Micro-XRF studies of environmental hot particles

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Intoduction

Radioactivity has been released into the environment not only in the form of evenly distributed gases and liquids but very often as inhomogeneous discrete particles of various size, composition and structure. Some of them – often called hot particles – represent high specific activities compared with that of the bulk material. The main sources of dispersed hot particles are the nuclear weapon tests and the nuclear fuel cycle. In releases of radioactive material to the environment from accidents with fire and explosions, the radioactivity will be associated to particles. For that reason it is of great importance to study the physical-chemical forms of the particles to be able to determine the fate of the radioactive material [1].

Materials and methods

The environmental particles studied have been separated from soil and sediments close to the epicenter of the contaminated area of the Thule accident (1968) site. Since the hot particles are dispersed into a natural environment their identification needs a lengthy separation procedure based on radioactivity of the material [2]. In an environmental sample one out of 10,000 or 1,000,000 particles is radioactive. Finally the separated are attached to an adhesive carbon tape and measured on digital image systems to identify the radioactive particle. With the digital image system it is possible to determine the position of the particle on the tape (the sizes of the particles are from 10 to 150 μm or a mass of about nanogram to microgram quantity. Figure 1. presents the electron micrographs and X-ray spectra of plutonium rich particles.

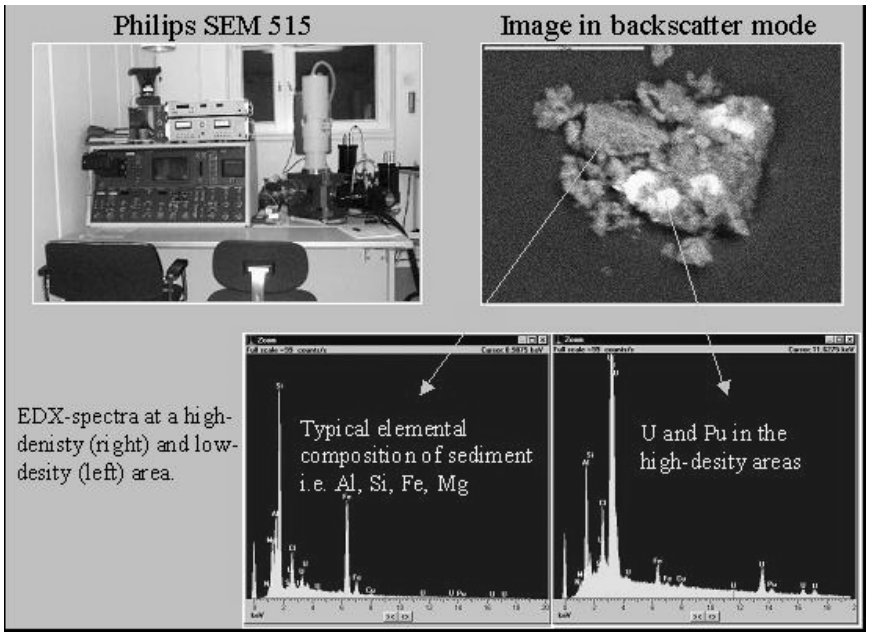

Figure 1. SEM analyses of radioactive particles.

The separated particles were measured on the ANKA Fluo beam line. The white beam of a bending magnet was monochromatized by a Ni-C multilayer double monochromator to 23.2 keV. The excitation energy was chosen in order to excite the L_1 edges of U and Pu, and to minimize the overlap between the L lines of the elements of interest and the Compton scatter peak. A polycapillary half-lens was employed for focusing a beam of 1×1 mm^2 down to a spot size of 20 µm diameter. The fluorescence yield was detected at an angle of 90° to the incoming beam using an OXFORD Si(Li) detector. Elemental maps were collected on selected particles using a step size of 20 µm and a spectral acquisition time of 20 s. From the particle areas having maximum U-Lα intensity, X-ray spectra were collected for longer acquisition time (100-400 s). U_3O_8 and UO_2 particles as well as NIST SRM 613 glass standard were also measured.

Results and discusssion

The scouting measurements performed so far have given an indication that in the sediment samples the particles stemming from Thule accident are containing traces of uranium and plutonium homogeneously distributed. The collected elemental maps of separated hot particles (Fig. 2) showed that the U/Pu Lα intensity ratio is homogeneous over the particles, but it is different for different particle diameters. On the bases of the hot particles analyzed it seems that uranium is preferentially leached than plutonium from the particles. Since the surface size of the particles was consistently different it indicates that the leaching rate of actinides is probably surface size dependent, and this effect is more pronounced for Plutonium. Such information is necessary to assess the environmental fate of the dispersed radioactivity close to the sea shore.

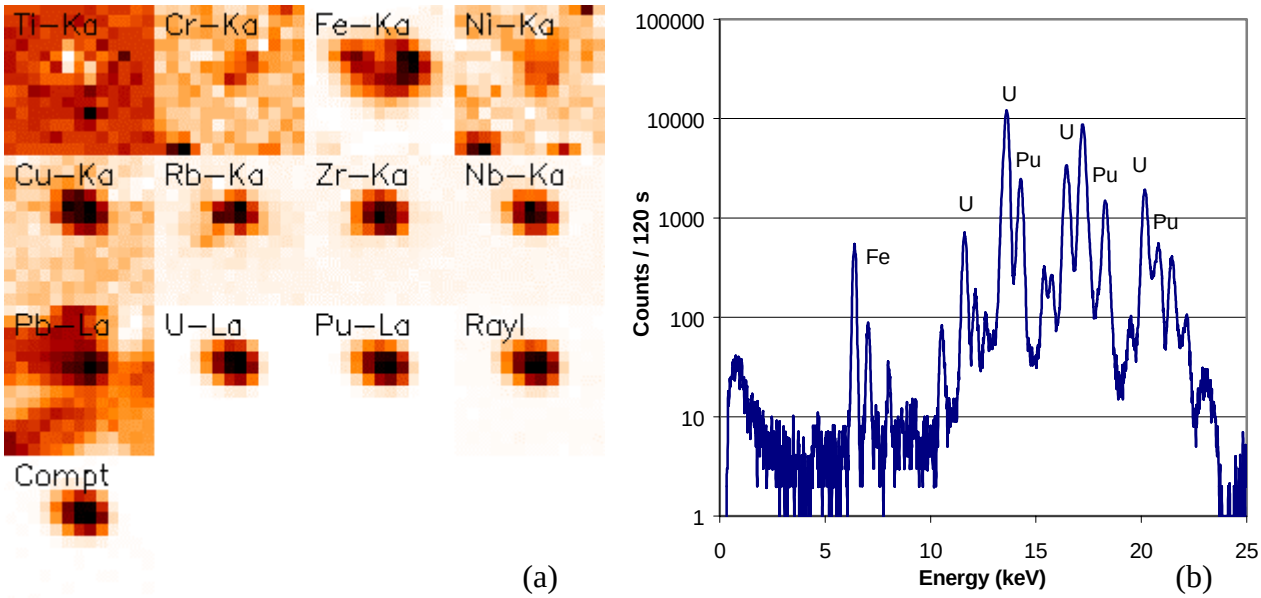

Figure 2. Pu-rich particle in sediment around Thule accident, (a) elemental maps of the particle, step size 20 µm, (b) X-ray spectrum collected at the position of the maximum U-Lα intensity

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