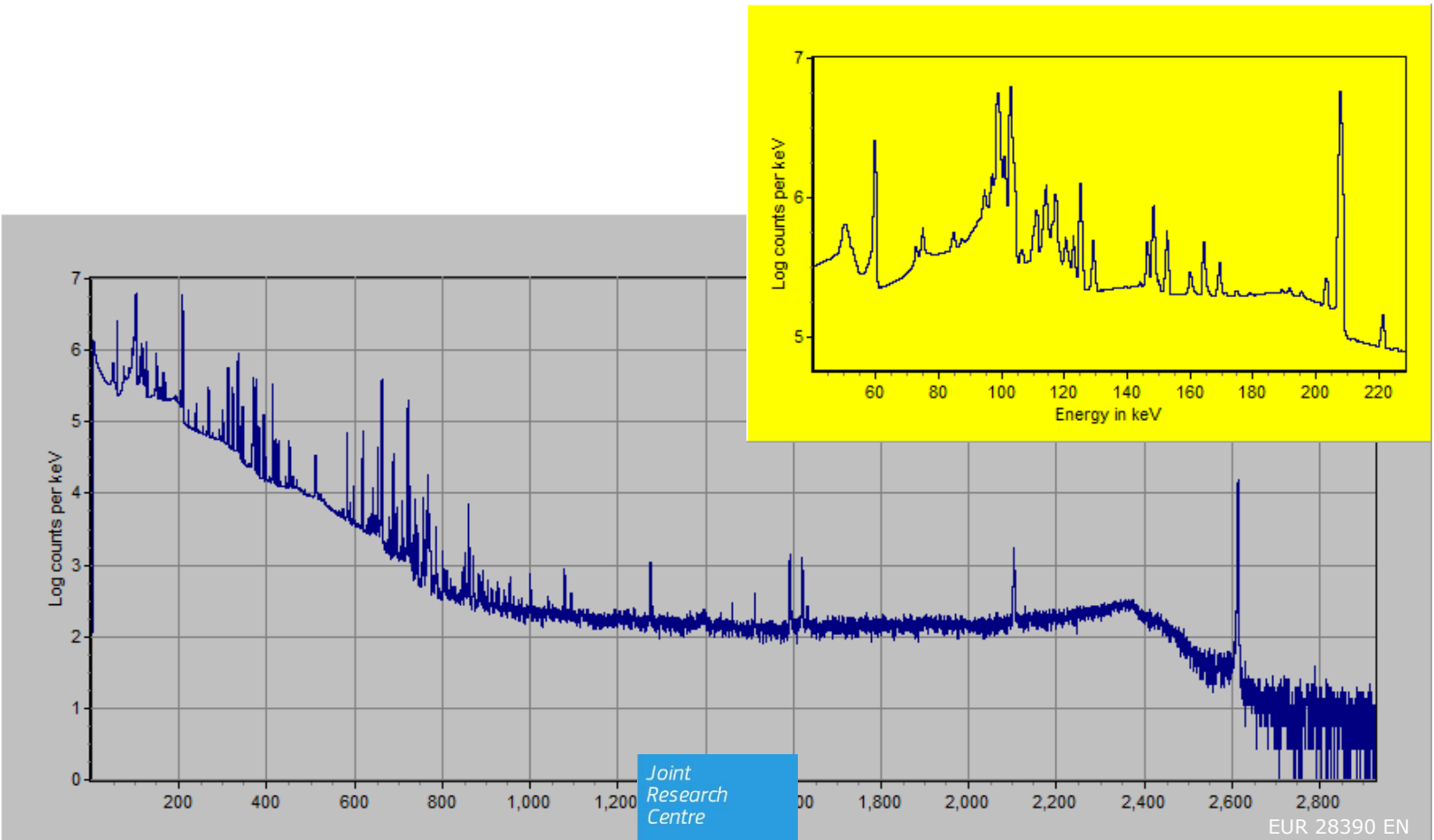
European Commission

JRC TECHNICAL REPORTS

Collection of high-resolution gamma spectra of certified Pu reference materials

Jozsef Zsigrai
Artur Mühleisen

2016



Joint Research Centre

EUR 28390 EN

Collection of high-resolution gamma spectra of certified Pu reference materials

This publication is a Technical report by the Joint Research Centre, the European Commission's in-house science service. It aims to provide evidence-based scientific support to the European policy-making process. The scientific output expressed does not imply a policy position of the European Commission. Neither the European Commission nor any person acting on behalf of the Commission is responsible for the use which might be made of this publication.

Contact information

Name: Jozsef Zsigrai, Artur Mühleisen

Address: European Commission, Joint Research Centre, Directorate for Nuclear Safety and Security, G.II.6

E-mail: Jozsef.Zsigrai@ec.europa.eu

Tel.: +49 (0)7247-951-871

JRC Science Hub
https://ec.europa.eu/jrc

JRC103886

EUR 28390 EN

ISBN 978-92-79-64836-6 (PDF)

ISSN 1831-9424 (online)

doi: 10.2789/359955 (online)

How to cite: Jozsef Zsigrai, Artur Mühleisen: Collection of high-resolution gamma spectra of certified Pu reference materials, JRC103886; European Atomic Energy Community, 2016

Table of contents

Abstract

A collection of high resolution gamma-ray spectra from well-characterized Pu certified reference materials has been recorded using a coaxial HPGe detector in the energy range up to 2.8 MeV. The spectra were measured for long acquisition times, ensuring very good counting statistics. The experimental setup assures that the measurement geometry is stable and reproducible, and that the spectra have minimum influence from background radiation and pile-up effects. The spectra will fill a gap in the data library of the International working group on gamma spectroscopy techniques for U and Pu isotopics. They can be used for testing different software algorithms for spectrum evaluation. They will also serve for comparison with spectra taken by medium-resolution detectors.

1. Introduction

The data library of the International working group on gamma spectrometry techniques for U and Pu isotopics (Ref. 1) contains spectra of U and Pu taken with various detectors under various conditions. These spectra are useful for testing the performance of software codes for spectrum evaluation. Recently spectra from medium-resolution detectors (LaBr3 and CdZnTe) have been added to the library (Ref. 2). The high-resolution spectra described in the current report were recorded by a HPGe detector in order to compare them to the medium-resolution spectra. However, they are also useful on their own for testing the performance of software codes for coaxial detectors in the energy range up to 2.8 MeV. The spectra are available upon request at JRC Directorate G, Department of Nuclear Safety and Security, Nuclear Safeguards and Forensics Unit, and will be offered to the data library of International working group on gamma spectroscopy techniques for U and Pu isotopics.

2. Experimental setup for Pu measurements

The spectra have been measured at EC JRC Directorate G in Karlsruhe (former Institute for Transuranium Elements), in a lab dedicated to HRGS (therefore with a slightly lower background radiation as in the case of MRGS measurements (Ref. 2) that have been performed in a lab where also other measurements take place). The equipment consisted of Canberra HPGe COAX detector (model GEM-50195-P), absorbers (3mm steel and/or 1mm Cd), Canberra Lynx electronics and PC with installed GENIE2000 software. The recorded spectra have 8192 channels with energy range up to 2.8 MeV. The electronics has been set up accordingly (see Attachment). Intentionally, instead of optimizing absorbers for HRGS, the same absorbers with the same thickness as for MRGS have been used.

A shielded experimental setup with well-defined geometry has been used for the gamma-spectrometric measurements (Figure 1 and Figure 2). The shield consists of 10 cm Pb, 2 mm of Cu and 4.5 mm PMMA. PMMA stands for poly(methyl methacrylate) ("plexiglass") with chemical composition $(C_5O_2H_8)_n$. Mechanical setup for holding the Plutonium samples at a fixed distance above the detector has been manufactured at the in-house workshop. The setup allows for introduction of absorbers of different thicknesses between the source and the detector without affecting the source-to-detector distance. The distances between elements of the experimental setup and the thicknesses of materials are presented in Table 1. The sample-to-detector distances are distances between the top of the detector and the lowest point of the sample's encapsulation. The sample holder plates ("top" in Table 1) have a round engraving at their middle to facilitate centred sample positioning on the detector axis. "Base plate" and "spacer" are PMMA tubing with inner diameter slightly smaller than outer diameter of the detector (but larger then active detector diameter). "Absorber cover" is PMMA element with a hole in the middle (the diameter of that hole is the same as for "spacer"). Both "top" and "absorber support plate" have a finite thickness across their widths and are made of machined PMMA.

Table 1: Setup distances and the thicknesses of materials (*PMMA="plexiglass")

PMMA elements	mm				
	distance	air	PMMA	Cd	Fe
base plate	4	4	0	0	0
spacer	195	195	0	0	0
absorber support plate	5.2	1.8	3.4	0	0
absorber cover (1mmCd)	10	9	0	1	0
absorber cover (Cd+Fe)	10	6	0	1	3
top	10	7	3	0	0
sum (1mm Cd)	224.2	216.8	6.4	1	0
sum (1mmCd+3mmFe)	224.2	213.8	6.4	1	3

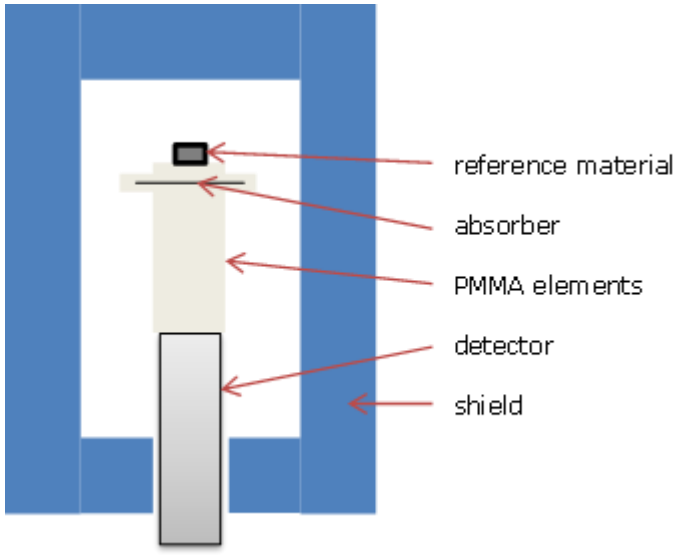


Figure 1: Schematics of experimental setup for the measurements.

Figure 2: Fig. 2 presents the photos of the experimental setup for COAX detector and Pu samples: a. outside view of the experimental setup (enclosure open and PMMA support for sample next to it), b. measurement enclosure as setup for measurements with 1mm Cd absorber and c. only enclosure with HPGe detector.

3. Pu reference materials

The reference materials measured have been PIDIE-1, 3, 5 and 7 (described in Refs. 3-5) and items Pu-61, 70, 84 and 93 from the CBNM Nuclear Reference Material set "271" (described in Refs. 6-7).

The PIDIE samples consist of ca. 0.425g Pu each in the form of a PuO_2 pressed pellet encapsulated in a welded steel container. The measured samples are from set number 4. The samples have been manufactured specifically for inter-comparison of the measurement capabilities of gamma spectrometry (Ref 4). The results of accompanying destructive analyses performed during that exercise are presented in Ref. 3. Unfortunately the exact mass of the pellets has not been recorded (Ref. 5). Isotopic composition of PIDIE samples based on data from Appendix A in Ref. 4, recalculated to weight % and renormalized to total Pu is presented in Table 2. The samples' drawing is presented in Figure 3.

Table 2: Isotopic composition of PIDIE samples in weight % (normalized to sum of Pu isotopes) with 2s absolute uncertainty for reference date 1.1.1988.

Reference sample		Isotope					
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
PIDIE 1	weight %	0.01101	93.7650	5.99025	0.19920	0.0346	0.2304
	2s	0.00033	0.0065	0.0052	0.00255	0.0015	0.0060
PIDIE 3	weight %	0.04716	84.5795	14.1442	0.9953	0.2338	0.6282
	2s	0.00038	0.0094	0.0052	0.0036	0.0075	0.0151
PIDIE 5	weight %	0.1314	75.8862	21.2169	2.0638	0.7017	1.7488
	2s	0.0011	0.0147	0.0115	0.0042	0.0015	0.0387
PIDIE 7	weight %	1.253	61.9848	25.5941	6.4919	4.6763	3.5287
	2s	0.016	0.0420	0.0195	0.0132	0.0081	0.1111

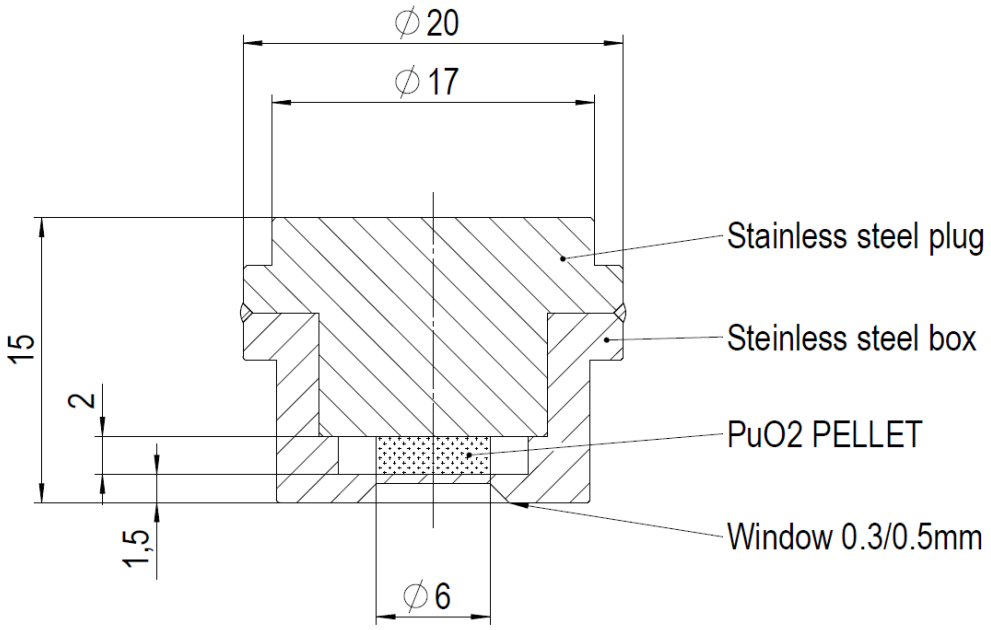


Figure 3: Drawing of a PIDIE sample (reproduced based on Ref. 3).

The CBNM Nuclear Reference Material "271" (Refs. 6 and 7) has been produced by Central Bureau for Nuclear Measurements of EC JRC. It consists of four plutonium oxide sintered pellets encased in stainless steel and protected by a plastic cap. Each pellet contains 6.65±0.06 g of PuO_2. The protective cap has been left on during the measurements. Plastic cap has a thickness of 2 mm, in addition there is a ca. 1mm air gap between the plastic and the metallic bottom of the sample (overall height of the sample is 24.1±0.1 mm). The samples' drawing and description are also presented in Refs. 6-7. The reference sample Pu93 is from set no. 0/10 and the reference samples Pu61, Pu70 and Pu84 are from set no. 0/12. The isotopic composition of the reference samples from Ref. 6 is for convenience presented in Table 3 and the samples' drawing in Figure 4.

Table 3: Isotopic composition of CBNM standard's reference samples in weight % with 2s absolute uncertainty and reference date 20.6.1986.

Reference sample		Isotope					
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
CBNM Pu93	weight %	0.0117	93.4123	6.3131	0.2235	0.0395	0.1047
	2s	0.00003	0.004	0.0039	0.0004	0.0003	0.0021
CBNM Pu84	weight %	0.0703	84.3377	14.2069	1.0275	0.3576	0.2173
	2s	0.0006	0.0084	0.0085	0.0018	0.001	0.0022
CBNM Pu70	weight %	0.8458	73.3191	18.2945	5.4634	2.0772	1.1705
	2s	0.0018	0.0098	0.0087	0.0034	0.0023	0.0117
CBNM Pu61	weight %	1.1969	62.5255	25.4058	6.6793	4.1925	1.4452
	2s	0.0025	0.0283	0.0241	0.0087	0.0064	0.0144

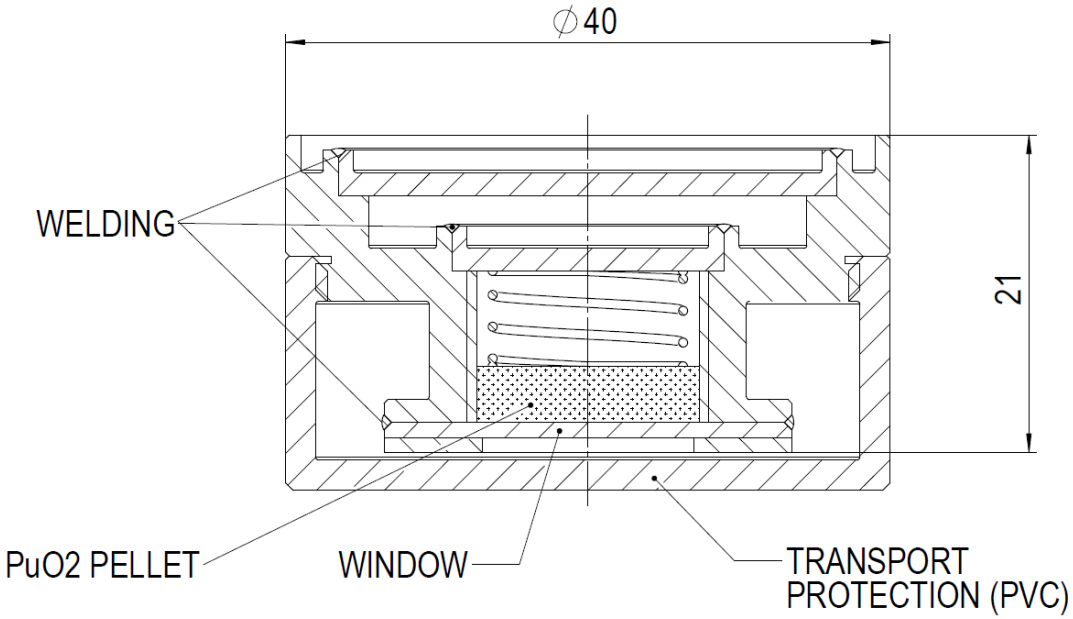


Figure 4: Drawing of a CBNM reference sample. (Reproduced based on Ref. 7)

4. Measurements and experimental conditions

Spectra of each reference sample with 50.000 to 150.000s live time per spectrum have been recorded. The logbook in the Attachment provides names of the recorded spectra, dates of the measurements, live time per particular spectrum, detector dead time as well as data on absorbers and detector-to-sample distances used. The spectra, converted to Ortec CHN format, are proposed for introduction to the data library of the International working group on gamma spectroscopy techniques for U and Pu isotopics (Ref. 1).

5. Conclusion

High quality high-resolution gamma spectra of Pu reference samples have been measured by HPGe detector. They will be used in support of phase I of the international exercise on medium resolution gamma spectrometry. The measured spectra are also suitable for validation of HRGS analysis software.

References

1. ESARDA website, Data library of The international working group on gamma spectroscopy techniques for U and Pu isotopics, https://esarda.jrc.ec.europa.eu/index.php?option=com_content&view=article&id=117&Itemid=255
2. J. Zsigrai, A. Mühleisen, M. R. Pascual: Collection of medium-resolution gamma spectra of certified Pu reference materials, JRC technical report 98337, http://publications.jrc.ec.europa.eu/repository/handle/JRC98337, 2015
3. R.J.S. Harry: PIDIE, plutonium isotopic determination inter-comparison exercise, Annual Meeting of the Institute of Nuclear Materials Management, Los Angeles (USA), ECN-RX--90-044, 1990; https://inis.iaea.org/search/search.aspx?orig_q=RN:22045752
4. J. Morel, B. Chauvenet: Intercomparaison des mesures de composition isotopique du plutonium par spectrometrie X et gamma. Resultats de l'action 'Pidie', rapport final, CEA Centre d'Etudes de Saclay, Gif-sur-Yvette (France). Dept. des Applications et de la Metrologie des Rayonnements Ionisants, 1991; https://inis.iaea.org/search/search.aspx?orig_q=RN:23077811
5. P.M.J. Chard, S. Croft, P.B. Sharp : Characterisation of the Harwell N95 high efficiency passive neutron counter, 17th annual ESARDA symposium, p. 551, Aachen, 1995; https://esarda.jrc.ec.europa.eu/index.php?option=com_jifile&filename=NzkwNzYwMjQyYTEwMzQ3OWYyN2ZjYWE5MThhODAyMDA=
6. CBNM Nuclear Reference Material 271, Certificate of Analysis, Commission of the European Communities, Joint Research Centre, Central Bureau for Nuclear Measurements, Geel, 1989
7. R.J. Friar, T.E. Sampson: Plutonium isotopic abundance measurements on CBNM NRM 271 analyzed with the FRAM and MGA codes, Los Alamos National Laboratory report LA 12309-MS, 1992; http://www.osti.gov/scitech/biblio/10140268

Attachment: List of spectra

List of spectra: Measurements of CBNM 271 and Pidie reference materials with HPGe COAX detector

Meas #	Sample	Replicate #	Distance, mm	Shield	LT, sec	DT, %	Meas date YYYY-MM-DD	Spectrum name
1	Pu-61	1	224.2	1 mm Cd + 3 mm steel	50000	18.42	09/12/2014	Pu61_COAX_1mmC_50000s.cnf
2	Bkg	1	-	1 mm Cd + 3 mm steel	64000	0.02	10/12/2014	Bkg1_COAX_1mmCd.cnf
3	Pu-70	1	224.2	1 mm Cd + 3 mm steel	50000	15.77	11/12/2014	Pu70_COAX_1mmCd_50000s.cnf
4	PIDIE-7	1	224.2	1 mm Cd + 3 mm steel	50000	3.38	12/12/2014	Pidie7_COAX1_1mmCd3mmSteel_50000s.cnf
5	PIDIE-7	2	224.2	1 mm Cd + 3 mm steel	50000	3.39	13/12/2014	Pidie7_COAX2_1mmCd3mmSteel_50000s.cnf
6	PIDIE-7	3	224.2	1 mm Cd + 3 mm steel	50000	3.38	13/12/2014	Pidie7_COAX3_1mmCd3mmSteel_50000s.cnf
7	PIDIE-7	4	224.2	1 mm Cd + 3 mm steel	50000	3.39	14/12/2014	Pidie7_COAX4_1mmCd3mmSteel_50000s.cnf
8	PIDIE-1	1	224.2	1 mm Cd	150000	0.92	15/12/2014	Pidie1_COAX_1mmCd_150000s.cnf
9	Bkg	2	224.2	1 mm Cd	50000	0.02	14/01/2015	Bkg2_COAX_1mmCd.cnf
10	Pu-84	1	224.2	1 mm Cd	50000	9.44	15/01/2015	Pu84_COAX_1mmCd_50000s.cnf
11	Pu-93	1	224.2	1 mm Cd	150000	6.61	19/01/2015	Pu93_COAX_1mmCd_150000s.cnf
12	PIDIE-3	1	224.2	1 mm Cd	150000	1.68	21/01/2015	Pidie3_COAX_1mmC_150000s.cnf
13	PIDIE-5	1	224.2	1 mm Cd	150000	3.29	23/01/2015	Pidie5_COAX_1mmCd_150000s.cnf
14	Bkg	3	-	1 mm Cd	150000	0.02	26/01/2015	Bkg3_COAX_1mmCd.cnf

Notes to List of spectra:

Date of measurement is date of start of the measurement (the same as stated within the spectrum)

Requirements: Pu240 peak at 642.37 keV should be measured with 1% uncertainty (not always possible)

Settings:

Detector HV:	3300
Amplifier polarity	positive
gain	x2
fine gain	1.10326
PUR guard	1.10x
offset	0

CBNM standards' dose rates at contact

Pu-61	15	mSv/h
Pu-70	10	mSv/h
Pu-84	3	mSv/h
Pu-93	1	mSv/h

Measured reference materials are from:

Pu-61	CBNM 271 set 0/12
Pu-70	CBNM 271 set 0/12
Pu-84	CBNM 271 set 0/12
Pu-93	CBNM 271 set 0/10
PIDIEs	are from set number 4

For all measurements the distance between the detector and reference material is the same

Overall distance:	224.2 mm
of which PMMA:	6.4 mm

air = overall distance - PMMA distance - absorber thickness

absorbers (=shield) used are the same as for MRGS measurements

List of abbreviations and definitions

HPGe COAX	High Purity Germanium Co-axial
HRGS	High Resolution Gamma Spectrometry
ITU	Institute for Transuranium Elements
MRGS	Medium Resolution Gamma Spectrometry
NDA	Non Destructive Analysis
PMMA	Poly(Methyl MethAcrylate)

List of figures

List of tables

LC-NA-28390-EN-N

JRC Mission

As the science and knowledge service of the European Commission, the Joint Research Centre's mission is to support EU policies with independent evidence throughout the whole policy cycle.

EU Science Hub
ec.europa.eu/jrc

@EU_ScienceHub

EU Science Hub - Joint Research Centre

Joint Research Centre

EU Science Hub

Publications Office

doi: 10.2789/359955

ISBN 978-92-79-64836-6