Characterisation of radioactive particles originating from U mining industry in Kazakhstan and Kyrgyzstan

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Following the cold war, extensive uranium mining and production took place at selected sites in Kazakhstan and Kyrgyzstan as a vital part of the nuclear weapon program in the former Soviet Union. The uranium deposit situated in the Southern Kazakhstan and the adjacent territory of Kyrgyzstan contains about 15 % of the world reserves of uranium. The deposit represents an area of roughly 60 000 km^2. The full industrial cycle of uranium recovery and processing of uranium ores have been undertaken in this region for about 50 years. Moreover, all types of uranium recovery have been undertaken: open-cast extraction; underground mining; in situ leaching with pumped sulphuric acid solutions. This densely populated territory was an administratively "closed area" within the USSR and information of the ecological state of this region is limited. Results available suggest that significant ecological damages, including radionuclide contamination and chemical pollution have occurred. Moreover, serious medical-pathological problems within the local population have been reported and these are claimed to be related to the environmental situation. Consequently, the Governments of Kazakhstan and Kyrgyzstan, and IAEA have expressed their concern about the radioecological situation within this region.

In the present work, we have characterised the terrestrial TENORM (Technologically Enhanced Naturally Occurring Radioactive Material) contamination at the former U mining sites Kadji-say, Kyrgyzstan and Kurday, Kazakhstan to obtain information on the solid state speciation and hence potential mobility of U from minerals. Digital autoradiography were utilised to document the presence of radioactive heterogenities in soils and minerals at both sites. Using light microscope and Environmental Scanning Electron Microscopy with Energy Dispersive X-ray analyser (ESEM) heterogeneities were isolated and identified as U containing particles. Following the ESEM characterisation these TENORM particles were subjected to synchrotron radiation based scanning micro-x-ray fluorescence (XRF)/micro- x-ray diffraction (XRD) at HASYLAB BL.

A focussed monochromatic X-ray microbeam of 10-15 μm diameter, having a divergence a ca 4 mrad was used for the investigations. This beam was obtained by employing a 200 period Mo/Si multilayer monochromator with mean layer thickness of 2.98 nm for energy band selection ($\Delta E/E=1\%$) and a single-bounce elliptical capillary for beam focusing [1]. A 1K Bruker CCD camera positioned behind the sample, was used for collecting diffraction patterns in transmission mode, along with a Si(Li) detector, positioned at 90 degrees relative to the primary X-ray beam, for simultaneous detection of the XRF signals.

Diffraction patterns of bright yellow Kadji-say mineral particles coincide with those of synthethic Na-zippeite ($Na_4(UO_2)_6[(OH)_{10}(SO_4)_3]\cdot 4H_2O$), pyrite ($Fe_2S$) and U_4O_{9+y} or UO_{2+x}, which may be interpreted as uraninite. In the latter case, the diffraction patterns are indistinguishable at the resolution obtained during these experiments thus frustrating the exact identification. Na-zippeite is a well-known secondary alteration product of uraninite (UO_2) in oxidized zones of U-rich deposits (e.g., mines), thus explaining the coexistence of these mineral species within the same particles. Uraninite (log K_{sp} of UO_2 = -60.6) and Na-zippeite (log K_{sp} = -116.5) is relatively insoluble. However, the possibility of mobilization, for example due to bacterial reductive dissolution of Na-zippeite, cannot be ruled out [2]. Only one U containing sample was isolated from Kurday soil.

Despite the fact that XRMA and XRF showed U to be present as a major element in this ~1 mm sized mineral grain, only quartz XRD signals were detected.

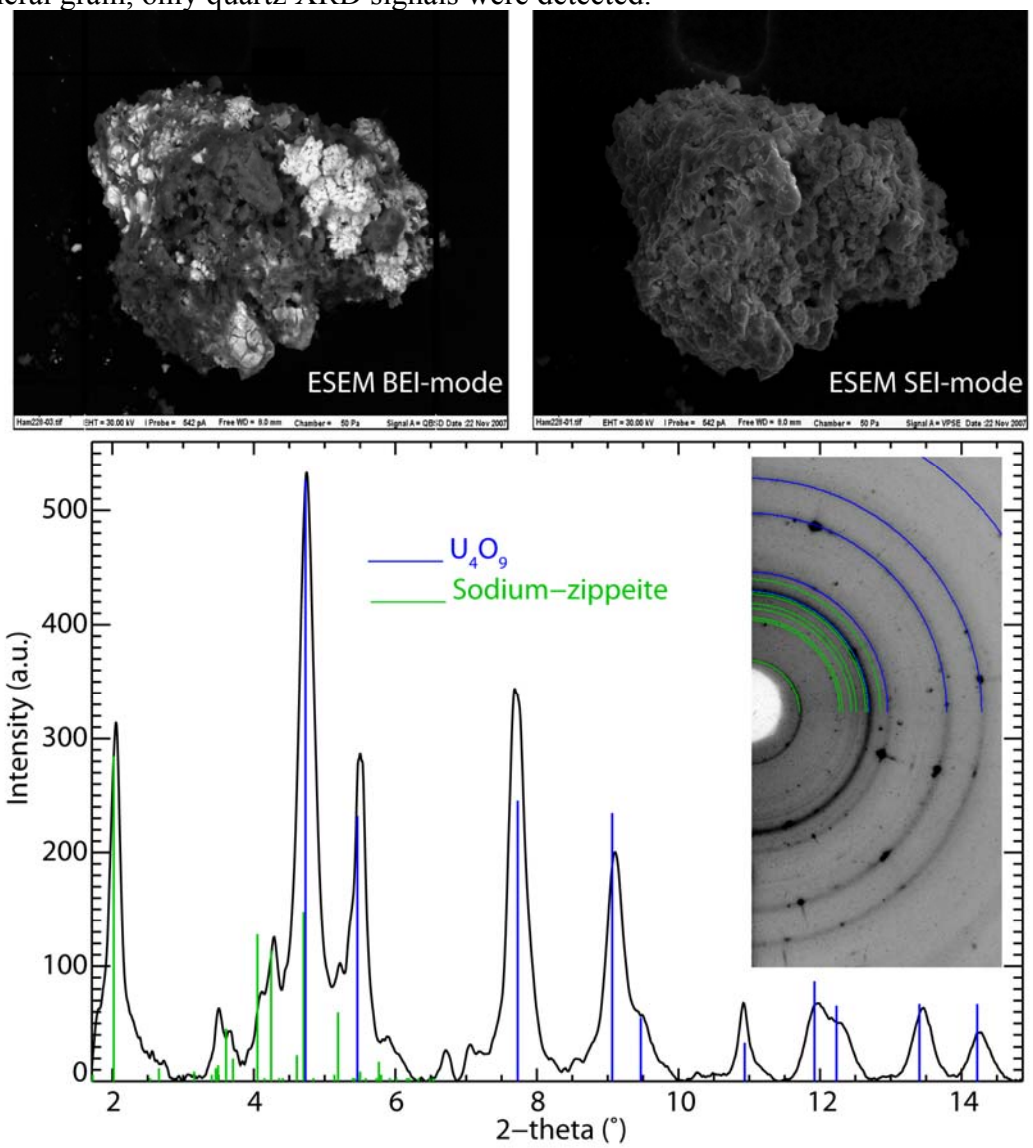

Figure 1: Electronmicrographs (backscattered electron imaging mode; upper left and secondary electron imaging mode; upper right) of a TENORM particle from the former Kadji-say U mining site in Kyrgyzstan. Diffractogram obtained from the same particle shows that U_4O_9 coexists with Na-zippeite ($Na_4(UO_2)_6[(OH)_{10}(SO_4)_3]\cdot 4H_2O$) within the particle.

To conclude, radioactive TENORM particles have successfully been identified, isolated and characterised using a combination of digital autoradiography, ESEM and synchrotron based μ-XRF/XRD. The results indicate that a large portion of the U at the two the former U mining sites Kadji-say, Kyrgyzstan and Kurday, Kazakhstan are associated with particles or large mineral grains. Furthermore, the results indicate that the solubility of the Kadji-say particles will be relatively low under oxidixzing conditions.

References

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