



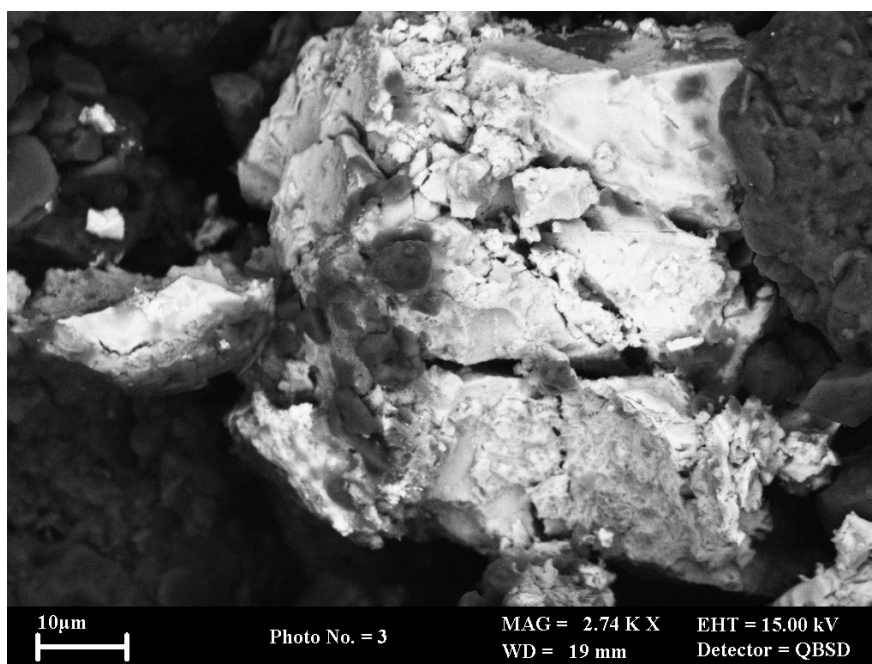
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Scanning electron microscopy of uranium particulate in two soil samples at a Depleted Uranium munitions strike site from the Kosovo Conflict

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Commercial Report CR/01/261N



BRITISH GEOLOGICAL SURVEY

COMMERCIAL REPORT CR/01/261 N

Scanning electron microscopy of uranium particulate in two soil samples at a Depleted Uranium munitions strike site from the Kosovo Conflict

AE Milodowski

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Front cover

Cover picture: backscattered scanning electron microscope image of a Depleted Uranium metal particle partly altered to secondary calcium-uranium-phosphate-sulphate phase (bright) in soil from Kosovo.

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Foreword

This report has been produced by the British Geological Survey (BGS) on behalf of the Defence Science and Technology Laboratories (DSTL, an agency of the UK Ministry of Defence, MoD) as part of an investigation into the environmental and health risk associated with the contamination by depleted uranium (DU) used in armour-piercing munitions fired by NATO aircraft at Serbian targets in Kosovo. In 2001 the UK MoD undertook sampling of soils and building debris associated with DU munitions strike sites in Kosovo. Two samples of DU-contaminated soils were studied in detail by BGS, using scanning electron microscopy (SEM), to characterise the nature of potential DU contamination. This is the second of two reports by the BGS describing SEM observations on uranium particulate in soils and building materials from Kosovo.

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Summary

Recent public debate has highlighted the concern over the potential long-term environmental and health effects arising from the use of depleted uranium (DU) munitions in recent military conflicts (Royal Society, 2001). DU munitions were used by NATO anti-tank (A-10) aircraft against Serbian targets during the recent conflict in Kosovo. In the United Kingdom, the Defence Science and Technology Laboratories (DSTL) are investigating the radiological and toxicological risk from DU munitions used in Kosovo. On behalf of the DSTL the British Geological Survey (BGS) undertook the analysis of two soil samples collected from a DU munitions strike site (the old VJ Barracks, near Podujevo) to characterise the nature of uranium-rich particulate material in DU-contaminated material. The samples were analysed by scanning electron microscopy techniques, using backscattered scanning electron microscopy (BSEM) to locate the uraniferous particles, and secondary electron imaging (SEM) to observe the morphology of the particles. Phase identification was supported by semi-quantitative microchemical information obtained from the phases by simultaneous energy-dispersive X-ray microanalysis (EDXA) during BSEM and SEM observation. This report details the analytical methods used and presents the observations and conclusions from this study.

BSEM proved very successful in locating dense uranium particulate on the surfaces of soil fragments because the very high atomic number of uranium produces high electron backscattering effects. Uraniferous particles were found covering the surfaces of soil fragments in both samples examined in this study. It is concluded that these particulate represents DU dust since no similar uraniferous material was found within the interior of the soil fragments. Uranium isotope analyses determined by inductively-coupled plasma – mass spectrometry (reported separately to DSTL by Gowing & Chenery, 2001) confirm that the soils contain uranium with an isotopic composition consistent with the presence of DU contamination.

Much of the uraniferous particulate material falls into two main size classes: (i) very fine material with a particle size less than 1 μm and generally less than 0.2 μm ; and (ii) coarser particles, typically 5-25 μm in size. As found in the earlier report (Milodowski, 2001) describing DU particulate in building debris, none of the uraniferous particles observed illustrated the typical spherical morphology of DU particulate reported previously from test firing of DU munitions against hard targets (cf. Patrick & Cornette, 1978). Instead, the observed DU particles from the soil samples are mainly flaky or irregular in morphology (single particles), or consist of colloform coatings and fine granular aggregates of secondary phases formed as secondary alteration products on the surfaces of primary uraniferous particles.

The coarser DU particulate material is mainly composed of uranium oxide, although some particles appear to relicts of uranium metal shards, which have been only partially oxidised. The fine-grained particles are composed mainly of uranium, calcium and oxygen, often with minor amounts of phosphorus and sulphur. They are interpreted as primarily a calcium-uranium carbonate or oxide/oxyhydroxide-like phase. The presence of sulphur and phosphorus suggest that the assemblage of phases may be more complex and may also include sulphate and phosphate phases. Material of a similar composition was also found as micro-granular and colloform coatings forming as alteration products on the surfaces of coarser uraniferous particles. The petrographical relationships observed by SEM indicate that the calcium-uranium-phosphorus-sulphur-bearing phases are formed by alteration of primary uranium oxide or uranium metal particulate within the soil environment. Uranium silicate has also been identified as an alteration product or the metallic uranium particles.

A small amount of titanium (estimated to be of the order of 1-3 % by weight) was observed in most of the particles and is most probably derived from titanium alloyed with uranium in the DU alloy.

Silicon, aluminium, potassium and iron are commonly observed in the EDXA spectra recorded from the uraniferous particles, particularly from the finer grained material. However, it is likely that these elements in the EDXA analyses are influenced by background X-ray emission from the soil matrix, which contains fine-grained quartz, illite and iron oxides.

Rarer, but much coarser, irregular fragments of uranium-rich material, up to 100 μm were also observed. They are usually fractured and often appear to be breaking down into smaller particles within the soil. EDXA indicates that these particles are partially oxidised metallic fragments of uranium that are altering to the same calcium-uranium-phosphorus-sulphur-rich phase observed in the finer grained uraniferous particles.

In addition to uraniferous particles, some fine-grained lead oxide rich particles and chromium-iron oxide particles were also observed. The origin of these particles is not clear, although it is conceivable that the lead is also munitions-derived.

1 Introduction

Recent public debate has highlighted the concern over the potential long-term health effects arising from the use of depleted uranium (DU) munitions in recent military conflicts (Royal Society, 2001). DU munitions were used by Allied forces in the Gulf War in 1991 and more recently by NATO in Bosnia and Kosovo. Reports of subsequent illnesses (e.g. ‘Gulf War Syndrome’) suffered by some the combatants involved in these conflicts has drawn attention to the possibility that they may be related to exposure to DU.

Uranium is a very dense metal (about twice as dense as lead) that exists in nature as three isotopes, ^{238}U , ^{235}U and ^{234}U . ^{238}U and ^{235}U are the most abundant isotopes and represent primordial uranium. ^{234}U is the radioactive decay product of ^{238}U . All three isotopes are unstable and are radioactive. Uranium is also a toxic heavy metal. DU is a by-product of the nuclear fuel manufacturing process, whereby nuclear fuel is enriched in the more radioactive ^{235}U . As a consequence, the residual uranium, which is depleted in ^{234}U and ^{235}U (i.e. DU), is about 40% less radioactive than natural uranium (Royal Society, 2001). However, the chemical toxicity of DU is the same as natural uranium.

As a by-product of nuclear fuel manufacture, DU is both abundant and cheap. Consequently, it has found cost-effective use in radiation shielding, rotors, and flywheels and in counterbalances for aircraft control surfaces. Its very high density and metallurgical properties also make DU ideal for military use as a kinetic energy ‘penetrator’ in munitions rounds designed to penetrate heavy armour of modern battle tanks and other armoured vehicles. These rounds are non-explosive but rely on the very high kinetic energy of a very dense DU projectile, travelling at high speed, to ‘punch’ holes in sophisticated modern armour plating. Unlike alternative tungsten penetrators, which blunt on impact with heavy armour, penetrators made of DU (alloyed with a small amount of titanium) are ‘self-sharpening’. Consequently, DU penetrators have considerably greater penetrative properties than alternative armour-piercing rounds. Furthermore, when a DU penetrator strikes tank armour it generates a dust cloud within the vehicle which spontaneously combusts, creating a fire which enhances damage to the target. DU can also be used in defensive armour plating of modern tanks, where it gives significantly increased protection against armour-piercing munitions.

There are concerns over the health risk posed by short-term exposure of military and civilian personnel to DU through handling the munitions during the conflict, or from inhalation or ingestion of the DU dust (created by impact of DU penetrators) under battle conditions or during post-conflict clean-up operations. Also, very little is known about the longer-term geochemical behaviour of DU contamination in the environment and the health risks that this may pose in the future. In the United Kingdom, the Defence Science and Technology Laboratories (DSTL, formerly DERA) are investigating the radiological and toxicological risk from DU munitions used in Kosovo. On behalf of DSTL, as part of this assessment, the British Geological Survey (BGS) has undertaken a series of investigations to characterise the DU contamination in building debris and soils from the old VJ Barracks area, near Podujevo in Kosovo – which was struck by DU munitions fired from NATO A-10 anti-tank aircraft. This is the second of two reports dealing with the mineralogical characterisation, by scanning electron microscopy, of uranium-rich particulate material in soils and building debris from this site. The first report described DU particulate material from two samples of building debris (Milodowski, 2001). The present report describes the nature of uraniferous particulate identified in two soil samples collected from the same area. The objectives were to:

- determine whether the presence of particulate uranium matter could be identified in the samples;

- identify the physical and chemical characteristics of uranium-rich particles in the samples;
- describe any evidence of alteration of uraniferous material in the samples.

2 Analytical method

2.1 SAMPLE DETAILS

Two samples of soil, collected by DSTL from the vicinity of the old VJ Barracks building, near Podujevo in Kosovo, were studied. Both samples were analysed by X-ray fluorescence spectrometry (XRF) for total uranium and lead concentration, and by inductively-coupled plasma - quadrupole mass spectrometry (ICP-MS) for uranium isotope composition, as part of a batch of soil samples being evaluated for DU contamination. These chemical and isotopic data are reported separately (Gowing & Chenery, 2001) but summary information for the two soil samples examined in detail by scanning electron microscopy (SEM) is given below:

DSTL Sample 3A (BGS SEM reference code H125)

Sample of soil, comprising rounded clasts or granules of brown soil up to 5 mm diameter and scattered root and other organic fibres. Originally thought to be potentially contaminated with asbestos but found during preliminary SEM observations to be free of any asbestos contamination.

Total uranium concentration = 149 ppm compared to typical batch soil values of 2-6 ppm

Total lead concentration = 165 ppm compared to background soil values of 13-80 ppm

Isotopic ratio $^{238}\text{U}/^{235}\text{U}$ = 488 to 490

DSTL Sample 3B (BGS SEM reference code H126)

Sample of soil, comprising rounded clasts or granules of brown soil up to 5 mm diameter and scattered root and other organic fibres. Originally thought to be potentially contaminated with asbestos but found during preliminary SEM observations to be free of any asbestos contamination.

Total uranium concentration = 983 ppm

Total lead concentration = 52 ppm

Isotopic ratio $^{238}\text{U}/^{235}\text{U}$ = 493

For comparison, the typical uranium and lead concentrations in the bulk of the batch of soils examined from Kosovo are 2-6 ppm and 13-80 ppm, respectively. The typical background isotopic ratio for $^{238}\text{U}/^{235}\text{U}$ in the Kosovo soil samples lies between 137 and 156. These data clearly indicate the presence of significant DU contamination in DSTL samples 3A and 3B, and a lead content in DSTL sample 3A that is significantly higher than in most of the other soils.

2.2 SCANNING ELECTRON MICROSCOPY

2.2.1 Sample preparation

Samples for analysis by scanning electron microscopy (SEM) were prepared as SEM stub mounts. The SEM stub mounts were prepared by hand-picking and fixing small (1-5 mm) 'rounded' grains of soil onto 13 mm diameter aluminium pin-type SEM stubs, using Leit CCC conducting carbon cement. Each sample was prepared and examined in duplicate. Prior to examination in the SEM instrument the stub mounts were coated with a thin film (approximately 25 nm thick) of carbon to make their surfaces electrically conductive. Carbon coating was carried out using an Emitech 950 vacuum evaporation-coating unit.

2.2.2 SEM examination

SEM analysis was carried out using a LEO 435VP variable pressure digital scanning electron microscope, fitted with an Oxford Instruments ISIS 300 digital energy-dispersive X-ray microanalysis system (EDXA) system and a KE-Developments four-element solid-state backscattered electron (BSEM) detector. Observations were typically made in conventional high vacuum SEM mode, using 10-20 kV electron beam accelerating potential and 50 to 500 pA beam currents, as required to achieve the desired observational resolution. The samples were imaged using both secondary electron imaging (SEI) and backscattered electron imaging (BSEM). However, where sample charging problems were experienced (due to poor adhesion of the carbon coating) the instrument was operated in variable pressure mode (at 0.1-0.3 torr) using backscattered imaging only. Phase identification was aided by semi-quantitative energy-dispersive X-ray microanalysis of features of interest using the EDXA system simultaneously during SEM/BSEM observation.

The surfaces of the samples were initially scanned under BSEM to locate uranium-rich particles. The BSEM image brightness is proportional to the average atomic number of the material, thus allowing the distribution of different minerals or phases to be determined on the basis of their chemistry (Goldstein et al., 1981). BSEM in conjunction with EDXA is a powerful tool, providing a combination of high-resolution textural information with semi-quantitative assessment of the mineral chemistry. In this way, the majority of phases - even trace amounts of very fine-grained or cryptocrystalline minerals - can be identified. The morphology of the uranium particles was subsequently examined in SEI mode. SE images are obtained by imaging of low-energy secondary electrons produced in, or returned from, material surfaces as a result of interaction of the high-energy electron beam with the valence electrons of atoms in the specimen surface (Goldstein et al., 1981). Image brightness and shadowing effects in SE images are dependent on the orientation of the sample surface with respect to both the incident electron beam and the electron detector. Consequently, SE images provide information on the morphology of the material being examined in the electron microscope.

3 Results

3.1 DSTL SAMPLE 3A

The soil comprises angular grains of quartz (SiO_2) of fine silt to sand grade (5-200 μm) in a porous clay matrix composed¹ of illite (idealised formula $\text{K}_{1-1.5}\text{Al}_4[\text{Si}_{7-6.5}\text{Al}_{1-1.5}\text{O}_{20}][\text{OH}]_4$), kaolinite ($\text{Al}_4[\text{Si}_4\text{O}_{10}][\text{OH}]_8$) and possibly chlorite (a complex clay mineral with an idealised structural formula of $[\text{Mg,Al,Fe}]_{12}[(\text{Si,Al})_8\text{O}_{20}][\text{OH}]_{16}$). Organic material, root and other plant fibres are also present. Minor to trace amounts of fine grained (<1 μm) iron and titanium oxides and occasional silt-grade grains of calcite (CaCO_3) are disseminated throughout the soil matrix. Rare monazite ($[\text{Ca,Th,Ce,La,Nd}]\text{PO}_4$) and zircon (ZrSiO_4) were also identified.

BSEM observations revealed that uranium-rich particles are very common on the surfaces of the granules or fragments of soil. They were easily distinguished in BSEM imaging mode as bright grains (high electron backscattering coefficient) against the dull (low electron backscattering coefficient) silicate and organic matrix groundmass of the soil (Figure 1). The abundance of

¹ The clay mineralogy is provisionally identified on the basis of qualitative compositional data from EDXA only, and the idealised formulae of these minerals are given as a rough indication of their likely chemistry. It should be noted that the actual chemical compositions of the clay minerals might differ from these idealised formulae. More definitive clay mineral identification and characterisation requires further assessment by X-ray diffraction analysis and quantitative microchemical analysis, which is beyond the scope of this study.

uranium-rich particles present, estimated by counting particles from representative SEM fields of view, is of the order of 2-12 particles/mm². This is lower than was observed on the surfaces of fragments of the building debris from this site (which were found to have up to 100 particles/mm² – cf. Milodowski, 2001) and is consistent with the lower uranium content determined by XRF in these soil samples. The uraniumiferous particles were not observed within the interior of soil clasts.

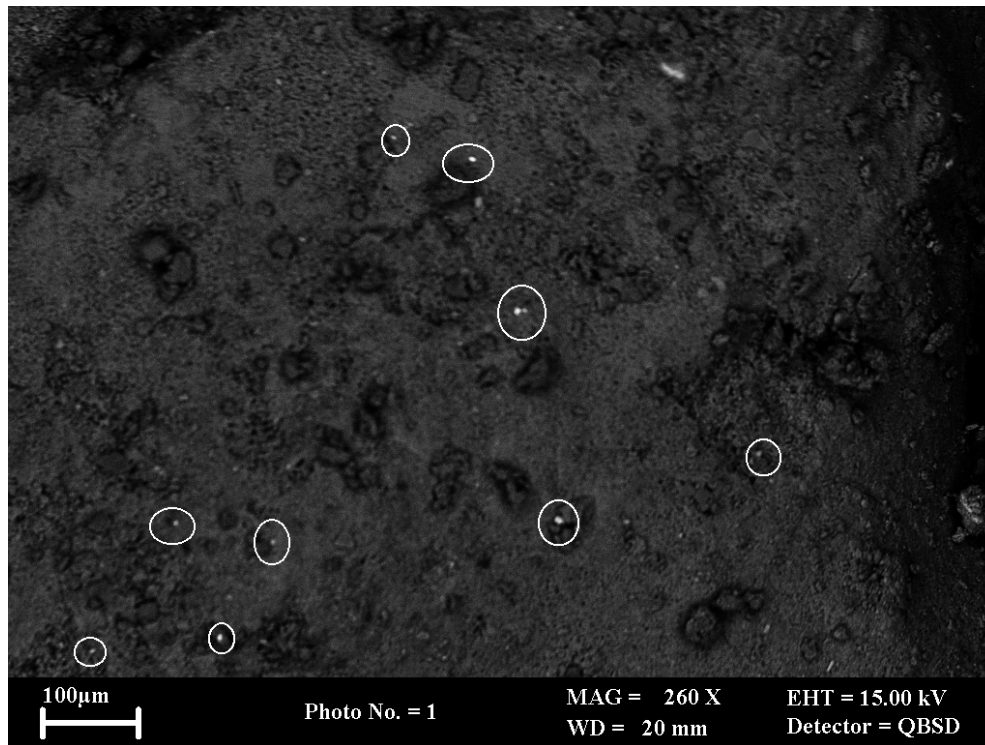


FIGURE 1. BSEM image of the surface of a soil fragment revealing the abundance of tiny particles of uranium-rich material (circled) resting on the surface, and which appear bright against the dark background of the soil matrix of quartz, clay minerals and organic matter under BSEM imaging. DSTL Sample 3A.

The uraniumiferous particles fall into two distinct size classes:

1. Fine grained particles - typically less than 1 μm and often much less than 0.2 μm diameter (Figure 1). This size class constitutes the most abundant type of uraniumiferous particle;
2. Particles varying from about 5 to 25 μm in size, although generally of the order of 10 μm (Figure 2).

The finer particles are generally flaky to needle-like or have very diffuse sub-rounded grain morphology. The coarser particles are more variable in morphology, and include blocky angular grains (Figure 3), irregular to angular flaky grains (Figure 2) or comprise complex granular aggregate particles (Figures 4 and 5). However, as was found in the previous study of DU particles in building debris (Milodowski, 2001) there is no evidence of uraniumiferous particles with the spheroidal morphology, produced by aerosolisation of DU that are reported from hard impact test firing of DU penetrators (Patrick & Cornette, 1978).

The uraniumiferous particles have variety of compositions. Qualitative EDXA spectra indicate that most of the fine-grained particles appear to contain major uranium, calcium and oxygen (Figure 6). This suggests that the particles are probably an oxide, oxyhydroxide or carbonate phase. Carbon is detected in the EDXA observations but it is not clear whether this is derived from the carbon coating used in sample preparation or a contribution from the organic components in the background matrix of the soil.



FIGURE 2. BSEM image of the surface of a soil fragment showing two distinct size classes of particles of uranium-rich material (circled), and which appear bright against the dark background of the soil matrix of quartz, clay minerals and organic matter under BSEM imaging. DSTL Sample 3A.

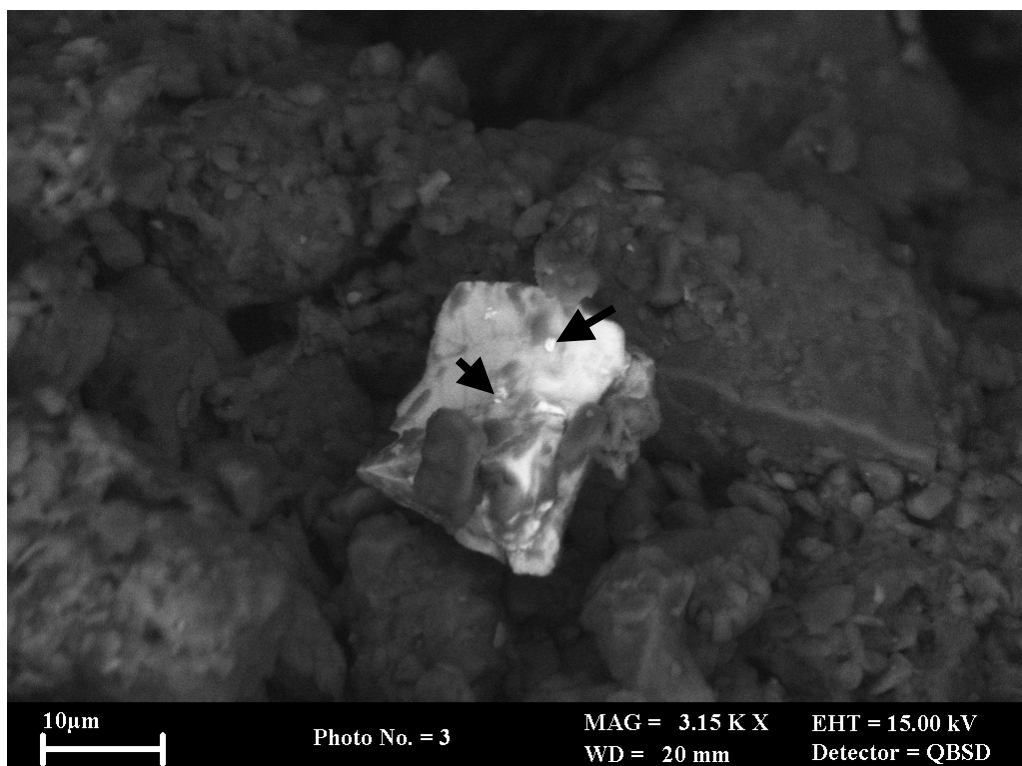


FIGURE 3. BSEM image showing bright blocky uranium-rich particle. The particle is composed mainly of calcium-uranium carbonate or oxide/oxyhydroxide phase with minor phosphorus and sulphur present. Tiny bright efflorescences of uranium oxide (arrowed) appear to be forming on the surface of the particle. DSTL Sample 3A.

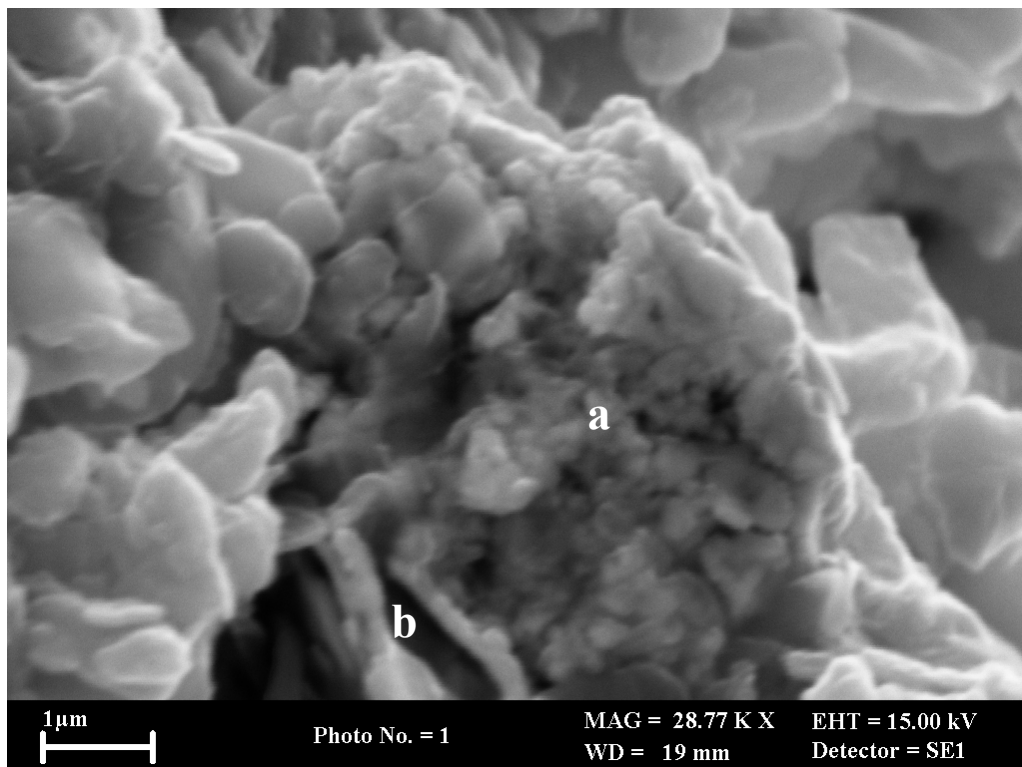


FIGURE 4. SE image of the showing the granular aggregate morphology of a relatively coarse uraniferous particle. The granular material (a) is a mixture of a calcium-uranium carbonate or oxide/oxyhydroxide phase and uranium silicate, and appears to have formed as an alteration of flakes of uranium metal or uranium oxide (b). DSTL Sample 3A.

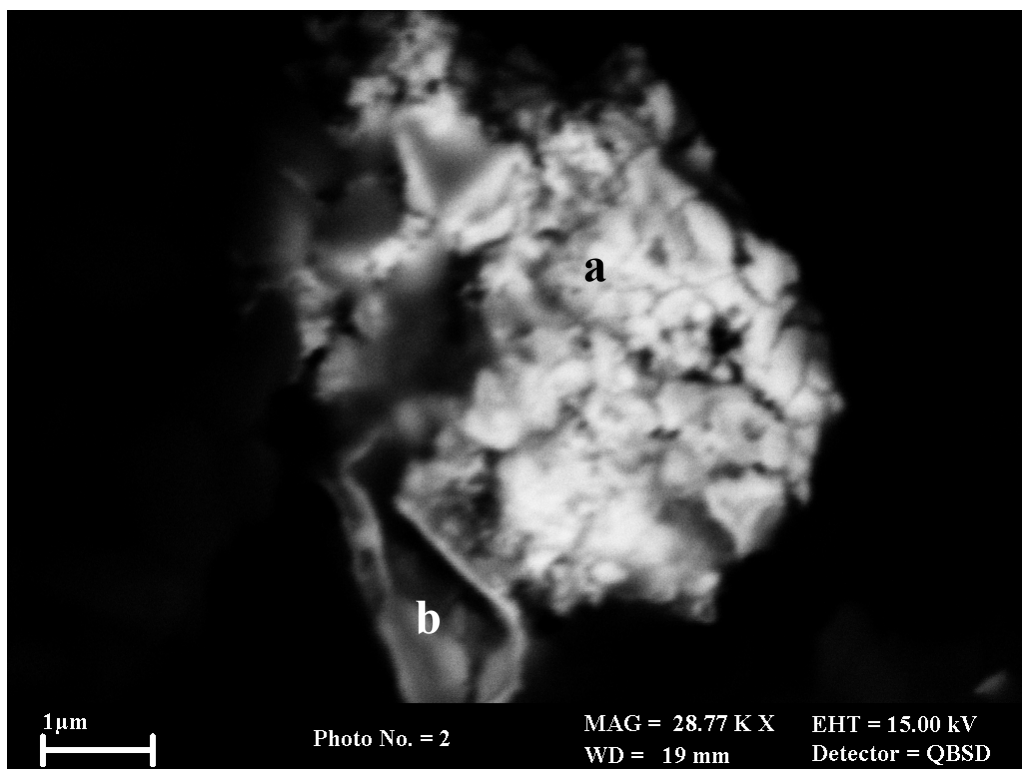


FIGURE 5. BSEM image of the same area as FIGURE 4, showing the granular calcium-carbonate or oxide/oxyhydroxide and uranium silicate forming as an alteration product (a) growing from the surfaces of flakes of uranium metal or uranium oxide (b). DSTL Sample 3A.

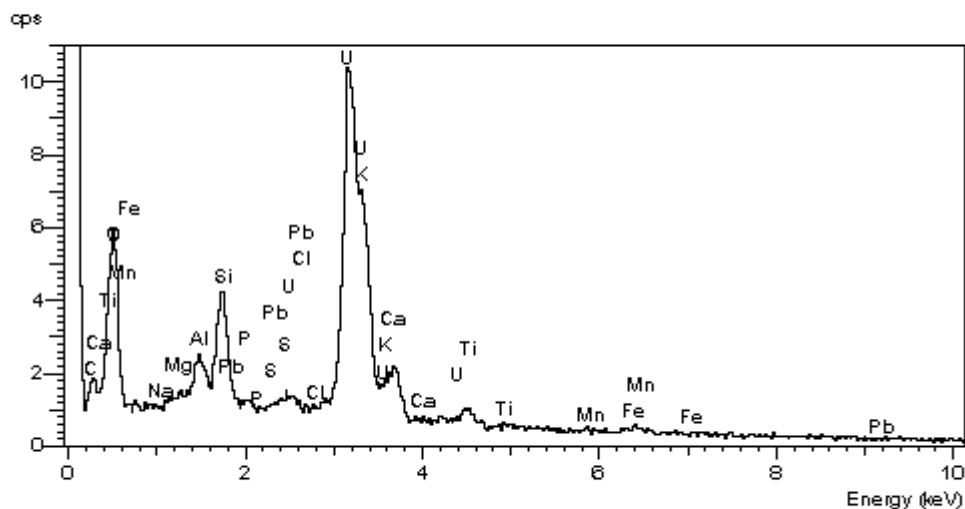


FIGURE 6. EDXA spectrum recorded from a typical fine grained uraniferous particle. The particle contains mainly uranium, oxygen and calcium. Silicon and aluminium reflect contribution to the X-ray emission from the background matrix of the soil. Titanium present is thought to be from the titanium originally present in the DU alloy. DSTL Sample 3A.

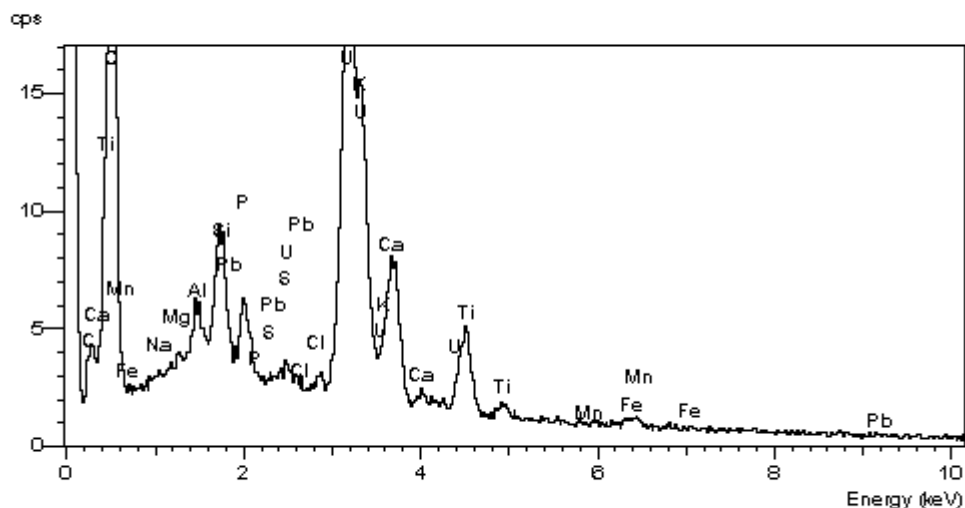


FIGURE 7. EDXA spectrum recorded from a coarse grained uraniferous particle. The particle contains uranium, oxygen, calcium, phosphorus and titanium. Silicon and aluminium reflect contribution to the X-ray emission from the background matrix of the soil. DSTL Sample 3A.

Particles containing minor to major amounts of phosphorus and sulphur are sometimes present. This suggests that there may be a more complex range of types of uranium particles including phosphate and sulphate phases. Aluminium and silicon present in most EDXA spectra are probably contributed from the background of aluminosilicate minerals in the soil matrix surface, although in some cases silicon appears to be intrinsic to the uranium-rich particles. Titanium is always a minor component in the EDXA analyses, and semi-quantitative estimates indicate that up to 2.5 % titanium (calculated as TiO_2) is present. This probably represents titanium derived from the original DU alloy.

The coarser uraniferous particles have complex fabrics. Some grains appear to be angular, blocky and shard-like (e.g. Figure 3). Some of the particles are similar in composition to the finer material (described above). Titanium is present in significant amounts (several percent TiO_2) in EDXA spectra (Figure 7) and is clearly intrinsic to the particle rather than as a contribution from the soil substrate. Many of the larger particles appear to be dominantly composed of uranium oxide (uranium and oxygen (with ubiquitous minor titanium) only are seen in EDXA spectra). In some cases, they appear to contain relict inclusions of actual metallic DU (EDXA spectra indicate that they essentially contain only uranium and minor titanium, and oxygen is largely absent). Other particles are complex composite grains, containing shard- or sheet-like cores of uranium oxide or uranium metal encrusted by micro-granular or colloform uranium silicate, uranium-calcium-oxide/oxyhydroxide or carbonate, or uranium-calcium-phosphate (\pm sulphate) alteration products (Figures 4 and 5)

In addition to uraniferous particles, small particles of lead or lead oxide are also present on the soil granule surfaces. These particles are up to 2 μm in size, and appear morphologically similar to the fine-grained uraniferous particles.

3.2 DSTL SAMPLE 3B

Soil sample 3B is very similar to sample 3A (described above) and contains very similar uraniferous particulate material. However, uraniferous particles are much more abundant in sample 3B. The abundance of uranium-rich particles present, estimated by counting particles from representative SEM fields of view, is of the order of 25-30 particles/ mm^2 . Furthermore, whereas lead-rich particles are relatively common in sample 3A, they were not observed in sample 3B. These observations are consistent with the bulk chemical analyses of the two soils – which show that sample 3B contains a significantly higher concentration of uranium but lower lead content than sample 3A.

Most of the uraniferous particles fall into the same size classes as observed in sample 3A. However, much coarser uraniferous particles, up to 100 μm in size are very common in sample 3B (e.g. Figure 8). They are often surrounded by clusters of finer uranium particles (Figure 8). In addition, the coarser particles are typically highly fractured (Figure 9). These observations indicate that the coarse particles are altering in the soil and are breaking down to finer material. These coarse particles appear to be shards of partially oxidised DU metal alloy (uranium and titanium, possibly with some manganese), or uranium oxide grains. They commonly display conchoidal and ribbed fracture surfaces. Typically, the particles exhibit the effects of in situ alteration within the soil environment (Figure 10), with dissolution cavities in the particle surfaces, oxidation of the uranium-titanium alloy to uranium oxide, and encrustation by later-formed uranium-calcium oxide /oxyhydroxide or carbonate and uranium-calcium-phosphate phases (Figure 10).

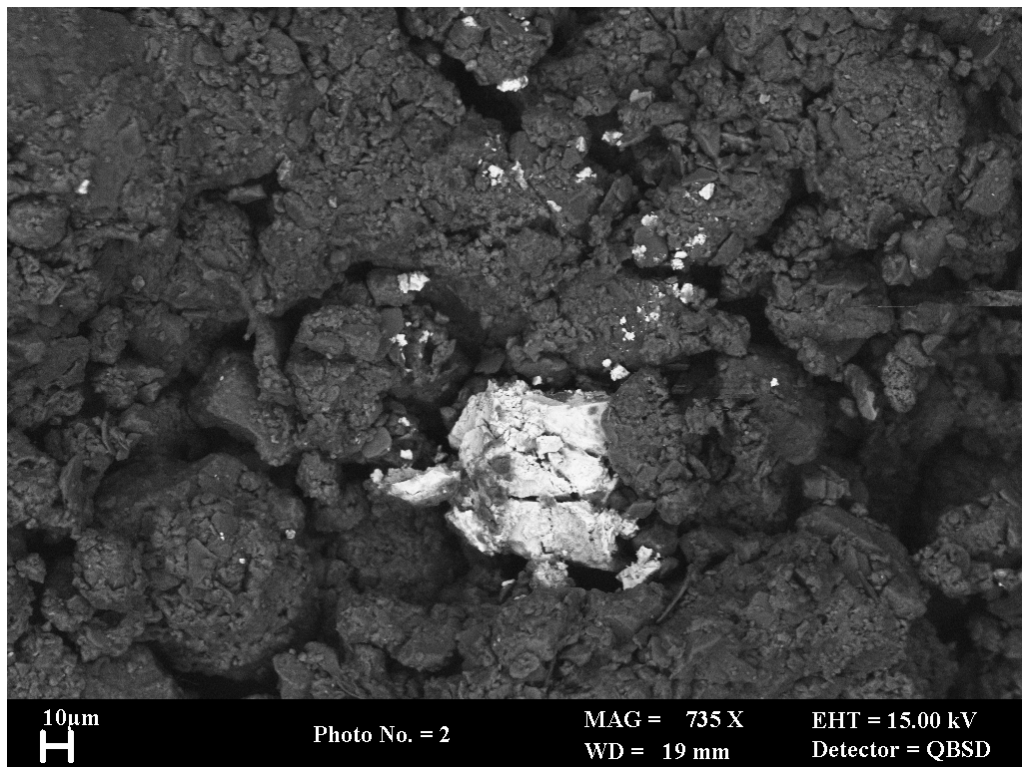


FIGURE 8. BSEM image of the showing a very coarse angular uraniumiferous fragment that is highly fractured and is closely associated with a cluster of finer uraniumiferous particles. The coarse uraniumiferous particle is partly altered uranium metal and uranium oxide, with secondary calcium-uranium carbonate or oxide/oxyhydroxide and calcium-uranium phosphate secondaries. DSTL Sample 3B

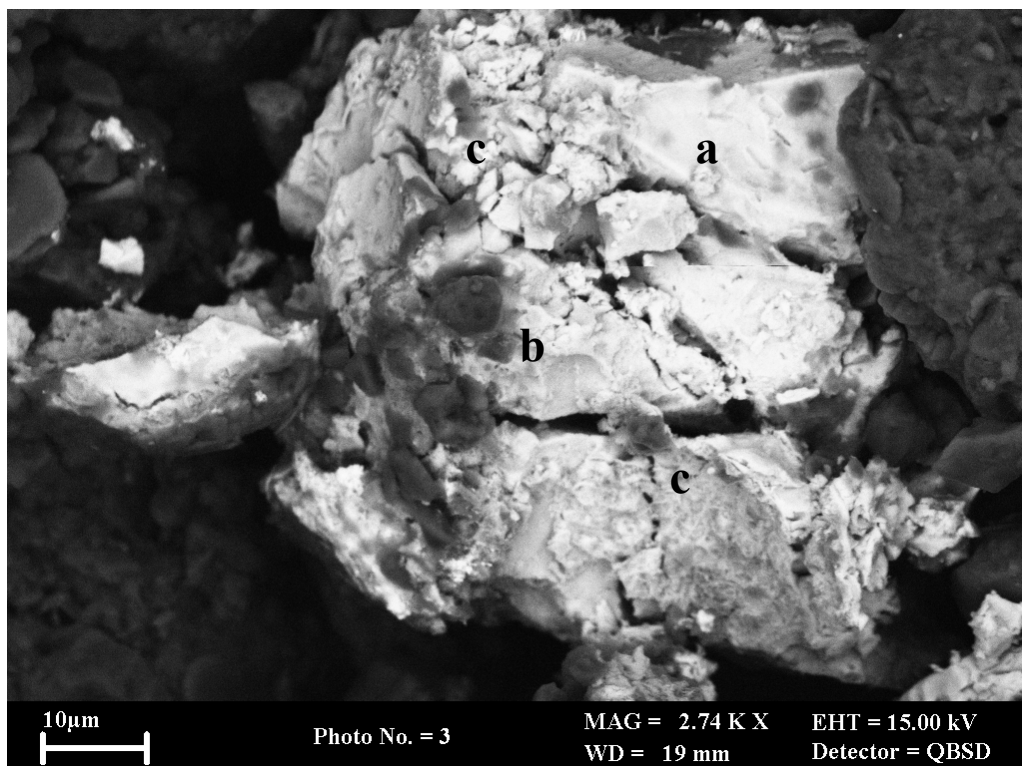


FIGURE 9. BSEM image of the showing detail of the very coarse angular uraniumiferous fragment in Figure 8. The coarse uraniumiferous particle is partly altered uranium metal (a) and uranium oxide (b), with flaky secondary calcium-uranium carbonate or oxide/oxyhydroxide and calcium-uranium phosphate secondaries forming on the surface (c). DSTL Sample 3B

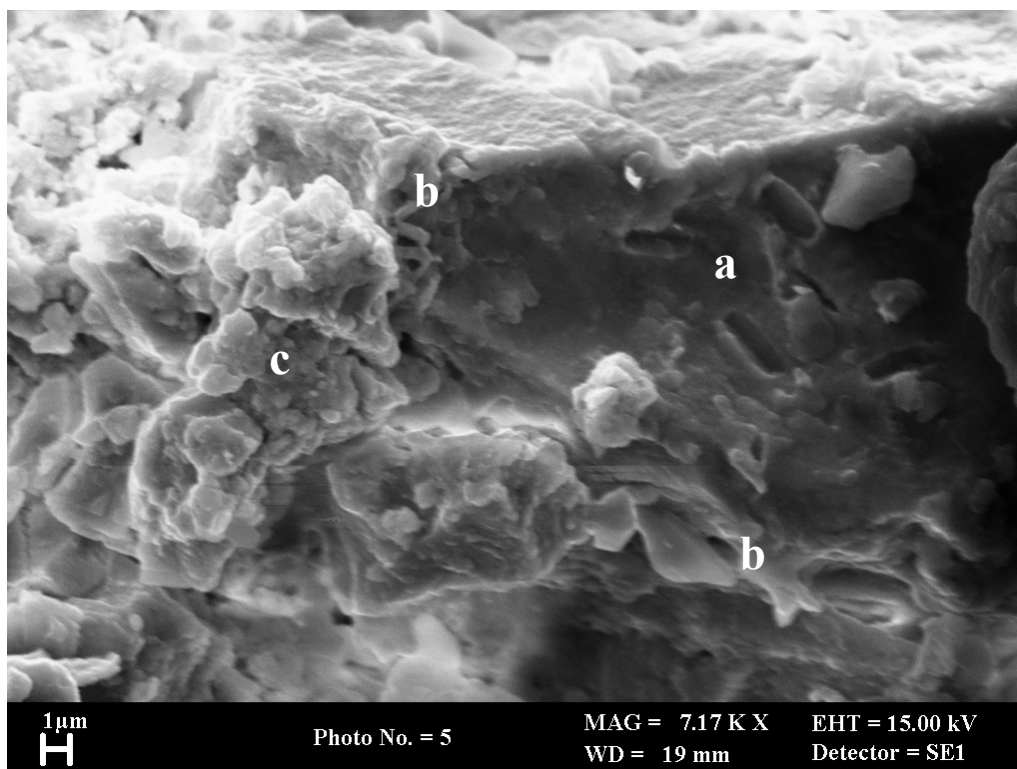


FIGURE 10. SE image of the showing detail of the surface morphology of the very coarse angular uraniferous fragment in Figure 8. The interior of the particle is exposed, showing uranium metal (a).displaying dissolution cavities developed in its surface. The surface of the metal is altering to uranium oxide (b), which in turn is encrusted by granular calcium-uranium-phosphate phase, or a calcium-uranium oxide/oxyhydroxide or carbonate phase (c). DSTL Sample 3B

4 Summary and conclusions

Two soil samples (DSTL sample numbers 3A and 3B) were examined in detail by scanning electron microscopy in order to characterise the nature of uranium-bearing phases. Both samples consist of organo-minerallic soil containing mainly quartz and minor calcite silt and fine sand grains in a matrix of illitic and kaolinitic clay with possible minor chlorite, and with very fine grained disseminated iron and titanium oxides. Uranium-rich particulate material is found on the external surfaces of the soil clasts or grains. However, no uranium-rich material was found within the interior of the soil fragments. This is consistent with the uranium particles representing externally-derived DU particulate. The presence of DU contamination is also confirmed by geochemical and isotopic analyses (Gowing & Chenery, 2001).

The uraniferous particulate in both samples mainly falls into two class sizes: (i) very fine material with a particle size less than 1 μm and generally less than 0.2 μm ; and (ii) coarser particles, typically 5-25 μm in size. However, sample 3B also contains much coarser material with grain sizes up to 100 μm . As found in the earlier report (Milodowski, 2001) describing DU particles in building debris, none of the uraniferous particles observed illustrated the typical spherical morphology of DU particulate reported previously from test firing of DU munitions against hard targets (cf. Patrick & Cornette, 1978).

The fine-grained particles are composed mainly of uranium, calcium and oxygen, often with minor amounts of phosphorus and sulphur. They are interpreted as primarily a calcium-uranium carbonate or oxide/oxyhydroxide-like phase. The presence of sulphur and phosphorus suggest

that the assemblage of phases may be more complex and may also include sulphate and phosphate phases. Material of a similar composition was also found as micro-granular and colloform coatings forming as alteration products on the surfaces of coarser uraniumiferous particles. The coarser DU particulate material is mainly composed of uranium oxide, although some particles appear to be relicts of uranium metal shards, which have been only partially oxidised. The petrographical relationships observed by SEM indicate that the calcium-uranium-phosphorus-sulphur-bearing phases are formed by alteration of primary uranium oxide or uranium metal particles within the soil environment. Uranium silicate has also been identified as an alteration product of the metallic uranium particles.

Rarer, but much coarser, irregular fragments of uranium-rich material, up to 100 µm were also observed in sample 3B. These are usually highly fractured and often appear to be breaking down into smaller particles within the soil. Petrographical observations show that these particles are partially oxidised metallic fragments of uranium that are altering to uranium oxide and the same calcium-uranium-phosphorus-sulphur-rich phases observed in the finer grained uraniumiferous particles. The metal fragments clearly show evidence of dissolution and corrosion, with the development of solution pitting in their surfaces. The calcium-uranium-phosphate alteration product appears to be late-forming and post-dates the alteration of the uranium metal to uranium oxide.

A small amount of titanium (estimated to be of the order of 1-3 % by weight) was observed in most of the particles and is most probably derived from titanium alloyed with uranium in the DU alloy.

Silicon, aluminium, potassium and iron are commonly observed in the EDXA spectra recorded from the uraniumiferous particles, particularly from the finer grained material. However, it is likely that these elements in the EDXA analyses are influenced by background X-ray emission from the soil matrix, which contains fine-grained quartz, illite and iron oxides.

In addition to uraniumiferous particles, some fine-grained lead oxide rich particles and chromium-iron oxide particles were also observed. The origin of these particles is not clear, although it is conceivable that the lead may also be munitions-derived.

References

Most of the references listed below are held in the Library of the British Geological Survey at Keyworth, Nottingham. Copies of the references may be purchased from the Library subject to the current copyright legislation.

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