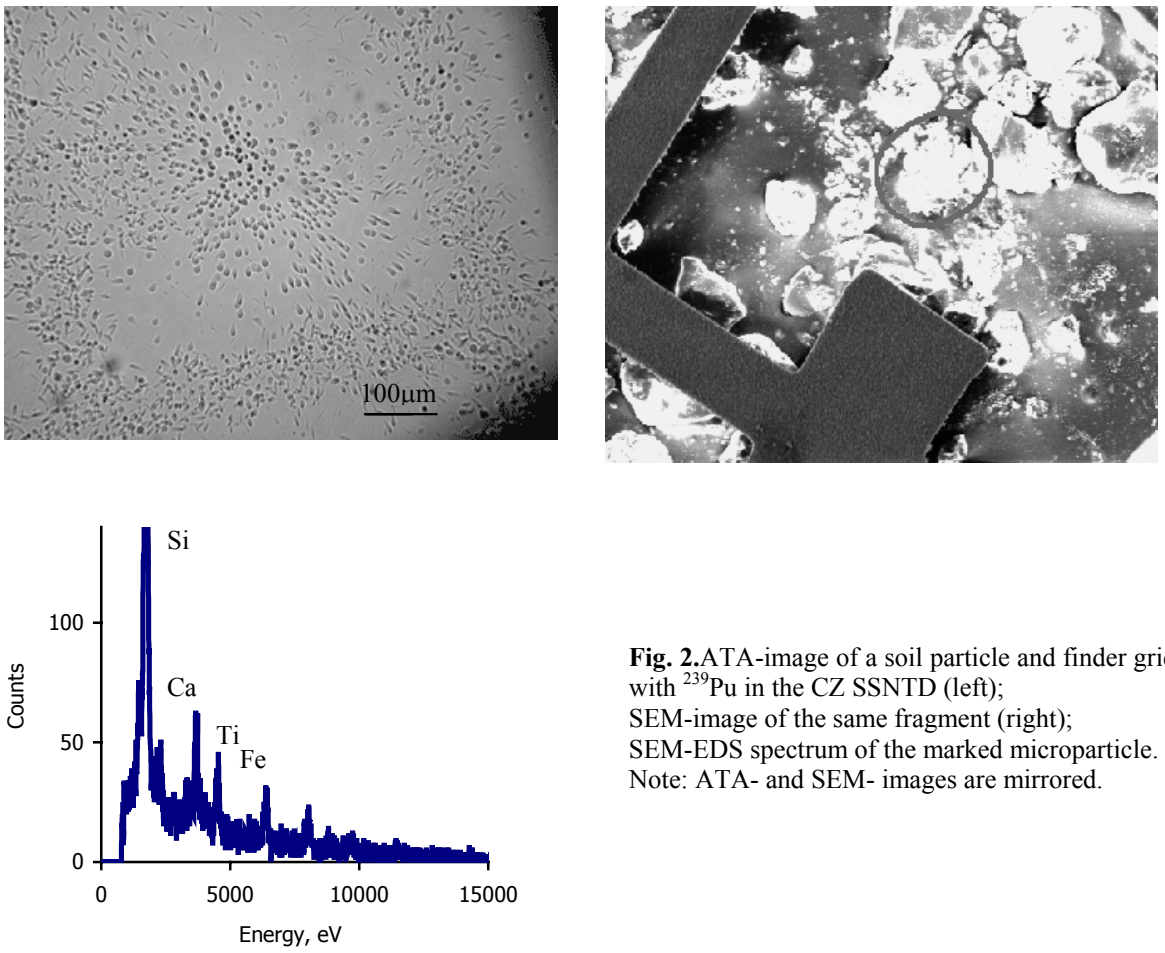

Fig. 2.ATA-image of a soil particle and finder grid with ^{239}Pu in the CZ SSNTD (left);
SEM-image of the same fragment (right);
SEM-EDS spectrum of the marked microparticle.
Note: ATA- and SEM- images are mirrored.

Results and Discussion

ATA results for anion exchange beads with natural uranium using two types of alpha-SSNTDs were the same as well as for FTA results (fig. 1a, b).

Track method has been developed to trace the location of alpha-emitters for the further SEM-EDS analysis.

Soil samples from the territory of Chernobyl NPP demonstrated irregular spatial distribution of alpha-emitting radionuclides. The location of the "hot" particles was fixed using SEM grid to which ^{239}Pu had been electrodeposited. As it was established by EDS that the major component of the samples was SiO_2. However particles that contained higher concentration of alpha-emitters were enriched in Fe and Ti if compared with the bulk composition (fig. 2).

References

1. *Kashkarov, L.L. et al.* (2001) Radionuclides and heavy metals in environment. NATO Science Series – IV, V.5, p.43.
2. *Kashkarov, L.L.; Kalinina, G.V.; Perelygin, V.P.* (2003) Rad. Meas., 36, 529-532.
3. *Kurosaki, H.; Lamont, S.; Filby, R.; Clark, S.B.; Peterman, D.R.* (2000) Los Alamos Science, 26, vol.2.

Electronic Scientific Information Journal "Herald of the Department of Earth Sciences RAS" № 1(22)'2004
Informational Bulletin of the Annual Seminar of Experimental Mineralogy, Petrology and Geochemistry – 2004
URL: http://www.scgis.ru/russian/cp1251/h_dgggms/1-2004/informbul-1_2004/geoecol-4e.pdf
Published on July, 1, 2004