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Preparation and Certification of IRMM-1027k, Large-Sized Dried (LSD) spike

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irmm
Institute for Reference Materials and Measurements

The mission of the IRMM is to promote a common and reliable European measurement system in support of EU policies.

European Commission
Joint Research Centre
Institute for Reference Materials and Measurements

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Contents

Summary

The concept of directly spiking samples of the solution with dissolved irradiated nuclear fuel for later dilution and measurement of the isotopic contents of uranium and plutonium has been developed successfully. Large sized dried spikes (LSD) have become a fundamental part of the fissile material control of irradiated nuclear fuel. In the frame of providing these spikes to the nuclear industry, a new set of LSD Spikes for the determination of uranium and plutonium by isotope dilution mass spectrometry in solutions of spent fuel from reprocessing plants has been prepared and certified for uranium and plutonium isotopic contents. The methodology followed was comparable to that of previous batches. The solution, made by dissolution of the starting materials in nitric acid, was dispensed directly into individual penicillin vials. However in this campaign an automated system was introduced to dispense and weigh the majority of vials. At the same time the system was validated by comparing the results with those of the traditional syringe dispensing and substitution weighing.

This new batch of large size dried spikes contains ca. 50 mg of uranium (^{235}U abundance = 19.1%) and ca. 1.8 mg of plutonium (^{239}Pu abundance = 97.8%) in each individual vial, covered with a light layer of organic material (cellulose acetate butyrate) as stabilizer.

The U and Pu amount content was certified based on values from mass metrology of the validated automated system or from the manual weighings. Verification of the amount contents of the spike was done by IDMS at IRMM. The values measured for the batch solution and of the dried covered spikes agreed well with those calculated from the weights of starting materials dissolved and the weights of the final solution.

Introduction

The series IRMM-1027k Large Size Dried (LSD) Spikes is being prepared to fulfil the existing requirement for reliable and traceable spikes in fissile material control of dissolved nuclear fuel. The amount content of the spikes is such that no dilution of a typical sample of dissolved fuel is needed before measurement by Isotope Dilution Mass Spectrometry (IDMS) using a single LSD spike. Because each spike is certified for amounts of plutonium and uranium in the vial, the only quantitative step needed at the reprocessing plant laboratory is to weigh as accurately as possible an aliquot of the dissolved fuel solution onto the spike and ensure complete mixing of spike and sample.

The plutonium component is highly enriched in ^{239}Pu and is used to measure the Pu content in the fuel. Approximately 1.8 mg Pu is contained in each LSD spike. The uranium component is a mixture of two uranium source materials, natural uranium and a highly enriched uranium component. These materials are mixed to arrive at a final enrichment of just under 20% in ^{235}U, which means for accountability purposes the uranium is classified as ‘low enriched’.

High purity metals are chosen as starting materials. For this campaign it was decided to use CETAMA MP2 plutonium metal and uranium metals EC NRM 101, CRM-116 as in most previous batches. This allows the isotopic contents of the LSD spike to be certified from the certificates of the metals (chemical purity and isotopic content), the weights of the metals and the solution. As a result the values of the uranium and plutonium isotopic contents of the final certified spike solution have low uncertainties which are directly traceability to the SI via the masses of the starting materials.

A single large volume of batch solution is made up; the first 192 were dispensed and weighed manually, the rest up to 1200 units by the automated system, into a number of penicillin vials. The solution in each vial is dried down and then covered with a light organic coating dried onto the spike material. The coating (cellulose acetate butyrate, CAB) was also used for previous batches. It provides a fixed layer to hold the dried spike material on the base of the vial, dissolves quickly in warm nitric acid and has no significant effect on the subsequent IDMS measurements.

Following the experience of previous series, in particular IRMM-1027e, 1027f, 1027g, 1027h, 1027i and 1027j, the isotopic contents of the batch solution and of a set of individual spike vials after drying are measured by isotope dilution to verify the values from the mass-metrology of the starting metals dissolved and the weight of the final solution.

Dissolution of standard materials

Pu Metal Cetama MP2

The metal standard is delivered in a flame-sealed vial with a certified mass of Pu metal. Four ampoules of MP2 were required for the preparation of this LSD spike. Each vial was cut open, the Pu removed with tweezers, weighed and placed in the 3 L borosilicate flask (see next paragraph). The total amount of Pu, calculated to obtain a solution of ca. 0.7 mg plutonium per gram solution when dissolved in 3 kg nitric acid, was weighed at IRMM. The results from the weighin of metal agreed well with the CETAMA certified mass of the MP2 metal.

Uranium metals EC NRM 101, CRM-116

Approximately 48.8 g EC NRM 101 (natural uranium) metal was etched with 1 M HNO_3 as recommended on the certificate to remove surface oxides, rinsed with de-ionised water then acetone and finally dried. The metal was accurately weighed and added to the flask containing the Pu solution. The same was done with 12.0 g NBL CRM-116 enriched uranium. The masses of the uranium were calculated so as to yield a solution of ca. 20 mg uranium per gram solution with an enrichment of ca. 19.8% in ^{235}U.

Making up the batch solution

The dissolution was carried out entirely in a 3 L long-necked borosilicate flask that had been cleaned in the IRMM MCL (Medium-Clean Chemistry Laboratory). All weighings were carried out as accurately as possible, with reference to a set of calibrated weights traceable to the international kilogram prototype at BIPM, Sèvres. The necessary corrections for air buoyancy effects, taking into account the ambient pressure, temperature, humidity and the density of the material were made.

The weighed Pu metal was transferred into the flask. Concentrated nitric acid and a few drops of conc. HF were added and the flask was warmed to about 90° C to dissolve the Pu. The dissolution was controlled visually and took several weeks to be complete. After cooling the solution was kept under controlled conditions to ensure complete Pu dissolution before the uranium was added. The uranium dissolved quickly and completely within a few days.

The complete dissolution of the metals and the solution homogeneity was ensured by allowing the solution to stand for at least 8 weeks after the starting materials had been adjudged to be completely dissolved.

After making up the solution to the prescribed mass of 3.1 kg, the solution was left for another 4 weeks to homogenise with occasional swirling by hand.

Measurement of isotopic abundances in the batch solution

A plastic syringe (50 mL) was filled from the batch solution and from this syringe 6 aliquots of 1 g were weighed into a set of glass vials. These were then spiked with 5 g each of IRMM-046b double spike (^{233}U+^{242}Pu) for isotope dilution mass spectrometry (IDMS). One extra vial containing ca. 1 g of solution was not spiked and was processed for measurement of isotopic ratios.

The chemical procedure prior to mass spectrometry as detailed in [1] was employed. A 1 M HNO_3 solution of uranium and of plutonium separated from the spiked and unspiked solutions were prepared for measurements of the isotopic ratios by TIMS.

The isotopic ratios of the uranium were measured on the Finnigan Triton and those of plutonium on the Finnigan MAT 262, following IRMM Quality Management procedures PR-077 for uranium and PR-075 for plutonium. The mass-spectrometers were calibrated for mass-fractionation by measuring IRMM-184 uranium isotopic reference material and IRMM-290 plutonium isotopic reference material during the procedure.

The measured ratios compared to the calculated values from the certificates are listed in Table 1 for uranium and Table 2 for plutonium. The certified ratios for uranium are taken from the Triton measurements and are compared to the ratios calculated from the mixing of the two metals and their certified isotopic abundances. The certified ratios for Pu are taken from the recertification of MP2 at IRMM (2007) as in the IRMM certificate in Annex 3 and decay corrected to 22 August 2007 date of verification measurements.

Table 1: Isotopic amount ratios of uranium in the batch solution. Values from certificates and metrological weighing are compared with abundances calculated from measurement of isotopic ratios in a sample of the batch solution. Expanded Uncertainties are given in brackets (coverage factor k=2).

	$n(^{234}U)/n(^{238}U)$	$n(^{235}U)/n(^{238}U)$	$n(^{236}U)/n(^{238}U)$
Calculated value	0.002 514(10)	0.237 382(54)	0.001 033 8(32)
Measured/Certified value	0.002 513 6(11)	0.237 407(77)	0.001 035 42(69)

Table 2: Isotopic amount ratios of plutonium in the batch solution. Values from the certificate are compared with certified values calculated from measurement of isotopic ratios in a sample of the batch solution. Expanded Uncertainties are given in brackets (coverage factor k=2).

	$n(^{238}Pu)/n(^{239}Pu)$	$n(^{240}Pu)/n(^{239}Pu)$	$n(^{241}Pu)/n(^{239}Pu)$	$n(^{242}Pu)/n(^{239}Pu)$
Certified value	0.000 030 68(29)	0.022 431 3(51)	0.000 230 5(30)	0.000 075 71(78)
Measured value	0.000 033 4(22)	0.022 428 6(45)	0.000 229 6(12)	0.000 076 0(18)

Verification of U and Pu amounts in the batch solution

The series of 6 spiked solutions described above together with the vial containing the unspiked sample were heated to dryness, then chemically conditioned and the U and Pu fractions separated by the standard ion-exchange method (*Work Instructions: 042 'Spiking, isotopic exchange and preliminary separation for mixtures of uranium and plutonium'; 041 'Separation and purification of uranium for measurement of isotopic ratios by TIMS'; 035 'Separation of Pu for TIMS measurements of isotopic ratios for IDMS or for isotopic abundances'*).

For the plutonium, a magazine was loaded with one filament for each blend; the remaining positions were filled by IRMM-290 standards. The plutonium was measured on the MAT 262 (*Work Instruction: 115 'Pu isotopic measurements in total evaporation using the MAT 262'*). The uranium measurements were carried out on the Triton (*Working Instruction 149 'measurement of uranium isotopic ratios by the TIMS TRITON'*). One filament per blend was loaded; the remaining positions per magazine were loaded with the unspiked sample (6x) and IRMM-184 U isotopic reference material. The Triton measurements were carried out using the Modified Total Evaporation technique. The method is also described in detail in [2].

The results of these IDMS measurements are given in Table 3 and Table 4 and shown in Figs. 1 and 2 compared with the U and Pu amount contents as calculated from the masses of the dissolved metals and solutions, taking into account the certified chemical purities of

the starting materials and making corrections for isotopic decay. There was good agreement between the isotopic amounts of U and Pu measured by IDMS and those calculated from the masses of the starting materials and the final solution.

Aliquots of the solution were subsequently dispensed into penicillin-type vials.

Aliquoting of batch solution

The solution in the flask was re-weighed and adjusted for the small evaporation losses during the time the verification of the batch measurements were done. Prior to dispensing, the vials were cleaned (*Working Instruction 071 ' Cleaning of glass penicillin vials for storage of LSD spikes)*, pre-engraved with the reference material name (IRMM-1027k) and an individual running number starting at 0001.

Manual aliquoting

Aliquots of the solution of about 2.5 g were transferred into penicillin-type vials using a commercial, manually operated dispenser. The first 192 aliquots were dispensed and weighed manually into a number of penicillin vials *(Working Instruction 185 'Mass determination substitution weighing')*. Each vial contained ca. 1.8 mg Pu and 50 mg U. Metrological weighings were carried out on an analytical balance (Mettler AT261 Delta range). The amount dispensed into the vial was measured by weighing the empty vial and weighing after adding the solution.

Automated system aliquoting

The automated system to produce LSD spikes has been installed in collaboration with Nucomat, a company with a recognized reputation in design and development of integrated automated systems. The major components of the system are a robot, two balances, a dispenser and a drying unit fitted into a glove box [3].

The robot is software driven and designed to control all movements inside the glove-box, to identify the penicillin vials with a barcode reader, to dispense the LSD batch solution into the vials and to weigh the amount dispensed. The weighing section is equipped with a semi-analytical balance (Sartorius TE124S) and a 5 kg balance (Sartorius TE6101) to monitor the mass of the mother solution during dispensing and to verify overnight losses by evaporation.

The system functionality has been evaluated and the performance validated by comparing the results from a series of samples dispensed and weighed by the automated system with the results by manual substitution weighing. A number of known sources of discrepancy needed to be corrected for. They involve a correct calibration of the balance, adjustment for the modified weighing pan, compensation of evaporation effects during the time elapsing between Nucomat and mass metrology service weighing, compensation for the assumed object density to make a direct readout from a balance comparable with the result of a substitution weighing,

After applying the proper correction factors to the data from the automated system balance no significant difference was observed between the two. However, an additional component of uncertainty of $3 \cdot 10^{-4}$ is introduced in the uncertainty budget for the certified weights provided by the automatic system.

The LSD spike solution was weighed into the vials over a period of about 5 days *(Working Instruction 299 'LSD automated system equipment manual')*. Batches of 48 vials were prepared and kept in a Perspex holder that fitted into a plastic box and each box was closed and stacked with the others ready for drying. The boxes with the penicillin vials were transferred into one of the drying glove-boxes for the next processes: drying and covering with CAB.

Drying solutions and addition and drying of CAB

The solutions were dried by gentle heating on a thermostatically controlled hot-plate at approx. 55° C. When the solutions had dried (typically 4-5 days continuous heating), about 0.7 mL of a 10% cellulose acetate butyrate (CAB) solution in acetone was added, the solution allowed to evaporate at room temperature for two hours and then heated at approx. 50° C for up to two hours to dry completely.

Two separate glove-boxes were used for the drying allowing up to 48 samples per week to be dried and covered with CAB. The vials containing dried samples were stacked horizontally and inspected regularly. If the material appeared to have flowed even slightly in the vial the vial was heated again to remove the last traces of solvent. The vials containing the dried material covered with CAB were closed with an iso-versilic stopper and an aluminium cap. The vials were then labelled and sealed in PVC packages for storage.

Drying, coating with CAB layer and packing were carried out over a period of several months.

Verification of U and Pu amount content in selected vials

After drying and CAB covering were complete, three series of six vials were randomly stratified chosen for verification measurements. The first series of six was selected from the vials manually dispensed, series 2 and 3 were selected from the vials delivered through the automated system. To each of these, 5 g of IRMM-046b spike was weighed in and the standard IDMS procedure and the working instructions above were used for the measurement of U and Pu amount content in the spikes. The uranium isotopic ratios $n(^{235}U)/n(^{233}U)$ were measured on the Triton, using the same procedure that was used for the batch solution verifications and the plutonium ratios $n(^{239}Pu)/n(^{242}Pu)$ were measured on the MAT 262.

The results of the verification measurements described above are given in Table 3 and Table 4 and shown in Figs. 1 and 2. These measurements gave values that agreed well with the values for uranium and plutonium amount content calculated from the amounts of dissolved metals and solution.

Table 3: Amount content of uranium in $mol \cdot g^{-1}$. Values from certificates and metrological weighing are compared with values calculated from measurement of samples of the batch solution and from vials. Expanded Uncertainties are given in brackets (coverage factor k=2).

Certificate	Batch	Vials series A	Vials series B	Vials series C
$8.174\,56(74) \cdot 10^{-5}$	$8.174\,8(44) \cdot 10^{-5}$	$8.170\,1(47) \cdot 10^{-5}$	$8.167\,0(60) \cdot 10^{-5}$	$8.168\,7(42) \cdot 10^{-5}$

Table 4: Amount content of plutonium in $mol \cdot g^{-1}$. Values from the certificate and metrological weighing are compared with values calculated from measurement of samples of the batch solution and from vials. Expanded Uncertainties are given in brackets (coverage factor k=2).

Certificate	Batch	Vials series A	Vials series B	Vials series C
$2.956\,7(13) \cdot 10^{-6}$	$2.958\,7(47) \cdot 10^{-6}$	$2.956\,3(47) \cdot 10^{-6}$	$2.955\,9(49) \cdot 10^{-6}$	$2.956\,3(46) \cdot 10^{-6}$

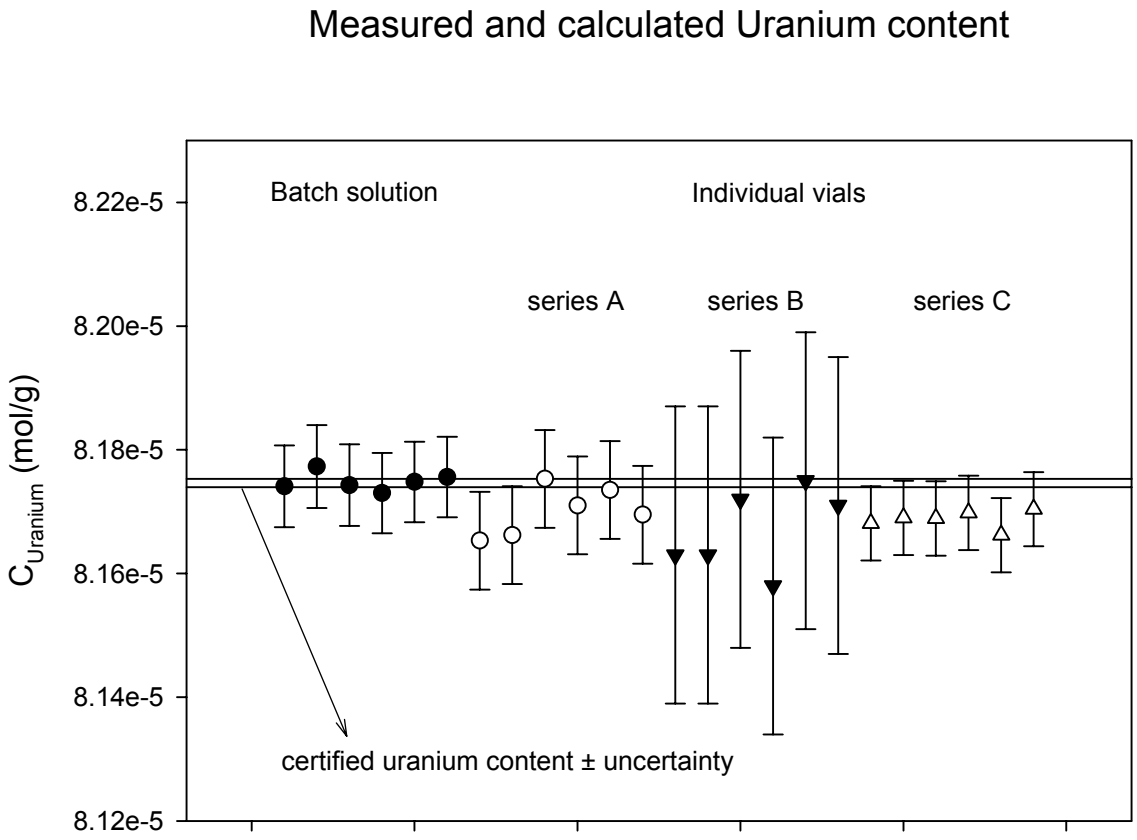

Figure 1: 'Metrological' concentration of uranium in IRMM-1027k (from the weights of metals and solution) compared with the measured values by IDMS.

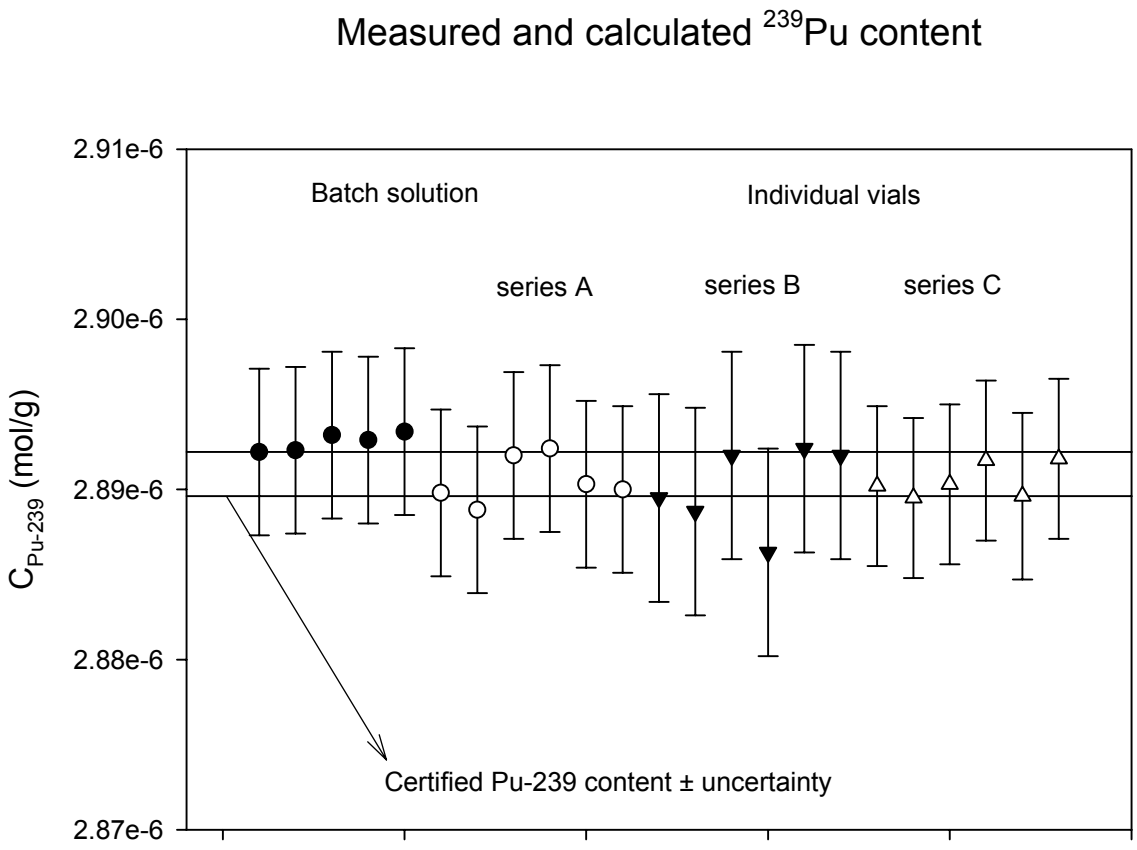

Figure 2: 'Metrological' concentration of plutonium in IRMM-1027k (from the weights of metals and solution) compared with the measured values by IDMS.

Conclusion

A new series of LSD spikes for IDMS determinations of uranium and plutonium contents in solutions of spent nuclear fuel from reprocessing plants has been prepared.

An automated system for the production of LSD spikes has been successfully taken into operation. The system was validated and proven to deliver reference materials with the same quality as those produced manually.

The certification of the spike is based on the metrological data, the certificate of the base materials and the verification measurements. The final certification values are established by mass-metrology of the metals and the solutions.

The verification of the certified values from the mass-metrology was accomplished by IDMS measurements on the batch solution and individual vials. The agreement was satisfactory.

The materials prepared are commercially available from IRMM, Geel as reference material IRMM-1027k for application in the nuclear safeguards measurements of uranium and plutonium in input solutions.

References

[1] Preparation and Certification of a new Type of Large Size Dried Spikes, Batch IRMM-1027f, A Alonso, R Eykens, F Kehoe, H Kühn, N Surugaya, A Verbruggen, R. Wellum, GE/R/IM/36/02

[2] New Procedures for Uranium Isotope Ratio Measurements using the new TRITON Thermal Ionisation Mass Spectrometer, S. Richter, A. Alonso, H. Kühn, R. Wellum, P.D.P. Taylor, Report EUR 21849

[3] An automated system for the preparation of Large Sized Dried (LSD) Spikes, A. Verbruggen, J. Bauwens, N. Van De Steene, U. Jakobsson, R. Eykens, R. Wellum, Y. Aregbe, ATALANTE Conference 2008, Montpellier (France), May 19-22, 2008

Annex 1: Certificate of uranium metal: EC NRM-101

Certified Nuclear Reference Material

Certificate of Analysis

EC NUCLEAR REFERENCE MATERIAL NO. 101

MATERIAL : URANIUM METAL

URANIUM MASS FRACTION : (999.85 ± 0.05) g·kg^{-1}

The uncertainty has been calculated by multiplying the estimated overall standard deviation by a factor of two. This corresponds to a confidence level of about 95 percent.

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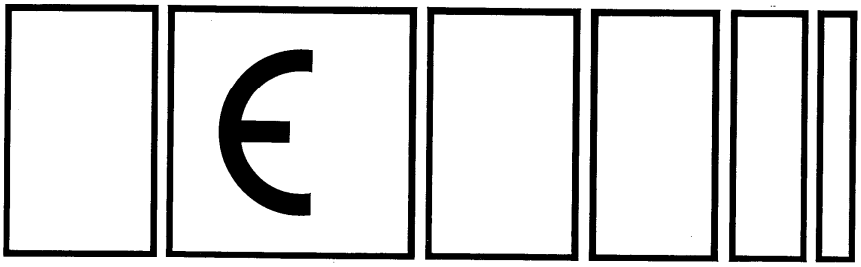
Joint Research Centre
Geel Establishment (CBNM)



Annex 2: Certificate of uranium metal: NBL CRM-116

U. S. Department of Energy
New Brunswick Laboratory

New Brunswick Laboratory
Certified Reference Materials
Certificate of Analysis

CRM 116

Uranium (Enriched) Metal
(Uranium and Uranium-235 Standard)

Uranium (etched metal basis) ..... $99.967_2 \pm 0.006_9$ Wt.%
($\alpha = 0.05$, n = 6)

Uranium-235 ..... $93.121_5 \pm 0.004_7$ Wt.%
($\alpha = 0.05$, n = 6)
$93.183_7 \pm 0.004_7$ At.%

Relative atomic weight 235.201

Metal must be etched in 1 + 1 HNO_3, rinsed in distilled-deionized water and acetone, and dried prior to use.

REFERENCE METHODS OF ANALYSIS: Titrimetry (high precision NBL method) verified with NBL CRM 112-A Uranium Metal Standard and thermal ionization mass spectrometry verified with NBL CRM U930 Uranium Isotopic Standard.

June 1978
Argonne, Illinois

Carleton D. Bingham
Director

Annex 3: Certificate of plutonium metal: Cetama MP2

CETAMA

COMMISSARIAT A L'ENERGIE ATOMIQUE

COMMISSION D'ETABLISSEMENT DES METHODES D'ANALYSE

cea

REFERENCE MATERIAL CERTIFICATE

PLUTONIUM METAL

"MP2"

Sample n° Axxx Mass : 0.xxxxxx ± 0.000012 g

(For the values x see page 4)

The reference material to which this certificate relates is intended for the calibration of chemical composition measurement. The overall chemical content of plutonium is certified. The confidence interval associated with the certified value for a single sample, takes into account uncertainties associated to with analysis and heteregenity of metal. This content, expressed as a percentage of mass, was the following on 12 march 2002 for a single sample with a probability level of 0.95.

99.90 ± 0.04 %

THE TRUE MASS OF THE SAMPLE A ± **12 µg**, RELATED TO A VACUUM, IS THAT INDICATED IN THIS CERTIFICATE AND ON THE AMPOULE.

The possibility of surface oxidation makes it impossible to envisage weighing at the time of use

Isotopique composition is certified on 12 march 2001 : see certificate IRMM page3

The preparation, analysis and certification of the plutonium to which this certificate relates was carried out by different units of the CEA group under the supervision of the Committee for Establishing Analysis Methods (CETAMA).

Le responsable MR

CETAMA
CEA VALRHO Marcoule
B. P. 17171
30207 BAGNOLS SUR CEZE CEDEX FRANCE
Téléphone (33) 4.66.79.69.88 - Télécopie (33) 4.66.79.69.89

CETAMA certifié ISO 9001:2000 par

Version : 06/2001

On 12/03/200, the metal contained around:

- by weight, 489 $mg.kg^{-1}$ of uranium,
- by weight, 438 $mg.kg^{-1}$ of américium..

UTILISATION

The sample, which consists of a piece of metal, is supplied in a double glass ampoule filled with pure nitrogen at a pressure of around 0.1 Pascal.

The ampoule must be opened with care inside a glove box. All the sample must be transferred to the dissolver.

Cover with 0.1 $mol.l^{-1}$ hydrochloric acid. The ampoule must be thoroughly washed with the same acid to recover any particles of metal which may have become separated. In 2 ml fractions, add the necessary quantity of 12 $mol.l^{-1}$ hydrochloric acid of guaranteed purity to obtain a 4 $mol.l^{-1}$ hydrochloric acid solution. Allow dissolving to proceed without heating for 10 to 15 minutes, then heat to boiling point. If there are still particles of plutonium at the bottom of the dissolver after heating for two hours, add 2 ml of 12 $mol.l^{-1}$ hydrochloric acid and 2 drops of 1 $mol.l^{-1}$ hydrofluoric acid and continue heating for another two hours. Repeat the operation if necessary until the material is totally dissolved.
If plutonium fluoride precipitates out, add a few drops of aluminium nitrate (approximately one $mol.l^{-1}$)..
Allow to cool and adjust to the required volume.

ADDITIONAL INFORMATION

The certified plutonium content has been deduced from analysis of impurities carried out by five laboratories and checked by chemical assay of the plutonium in two different laboratories using three different methods of analysis.
Spark Source Mass Spectrometry has given a full analysis of the impurities and, where concentration levels allowed, inductively-coupled plasma atomic emission spectrometry has been used to establish the concentrations of some of them.
The uranium was determined by laser spectrofluorimetry and the americium by gamma spectrometry.Carbon was determined by coulometry, after transformation into gaseous form by combustion in oxygen.
The gases were analysed by chromatography in the aqueous phase:

- for nitrogen and oxygen after extraction by high temperature stream under an inert gas,
- for hydrogen after diffusion in a vacuum.

Annex 4: Certificate of plutonium metal: isotopic abundances IRMM

EUROPEAN COMMISSION
DIRECTORATE GENERAL JRC
JOINT RESEARCH CENTRE
IRMM
Institute for Reference Materials and Measurements

CERTIFICATE of a reference measurement

IM/MeaC/07/116

11 April 2007

SUBJECT : Recertification of CEA CETAMA MP2

1. Applicant: A. Verbruggen

2. Sample Identification:
   - CEA/CETAMA/MP2
   - Chemical form: Pu metal provided by CEA/CETAMA

3. Measurands:
   - Isotopic composition

isotope amount ratio(s)	
$n(^{238}Pu)/\ n(^{239}Pu)$	0.000 030 83(29)
$n(^{240}Pu)/\ n(^{239}Pu)$	0.022 432 4(51)
$n(^{241}Pu)/\ n(^{239}Pu)$	0.000 237 8(31)
$n(^{242}Pu)/\ n(^{239}Pu)$	0.000 075 70(78)

amount fraction (·100)		mass fraction (·100)	
$n(^{238}Pu)/n(Pu)$	0.003 015(29)	$m(^{238}Pu)/m(Pu)$	0.003 002(28)
$n(^{239}Pu)/n(Pu)$	97.773 05(58)	$m(^{239}Pu)/m(Pu)$	97.763 80(59)
$n(^{240}Pu)/n(Pu)$	2.193 28(49)	$m(^{240}Pu)/m(Pu)$	2.202 27(49)
$n(^{241}Pu)/n(Pu)$	0.023 25(30)	$m(^{241}Pu)/m(Pu)$	0.023 44(31)
$n(^{242}Pu)/n(Pu)$	0.007 402(76)	$m(^{242}Pu)/m(Pu)$	0.007 494(77)

molar mass: 239.074 790 8(91) $g \cdot mol^{-1}$

4. Date of sample receipt : n.a.
   Date of completion of measurement : 7 November 2006

5. All uncertainties indicated are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty estimated following the ISO/BIPM guide[1]. They are given in parentheses and include a coverage factor k=2. They apply to the last two digits of the value. The values certified are traceable to the SI. The primary certified values are the isotope amount ratio ; other values are derived from them. Reproducing the derived values may result in difference due to rounding errors.

[1] International Organisation for Standardisation, Guide to the expression of Uncertainty in Measurement, ©ISO, ISBN 92-67-10188-9, Geneva, Switserland, 1993

Uncertainty budget :

Quantity	Value	Standard Uncertainty	Index
Atomic mass ^{239}Pu	239.05215760 g/mol	$5.1 \cdot 10^{-6}$ g/mol	59.6 %
Measurement ratio 240/239	0.02243535 mol/mol	$3.81 \cdot 10^{-6}$ mol/mol	14.9 %
Measurement ratio 241/239	$240 \cdot 10^{-6}$ mol/mol	$450 \cdot 10^{-9}$ mol/mol	0.9 %
Measurement ratio 242/239	$75 \cdot 10^{-6}$ mol/mol	$175 \cdot 10^{-9}$ mol/mol	0.4 %
variability$_{241/239}$	0.0 mol/mol	$2.65 \cdot 10^{-6}$ mol/mol	21.0 %
variability$_{242/239}$	0.0 mol/mol	$650 \cdot 10^{-9}$ mol/mol	3.0 %
M_{Pu}	239.07478500 g/mol	$6.46 \cdot 10^{-6}$ g/mol	

6. The traceability to SI is established through standards from IRMM-290.

7. Analytical measurement procedure
   - Mass spectrometric measurments were performed by H Kühn an F Kehoe for the $[n(^{238}Pu)/n(^{239}Pu)]$, $[n(^{240}Pu)/n(^{239}Pu)]$, $[n(^{241}Pu)/n(^{239}Pu)]$ and $[n(^{242}Pu)/n(^{239}Pu)]$ using the MAT262 TIMS, sample solutions were prepared for TIMS analysis by F Kehoe. A. Verbruggen was responsible for preparation and issuance of the certificate.
   - The atomic masses, used in the calculation are from G. Audi and A.H. Wapstra.[2]
   - Reference numbers of the measurement data: measurements number T26629, T26A03, T26B07, logged in S:\D04-IM\Secure Data\Project Data\MP2 (based on 081a and LSD1027i)\MP2 IA Summary MAT262 measurements.
   - Full details of the preparation and the certification procedure can be found in certification report EUR*****.

8. These samples will be stored for a minimum period of six months from the date of this certificate

André Verbruggen
Group leader Nuclear Chemistry

Stephan Richter
Group leader Nuclear Mass Spectrometry

Copies
P Taylor, IM unit head
Y Aregbe, Action leader Nuclear Safeguards
F Kehoe
H Kühn

[2] G. Audi and A.H. Wapstra, The 2003 atomic mass evaluation, Nucl Phys A729 (2003) 337-676

Annex 5: Mass Metrology certificate: base materials

EUROPEAN COMMISSION DIRECTORATE GENERAL Joint Research Centre	**Certificate of weighing**	Institute for Reference Materials and Measurements

E. 3642 **Issued date**: 11 September 07 **Page** 1 of 1

Applicant: Verbruggen **Group:** IM-Nuclear

Project: IRMM-1027 K **IM-unit ref.:**

Description: Preparation mother solution IRMM-1027 K.

Date of receipt of request: 1 June 2006 **Weighing date**: 27 September 2006

The reported results applies only to the objects / samples described in this certificate

	Weight in g
Mass of Pu metal (MP 2 BC 2701)	2.2142 (2)
Mass of U metal (NBL-CRM-116)	11.983 (1)
Mass of U metal (EC-NRM-101)	48.771 (4)
Mass of IRMM-1027 K	3129.17 (5)

Observations:

The measurements and uncertainty estimates, were performed according to working instruction WI-0185, "Mass determination by substitution weighing"on balances AT 261 and At 201 with IRMM inventory No 1999 00337 27 and 1996 00547 73.

Traceability:

The certified mass values are traceable to the International Kilogram Prototype via regular calibrations of the IRMM principal kilogram. The sets of working mass standards M 3 and M 10 were used as reference in the mass determination.

Uncertainty:

All reported uncertainties are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM Guide to the expression of Uncertainty in Measurement. The coverage factor $k = 2$ corresponds to a coverage probability of about 95%. U applies to the last digit of the value of the measurement result and is given in parentheses ().

Annexes:

Signature
Mass Metrology Service

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Annex 6: Mass Metrology certificates: verification measurements

EUROPEAN COMMISSION DIRECTORATE GENERAL Joint Research Centre	**Certificate of weighing**	Institute for Reference Materials and Measurements

E. 3641 REV 1 **Issued date:** 10/09/2007 *03/10/2008* **Page** 1 **of** 1

Applicant: Verbruggen **Group:** IM-Nuclear

Project: 1027 K **IM-unit ref.:**

Description: Verification of 1027 K on ~~individual vials~~ *batch solution.*

Date of receipt of request: 01/06/2006 **Weighing date:** 30/05/2007

The reported results applies only to the objects / samples described in this certificate

Vial Number	U-Pu solution in g	IRMM-046 b in g
1027 K-1	0.9742 (5)	5.0304 (5)
1027 K-2	0.9731 (5)	4.9744 (5)
1027 K-3	0.9900 (5)	5.0011 (5)
1027 K-4	0.9974 (5)	5.0055 (5)
1027 K-5	1.0013 (5)	5.0276 (5)
1027 K-6	1.0006 (5)	5.0065 (5)
1027 K-IA	0.9999 (5)	

Observations:
The measurements and uncertainty estimates, were performed according to working instruction WI-0185, "Mass determination by substitution weighing"on balances AT 261 with IRMM inventory No 1999 00337 27.

Traceability:
The certified mass values are traceable to the International Kilogram Prototype via regular calibrations of the IRMM principal kilogram. The set of working mass standards M 3 was used as reference in the mass determination.

Uncertainty:
All reported uncertainties are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM Guide to the expression of Uncertainty in Measurement. The coverage factor $k = 2$ corresponds to a coverage probability of about 95%. U applies to the last digit of the value of the measurement result and is given in parentheses ().

Annexes:

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EUROPEAN COMMISSION DIRECTORATE-GENERAL Joint Research Centre	**Certificate of weighing**	Institute for Reference Materials and Measurements

E. 3673 **Issued date:** 13 March2008 **Page** 1 **of** 1

Applicant: Verbruggen **Group:** IM-Nuclear

Project: 1027 K **IM-unit ref.:**

Description: Verification of 1027 K on individual vials.

Date of receipt of request: 21 December 2008 **Weighing date:** 22 January 2008

The reported results applies only to the objects / samples described in this certificate

Vial Number	U-Pu solution in g	IRMM-046 b in g
1027 K-15	2.5270 (5)	4.9917 (5)
1027 K-50	2.4136 (5)	5.0287 (5)
1027 K-85	2.5028 (5)	5.0125 (5)
1027 K-110	2.5014 (5)	5.0239 (5)
1027 K-145	2.5815 (5)	5.0051 (5)
1027 K-180	2.5150 (5)	5.0005 (5)

Observations:

The measurements and uncertainty estimates, were performed according to working instruction WI-0185, "Mass determination by substitution weighing"on balances AT 261 with IRMM inventory No 1999 00337 27.

Traceability:

The certified mass values are traceable to the International Kilogram Prototype via regular calibrations of the IRMM principal kilogram. The set of working mass standards M 3 was used as reference in the mass determination.

Uncertainty:

All reported uncertainties are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM Guide to the expression of Uncertainty in Measurement. The coverage factor $k = 2$ corresponds to a coverage probability of about 95%. U applies to the last digit of the value of the measurement result and is given in parentheses ().

Annexes:

Signature
Mass Metrology Service

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EUROPEAN COMMISSION DIRECTORATE GENERAL Joint Research Centre	**Certificate of weighing**	Institute for Reference Materials and Measurements

E. 3675 **Issued date:** 15 May 2008 **Page** 1 **of** 1

Applicant: Verbruggen **Group:** IM-Nuclear

Project: 1027 K **IM-unit ref.:**

Description: Verification of 1027 K on individual vials series B.

Date of receipt of request: 21 December 2008 **Weighing date:** 12 februari 2008

The reported results applies only to the objects / samples described in this certificate

Vial Number	U-Pu solution in g	IRMM-046 b in g
1027 K-250	2.6162 (6)	4.9673 (5)
1027 K-410	2.6041 (6)	5.0484 (5)
1027 K-580	2.5977 (6)	4.9843 (5)
1027 K-740	2.6050 (6)	4.9903 (5)
1027 K-910	2.5749 (6)	5.0036 (5)
1027 K-1070	2.5424 (6)	5.0014 (5)

Observations:

The measurements and uncertainty estimates, were performed according to working instruction WI-0185, "Mass determination by substitution weighing"on balances AT 261 with IRMM inventory No 1999 00337 27.

Traceability:

The certified mass values are traceable to the International Kilogram Prototype via regular calibrations of the IRMM principal kilogram. The set of working mass standards M 3 was used as reference in the mass determination.

Uncertainty:

All reported uncertainties are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM Guide to the expression of Uncertainty in Measurement. The coverage factor $k = 2$ corresponds to a coverage probability of about 95%. U applies to the last digit of the value of the measurement result and is given in parentheses ().

Annexes:

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Mass Metrology Service

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EUROPEAN COMMISSION DIRECTORATE-GENERAL Joint Research Centre	**Certificate of weighing**	Institute for Reference Materials and Measurements

E. 3676 **Issued date:** 15 May 2008 **Page** 1 **of** 1

Applicant: Verbruggen **Group:** IM-Nuclear

Project: 1027 K **IM-unit ref.:**

Description: Verification of 1027 K on individual vials series C.

Date of receipt of request: 21 December 2008 **Weighing date:** 12 februari 2008

The reported results applies only to the objects / samples described in this certificate

Vial Number	U-Pu solution in g	IRMM-046 b in g
1027 K-320	2.6106 (6)	4.9991 (5)
1027 K-500	2.6154 (6)	4.9829 (5)
1027 K-660	2.5882 (6)	5.0011 (5)
1027 K-820	2.5811 (6)	4.9612 (5)
1027 K-990	2.5767 (6)	5.0492 (5)
1027 K-1150	2.5525 (6)	5.0535 (5)

Observations:

The measurements and uncertainty estimates, were performed according to working instruction WI-0185, "Mass determination by substitution weighing"on balances AT 261 with IRMM inventory No 1999 00337 27.

Traceability:

The certified mass values are traceable to the International Kilogram Prototype via regular calibrations of the IRMM principal kilogram. The set of working mass standards M 3 was used as reference in the mass determination.

Uncertainty:

All reported uncertainties are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM Guide to the expression of Uncertainty in Measurement. The coverage factor $k = 2$ corresponds to a coverage probability of about 95%. U applies to the last digit of the value of the measurement result and is given in parentheses ().

Annexes:

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Signature
Mass Metrology Service

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Annex 7: Certificate of IRMM-1027k

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for reference materials and measurements
Isotope Measurements (Geel)

CERTIFICATE
SPIKE ISOTOPIC REFERENCE MATERIAL IRMM-1027k

This Spike Isotopic Reference Material consists of a certified mass of approximately 2.5 g of solution subsequently evaporated to dryness and covered with a dry layer of circa 50 mg cellulose acetate butyrate (CAB) to ensure spike integrity.

Each unit is identified by a vial number. The sample mass of the solution for each vial is listed in table 1.

The Isotopic Reference Material (Spike) is supplied with amount concentrations of ^{235}U, ^{238}U and ^{239}Pu certified to be

1.563 74(29) 10^{-5} mol (^{235}U) · g^{-1} (solution)
6.587 44(73) 10^{-5} mol (^{238}U) · g^{-1} (solution)
2.890 9(13) 10^{-6} mol (^{239}Pu) · g^{-1} (solution)

Other uranium and plutonium isotopes present are related to the ^{238}U and ^{239}Pu concentration through the following certified amount ratios:

$n(^{234}U)/n(^{238}U)$	:	0.002 514(10)
$n(^{235}U)/n(^{238}U)$	:	0.237 382(54)
$n(^{236}U)/n(^{238}U)$	:	0.001 033 8(32)

$n(^{238}Pu)/n(^{239}Pu)$	:	0.000 030 59(29)
$n(^{240}Pu)/n(^{239}Pu)$	:	0.022 430 7(51)
$n(^{241}Pu)/n(^{239}Pu)$	:	0.000 226 5(30)
$n(^{242}Pu)/n(^{239}Pu)$	:	0.000 075 71(78)

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This corresponds to isotopic compositions of uranium and plutonium with the following abundances:

amount fraction (·100)		mass fraction (·100)	
$n(^{234}U)/n(U)$	0.202 62(82)	$m(^{234}U)/m(U)$	0.199 70(81)
$n(^{235}U)/n(U)$	19.129 4(36)	$m(^{235}U)/m(U)$	18.934 3(36)
$n(^{236}U)/n(U)$	0.083 31(26)	$m(^{236}U)/m(U)$	0.082 81(25)
$n(^{238}U)/n(U)$	80.584 7(32)	$m(^{238}U)/m(U)$	80.783 2(32)

amount fraction (·100)		mass fraction (·100)	
$n(^{238}Pu)/n(Pu)$	0.002 991(28)	$m(^{238}Pu)/m(Pu)$	0.002 978(28)
$n(^{239}Pu)/n(Pu)$	97.774 32(58)	$m(^{239}Pu)/m(Pu)$	97.765 07(58)
$n(^{240}Pu)/n(Pu)$	2.193 14(49)	$m(^{240}Pu)/m(Pu)$	2.202 13(49)
$n(^{241}Pu)/n(Pu)$	0.022 15(29)	$m(^{241}Pu)/m(Pu)$	0.022 33(29)
$n(^{242}Pu)/n(Pu)$	0.007 402(76)	$m(^{242}Pu)/m(Pu)$	0.007 495(77)

The molar mass of the uranium in this sample is 237.465 794(96) $g \cdot mol^{-1}$
The molar mass of the plutonium in this sample is 239.074 759 8(91) $g \cdot mol^{-1}$

From the certified values, the following amount contents are derived:

$8.174\,56(74) \cdot 10^{-5}$	mol (U) · g^{-1} (solution)
$3.675\,48(68) \cdot 10^{-3}$	g (^{235}U) · g^{-1} (solution)
$15.681\,5(17) \cdot 10^{-3}$	g (^{238}U) · g^{-1} (solution)
$19.411\,8(18) \cdot 10^{-3}$	g (U) · g^{-1} (solution)

$2.956\,7(13) \cdot 10^{-6}$	mol (Pu) · g^{-1} (solution)
$6.910\,6(31) \cdot 10^{-4}$	g (^{239}Pu) · g^{-1} (solution)
$7.068\,6(32) \cdot 10^{-4}$	g (Pu) · g^{-1} (solution)

NOTES

1. This Spike Isotopic Reference Material is traceable to the SI in the shortest possible way. The values of the U and Pu isotope ratios were measured at IRMM and are traceable to the SI via the values of the isotope ratios of the isotopic reference materials IRMM-183, 184, 185, 186, 187 for uranium and IRMM-290 for plutonium. The U and Pu content of this spike are traceable to the SI via reference materials NBL CRM-116, EC NRM-101 and CETAMA MP2. Measurements calibrated by this Isotopic Reference Material have therefore the potential of being traceable to the SI.

2. All uncertainties indicated in this certificate are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty estimated following the ISO/BIPM Guide to the Expression of Uncertainty in Measurement. They are given in parentheses and include a coverage factor k=2. They apply to the last two digits of the value.

3. The IRMM-1027k was prepared by metrological weighing of U metals (NBL CRM 116, EC NRM 101) and Pu metal (CETAMA MP2), dissolution in HNO_3, subsequently dispensing by metrological weighing into individual units, drying and conditioning in cellulose acetate butyrate (CAB).

4. IRMM-1027k is delivered in individual glass (penicillin) vials each containing about 45 mg U and 1.8 mg Pu.

5. Values for isotope amount ratios, isotopic compositions and concentrations are valid for 01 January 2008. This certificate is valid until September 2010; the validity may be extended after further tests on the stability of the spike material are carried out.

6. It is recommended to store the vials in vertical position.

7. The half lives used in the calculations are

^{238}Pu : 8.77 (03) $\cdot 10^1$ a[(1)]
^{239}Pu : 2.411 (03) $\cdot 10^4$ a[(1)]
^{240}Pu : 6.563 (07) $\cdot 10^3$ a[(1)]
^{241}Pu : 1.432 5(24) $\cdot 10^1$ a[(2)]
^{242}Pu : 3.735 (11) $\cdot 10^5$ a[(1)]
^{244}Pu : 8.00 (09) $\cdot 10^7$ a[(1)]

8. The atomic masses, used in the calculations, are[(3)]

^{233}U : 233.039 627 0 (60) g·mol^{-1}
^{234}U : 234.040 944 7 (44) g·mol^{-1}
^{235}U : 235.043 922 2 (42) g·mol^{-1}
^{236}U : 236.045 561 0 (42) g·mol^{-1}
^{238}U : 238.050 783 5 (44) g·mol^{-1}

^{238}Pu : 238.049 559 9 (40) g·mol^{-1}
^{239}Pu : 239.052 163 4 (40) g·mol^{-1}
^{240}Pu : 240.053 813 5 (40) g·mol^{-1}
^{241}Pu : 241.056 851 5 (40) g·mol^{-1}
^{242}Pu : 242.058 742 6 (40) g·mol^{-1}
^{244}Pu : 244.064 204 (10) g·mol^{-1}

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(1) IAEA, Decay data of the Transactinium Nuclides, Technical Reports Series No. 261, 1986
(2) A. Verbruggen, R. Wellum, Progress in re-measuring the half-life of ^{241}Pu, 49th INMM conference, Nashville, USA, July 2008
(3) G. Audi and A.H. Wapstra, The 2003 atomic mass evaluation, Nucl Phys A729 (2003) 337-676.

9. The vials should be handled with great care and by experienced personnel in a laboratory environment suitably equipped for the safe handling of radioactive materials.

10. Full details of the certification procedure can be found in the Preparation and Certification Report.[4]

Chemical preparation and ampouling of this IRM were accomplished by J Bauwens, F Kehoe and R Eykens.

The isotopic verification measurements were carried out by F Kehoe, S Richter and H Kühn for uranium and plutonium on samples chemically prepared by F Kehoe. Measurements of isotopic ratios were calibrated against synthetic isotopic mixtures prepared by R Eykens for uranium and J Broothaerts for plutonium.

Metrological weighings required in the preparation and certification were performed by F Kehoe and R Eykens.

The overall co-ordination leading to the establishment, certification and issuance of this Spike Isotopic Reference Material was performed by A Verbruggen.

B-2440 GEEL
September 2008

Y Aregbe
IRMM Safeguards Coordinator

P Taylor
Head
Isotope Measurements Unit

[4] A. Verbruggen, R. Eykens, F. Kehoe, H. Kühn, S. Richter, Y. Aregbe, Preparation and Certification of IRMM-1027k, Large-Sized Dried (LSD) spike, report EUR23539 EN

Table 1: list of vial numbers, mass of solution before drying

N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)
1	2.7279	51	2.5035	101	2.5066	151	2.5115	201	2.6016	251	2.6152	301	2.6221
2	2.7081	52	2.5033	102	2.5060	152	2.5170	202	2.6342	252	2.5977	302	2.6143
3	2.5876	53	2.4992	103	2.5057	153	2.4594	203	2.6152	253	2.6228	303	2.6137
4	2.4765	54	2.5039	104	2.5042	154	2.5143	204	2.6017	254	2.6229	304	2.6204
5	2.4917	55	2.5048	105	2.4568	155	2.5117	205	2.6351	255	2.6161	305	2.6008
6	2.5295	56	2.4957	106	2.5104	156	2.5137	206	2.6123	256	2.6128	306	2.6191
7	2.5321	57	2.4943	107	2.5039	157	2.5111	207	2.6195	257	2.6152	307	2.6115
8	2.5288	58	2.4729	108	2.4998	158	2.5136	208	2.6138	258	2.6184	308	2.6167
9	2.5329	59	2.5013	109	2.5042	159	2.5140	209	2.5972	259	2.5990	309	2.6179
10	2.5307	60	2.5053	110	2.5014	160	2.5164	210	2.6191	260	2.6292	310	2.6182
11	2.5341	61	2.4777	111	2.5026	161	2.5130	211	2.6362	261	2.6031	311	2.6154
12	2.5244	62	2.5092	112	2.4989	162	2.5171	212	2.6123	262	2.6143	312	2.6142
13	2.5204	63	2.5023	113	2.5045	163	2.5138	213	2.6186	263	2.6144	313	2.5980
14	2.5175	64	2.5042	114	2.5063	164	2.5147	214	2.6130	264	2.6093	314	2.6309
15	2.5270	65	2.5152	115	2.5087	165	2.6165	215	2.6210	265	2.6420	315	2.6124
16	2.5202	66	2.5005	116	2.5082	166	2.5130	216	2.6173	266	2.6073	316	2.5943
17	0.0005	67	2.5050	117	2.4727	167	2.5180	217	2.6073	267	2.6088	317	2.6164
18	2.5211	68	2.5114	118	2.5067	168	2.5191	218	2.6248	268	2.6302	318	2.6315
19	2.5142	69	2.5014	119	2.4993	169	2.5059	219	2.6162	269	2.6069	319	2.6064
20	2.4896	70	2.5028	120	2.4940	170	2.5272	220	2.6118	270	2.6107	320	2.6106
21	2.5172	71	2.4961	121	2.4933	171	2.5245	221	2.6251	271	2.6087	321	2.6254
22	2.5149	72	2.4498	122	2.4985	172	2.5148	222	2.6136	272	2.6195	322	2.6045
23	2.5039	73	2.4973	123	2.4908	173	0.0005	223	2.6166	273	2.6208	323	2.6301
24	2.5152	74	2.5073	124	2.4955	174	2.5211	224	2.6026	274	2.6195	324	2.6172
25	2.5085	75	2.5027	125	2.4979	175	2.5191	225	2.6302	275	2.6168	325	2.6055
26	2.5148	76	2.5057	126	2.5036	176	2.5196	226	2.5930	276	2.6078	326	2.6139
27	2.5206	77	2.5039	127	2.5004	177	2.5143	227	2.6342	277	2.6168	327	2.6152
28	2.5125	78	2.4989	128	2.4954	178	2.5213	228	2.6168	278	2.6059	328	2.6138
29	2.5127	79	2.5065	129	2.4584	179	2.5108	229	2.6107	279	2.6297	329	2.6057
30	2.5071	80	2.5009	130	2.5007	180	2.5150	230	2.6237	280	2.6138	330	2.6248
31	2.4802	81	2.5029	131	2.5089	181	2.5137	231	2.6103	281	2.6084	331	2.5992
32	2.5142	82	2.5007	132	2.5037	182	2.5177	232	2.6173	282	2.6142	332	2.6254
33	2.5178	83	2.5035	133	2.5080	183	2.5172	233	2.6031	283	2.6183	333	2.6203
34	2.5080	84	2.4749	134	2.4978	184	2.5195	234	2.6326	284	2.6178	334	2.6011
35	2.5195	85	2.5028	135	2.5050	185	2.5213	235	2.6094	285	2.6099	335	2.6273
36	2.4957	86	2.5034	136	2.5052	186	2.5161	236	2.6142	286	2.6050	336	2.6123
37	2.5060	87	2.5001	137	2.4982	187	2.5150	237	2.6161	287	2.6128	337	2.6189
38	2.5172	88	2.5017	138	2.5032	188	2.5144	238	2.6189	288	2.6347	338	2.6153
39	2.5175	89	2.5016	139	2.5065	189	2.5153	239	2.6154	289	2.5953	339	2.6117
40	2.4733	90	2.4953	140	2.5014	190	2.5177	240	2.6160	290	2.6361	340	2.5987
41	2.5205	91	2.4956	141	2.4997	191	2.5150	241	2.6055	291	2.6064	341	2.6100
42	2.5214	92	2.4938	142	2.5025	192	2.5121	242	2.6232	292	2.5960	342	2.6298
43	2.5003	93	2.5025	143	2.4990	193	2.5271	243	2.6069	293	2.6371	343	2.6161
44	2.5158	94	2.4943	144	2.4722	194	2.5759	244	2.6252	294	2.6130	344	2.6100
45	2.5153	95	2.4932	145	2.5815	195	2.6180	245	2.6132	295	2.6152	345	2.6216
46	0.0005	96	2.4972	146	2.4880	196	2.6004	246	2.5969	296	2.6223	346	2.6183
47	2.5179	97	2.4394	147	2.4909	197	2.6330	247	2.6386	297	2.6117	347	2.5959
48	2.5252	98	2.4641	148	2.5001	198	2.6032	248	2.6155	298	2.6070	348	2.6222
49	2.4894	99	2.5048	149	2.5103	199	2.6302	249	2.6168	299	2.6087	349	2.6100
50	2.4136	100	2.4720	150	2.5095	200	2.6149	250	2.6162	300	2.6189	350	2.6254

Table 1: list of vial numbers, mass of solution before drying (continued)

N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)
351	2.5946	401	2.6160	451	2.6145	501	2.6069	551	2.6275	601	2.5961	651	2.5939
352	2.6208	402	2.6147	452	2.6166	502	2.5962	552	2.6006	602	2.6080	652	2.5835
353	2.6327	403	2.6128	453	2.6128	503	2.6276	553	2.6061	603	2.5870	653	2.6210
354	2.5948	404	2.6125	454	2.6052	504	2.5939	554	2.6148	604	2.5996	654	2.5850
355	2.6290	405	2.6139	455	2.6239	505	2.6229	555	2.6141	605	2.5932	655	2.6039
356	2.6167	406	2.5912	456	2.6094	506	2.6044	556	2.5869	606	2.6151	656	2.5953
357	2.6032	407	2.6319	457	2.6207	507	2.6232	557	2.6200	607	2.5795	657	2.5915
358	2.6313	408	2.6170	458	2.6056	508	2.6056	558	2.6182	608	2.6125	658	2.6090
359	2.6125	409	2.5990	459	2.6142	509	2.6060	559	2.6000	609	2.5888	659	2.5948
360	2.5969	410	2.6145	460	2.6151	510	2.6115	560	2.6152	610	2.6189	660	2.5882
361	2.6265	411	2.6166	461	2.6157	511	2.6184	561	2.5943	611	2.5910	661	2.6061
362	2.6118	412	2.6128	462	2.6096	512	2.5997	562	2.6264	612	2.6001	662	2.5992
363	2.6194	413	2.6052	463	2.6007	513	2.6189	563	2.6130	613	2.6116	663	2.5920
364	2.6017	414	2.6239	464	2.6228	514	2.6101	564	2.6045	614	2.5870	664	2.6019
365	2.6271	415	2.6094	465	2.6063	515	2.6019	565	2.6111	615	2.6123	665	2.5959
366	2.6124	416	2.6207	466	2.6169	516	2.6119	566	2.6034	616	2.5853	666	2.5986
367	2.6134	417	2.6056	467	2.6024	517	2.6220	567	2.6130	617	2.6150	667	2.5947
368	2.6094	418	2.6142	468	2.6276	518	2.6036	568	2.6126	618	2.5988	668	2.6052
369	2.6051	419	2.6151	469	2.6113	519	2.6073	569	2.6090	619	2.5945	669	2.5917
370	2.6168	420	2.6157	470	2.6152	520	2.6309	570	2.6041	620	2.5985	670	2.6007
371	2.6096	421	2.6096	471	2.6100	521	2.6087	571	2.6126	621	2.5926	671	2.6025
372	2.6203	422	2.6007	472	2.6156	522	2.6100	572	2.6028	622	2.6077	672	2.6041
373	2.6116	423	2.6228	473	2.6071	523	2.6050	573	2.6150	623	2.5854	673	2.5988
374	2.6162	424	2.6063	474	2.5998	524	2.6173	574	2.6059	624	2.5951	674	2.6040
375	2.6215	425	2.6169	475	2.6312	525	2.6090	575	2.5978	625	2.6013	675	2.6013
376	2.6183	426	2.6024	476	2.5953	526	2.6109	576	2.6178	626	2.5942	676	2.6040
377	2.6122	427	2.6276	477	2.6268	527	2.6108	577	2.6157	627	2.6127	677	2.5966
378	2.6201	428	2.6113	478	2.6054	528	2.6007	578	2.5979	628	2.5983	678	2.5996
379	2.6064	429	2.6152	479	2.6269	529	2.6213	579	2.6189	629	2.5862	679	2.6101
380	2.6066	430	2.6100	480	2.6078	530	2.6085	580	2.5977	630	2.6047	680	2.5888
381	2.6232	431	2.6156	481	2.6141	531	2.6094	581	2.6171	631	2.5917	681	2.6059
382	2.6107	432	2.6071	482	2.6112	532	2.6032	582	2.5982	632	2.6004	682	2.5988
383	2.5974	433	2.5998	483	2.6086	533	2.6098	583	2.6216	633	2.6017	683	2.5951
384	2.6275	434	2.6312	484	2.6127	534	2.6283	584	2.6099	634	2.5901	684	2.6072
385	2.6214	435	2.5953	485	2.6103	535	2.6050	585	2.6011	635	2.5906	685	2.5966
386	2.6164	436	2.6268	486	2.6144	536	2.6081	586	2.6057	636	2.6038	686	2.6032
387	2.5954	437	2.6054	487	2.6116	537	2.6126	587	2.5943	637	2.5918	687	2.6003
388	2.6284	438	2.6269	488	2.6128	538	2.6074	588	2.6175	638	2.5920	688	2.5839
389	2.6081	439	2.6078	489	2.6167	539	2.6164	589	2.6076	639	2.6100	689	2.6172
390	2.6053	440	2.6141	490	2.5930	540	2.6085	590	2.5901	640	2.5870	690	2.5983
391	2.6384	441	2.6112	491	2.6266	541	2.6153	591	2.6267	641	2.5757	691	2.5999
392	2.5931	442	2.6086	492	2.6097	542	2.6048	592	2.6041	642	2.6170	692	2.6073
393	2.6183	443	2.6127	493	2.6043	543	2.6044	593	2.5904	643	2.5857	693	2.6021
394	2.6202	444	2.6103	494	2.6196	544	2.6101	594	2.6198	644	2.6016	694	2.5947
395	2.6072	445	2.6144	495	2.6068	545	2.6203	595	2.5987	645	2.6035	695	2.5979
396	2.6108	446	2.6116	496	2.6195	546	2.5971	596	2.6023	646	2.5975	696	2.5995
397	2.6236	447	2.6128	497	2.5965	547	2.6259	597	2.6100	647	2.5982	697	2.5967
398	2.6047	448	2.6167	498	2.6153	548	2.6090	598	2.6006	648	2.5803	698	2.6002
399	2.6104	449	2.5930	499	2.6176	549	2.6132	599	2.6021	649	2.6011	699	2.5983
400	2.6189	450	2.6266	500	2.6154	550	2.5940	600	2.5946	650	2.6033	700	2.5902

Table 1: list of vial numbers, mass of solution before drying (continued)

N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)	N°	Mass (g)		Mass (g)
701	2.5967	751	2.5920	801	2.5873	851	2.5699	901	2.5779	951	2.5775	1001	2.5905
702	2.5892	752	2.5928	802	2.5795	852	2.5929	902	2.5956	952	2.5624	1002	2.5364
703	2.5977	753	2.6030	803	2.5909	853	2.5925	903	2.5742	953	2.5668	1003	2.6025
704	2.6026	754	2.5852	804	2.5759	854	2.5747	904	2.5752	954	2.5674	1004	2.5613
705	2.6067	755	2.6107	805	2.5991	855	2.5912	905	2.5781	955	2.5805	1005	2.5694
706	2.5942	756	2.5999	806	2.5820	856	2.5777	906	2.5805	956	2.5524	1006	2.5646
707	2.6143	757	2.5834	807	2.5940	857	2.5891	907	2.5646	957	2.5642	1007	2.5328
708	2.5976	758	2.6075	808	2.5838	858	2.5762	908	2.5763	958	2.5790	1008	2.5790
709	2.5983	759	2.5855	809	2.5745	859	2.5682	909	2.5972	959	2.5766	1009	2.5640
710	2.5878	760	2.5948	810	2.5996	860	2.5822	910	2.5749	960	2.5551	1010	2.5886
711	2.6060	761	2.6018	811	2.5990	861	2.5833	911	2.5650	961	2.5783	1011	2.5403
712	2.6042	762	2.6015	812	2.5714	862	2.5817	912	2.5997	962	2.5759	1012	2.5744
713	2.5986	763	2.5871	813	2.5939	863	2.5666	913	2.5804	963	2.5849	1013	2.5340
714	2.5980	764	2.6032	814	2.5847	864	2.5898	914	2.5924	964	2.5692	1014	2.5901
715	2.5924	765	2.5857	815	2.5751	865	2.5587	915	2.5792	965	2.5660	1015	2.5749
716	2.6030	766	2.6064	816	2.5943	866	2.5829	916	2.5674	966	2.5946	1016	2.5590
717	2.5866	767	2.5917	817	2.5791	867	2.5777	917	2.5839	967	2.5727	1017	2.5595
718	2.6010	768	2.6010	818	2.5903	868	2.5913	918	2.5844	968	2.5557	1018	2.5593
719	2.5880	769	2.5900	819	2.5793	869	2.5766	919	2.5737	969	2.5766	1019	2.5593
720	2.5996	770	2.5913	820	2.5811	870	2.5897	920	2.5824	970	2.5484	1020	2.5615
721	2.6066	771	2.6001	821	2.5820	871	2.5725	921	2.5865	971	2.5766	1021	2.5587
722	2.5855	772	2.5986	822	2.5924	872	2.6000	922	2.5752	972	2.5466	1022	2.5844
723	2.6001	773	2.5862	823	2.5743	873	2.5719	923	2.5710	973	2.5826	1023	2.5697
724	2.5957	774	2.5877	824	2.5897	874	2.5779	924	2.5874	974	2.5704	1024	2.5577
725	2.5881	775	2.6072	825	2.5884	875	2.5853	925	2.5793	975	2.5804	1025	2.5667
726	2.5967	776	2.5958	826	2.5775	876	2.5800	926	2.5603	976	2.5698	1026	2.5576
727	2.5976	777	2.5914	827	2.5821	877	2.5583	927	2.5936	977	2.5712	1027	2.5665
728	2.6068	778	2.5908	828	2.5699	878	2.6020	928	2.5631	978	2.5817	1028	2.5533
729	2.5936	779	2.5832	829	2.5852	879	2.6024	929	2.5707	979	2.5556	1029	2.5664
730	2.6046	780	2.5969	830	2.5945	880	2.5891	930	2.5885	980	2.5586	1030	2.5537
731	2.5804	781	2.5725	831	2.5740	881	2.5783	931	2.5748	981	2.5629	1031	2.5688
732	2.6073	782	2.6040	832	2.5944	882	2.6001	932	2.5690	982	2.6014	1032	2.5528
733	2.6077	783	2.5869	833	2.5788	883	2.5739	933	2.5602	983	2.5731	1033	2.5574
734	2.6007	784	2.5983	834	2.5667	884	2.5889	934	2.5936	984	2.5728	1034	2.5581
735	2.5912	785	2.5935	835	2.5970	885	2.5763	935	2.5556	985	2.5880	1035	2.5640
736	2.6003	786	2.5923	836	2.5851	886	2.5899	936	2.5700	986	2.5640	1036	2.5543
737	2.5951	787	2.5972	837	2.5748	887	2.5497	937	2.5691	987	2.5775	1037	2.5503
738	2.6028	788	2.5931	838	2.5907	888	2.6005	938	2.5892	988	2.5714	1038	2.5438
739	2.5884	789	2.5798	839	2.5772	889	2.5876	939	2.5736	989	2.5600	1039	2.5768
740	2.6050	790	2.6005	840	2.5874	890	2.5843	940	2.5809	990	2.5767	1040	2.5605
741	2.6023	791	2.5836	841	2.5864	891	2.5647	941	2.5716	991	2.5649	1041	2.5665
742	2.5974	792	2.5990	842	2.5837	892	2.6019	942	2.5702	992	2.5721	1042	2.5506
743	2.5936	793	2.5847	843	2.5829	893	2.5823	943	2.5543	993	2.5481	1043	2.5635
744	2.5908	794	2.5937	844	2.5858	894	2.5818	944	2.5945	994	2.5913	1044	2.5188
745	2.6134	795	2.5826	845	2.5674	895	2.5618	945	2.5709	995	2.5636	1045	2.5845
746	2.5951	796	2.5945	846	2.5952	896	2.5915	946	2.5538	996	2.5677	1046	2.5578
747	2.5985	797	2.5867	847	2.5749	897	2.5875	947	2.5831	997	2.5397	1047	2.5567
748	2.5897	798	2.5899	848	2.5834	898	2.5718	948	2.5557	998	2.5985	1048	2.5643
749	2.5924	799	2.5857	849	2.5727	899	2.5778	949	2.5571	999	2.5669	1049	2.5424
750	2.6000	800	2.5895	850	2.5847	900	2.5867	950	2.5932	1000	2.5575	1050	2.5761

Table 1: list of vial numbers, mass of solution before drying (continued)

N°	Mass (g)	N°	Mass (g)	N°	Mass (g)
1051	2.5706	1101	2.5592	1151	2.5476
1052	2.5638	1102	2.5134	1152	2.5594
1053	2.5714	1103	2.5851	1153	2.5451
1054	2.5275	1104	2.5541	1154	2.5723
1055	2.5763	1105	2.5131	1155	2.5430
1056	2.5347	1106	2.5524	1156	2.5610
1057	2.5841	1107	2.5668	1157	2.5542
1058	2.5592	1108	2.5696	1158	2.5548
1059	2.5321	1109	2.5281	1159	2.5391
1060	2.5826	1110	2.5653	1160	2.5488
1061	2.5585	1111	2.5735	1161	2.5510
1062	2.5384	1112	2.5528	1162	2.5452
1063	2.5747	1113	2.5137	1163	2.5423
1064	2.5670	1114	2.5797	1164	2.5469
1065	2.5175	1115	2.5158	1165	2.5600
1066	2.5688	1116	2.5753	1166	2.5469
1067	2.5775	1117	2.5550	1167	2.5430
1068	2.5616	1118	2.5596	1168	2.5413
1069	2.5787	1119	2.5357	1169	2.5427
1070	2.5424	1120	2.5464	1170	2.5422
1071	2.5568	1121	2.5498	1171	2.5500
1072	2.5883	1122	2.5459	1172	2.5374
1073	2.5650	1123	2.5242	1173	2.5662
1074	2.5488	1124	2.5957	1174	2.5627
1075	2.5473	1125	2.5244	1175	2.5451
1076	2.5633	1126	2.5437	1176	2.5737
1077	2.5730	1127	2.5467	1177	2.5548
1078	2.5599	1128	2.5627	1178	2.5500
1079	2.5586	1129	2.5104	1179	2.5427
1080	2.5546	1130	2.5346	1180	2.5569
1081	2.5305	1131	2.5624	1181	2.5435
1082	2.5728	1132	2.5665	1182	2.5440
1083	2.5650	1133	2.5373	1183	2.5600
1084	2.5533	1134	2.5579	1184	2.5579
1085	2.5582	1135	2.5336	1185	2.5569
1086	2.5567	1136	2.5357	1186	2.5453
1087	2.5266	1137	2.5802	1187	2.5465
1088	2.5785	1138	2.5203	1188	2.5419
1089	2.5230	1139	2.5625	1189	2.5562
1090	2.5856	1140	2.5345	1190	2.5580
1091	2.5049	1141	2.5528	1191	2.5575
1092	2.5669	1142	2.5204	1192	2.5581
1093	2.5695	1143	2.5575	1193	2.5465
1094	2.5509	1144	2.5685	1194	2.5623
1095	2.5696	1145	2.5331	1195	2.5779
1096	2.5463	1146	2.5605	1196	2.5503
1097	2.5509	1147	2.5212	1197	2.5534
1098	2.5633	1148	2.5881	1198	2.5589
1099	2.5577	1149	2.5524	1199	2.5395
1100	2.5457	1150	2.5525	1200	2.5734

European Commission

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Abstract

The concept of directly spiking samples of the solution with dissolved irradiated nuclear fuel for later dilution and measurement of the isotopic contents of uranium and plutonium has been developed successfully. Large sized dried spikes (LSD) have become a fundamental part of the fissile material control of irradiated nuclear fuel. In the frame of providing these spikes to the nuclear industry, a new set of LSD Spikes for the determination of uranium and plutonium by isotope dilution mass spectrometry in solutions of spent fuel from reprocessing plants has been prepared and certified for uranium and plutonium isotopic contents. The methodology followed was comparable to that of previous batches. The solution, made by dissolution of the starting materials in nitric acid, was dispensed directly into individual penicillin vials. However in this campaign an automated system was introduced to dispense and weigh the majority of vials. At the same time the system was validated by comparing the results with those of the traditional syringe dispensing and substitution weighing.

This new batch of large size dried spikes contains ca. 50 mg of uranium (^{235}U abundance = 19.1%) and ca. 1.8 mg of plutonium (^{239}Pu abundance = 97.8%) in each individual vial, covered with a light layer of organic material (cellulose acetate butyrate) as stabilizer.

The U and Pu amount content was certified based on values from mass metrology of the validated automated system or from the manual weighings. Verification of the amount contents of the spike was done by IDMS at IRMM. The values measured for the batch solution and of the dried covered spikes agreed well with those calculated from the weights of starting materials dissolved and the weights of the final solution.

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