Results from micro-XRF and micro-Tomographic measurements of environmental released Pu/U hot particles at ANKA

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General

In recent years the interest for so called hot radioactive particles have increased rapidly as for radiation protection point of view as well as in nuclear environmental forensic. It have been shown that such particles, in case of accidental release of radioactivity "correspond" to the major part of the released activity and will act like small point radioactive sources in the environment. Therefore, it is very important to study their environmental impact in terms of mobility, weathering and corrosion. For this scope, the elemental mapping distribution, the oxidation states of U and Pu as well as their 3D distribution should be studied.

In the present report the results obtained by micro-XRF with two beam diameter size (20 μm and for one particle also a beam of 1-2 μm). In addition on one particle micro-tomography was performed. The results obtained were very promising and it would be worthwhile to perform further investigation on more particles.

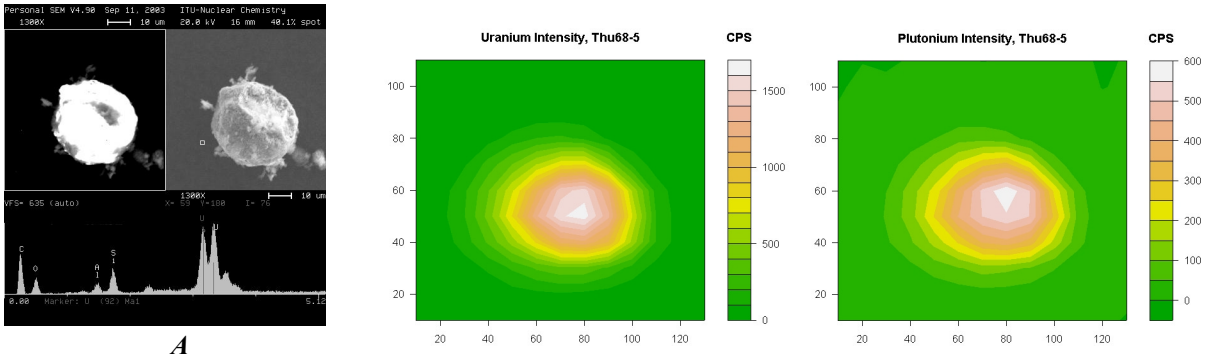
Material and methods

The radioactive hot particles investigated were separated from contaminated sediment samples. The samples were collected during two different campaigns at Thule nuclear accident site (Greenland). The single radioactive hot particles were localised and identified with a digital autoradiograph, the Beta Camera. All the particles were then analysed as for their size, morphology and surface elemental composition with a SEM with an EDX-system. The identified radioactive particles were measured at the Fluo-Topo beam line at ANKA. The white beam of a bending magnet was monochromatised by a W-BC_4 multilayer double monochromator to 23.2±0.25 keV. The excitation energy was chosen in order to excite the L1 edges of U and Pu, and to minimise the overlap between the L lines of the elements of interest and the Compton scattering peak. An ellipsoidal glass monocapillary (IfG, Germany) was employed for focusing a beam of 1x1 mm^2 down to a spot size of 20 um diameter. An energy dispersive OXFORD Si(Li) detector with a FWHM of 133 eV at 5.9 keV was placed in 90˚ geometry with respect to the incoming beam to minimise the amount of scattered photons in the detector. Elemental maps were collected on selected particles using a step size of 10-20 mm and a spectral acquisition time of 20 s. In addtion one particle were measured with a x-ray lens instead of the monocapillary with beam size of ~1 um. In addition tomography was carried out for one particle using the same x-ray lens set-up. The spectra were deconvoluted with the software program AXIL, using non linear least-squares fitting. The energies and relative intensities of the L and M line series of plutonium have been added to the X-ray library of AXIL.

Results

The SEM backscattered image of a U/Pu particle is reported in Fig. 1A. Fig 1B shows the mapping of U and Pu obtained with a beam spot was 20 μm. As it can be seen U and Pu appear as homogenous distributed in the particles. However, this beam size is to large compared to the particle to observed fine structure in the particles. In fact when using a beam size of about 1-2

µm it has been observed that the fine distribution of the two elements is not more as homogeneous as found in the previous experiments.



A

Fig. 1: *A: SEM-EDX image; B: Example of micro-XRF map obtained for the same particle with a beam size of 20 um (B(a): U mapping; B(b): Pu mapping)*

In Fig 2, the ratio of the mapping obtained for U and Pu with 1-2 µm size beam is shown and hetereogenities in the U/Pu distribution over an area of 100µm x 120µm of the sample surface can be noted. The Pu/U intensity ratio in the investigated particles was found to vary between 0.22-0.36. This variation may reflect preferential leaching of one element and/or that the particles originate from different components of the atomic bombs or from batches from different bombs.

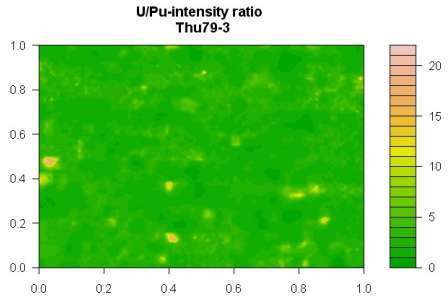
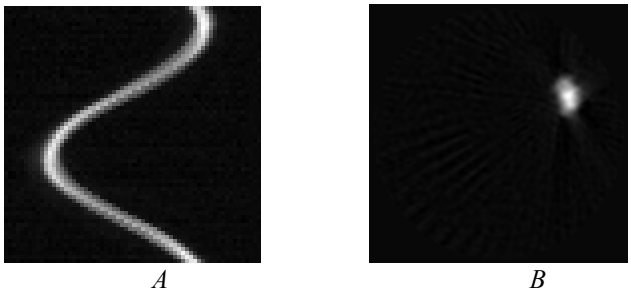


Fig. 2. *U/Pu ratio mapping obtained for a sample surface of 100 µm x 120µm with a beam size of about 1-2 µm*

The experiments performed by tomography confirmed that the 3D distribution of U and Pu was not homogeneous. In Fig 3 a corrected Pu sinogram together with the reconstructed image (one slice of the particle) can be seen. The tomographic data is still preliminary, as further corrections must be done in order to compare the Pu/U distribution.



A *B*

Fig 3*. A. Pu sinogram of one slice of the particle, B: reconstructed Pu image of the slice*

.References

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