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Characterisation of projectiles composed of depleted uranium

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Abstract

Projectiles suspected to be composed of depleted uranium (DU) were found in Kosovo. Their properties were analysed using alpha and gamma ray spectrometry, mass spectrometry and electron microscopy. They were found to be composed of DU with small amounts of other elements such as Ti. ^{236}U was detected in the penetrators, reflecting the use of reprocessed fuel. No transuranium elements were detected. The typical external dose rate meter is not the best option for mapping the location of penetrators from the ground. Monte Carlo calculations were performed in estimating possible skin doses. Penetrators in long-lasting contact with skin may cause a notable equivalent dose to skin.

Keywords: Penetrator; Depleted uranium; Plutonium; Skin dose

1. Introduction

At the end of 2000, STUK and the Finnish Defence Forces Technical Research Centre received three projectiles suspected of being composed of depleted uranium

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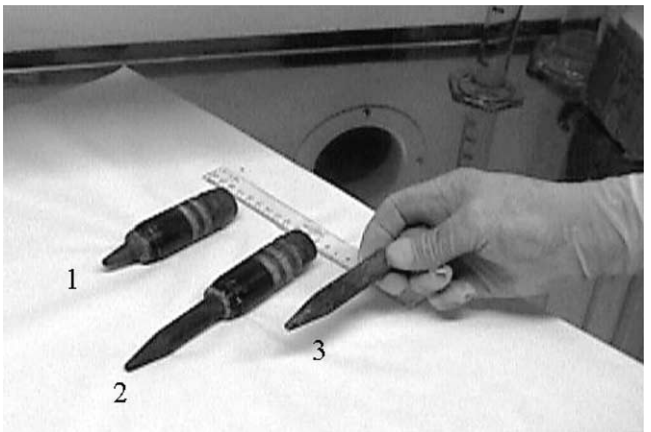

Fig. 1. Three rounds are shown on a laboratory table. The penetrator of round 1 is still inside the jacket whereas the penetrator of round 2 is partly outside. The most detailed analyses here were performed for penetrator 3. Penetrator 1 was found in Kokouce and penetrators 2 and 3 in Malopoljce (penetrators 2 and 3 were found on the ground, indicating manual transfer).

(DU). The projectiles were found in Kosovo by the UNEP team (UNEP, 2001) and the Finnish Peacekeeper Battalion in the areas where the North Atlantic Treaty Organisation had used DU munitions.

The objects considered here were not the entire rounds, but the parts known as penetrators (object 3 in Fig. 1). The dimensions of penetrator 3 are shown in Fig. 2. The density of penetrator 3 calculated from the data presented in Fig. 2 is 18 g cm^{-3}, which is slightly less than the nominal density of U metal (19.07 g cm^{-3}).

The aim of this study was to identify and quantify the radioactive materials present in the penetrators and to estimate the possible skin dose produced by a penetrator in contact with the skin. Comprehensive analyses of the three penetrators were performed and different complementary methods, such as alpha spectrometry, gamma-ray spectrometry, mass spectrometry and electron microscopy, were used in order to verify the results obtained. From the point of view of the present special issue, the analysis of penetrator properties is relevant for the following reasons:

1. DU-penetrators, as such, may pose a potential radiological hazard.
2. Characterisation of the penetrator properties is a prerequisite for assessing the consequences of DU dispersed into the environment.
3. Published data on penetrator properties are scarce or sometimes even misleading. Thus, independent and reliable data is needed.

2. External dose rate measurement

The typical external dose rate prevailing in the area where the Finnish peacekeeper battalion was located in Kosovo was of the order of 0.05–0.12 μSv h^{-1} (Ministry

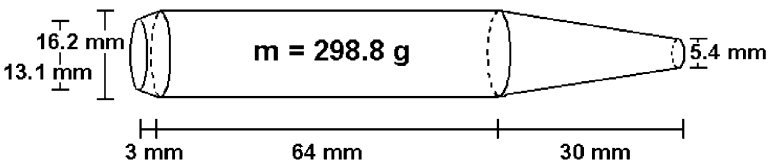

Fig. 2. Dimensions of penetrator 3. The figures are only indicative owing to the variable surface roughness and somewhat round-edged shape of the penetrator. (The penetrator was stuck in the ground, causing the possible deformation.)

of Defence, 2000). Here, the external dose rate, i.e. the ambient dose equivalent rate at a depth of 10 mm, $H*(10)$, was measured in the laboratory as a function of distance from penetrator 3 (Fig. 3). The purpose was to find out whether it is possible to locate penetrators in the environment using a typical portable GM dose-rate meter used in field conditions. The external dose rate near the surface of the penetrator was in the order of 10 μSv h^{-1}, whereas at distances greater than approximately 0.5 m, the external dose rate was almost equal to the background dose rate. Thus, portable dose rate meters are not necessarily the best option for mapping the location of penetrators in the ground. Visual signs or other on site information at a suspected location for finding rounds on the ground are essential to locate penetrators.

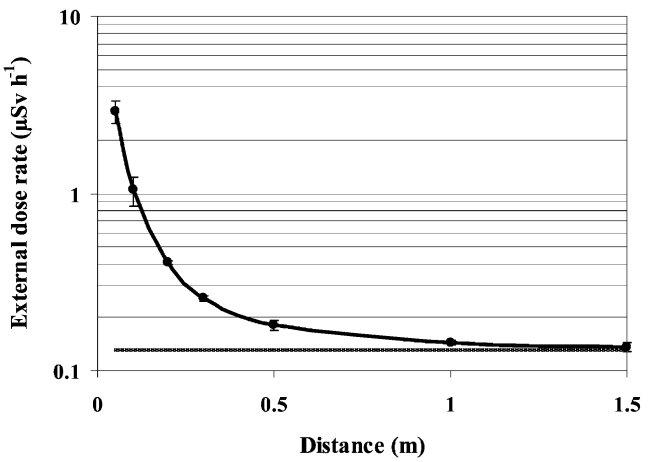

Fig. 3. External dose rate, i.e. ambient dose equivalent rate at a depth of 10 mm, $H*(10)$, as a function of distance from penetrator 3. The first two measurements (distances 5 and 10 cm) were done using a Rados RDS-120 portable Geiger counter and the others using a PIC (the minimum distance for PIC is 20 cm). Each point refers to a counting time of approximately 1 min and error bars indicate the 95% confidence level. The horizontal line indicates the background dose rate in the laboratory.

3. Gamma-ray analysis using HPGe spectrometers

HPGe spectrometers were used for analysing the amount of gamma-emitting radionuclides in penetrator 3. A typical spectrum is presented in Fig. 4. Because the shape

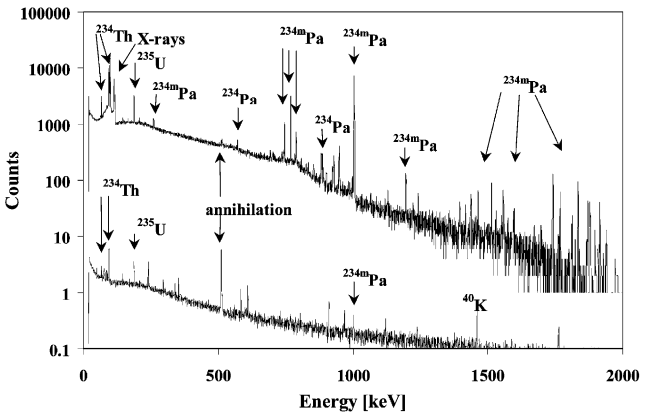

Fig. 4. Gamma-ray spectrum of penetrator 3 (above) measured by a HPGe detector (40% rel. eff. @1.3 MeV) compared to the background (below) with the same detector and counting time (38 min).

and density of the penetrator is not ideal for gamma-ray analysis, the activities could not be accurately estimated. The activity of ^{238}U (3.8 MBq) was calculated on the basis of ^{234m}Pa, whereas the activity of ^{235}U (0.06 MBq) was measured directly (Table 1). These numbers gave the ^{235}U/^{238}U mass ratio of 0.0025±0.0005. The total mass of the penetrator could be back-calculated from the specific activities of ^{238}U (12440 Bq g^{-1}) and ^{235}U (79980 Bq g^{-1}), giving the value 305 g for ^{238}U and 0.75 g for ^{235}U. The sum of these numbers was somewhat higher than the measured mass of the penetrator, i.e. about 300 g (Fig. 2), leading to the conclusion that the penetrator was composed almost entirely of U. The amount of other possible elements was negligible.

The U (and possible Pu) isotope ratios for penetrator 3 were also measured using the U-Pu InSpector, which uses X-rays and low-energy gamma rays for analysing the composition of macroscopic objects (Canberra, 1995). Using 27-h counting time,

Table 1
Activities of gamma-emitting nuclides in penetrator 3. The activities of ^{238}U and ^{234}Th were calculated from ^{234m}Pa by assuming the equilibrium in the decay chain ^{238}U→^{234}Th→^{234m}Pa. However, the gamma-peak of ^{238}U (49.5 keV), as well as those of ^{234}Th (63.3 and 92.6 keV), ^{234}U (53.3 keV) and ^{231}Th (25.7 keV) were directly detected (especially using a broad energy range germanium detector), but quantitative estimation of the activities in this energy region was not possible mainly because of the high attenuation in the penetrator

Nuclide	Activity (Bq)	Uncertainty (1σ)
^{238}U	3,800,000	~15%
^{235}U	60,000	~20%
^{234}Th	3,800,000	~15%
^{234m}Pa	3,800,000	~15%
^{234}Pa	11,000	~20%

no Pu was detected and the abundance of ^{238}U was 99.79% (uncertainty±0.009) and the abundance of ^{235}U was 0.213% (uncertainty±0.009). The amount of ^{234}U was below the detection limit.

4. Scanning electron microscopy (SEM) and X-ray microanalysis (EDS)

Small pieces from the penetrators were taken using a bore with a normal bore bit composed of steel. The material of the projectiles was found to be hard, referring to the hardness of steel typically used in tanks. The boring flakes were recovered and analysed using SEM equipped with EDS (LEO 1450 VP, Oxford Instruments Inca Energy 400), alpha spectrometry and Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

Some boring flakes from penetrator 3 were substituted onto a metallic disc using adhesive tape. SEM-micrographs were taken to verify the presence of U and other elements, as well as to obtain a view of the structure of the pieces. A typical flake was a large (visible with the naked eye) and comprised of an irregularly shaped piece of the projectile (Fig. 5) containing U, Ti and minor amounts of other elements. U and Ti seemed to be homogeneously distributed in the boring chips. However, the overall homogeneity of the penetrator was not analysed in this study.

5. Alpha spectrometry and ICP-mass spectrometry

At first, the alpha spectra of the boring flakes were counted without tedious dissolution-separation-electrodeposition processes in order to verify the presence of uran-

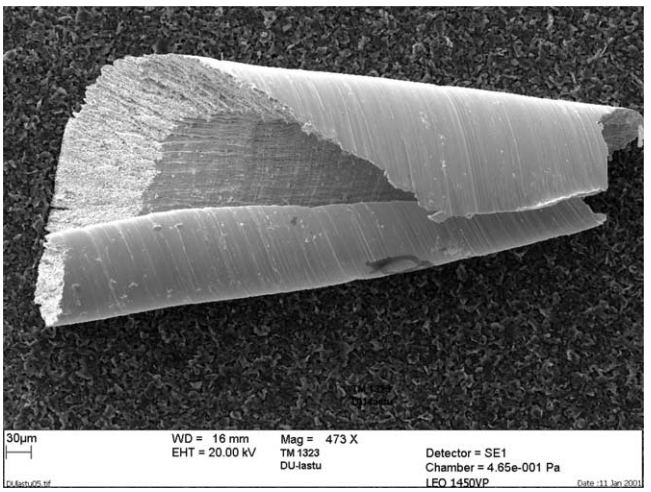

Fig. 5. A typical boring flake from penetrator 3.

ium in the flakes rapidly (dashed line in Fig. 6). Alpha-active materials were definitely present in the flakes with a large amount of ^{238}U visible from the broad peak region in the lower-energy part of the spectrum.

Self-absorption of alpha particles in the boring chips prevented a quantitative analysis. Therefore, a known amount of chips was dissolved into nitric acid and an aliquot of dissolution was used for the ICP-MS (VG Plasma Quad 2+) (Rosenberg & Zilliacus, 1997). A routine (trans)uranium analysis using chromatographic resins for the separation of U and possible Pu and Am+Cm was performed on the remaining sample (Moring et al., 2001). ^{238}U, ^{236}U, ^{235}U and ^{234}U were identified in the alpha spectra of the U-fraction (solid line in Fig. 6). Mass ratios are shown in Table 2. Neither Pu nor other transuranium elements were detected in penetrators 3 and 2. The detection limit for $^{239+240}$Pu was 0.1 ng for penetrator 3, indicating an activity of 0.24 Bq (^{239}Pu). The respective detection limit for penetrator 2 was 0.46 Bq.

Let us assume that the amount of ^{239}Pu in the penetrators equals the above-mentioned detection limit of 0.24 Bq. By assuming that 1000 penetrators were shot in one operation and that all penetrator material was evenly deposited within 100 m^2, the ^{239}Pu deposition would be 2.4 Bq m^{-2}. The typical ^{239}Pu fallout from nuclear weapon tests in Finland is about 60 Bq m^{-2} (Hardy et al., 1973). The Chernobyl-derived deposition of ^{239,240}Pu, even in the most contaminated regions in Finland, was about 10% of the global fallout from weapon tests (Paatero, 2000).

The dissolved samples from penetrators 2 and 3 were subsequently analysed using an ICP mass spectrometer. In addition to U-isotopes (Table 3), Ti was found to be present in the samples, as well as small amounts (<0.1%) of impurities such as Zn, Zr, Cu and Ni. The presence of Ti was an expected result (UNEP, 1999). Both alpha spectrometry and ICP-mass spectrometry gave almost equal results for the U-isotope ratios.

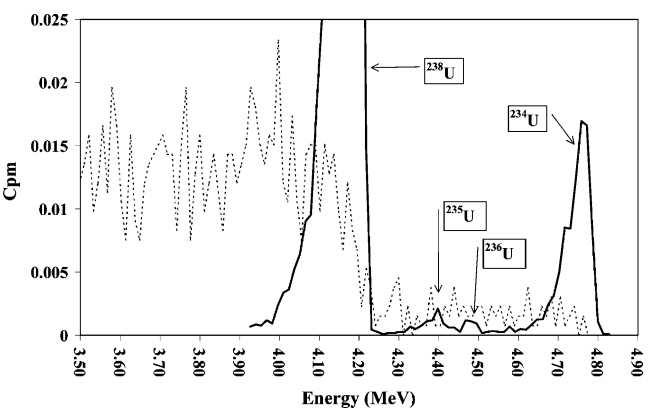

Fig. 6. Alpha spectra measured from boring chips (dashed line, data acquisition time 22 h) and U-fraction (solid line, data acquisition time 49.3 h) of penetrator 3.

Table 2
U-isotope mass ratios of the penetrators detected by alpha spectrometry. Unc. refers to the uncertainties of the areas of the alpha-peaks

Nuclides	Penetrator 1	Unc.	Penetrator 2	Unc.	Penetrator 3	Unc.
$^{234}U/^{238}U$	0.0000068	0.0000003	0.0000059	0.0000010	0.0000066	0.0000002
$^{235}U/^{238}U$	0.00138	0.00023	0.0019	0.00098	0.0017	0.00018
$^{236}U/^{238}U$	0.000017	0.000004	0.000032	0.000023	0.000024	0.000004

Table 3
U-isotope mass ratios detected by ICP-mass spectrometry. The amount of Ti in the samples from the penetrators 2 and 3 was 0.6±0.1 w-% and approximately 1 w-%, respectively

Nuclides	Penetrator 2	Unc.	Penetrator 3	Unc.
$^{234}U/^{238}U$	–[a]	–	0.0000086	0.0000006
$^{235}U/^{238}U$	0.0019	0.0001	0.0022	0.0001
$^{236}U/^{238}U$	–	–	0.000031	0.000003

[a] Not analysed.

6. Monte Carlo analysis and the measurement of possible skin doses

The penetrators were modelled as an 8-cm long cylinder with a diameter of 1.5 cm. Radioactive materials were assumed to be homogeneously distributed in the penetrator in the amounts presented in Table 4. The cylinder was assumed to be in contact with the skin or at variable distances from the skin. EGS4 Monte Carlo code (Nelson et al., 1985) was used for the calculations. This code enables calculation of the doses caused by beta particles and photons (the beta spectra, as well as photon transitions were taken from Chu et al., 1999). The code does not allow calculation of the doses caused by neutrons and alpha particles. Alpha particles do not cause external doses (they are strongly absorbed in the penetrator itself and in the surface of the skin).

The absorbed dose depends strongly on the depth of the tissue, as well as the type of radiation (Fig. 7). The dose caused by beta particles (almost entirely from ^{234m}Pa) dominates at lesser depths (less than about 0.6 cm), whereas at greater depths, the dose originates from the gamma radiation and bremsstrahlung which is produced in the penetrator itself (from ^{234m}Pa).

To avoid deterministic effects on the basal cell layer of the skin (nominal depth 0.07 mm), ICRP (1990) recommends an annual equivalent dose limit of 50 mSv for

Table 4
Nuclides assumed to be present in the penetrators and taken into account in the EGS4 calculations. About ten major peaks of the photon spectra of each nuclide were taken into account in the calculations. The activities are those obtained from the gamma-ray analysis of penetrator 3 (Table 1) added to those of ^{234}U and ^{236}U from the alpha analysis

Nuclide	Activity (Bq)	Number of photons per disintegration
^{238}U	3,800,000	0.081
^{236}U	17,100	0.10
^{235}U	60,000	2.05
^{234}U	462,000	0.11
^{234}Th	3,800000	0.237
^{234m}Pa	3,800,000	0.0287
^{234}Pa	11,000	3.532

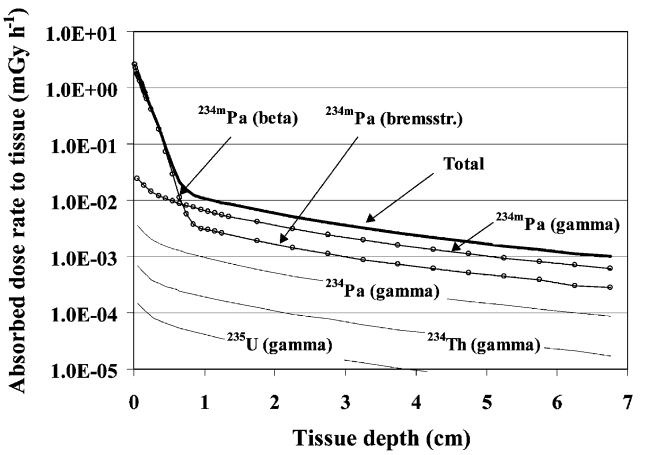

Fig. 7. Absorbed dose rate (mGy h^{-1}) as a function of tissue (water) depth calculated by the EGS4 Monte Carlo code (only the most significant nuclides are presented). The penetrator was assumed to be in contact with skin. ^{234m}Pa was the dominating nuclide with respect to the absorbed dose. The contribution of ^{234}Pa gammas was about 7% of the total dose at depths greater than 1 cm. The contribution of other nuclides, such as ^{235}U and ^{234}Th was small (^{238}U, ^{236}U and ^{234}U less than 1%).

the public, averaged over any 1 cm^2, regardless of the area exposed. For workers the limit is 500 mSv.

Equivalent skin doses were calculated as a function of distance from the penetrator. When the penetrator was assumed to be in contact with the skin, the calculated equivalent dose rate at a skin depth of 0.07 mm, averaged over 1 cm^2, was approximately 2 mSv h^{-1} (see Table 5, for beta and gamma radiation 1 mGy=1 mSv). By keeping the penetrator in the same position in the pocket, the annual equivalent dose limit for the public (50 mSv) would be exceeded in 1 exposure day (i.e. the penetrator-in-the-pocket scenario is assumed). The limit for workers would be exceeded within approximately ten days.

Table 5
Calculated and measured absorbed dose rate into soft tissue at depth of 0.07 mm and averaged over 1 cm^2 as a function of distance (distance between the surface of the penetrator and the probe). The dose is mainly caused by beta-active ^{234m}Pa

Distance (cm)	Monte Carlo calculation (mGy h^{-1})	Measurement (mGy h^{-1})
0	1.96	–
0.3	1.28	–
5	0.144	–
10	0.046	0.048
20	0.013	0.015
30	0.0059	0.0069
40	0.0035	0.0040

In order to verify the results of the Monte Carlo calculations, a portable scintillation detector (Electra DP2R) was used to determine the personal dose equivalent rate at a depth of 0.07 mm in the skin as a function of the distance from the penetrator. The detector was calibrated for absorbed dose rate in soft tissue (depth 0.07mm) using a $^{90}Sr/^{90}Y$ beta source. The reference absorbed dose rate in soft tissue in this calibration was determined by an extrapolation ionisation chamber. Differences between the calculations and the measurements were small (Table 5).

7. Conclusions

The properties of the penetrators were analysed using different methods. The penetrators were composed of DU ($^{235}U/^{238}U$ mass ratio about 0.002) with small amounts of Ti and impurities. The presence of ^{236}U reflects the use of reprocessed nuclear fuel in the enrichment process. Neither Pu nor other transuranium elements were detected. However, these results represent small subsamples and the overall homogeneity of the penetrators was not studied.

A typical portable GM dose-rate meter is not the best detector for mapping the location of the penetrators in the environment. A penetrator in contact with skin may cause an equivalent dose to skin that is not negligible. However, deterministic effects do not seem to be possible. At lesser tissue depths (<0.6 cm), the dose was attributed to mainly beta particles of ^{234m}Pa, whereas photons dominate at greater depths.

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