Identification of the U-mineral schoepite in DU particles using a combination of µ-XRF and µ-XRD

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We have previously reported that characteristics of depleted uranium (DU) particles in soil/sand from Kosovo and Kuwait contaminated during the Balkan conflict and Gulf war varied significantly depending on the release scenario (Lind, 2006; Salbu et al., 2004; Salbu et al., 2003). Bright yellow, highly oxidised DU particles (oxidation state +5 to +6) with a wide size distribution from submicron to several hundred micrometers were associated with a fire in an ammunition storage facility. DU in sand samples contaminated with these particles dissolved quite rapidly in simulated gastrointestinal fluid (0.16 M HCl). In contrast, the initial extraction of soil and sand samples contaminated with smaller and less oxidised DU (interpreted as UO₂, U₃O₈ or a mixture of these oxidized forms) particles, originating from the corrosion of DU penetrators or collected in tanks hit by DU ammunition, appeared to be much slower. In these previous experiments, individual DU particles isolated from aliquots of the above mentioned samples were characterised with respect to structure and elemental composition by scanning electron microscopy with X-ray microanalysis (SEM/XRMA). Furthermore, information on composition and oxidation states of matrix elements in particles was attained from µ-XRF (x-ray fluorescence) and µ-XANES (x-ray absorption near edge spectrometry) by means of Synchrotron Radiation based µ-X-ray techniques performed at the HASYLAB synchrotron facility, Hamburg, Germany. Recently, µ-XRF/µ-XRD experiments were carried out at HASYLAB Beamline L to provide more information on the crystalline phase(s) in which U is present in the individual DU particles.

In the present work, micro-XRF spectra and micro-XRD diffraction patterns were recorded simultaneously from the same location on the DU particles using a scanning micro-XRF/XRD set-up. This set-up was also utilised to obtain µ-XRF/µ-XRD maps (2 D) and line scans of the particles. A focussed monochromatic X-ray microbeam of 10-15 μm diameter, having a divergence of ~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 (ΔE/E=1%) and a single-bounce elliptical capillary for beam focussing (Falkenberg et al., 2004). A 1K Bruker CCD camera positioned behind the sample, was used for collecting diffraction patterns in transmission mode, along with a silicon drift detector (SDD), positioned at 90 degrees relative to the primary X-ray beam, for simultaneous detection of the XRF signals.

The present investigation shows that U in individual DU particles from Ceja Mountain is present as UO₂ and U₃O₇. The peak resolution obtained in the present experiments does not allow to distinguish UO₂ from U₃O₇. DU particles originating from the impact of DU ammunitions with tanks in Kuwait was similar to U in Kosovo particles (impact) with UO₂ and/or U₃O₇ identified as the main species present. U in DU particles released during the fire in the ammunition storage was present as schoepite [(UO₂)₆O₂(OH)₁₂·12(H₂O)] and metaschoepite [UO₂·1.5(H₂O)] (Fig. 1). Schoepite is found as an alteration product of uraninite (UO₂) and alters spontaneously to metaschoepite and paraschoepite [UO₃·2(H₂O)]. It
is a relatively soluble mineral which accounts for the rapid extraction observed in leaching experiments.

![XRD patterns of a DU particle](image)

**Figure 1.** XRD patterns of a DU particle originating from the ammunition storage fire in Kuwait, 1991 coincide with XRD patterns for schoepite and metaschoepite from the JCPDS ICDD database plotted schematically below the pattern.

The presence of respiratory U particles with oxidised forms higher than UO$_2$ and exhibiting high weathering rates, indicates that environmental or health impact assessments for areas affected by DU munitions should take into account oxidation states, their corresponding weathering rates and the subsequent mobilisation of U from oxidized DU particles. These results should be of importance for health impact assessments in ecosystems contaminated by a toxic heavy metal such as U.

**References**

