



Measurement of production date (age) of nanogram amount of uranium

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Abstract

JRC-Karlsruhe obtained a swipe sample from a highly enriched uranium seizure, which had taken place in 2011. Due to the very low amount of uranium (nanograms) a new method needed to be developed to determine the U production date (age). The particles on the swipe were collected on a pyrolytic graphite planchet using a vacuum impactor and they were subsequently leached with *cc*HNO₃. The “bulk” U isotopic composition (²³⁵U: 72.51 ± 0.03 wt%) and the production date (December 1992 ± 1 year) determined by MC-ICP-MS indicated that the material showed similarity with two other HEU cases seized earlier in Europe.

Keywords Nuclear forensics · Production date · Uranium · Inductively coupled plasma mass spectrometry · Swipe sample

Introduction

The nuclear forensics methods aim at providing hints on the intended use, origin, production time and history of nuclear and other radioactive materials [1–3]. The scientific results, obtained in a timely manner, support the nuclear forensic findings and may serve law enforcement as investigative leads or as evidence. Several characteristic parameters (signatures) such as physical dimensions of the material, isotopic composition of U and Pu, impurities or production date can be used to re-establish the material history, hence link the material in question to a production process or even a facility. Moreover, traditional forensic evidence associated with the material may help to identify individuals who handled the material [1, 4–9]. Determination of the production date (age dating) is based on the radioactive decay of the material and the measurement of the formed daughter

products relative to the parent nuclides. The (model) age of the material is a prominent signature as it is a so-called *predictive signature* and does not require any reference information [10, 11]. Uranium age dating is typically carried out using the decay of ²³⁴U to ²³⁰Th, achieving the quantification of both nuclides by isotope dilution mass spectrometry. Age dating measurements, however, require typically milligram amounts of material and a tedious separation due to the low abundance of the decay products [3, 12]. Using lower sample amount can result in high uncertainties due to the small quantity of the daughter nuclides and lower measurement precision, e.g. due to the higher contribution from the background. Age dating of U particles by secondary ion mass spectrometry (SIMS) would require relatively large particles (micrometer-sized) of an old material [13].

In June 2011 the Moldavian Police arrested six suspects alleged to sell kilograms of highly enriched uranium (HEU) in Chisinau, Moldova. During the arrest a glass vial, containing 4.4 g of uranium, was found (Fig. 1a). The sample was analyzed by portable gamma spectrometry (ORTEC, Micro-Detective) in the National Agency for Regulation of Nuclear and Radiological Activities (NARNRA) in Moldova. The result indicated that the sample had a ²³⁵U enrichment of more than 72%. JRC-Karlsruhe obtained a sample (a piece of paper that had been in contact with the HEU powder; referred later as a “swipe”) from the confiscated material for nuclear forensic analysis in 2017 (Fig. 1b). The HEU powder was put on a paper swipe without further alteration.

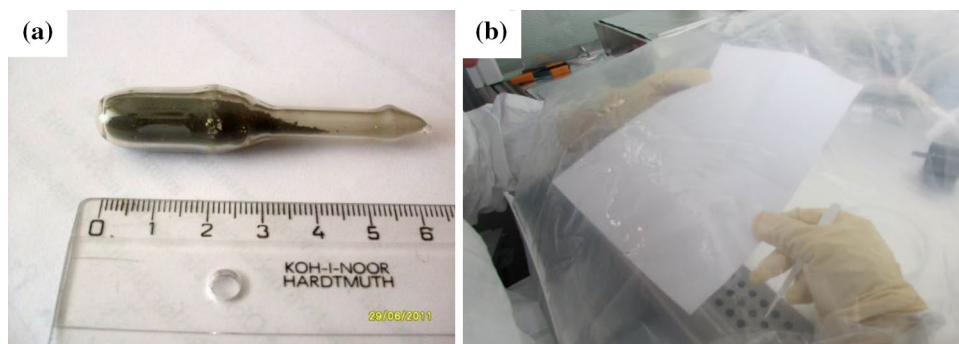
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Fig. 1 The confiscated HEU sample in Moldova in 2011 (a), a swipe sample received in JRC-Karlsruhe in 2017 (b)



As the sample amount was very low (the material was not even visible on the swipe) and the particles were small (based on the scanning electron microscopy their size was between 100 and 200 nm), only a limited number of analysis could be performed. It was decided to look into the production date (age) of the material besides the determination of the isotopic composition of U, which was performed by Large-Geometry SIMS (LG-SIMS) as a primary technique for particle analysis [14]. Due to the small size of the particles production date measurement could not be performed by LG-SIMS. Thus, a new method had to be developed, which combined the measurement of “bulk” (i.e. the average of all the particles) U isotopic composition and the production date determination of nanogram amount of a uranium sample by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS).

Theory of direct production date measurement

The ^{230}Th and ^{234}U signals are clearly visible in the MC-ICP-MS spectrum for highly enriched uranium (Fig. 2) without chemical separation, even though the lower abundant ^{230}Th

is at $\mu\text{g g}^{-1}$ level relative to the total U and no Th and U separation was done. This is due to the fact that the “minor” ^{234}U abundance is high in HEU in contrast to natural or low-enriched U samples. However, the quantification of the $^{230}\text{Th}/^{234}\text{U}$ ratio is difficult due to the different ionisation efficiencies of Th and U, the matrix effect and without using isotope dilution (i.e. use of ^{229}Th or ^{233}U as a spike).

The relation between the ^{234}U decaying to ^{230}Th can be expressed with the radioactive decay rules (Bateman equation):

$$\frac{N_{230\text{Th}}}{N_{234\text{U}}} = \frac{\lambda_{234\text{U}}}{\lambda_{230\text{Th}} - \lambda_{234\text{U}}} (e^{-\lambda_{234\text{U}}t} - e^{-\lambda_{230\text{Th}}t}) + \frac{N_{230\text{Th}}^0}{N_{234\text{U}}} e^{-\lambda_{230\text{Th}}t} \quad (1)$$

where the N refers to the amount of the nuclides, λ is the decay constant. Assuming that the U purification resulted in a complete separation of the ^{230}Th decay product, in consequence, there is no ^{230}Th in the sample at time zero:

$$\frac{N_{230\text{Th}}}{N_{234\text{U}}} = \frac{\lambda_{234\text{U}}}{\lambda_{230\text{Th}} - \lambda_{234\text{U}}} (e^{-\lambda_{234\text{U}}t} - e^{-\lambda_{230\text{Th}}t}) \quad (2)$$

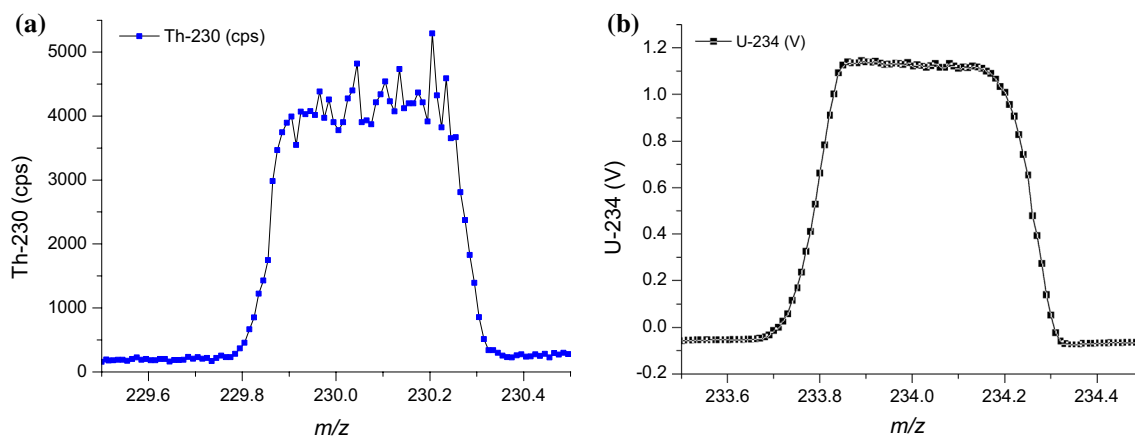


Fig. 2 Simultaneously detected signals of ^{230}Th (ion counter) and ^{234}U (Faraday detector) by MC-ICP-MS using 300 ng g^{-1} of CRM U630

As the decay constants are small, using $e^x \approx 1 + x$ approximation if $x \ll 1$ (Taylor-series), one can re-write the equation:

$$\frac{N_{230\text{Th}}}{N_{234\text{U}}} = \frac{\lambda_{234\text{U}}}{\lambda_{230\text{Th}} - \lambda_{234\text{U}}} (\lambda_{230\text{Th}} - \lambda_{234\text{U}}) t \approx \lambda_{234\text{U}} t \quad (3)$$

For a 20-year-old sample, the simplification results in -0.13% bias. The equation implies that the ingrowth of ^{230}Th is approximately linear and depends on the decay constant of ^{234}U . Thus, measuring the $^{230}\text{Th}/^{234}\text{U}$ ratio the production date can be calculated easily. The directly obtained $^{230}\text{Th}/^{234}\text{U}$ intensity ratio measured by MC-ICP-MS, however, is not equal to the “true” ^{230}Th and ^{234}U ratio in the sample due to U/Th ionisation difference, the mass bias effects and relative detector efficiencies. Within one sequence these values can be considered constant, the measured $^{230}\text{Th}/^{234}\text{U}$ intensity ratio thus being proportional to the $^{230}\text{Th}/^{234}\text{U}$ amount ratio in the sample:

$$\frac{N_{230\text{Th}}}{N_{234\text{U}}} \approx \lambda_{234\text{U}} t = f \frac{I_{230\text{Th}}}{I_{234\text{U}}} \quad (4)$$

where $I_{\text{Th-230}}$ and $I_{\text{U-234}}$ are the measured intensities by MC-ICP-MS for ^{230}Th and ^{234}U , respectively. Merging all the constants together, the measured $^{230}\text{Th}/^{234}\text{U}$ intensity ratio is proportional to the production date (time elapsed since the last chemical purification):

$$\frac{I_{230\text{Th}}}{I_{234\text{U}}} = f' t \quad (5)$$

where f' constant includes Th/U ionisation difference, detector relative efficiencies, mass bias of ^{230}Th and ^{234}U and the ^{234}U decay constant. Thus, the f' constant can be calculated using a reference material with certified production date (in our case CRM U630 was used) after the measurement of the $^{230}\text{Th}/^{234}\text{U}$ intensity ratio. This f' value can be then applied to the unknown sample to calculate the production date after the measurement of its $^{230}\text{Th}/^{234}\text{U}$ intensity ratio.

The developed method is applicable for highly enriched uranium (i.e. containing relatively high amount of ^{234}U), which is reasonably pure from other elements. For instance, “dirty” swipe samples can contain high amounts of Pb, e.g. from lead shielding, and due to the possible molecular interferences (e.g. $^{204}\text{Pb}^{12}\text{C}^{14}\text{N}^+$) erroneous results can be obtained. This analysed sample contained HEU particles on a piece of paper, which had been in touch only with the bulk HEU powder. Therefore the blank level was expected to be low.

In addition to that, the method is obviously applicable only for HEU with single U composition and not for

mixtures containing e.g. natural uranium. This sample was analysed prior to the age determination by SIMS, which showed that the HEU particles had all the same isotopic composition and no other uranium particles, with different composition, were detected.

Experimental

Reagents and materials

All labware was thoroughly cleaned before use. Suprapur grade nitric acid (Merck, Darmstadt, Germany) was used for the sample preparation, which was further purified by sub-boiling distillation (AHF analysentechnik AG, Tübingen, Germany). For dilutions ultrapure water was used (Elga LabWater, Celle, Germany). Perfluoralkoxy (PFA) vials with a diameter of 22 mm (volume: 3 mL) were purchased from AHF analysentechnik AG (Tübingen, Germany). Polished pyrolytic graphite planchets were obtained from ANAME Instrumentación Científica (Madrid, Spain).

Instrumentation

A NuPlasmaTM (NU Instruments, Wrexham, United Kingdom) double-focusing multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS), equipped with 11 Faraday detectors and 3 discrete dynode electrode multipliers was used for the U isotope abundance and age dating measurements. The resistors fitted to the Faraday detectors were $10^{11} \Omega$. The instrument was operated in low mass resolution mode ($R = 300$). The samples were introduced using an Aridus II desolvation unit (CETAC Technologies Inc., Omaha, NE, USA) as a solution. Detailed optimised instrumental settings and data acquisition parameters are given in Table 1.

The ICP-MS was optimised daily (torch position, gas flows, voltages) using a 50 ng g^{-1} multi-elemental solution (Inorganic Ventures, Christiansburg, USA). The optimisation aimed at achieving highest sensitivity and stability of the acquired U signal. The ^{234}U signal was aimed to about 1 V intensity for the age dating measurement and only the ^{230}Th and ^{234}U signals were measured.

For the U isotopic composition determination the isotopes ^{234}U , ^{235}U , ^{236}U and ^{238}U were measured on the Faraday detectors, while for the age dating ^{230}Th was measured on the ion counter equipped with a retardation filter and ^{234}U was measured simultaneously on a Faraday detector. The retardation filter improves the abundance sensitivity on $m/z = 230$ by a factor of ~ 10 . The measurement of the age dating was done in one sequence to make sure that Th and U sensitivities, detector efficiencies and mass bias (i.e. f' in Eq. 5) were constant. Calibrant to calculate the f' was

Table 1 MC-ICP-MS operating parameters

MC-ICP-MS instrument settings	
Forward power (W)	1300
Cooling gas flow rate (L min ⁻¹)	14.0
Auxiliary gas flow rate (L min ⁻¹)	0.98–1.05
Nebulizer gas flow rate (L min ⁻¹)	0.9–0.98
Sample introduction conditions	
Solution uptake rate (μL min ⁻¹)	80
Spray chamber temperature (°C)	110
Membrane temperature (°C)	160
Sweep gas flow rate (L min ⁻¹)	6.80
Data acquisition	
Mass resolution	~300
Number of spectra acquired	6×5
Magnet delay between blocks (s)	2
Scan type	Static multi-collection
Cup configuration—U isotopic measurements ^a	L1: ²³⁴ U, Axial: ²³⁵ U, H1: ²³⁶ U, H3: ²³⁸ U
Cup configuration—age dating measurements ^a	IC0: ²³⁰ Th, H1: ²³⁴ U

^aL1, Axial, H1, and H3 denote Faraday detectors; IC0 denotes a discrete dynode electron multiplier operated in pulse counting mode equipped with a retardation filter

measured before each sample. The MC-ICP-MS measurement was performed in the following order: Calibrant—Quality control—Calibrant—Sample—Calibrant—Quality control. Dilute HNO₃/HF mixture between the samples was used to remove the Th and U traces.

Measured U samples

The used certified reference materials (CRMs), U500, U850 and CRM U630, are U standard reference materials in the form of U₃O₈ (New Brunswick Laboratory Argonne, IL, USA) and they have 50, 85 and 63% nominal enrichment of ²³⁵U, respectively. All CRMs are certified for their U isotopic composition, while the CRM U630 is certified for model purification date as well (certified model date: 6 June, 1989 with an uncertainty of 190 days). For the U isotopic measurements the mass bias was determined using the U500 CRM whereas for the quality control (to check the U isotopic abundances) U850 CRM was used. For the production date measurement the ²³⁰Th/²³⁴U amount ratio and the age was calculated after calibrating with the CRM U630 radiochronometric standard. For quality control purpose a 70% highly enriched uranium (HEU-70) was used (known production date: 19 July, 2011), which has been prepared in the JRC-Karlsruhe, afterwards analysed and validated by several international laboratories [10]. The concentration of the samples for age measurement was approximately 250 ng g⁻¹ U.

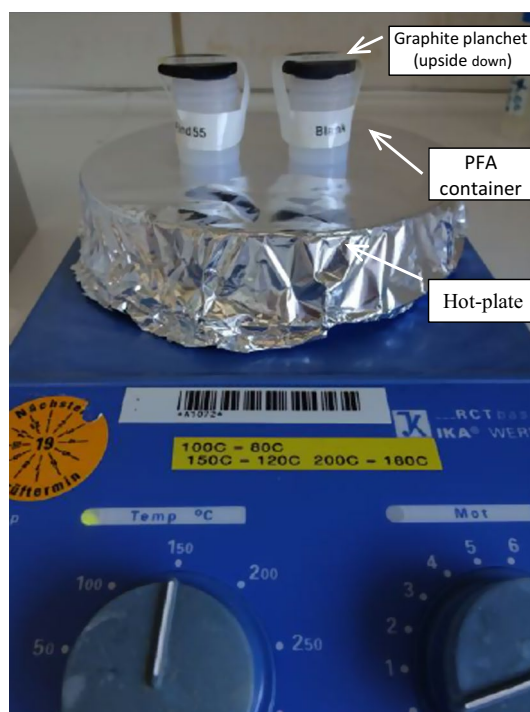


Fig. 3 U leaching from the pyrolytic graphite planchet

Sample preparation

As the U amount in the sample was limited, chemical separation or spiking was not possible. After careful considerations and performing tests using a blank and standard, the following method was chosen: the particles on the swipe sample were collected on a pyrolytic graphite planchet using the vacuum impactor method in a clean laboratory as done for SIMS analysis routinely [14–16]. The sample was vacuumed along the whole swipe to collect as many particles as possible. For fixing the particles on the graphite planchet, polyisobutylene (PIB) was added. The planchet was baked after collection to remove the PIB to avoid any interference during the MC-ICP-MS measurement. The U particles were then leached out from the planchet and dissolved in *cc*HNO₃ using a specially developed procedure to minimize the background and consumption of chemicals (Fig. 3). The graphite planchet was placed and fixed on the top of a PFA vial containing 3 mL of *cc*HNO₃ facing the liquid phase. The planchet was not immersed in *cc*HNO₃ to minimize the background and avoid graphite residues. Covering the PFA vial with the graphite planchet separated the leachate from the atmospheric environment. The diameter of the PFA vial was chosen in such a way that its diameter was slightly larger than the center (i.e. the U particles containing) part of the planchet. The vial was placed on a hot plate for 24 h to make sure that all U was leached out from the planchet and was got in the liquid phase. The temperature of the hot plate was

slightly below the boiling point of the HNO_3 , thus it resulted in reflux, which dissolved the particles on the planchet and the nitric acid dropped back in the vial. As U is not volatile, the U solution remained in the PFA container. A blank was prepared together with the sample. The dissolution was tested beforehand with particles of known U isotopic composition (i.e. U010 CRM) to optimize the leaching time, to measure the background and check the leaching efficiency. Tests showed that more than 90% of the U could be recovered with the above described method, while the U background was approximately 17 pg. Lower recoveries were obtained if the amount of ccHNO_3 was lower, reflux time was insufficient or the temperature was low. During testing the isotopic composition of the leached U010 CRM was measured by MC-ICP-MS and compared to SIMS results on individual particles. The results agreed well, thus no cross-contamination took place during the sample preparation procedure.

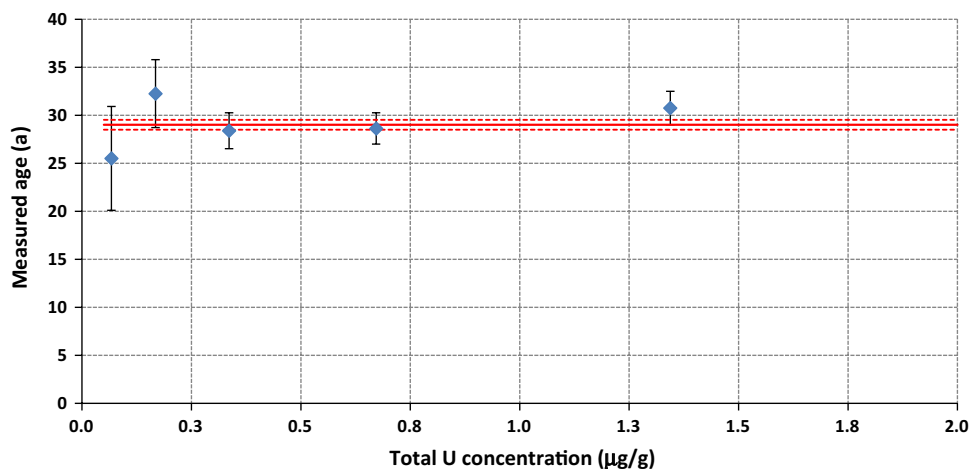
The same procedure was applied to the “real” sample. It was evaporated and taken up in 1.5 mL 4% HNO_3 . About 390 ng U could be recovered from the sample. The sample was split in two fractions in order to measure the “bulk” (i.e. the average of the particles) U isotopic composition (about 10% of the aliquot) and the production date (about 90% of the aliquot) by MC-ICP-MS.

Results and discussion

Minimum U amount for age dating

Using U630 one can estimate the minimum amount needed for the age dating using the direct measurement by MC-ICP-MS (Fig. 4). Measuring increasing concentration of U, the required U amount for an accurate and precise measurement can be determined.

Fig. 4 Age dating results with expanded uncertainties ($k=2$) using a CRM U630 by MC-ICP-MS with increasing U concentration. The solid line is the certified model purification age (29.01 ± 0.52 years). Reference date: 28 June, 2018



Using about 300 ng of U (with about 0.15 pg ^{230}Th) one can get an age result, which agrees with the model purification date and which is precise enough. Above this U amount the precision of the measurement, which is about 6% relative to the measured value, will not increase significantly.

Results of the swipe sample

“Bulk” U isotopic composition

The U isotopic composition of the swipe sample is given in Table 2. The MC-ICP-MS results for the U “bulk” isotopic composition agreed well with the LG-SIMS results (average $^{235}\text{U} = 72.6\%$) performed on 41 individual particles. This indicated that no cross-contamination had taken place during the sample preparation. The blank was negligible.

Age dating result

The measured production date for the sample together with the HEU-70 results is summarized in Table 3. The reported uncertainties are expressed as expanded uncertainties with a coverage factor of $k=2$. The half-lives of ^{234}U and ^{230}Th applied are $245,250 \pm 490$ years and $75,690 \pm 230$ years, respectively. The uncertainties took into account the calibrant model age uncertainty as well as the measured calibrant and sample signal uncertainties. The f' values

Table 2 “Bulk” U isotopic composition of the swipe sample by MC-ICP-MS

	Amount (%)	$U (k=2)$	Relative U (%)
^{234}U	1.1740	0.0008	0.07
^{235}U	72.67	0.03	0.04
^{236}U	12.11	0.01	0.08
^{238}U	14.040	0.003	0.02

Table 3 Age dating measurement sequence and obtained results

Sample	f' (1/a) of CRM U630	Model age (a)	Production date
HEU-70 No.1 ^a	0.53 ± 0.01	6.8 ± 0.2	September 2011 ± 0.2 years
Swipe	0.53 ± 0.01	25.6 ± 1.0	December 1992 ± 1.0 years
HEU-70 No.2 ^a	0.52 ± 0.01	7.0 ± 0.2	June 2011 ± 0.2 years

^aThe known production date is 19 July, 2011, which corresponds to 6.941 years. The production date has < 0.02 years uncertainty [10]. Reference date and date of measurement: 28 June, 2018

calculated from the CRM U630, which were measured always right before the samples (HEU-70 or the swipe sample), were stable throughout the measurement sequence, and showed no change during the analysis. The results obtained for the HEU-70, before and after the swipe sample measurement, (September 2011 and June 2011, respectively) agreed well with the known production date of 19 July, 2011 [10]. The individual results are shown in the Supplementary Information.

The age dating result for the swipe sample is December 1992 with an uncertainty of 1.0 years. The uncertainty is mainly determined by the ²³⁰Th/²³⁴U ratio measurements and CRM U630 model purification date uncertainty making up > 95% of the overall uncertainty, other contributions (e.g. from the half-life uncertainties) are minimal.

Conclusions

A novel sample preparation and measurement procedure was developed and validated for the “bulk” U isotopic composition and age determination from a swipe sample containing only nanogram amounts of U by MC-ICP-MS. The U isotopic information could be used to complement the LG-SIMS results performed on individual U particles. Using these two main characteristics, i.e. isotopic composition and age of uranium, one could link the HEU seized in Moldova to two other similar illicit trafficking cases, which had taken place around 10 years earlier in Bulgaria [17] and in France [18]. First of all, the packaging of the sample in all cases was very similar: cylindrical lead container lined with yellow paraffin wax and the HEU powder was inside a flamed-sealed glass ampoule. The U isotopic composition in the Bulgarian case was ²³⁴U: 1.2%, ²³⁵U: 72.7%, ²³⁶U: 12.1% and ²³⁸U: 14.0% (all values are atom-%, uncertainties are less than 0.5%) [17], while in the French seizure the U isotopic composition was ²³⁴U: 1.17 ± 0.02, ²³⁵U: 72.57 ± 0.86, ²³⁶U: 12.15 ± 0.14 and ²³⁸U: 14.11 ± 0.08 [18]. Therefore, both the Bulgarian and French HEU cases agree well with the HEU seized in Moldova. The age dating results differed about one year between the Bulgarian and French cases being 30 October, 1993 ± 50 days, and November 1994 ± 100 days, respectively [19]. Thus, the age dating result for the HEU found in Moldova is in agreement

with the Bulgarian seizure. In conclusion, the HEU material found in three different European countries within the time span of 12 years is very likely coming from the same source material. Whether the HEU in different cases originates from the same batch, i.e. was purified at the same time, can be questioned. However, the link between the cases is evident.

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References

1. Mayer K, Wallenius M, Varga Z (2013) Nuclear forensic science: correlating measurable material parameters to the history of nuclear material. *Chem Rev* 113(2):884–900. <https://doi.org/10.1021/cr300273f>
2. Keegan E, Kristo MJ, Toole K, Kips R, Young E (2016) Nuclear forensics: scientific analysis supporting law enforcement and nuclear security investigations. *Anal Chem* 88(3):1496–1505. <https://doi.org/10.1021/acs.analchem.5b02915>
3. Mayer K, Wallenius M, Varga Z (2015) Interviewing a silent (radioactive) witness through nuclear forensic analysis. *Anal Chem* 87(23):11605–11610. <https://doi.org/10.1021/acs.analchem.5b01623>
4. Varga Z, Krajčák J, Peňkin M, Novák M, Eke Z, Wallenius M, Mayer K (2017) Identification of uranium signatures relevant for nuclear safeguards and forensics. *J Radioanal Nucl Chem* 312(3):639–654. <https://doi.org/10.1007/s10967-017-5247-5>
5. Tandon L, Hastings E, Banar J, Barnes J, Beddingfield D, Decker D, Dyke J, Farr D, FitzPatrick J, Gallimore D, Garner S, Gritzko R, Hahn T, Havrilla G, Johnson B, Kuhn K, LaMont S, Langner D, Lewis C, Majidi V, Martinez P, McCabe R, Mecklenburg S, Mercer D, Meyers S, Montoya V, Patterson B, Pereyra RA, Porterfield D, Poths J, Rademacher D, Ruggiero C, Schwartz D, Scott M, Spencer K, Steiner R, Villarreal R, Volz H, Walker L, Wong A, Worley C (2008) Nuclear, chemical, and physical characterization of nuclear materials. *J Radioanal Nucl Chem* 276:467–473
6. Mayer K, Wallenius M, Fanghänel T (2007) Nuclear forensic science—from cradle to maturity. *J Alloys Compd* 444–445:50–56
7. Wallenius M, Mayer K, Ray I (2006) Nuclear forensic investigations: two case studies. *Forensic Sci Int* 156:55–62

8. Varga Z, Wallenius M, Mayer K, Meppen M (2011) Analysis of uranium ore concentrates for origin assessment. *Radiochim Acta* 1:1–4
9. Keegan E, Richter S, Kelly I, Wong H, Gadd P, Kuehn H, Alonso-Munoz A (2008) The provenance of Australian uranium ore concentrates by elemental and isotopic analysis. *Appl Geochem* 23:765–777
10. Varga Z, Mayer K, Bonamici CE, Hubert A, Hutcheon I, Kinman W, Kristo M, Pointurier F, Spencer K, Stanley F, Steiner R, Tandon L, Williams R (2015) Validation of reference materials for uranium radiochronometry in the frame of nuclear forensic investigations. *Appl Radiat Isot* 102:81–86. <https://doi.org/10.1016/j.apradiso.2015.05.005>
11. Kristo MJ, Tumey SJ (2013) The state of nuclear forensics. *Nucl Instrum Methods Phys Res Sect B* 294:656–661
12. Wallenius M, Morgenstern A, Apostolidis C, Mayer K (2002) Determination of the age of highly enriched Uranium. *Anal Bioanal Chem* 374:379–384
13. Fauré AL, Dalger T (2017) Age dating of individual micrometer-sized uranium particles by secondary ion mass spectrometry: an additional fingerprint for nuclear safeguards purposes. *Anal Chem* 89(12):6663–6669. <https://doi.org/10.1021/acs.analchem.7b00887>
14. Ranebo Y, Hedberg PML, Whitehouse MJ, Ingeneri K, Littmann S (2009) Improved isotopic SIMS measurements of uranium particles for nuclear safeguard purposes. *J Anal At Spectrom* 24(3):277–287. <https://doi.org/10.1039/b810474c>
15. Esaka F, Watanabe K, Fukuyama H, Onodera T, Esaka KT, Magara M, Sakurai S, Usuda S (2004) Efficient isotope ratio analysis of uranium particles in swipe samples by total-reflection X-ray fluorescence spectrometry and secondary ion mass spectrometry. *J Nucl Sci Technol* 41(11):1027–1032. <https://doi.org/10.1080/18811248.2004.9726327>
16. Hedberg PML, Peres P, Cliff JB, Rabemananjara F, Littmann S, Thiele H, Vincent C, Albert N (2011) Improved particle location and isotopic screening measurements of sub-micron sized particles by secondary ion mass spectrometry. *J Anal At Spectrom* 26(2):406–413. <https://doi.org/10.1039/c0ja00181c>
17. Moody KJ, Hutcheon ID, Grant PM (2005) *Nuclear forensic analysis*. CRC Press, Boca Raton
18. Baude S, Chartier B, Kimmel D, Mariotte F, Masse D, Peron H, Tilly D (2007) The French response in cases of illicit nuclear trafficking lessons learned from a real case (IAEA-CN-154/062). In: International conference on illicit nuclear trafficking: collective experience and the way forward, Edinburgh, Scotland, 2007. International Atomic energy Agency, Vienna
19. Kristo MJ, Gaffney AM, Marks N, Knight K, Cassata WS, Hutcheon ID (2016) Nuclear forensic science: analysis of nuclear material out of regulatory control. *Annu Rev Earth Planet Sci* 44:555–579

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