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M. Marouli, G. Lutter, S. Pommé, R. van Ammel, M. Hult, et al.. Measurement of absolute γ -ray emission probabilities in the decay of $^{227}\mathrm{Ac}$ in equilibrium with its progeny. Applied Radiation and Isotopes, 2019, 144, pp.34-46. 10.1016/j.apradiso.2018.08.023 . cea-04550280

HAL Id: cea-04550280 https://cea.hal.science/cea-04550280v1

Submitted on 17 Apr 2024 $\,$

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Contents lists available at ScienceDirect

Applied Radiation and Isotopes



journal homepage: www.elsevier.com/locate/apradiso

Measurement of absolute γ -ray emission probabilities in the decay of ²²⁷Ac in equilibrium with its progeny



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HIGHLIGHTS

• Measured gamma-ray emission probabilities in ²²⁷Ac decay series.

• Mean values taken from 4 laboratories.

· Source activity determined by primary standardisation.

• Results reported for 87 γ-ray lines.

· Generally in good agreement with literature values.

ARTICLE INFO	A B S T R A C T
Keywords: γ-ray intensities ²²⁷ Ac ²²⁷ Th ²²³ Ra Decay data γ-ray spectrometry	The emission probabilities of γ rays produced in the ²²⁷ Ac decay series were determined by high-resolution γ -ray spectrometry of sources with standardised activity. The sources were prepared quantitatively on glass discs by drop deposition of a solution with ²²⁷ Ac in radioactive equilibrium with its daughter nuclides. Their activity was measured by a primary standardisation technique based on alpha-particle counting at a defined low solid angle. Four laboratories performed γ -ray spectrometry and derived absolute γ -ray intensities. Mean values were calculated and compared with literature data and the currently recommended evaluated data. New values on control were emphasibilities are prepared.

1. Introduction

The ²²⁷Ac decay series has been the subject of increased metrological interest in recent years. Firstly, it is part of the ²³⁵U decay chain, which is ubiquitously present in nature and considered a health risk where industry processing natural resources enhances the concentration of natural occurring radionuclides (NOR) in its products, byproducts and waste. Of particular concern are the alpha decays from ²²⁷Ac and progeny at the end of the ²³⁵U decay chain, which can cause a high internal dose after incorporation in the human body. Secondly, the daughter nuclides ²²⁷Th and ²²³Ra and their progeny are becoming increasingly important for radioimmunotherapy (Seidl, 2014). The capacity of alpha particles to cause DNA double-strand breaks which trigger cell death can be successfully applied in treatment of disseminated cancer cells and small tumour cell clusters (Humm et al., 2015; Kratochwil et al., 2016).

Accurate decay data of the ²²⁷Ac decay series are needed to provide the necessary metrological tools to verify exemption levels in the NORM industry (EURATOM, 2014) by γ -ray spectrometry and to calibrate ionisation chambers for the quantified therapeutic use of ²²⁷Th-²²³Ra in nuclear medicine. In recent work, improved γ -ray intensities in the decay of ²³⁵U have been published in support of SI-

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https://doi.org/10.1016/j.apradiso.2018.08.023

Received 20 March 2018; Received in revised form 27 July 2018; Accepted 28 August 2018 Available online 30 August 2018 0969-8043/ © 2018 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (http://creativecommons.org/licenses/BY/4.0/).

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Ellapsed time from purification (days)

Fig. 2. Theoretical relative activities in the 227 Ac decay chain. A₀ is the initial ^{227}Ac activity and A_i the activity of the decay product.



Fig. 3. Alpha-particle spectra of a $^{\rm 227}Ac_{\rm eq}$ source and a background spectrum with decays from recoiled atoms in the PIPS® detector taken at JRC.

traceability of radiological measurements (Marouli et al., 2018), in particular in the context of NORM. Mostly in support of alpha immunotherapy, specific studies have been performed on the half-lives of ²²⁷Th (Collins et al., 2015c) and ²²³Ra (Bergeron and Fitzgerald, 2015; Collins et al., 2015a; Kossert et al., 2015), primary standardisation of their activity and determination of the γ -ray emissions in their decay



Fig. 4. Alpha spectrum of a ²²⁷Ac_{eq} source (Ac227G1504), measured at JRC-Geel with the defined solid angle alpha counting set-up.

Table 1				
Activity of 227	Ac in the s	source	s standard	ised by DSA
alpha-particle	counting	and	standard	uncertainty
(k = 1).				

Source	Activity $(Bq)^{\dagger}$
Ac227G1502	1183 (9)
Ac227G1507	1077 (7)
Ac227S1508	945 (8)
Ac227S1509	1103 (6)

* Reference date: 17th June 2015.

Table 2

.. .

Typical uncertainty components of the ²²⁷Ac activities (k = 1).

Component	Relative uncertainty (%)
Counting statistics	0.10
Dead time (+pile-up)	0.08
Background	0.01
Detection efficiency	0.15
Extrapolation to zero energy	0.50
Absorption	0.10
Scattering	0.02
Non-equilibrium	0.15
Impurities	0.10
Total	0.57



Fig. 5. $\gamma\text{-ray}$ spectrum of a $^{227}\text{Ac}_{eq}$ source measured at JRC-Geel. The selected $\gamma\text{-}$ ray peaks for the estimation of the source activity are colour and shape coded.

Table 3

Activity measurement of the Ac227G1509 source by	γ-ray	spectrometry.
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	Ge-T5	Ge-5
Radionuclide	Activit	ty (Bq)
²²³ Ra	1110(50)	1077(51)
²¹⁹ Rn	1070(60)	1082(53)
²¹¹ Pb	1115(55)	1156(57)
²¹¹ Bi	1130 (60)	1145(57)
²²⁷ Ac	1107 (50)	1115(51)
	1110	(50)*

* Power moderated mean value.

(Keightley et al., 2015; Kossert et al., 2015; Collins et al., 2015b; Zimmerman et al., 2015; Pibida, 2015), and methodology for age dating of a ²²⁷Th-²²³Ra material (Pommé et al., 2016). In previous studies, the decay characteristics of the ²³⁰U (Pommé et al., 2012a, 2012b; Marouli et al., 2012; Suliman et al., 2012) and ²²⁵Ac (Pommé et al., 2012c; Marouli et al., 2013; Suliman et al., 2013) decay chains have been investigated for the same purpose.

Actinium-227 (21.77 a) decays to stable ²⁰⁷Pb through a series of twelve alpha and beta emitters (DDEP, 2004-2016) (Fig. 1). Following the main decay path, a plethora of γ rays is emitted predominantly from the 227 Th \rightarrow 223 Ra and 223 Ra \rightarrow 219 Rn decays. Taking into consideration initially only evaluated nuclear decay data by the Decay Data Evaluation Project (DDEP) and Nuclear Data Sheets (NDS), ²²⁷Ac decays mainly to ²²⁷Th by β^- decay followed by γ rays which have not been measured directly but deduced from the absolute β^- emission probabilities and the total conversion coefficients (Kondev et al., 2016). Recommended γ -ray intensities for the 227 Th \longrightarrow 223 Ra decay come from measurements dating back to 1969, with the most recent one from G. Ardisson and published in a private communication by Abdul-Hadi et al. (1993). The levels of ²¹⁵Po are fed from the ²¹⁹Rn \rightarrow ²¹⁵Po alpha decay. Evaluations of the γ -ray emission probabilities by DDEP and NDS (Lee, 2013) include results dating back to 1967. The latest one is from 1999 by Liang et al. (1999) who observed the alpha decay of ²¹⁹Rn and coincident γ rays and electrons. In many cases, the results from the different studies are not in agreement. For the only γ ray observed from the alpha decay of ²¹⁵Po, two discrepant experimental values exist at 0.048 (5) % (Briancon et al., 1968) and 0.064 (2) % (Davidson, Connor,

Table 4

Experimental conditions of the γ -ray intensity measurements.

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Table 5 Absolute $\gamma\text{-ray emission probabilities measured in the <math display="inline">^{227}\text{Ac}{\longrightarrow}^{227}\text{Th}$ decay.

	-					•
Fnergy (keV)	DDFP	IRC	FNFA	CEA	CMI	This work
	DDD	DICC.	LINL/1	0.041	CIVIL .	THIS WOLK

0, 1	•		
37.9	0.049	0.050 (5)	0.050 (5)

1970). ²¹¹Pb \rightarrow ²¹¹Bi is a well-studied decay with values on 22 γ -ray emission probabilities published in 12 papers. The subsequent alpha decay (99.724 %) of ²¹¹Bi \rightarrow ²⁰⁷Tl is followed by a single γ ray with an absolute emission probability in the most recent evaluations being calculated using the total internal conversion coefficient. The latest experimental value is provided by Momeni (1982). Both calculated (13.0 %) and experimental (13.3 %) values are in agreement. One γ ray is emitted in the ²⁰⁷Tl \rightarrow ²⁰⁷Pb β ⁻ decay. The γ -ray intensity is deduced from the intensity ratio of the 897.77 keV ²⁰⁷Tl γ -ray line to the 351 keV from the ²¹¹Bi \rightarrow ²⁰⁷Tl decay as discussed by Hindi et al. (1988).

One of the objectives of the EMRP project 'JRP IND57 MetroNORM: Metrology for processing materials with high natural radioactivity' was to improve the quality of available data for γ -ray emission probabilities in the decay of the ²²⁷Ac and its progeny. Experimental work from the four out of the five laboratories participating in the project is presented in this paper. The fifth laboratory, Laboratorio de Metrología de Radiaciones Ionizantes CIEMAT in Spain, has withdrawn its results at the final stage for technical reasons. Most of the emission probabilities derived from this work agree with published values and uncertainties have been reduced in some cases.

2. Source preparation

2.1. Source production of ²²⁷Ac

The $^{227}Ac_{eq}$ (i.e., ^{227}Ac in equilibrium with its daughter products) solution was provided to JRC-Geel (JRC-IRMM before 2016) by JRC-Karlsruhe, four years after its production, (JRC-ITU before 2016) in a tip vial containing 0.5 mL (0.1 M HCl) solution with an activity of 4.5 MBq (on the reference date of March 2011). The material was originally produced by neutron irradiation of 226 Ra at the BR2 reactor at the Belgian nuclear research institute (SCK•CEN, 2015). The separation of

	JRC	ENEA	CEA	CMI
Detector	HPGe, Planar BEGe, Thin dead layer, Underground lab (Hades)	HPGe (p-type)	HPGe (n-type), Underground lab	HPGe
FWHM	0.91 keV @ 122 keV, 1.93 keV @ 1332.5 keV	0.9 keV @ 122 keV, 1.9 keV @ 1332.5 keV	0.99 keV @ 122 keV, 2.05 keV @ 1332.5 keV	1.8 keV @ 1332.5 keV
Relative efficiency	50 %	40 %	52 %	40 %
Live time (days)	41	5	4	6
Source-detector distance (mm)	120	100	50	100
FEP Efficiency	PTB point sources + EGSnrc ^I	GESPECOR 4.2 ^{II}	Point sources + AÇORES ^{III}	MCNP ^{IV}
Coincidence corrections	EGSnrc	GESPECOR 4.2	ETNA ^V	MCNP

^I Kawrakow et al. (2015).

^{II} Sima et al. (2001).

^{III} LNHB (2008).

^{IV} Goorley et al. (2012).

^v Piton et al. (2000).

²²⁷Ac from ²²⁶Ra was performed by repeated extraction chromatography using RE resin (Apostolidis et al., 2005). The aliquot was further purified by extraction chromatography using DGA-branched resin (TEHDGA) as described by Zielinska et al. (2007).

2.2. Sources for activity measurement

 227 Ac had reached equilibrium with the decay products at the start of the source preparation, (Fig. 2). One of the decay products, the noble gas 219 Rn ($T_{1/2} \approx 4$ s), poses a risk of contamination during the handling of the material. This risk is also present when manipulating deposited open sources. Additionally, alpha-emitters gain recoil energy of roughly 100 keV during decay, which is enough for some decay products to escape from an open source and spread in the air. Safety measures were taken to reduce the risks of radiation contamination by

Table 6 Absolute γ -ray emission probabilities (%) measured in the ²²⁷Th \rightarrow ²²³Ra decay inhalation. To avoid possible contamination, source preparation was carried out in glove boxes.

To obtain thin sources with small crystals, the 34-mm-diameter glass source support discs were pre-treated before drop deposition. A solution of wetting agent (Tween[®] 1/40 diluted) and seeding agent (Ludox 1/100 diltued) was sprayed on the discs in a circular shape of 6 mm diameter using an airbrush pencil (Van Ammel et al., 2011) and dried in air. From the original solution, $20 \,\mu\text{L}$ (180 kBq) was transferred to a double-sealed ampoule. Carrier solution (3.6 mL 0.1 M HCl) was added, resulting in an approximate activity concentration of 1.0 kBq/20 μ L. From this ampoule, 18 sources were prepared on the glass discs and one (Ac227SS1508) on a 25.4 mm stainless steel disc. After the deposition, the sources were dried in open air without the aid of a heating device. The sources were subsequently covered with two VYNS (vinyl chloride vinyl acetate copolymer) foils of $35 \,\mu\text{g/cm}^2$ thickness.

91.580.688 (12)0.071 (6)0.071 (6)40.20.050 (3)0.067 (S)0.067 (S)0.067 (S)0.067 (S)0.065 (S)0.065 (S)0.065 (S)0.065 (S)0.065 (S)0.065 (S)0.056 (S)0.052 (S)0.053 (S)0.052 (S)0.053 (S)0.052 (S)0.053 (S)0.053 (S)0.056 (S)0.056 (S)0.056 (S)0.056 (S)0.056 (S)0.056 (S)0.056 (S)0.056 (S)0.011 (S)0.017 (S)0.011 (S)0.017 (S)0.010 (S)0.017 (S) </th <th>Energy (keV)</th> <th>NDS</th> <th>JRC</th> <th>ENEA</th> <th>CEA</th> <th>СМІ</th> <th>This work</th>	Energy (keV)	NDS	JRC	ENEA	CEA	СМІ	This work
40.20.057 (a)0.067 (b)0.067 (b)0.063 (b)61.4110.090 (13)0.663 (b)0.663 (b)0.663 (b)113.110.700.82 (b) ^b 0.208 (2)0.82 (d)117.20.199 (22)0.201 (12)0.208 (12)0.203 (0)123.580.014 (b)0.0068 (13)0.017 (2)0.017 (2)156.190.0053 (2)0.017 (2)0.017 (2)0.017 (2)166.360.015 (3)0.017 (2)0.017 (2)0.017 (2)166.360.015 (3)0.012 (1)0.020 (1)0.019 (5)169.950.005 (22)0.008 (1)0.020 (1)0.020 (1)199.950.015 (3)0.020 (2)0.020 (1)0.020 (1)204.940.043 (3)0.020 (2)0.020 (1)0.020 (1)204.940.043 (3)0.020 (2)0.020 (1)0.200 (1)204.940.055 (22)0.036 (1)0.020 (1)0.020 (1)204.940.043 (3)0.020 (2)0.020 (1)0.020 (1)204.940.023 (3)0.280 (1)0.260 (1)0.250 (1)204.940.13 (1)0.280 (1)1.26 (6)1.20 (4)1.31 (6)212.7 + 212.70.98 (13)0.97 (5)1.20 (4)1.31 (6)1.21 (5)213.960.101 (14)0.021 (2'0.021 (2')0.021 (2')0.021 (2')214.90.101 (14)0.021 (2')0.014 (6)0.017 (2)215.90.101 (14)0.021 (2')0.014 (6)0.017 (2)215.90.101 (1)0.261 (2')<	31.58	0.068 (12)	0.071 (6)				0.071 (6)
61.4110.090 (13)0.063 (5)°0.063 (5)°0.16 (1)113.120.21 (3)0.22 (5)°0.28 (1)°0.83 (5)°0.82 (3)°117.20.199 (22)0.201 (12)0.28 (12)0.200 (13)0.238 (13)123.580.014 (5)0.0066 (13)0.203 (12)0.203 (12)0.203 (13)141.420.119 (23)0.123 (6)10.110 (14)0.118 (8)150.140.011 (3)0.017 (2)10.017 (2)0.017 (2)0.017 (2)162.190.008 (3)0.012 (1)10.0109 (4)0.012 (1)169.950.055 (22)0.008 (1)10.020 (1)0.020 (1)197.560.055 (22)0.002 (2)0.020 (1)0.020 (1)0.020 (1)201.640.024 (3)0.202 (2)0.020 (1)0.020 (1)0.020 (1)204.14204.930.33 (16)10.262 (1)0.256 (1)0.200 (1)204.14204.930.33 (16)10.262 (1)0.265 (1)0.266 (1)204.141.26 (3)0.282 (13)0.262 (1)0.262 (1)0.263 (1)204.141.26 (3)1.25(5)1.20(4)1.21 (5)1.21 (5)1.20 (1)204.141.29 (1)1.28 (6)1.20 (1)1.26 (1)1.26 (1)0.27 (7)215.940.101 (4)0.21 (2)°1.20 (1)1.26 (1)1.26 (1)0.27 (7)215.940.101 (4)0.21 (2)°1.20 (1)1.83 (3)0.77 (7)215.950.101 (1)0.21 (2)°1.36 (1) <td>40.2</td> <td>0.015 (4)</td> <td>0.067 (5)</td> <td></td> <td></td> <td></td> <td>0.067 (5)</td>	40.2	0.015 (4)	0.067 (5)				0.067 (5)
62.45 + 62.680.13 (1)0.16 (1)0.83 (5)*0.83 (5)113.110.700.82 (5)*0.83 (5)*0.83 (5)*0.203 (9)123.580.014 (5)0.201 (12)0.208 (12)0.200 (13)0.203 (9)123.580.119 (23)0.123 (6)10.110 (14)0.118 (8)150.140.119 (23)0.017 (2)10.017 (2)0.008 (13)0.017 (2)166.360.005 (3)0.012 (1)10.019 (2)0.012 (1)0.012 (1)166.360.013 (4)0.009 (15)0.009 (15)0.000 (15)0.000 (15)201.640.024 (3)0.202 (2)0.020 (1)0.200 (1)0.200 (1)204.140.23 (3)0.202 (2)0.020 (1)0.200 (1)0.200 (1)204.140.23 (3)0.326 (15)1.20 (1)0.200 (1)0.200 (1)204.140.23 (3)0.326 (15)1.20 (1)0.200 (1)0.200 (1)204.140.23 (3)0.326 (15)1.20 (1)0.200 (1)0.200 (1)204.140.23 (3)0.326 (1)1.20 (1)0.200 (1)0.200 (1)204.140.39 (5)1.20 (1)0.200 (1)0.200 (1)0.200 (1)204.1420.49 (3)0.326 (1)1.20 (1)0.200 (1)0.200 (1)204.1420.99 (3)0.29 (1)1.26 (1)1.26 (1)1.26 (1)204.1420.99 (3)0.29 (1)1.26 (1)1.26 (1)1.26 (1)204.1420.99 (3)0.21 (1)1.26 (1)1.26 (1)1.26 (1) <tr< td=""><td>61.441</td><td>0.090 (13)</td><td>0.063 (5)^a</td><td></td><td></td><td></td><td>0.063 (5)</td></tr<>	61.441	0.090 (13)	0.063 (5) ^a				0.063 (5)
113.110.700.82 (s) ^b 0.82 (s)0.82 (s)0.82 (s)117.20.199 (22)0.201 (2)0.208 (2)0.200 (13)0.208 (3)123.580.014 (5)0.006 (13)0.110 (14)0.118 (8)141.420.119 (23)0.123 (6)1.10 (14)0.117 (2)150.140.001 (3)0.017 (2)0.017 (2)0.017 (2)164.360.005 (3)0.017 (2)0.010 (1)0.017 (2)166.360.005 (3)0.012 (1)0.0109 (5)0.012 (1)169.950.005 (22)0.006 (1)0.0109 (4)0.020 (17)201.640.024 (3)0.202 (2)0.0200 (1)0.020 (17)204.140.22 (3)0.202 (2)0.020 (1)0.200 (17)204.14 + 204.980.39 (5)1.25 (5)1.20 (4)1.16 (5)210.621.25 (14)1.21 (6)1.25 (6)1.20 (4)1.16 (5)211.621.25 (13)0.097 (5)1.16 (16)1.21 (5)1.21 (5)216.630.110 (14)0.097 (5)1.26 (6)1.26 (1)1.26 (7)215.951.11 (18)0.093 (5)1.16 (16)1.10 (5)1.17 (2)252.550.110 (12)0.124 (6)1.29 (1)0.29 (1)0.21 (2)252.570.51 (1)0.124 (6)1.29 (1)0.21 (2)0.23 (1)254.630.71 (4)0.71 (4)0.74 (6)1.87 (7)0.31 (8)252.570.51 (10)0.124 (6)1.51 (18)0.126 (6)1.29 (1)254.630.51 (4)0.53 (62.45 + 62.68	0.21 (3)	0.16 (1)				0.16 (1)
117.20.199 (2)0.201 (2)0.208 (2)0.200 (13)0.203 (9)123.580.014 (5)0.0066 (13)0.0066 (13)0.113 (0)0.017 (2)0.110 (14)0.017 (2)150.140.010 (3)0.017 (2)0.017 (2)0.017 (2)0.017 (2)163.360.005 (3)0.012 (1)0.012 (1)0.008 (1)0.017 (2)169.950.005 (22)0.008 (1)0.019 (4)0.008 (1)201.640.204 (3)0.020 (2)0.020 (1)0.020 (1)0.020 (1)204.140.23 (3)0.206 (1)0.236 (1)0.236 (1)204.980.39 (5)0.256 (1)0.236 (1)0.236 (1)204.090.39 (5)0.256 (1)0.236 (1)0.267 (1)204.090.39 (5)0.256 (1)0.236 (1)0.267 (1)215.70.098 (13)0.097 (5)1.20 (4)1.18 (5)1.21 (5)215.90.101 (1)1.28 (6)1.23 (6)1.26 (1)1.33 (6)1.77 (7)255.250.111 (18)0.93 (5)0.43 (13)0.77 (7)255.430.71 (4)0.23 (2)7.6 (4)6.88 (18)7.04 (22)7.0 (3)255.230.51 (4)0.534 (25)0.513 (18)0.23 (2)256.230.51 (4)0.54 (2)7.6 (4)6.88 (18)7.04 (22)7.0 (3)255.230.51 (4)0.534 (25)0.513 (18)0.53 (12)0.23 (1) <tr <td="">256.230.51 (4)</tr>	113.11	0.70	0.82 (5) ^b			0.83 (5) ^b	0.82 (4)
123.580.014 (5)0.0068 (13)0.0068 (13)0.010 (0)0.010 (0)0.010 (0)0.011 (0)0.017 (2)0.010 (1)0.017 (2)0.008 (1)0.008 (1)0.008 (1)0.008 (1)0.008 (1)0.009 (1)0.009 (1)0.000 (1) <td>117.2</td> <td>0.199 (22)</td> <td>0.201 (12)</td> <td>0.208(12)</td> <td></td> <td>0.200 (13)</td> <td>0.203 (9)</td>	117.2	0.199 (22)	0.201 (12)	0.208(12)		0.200 (13)	0.203 (9)
141.420.119 (23)0.213 (6)0.110 (14)0.118 (8)150.140.011 (3)0.017 (2)0.017 (2)0.017 (2)166.360.015 (3)0.012 (1)0.012 (1)169.950.0055 (22)0.008 (1)0.019 (2)0.019 (1)197.560.013 (4)0.020 (2)0.020 (1)0.020 (1)201.640.024 (3)0.020 (2)0.020 (1)0.200 (17)204.140.23 (3)0.200 (2)0.200 (1)0.200 (17)204.980.39 (5)0.256 (11)0.366 (16)206.080.25 (3)0.288 (14)2.66(27)0.256 (11)206.080.25 (3)0.288 (14)2.0(4)1.20(4)1.21 (5)216.71.25 (14)1.21 (6)1.25(6)1.20(4)1.21 (5)218.90.101 (14)0.037 (5)0.997 (5)0.997 (15)1.21 (5)218.90.101 (14)0.021 (2)*0.997 (5)0.997 (15)1.31 (5)1.30 (5)246.120.0123 (14)0.21 (2)*0.997 (15)1.30 (5)1.21 (14)1.30 (5)1.21 (14)255.50.111 (18)0.93 (12)*0.83 (3)0.77 (7)1.55 (3)0.53 (2)1.26 (1)256.230.71 (4)0.54 (4)0.54 (2)7.0 (3)0.52 (2)1.29 (15)0.53 (2)256.340.71 (4)0.54 (1)0.54 (2)0.513 (18)0.523 (2)256.350.71 (4)0.54 (1)0.53 (2)1.16 (5)1.16 (6)1.17 (8)279.80.56 (10)0.54 (1)0.54 (1)	123.58	0.014 (5)	0.0068 (13)				0.0068 (13)
150.140.011 (3)0.017 (2) 0.017 (2)162.190.008 (3)0.017 (2)0.017 (2)168.360.015 (3)0.012 (1)0.012 (1)169.950.013 (2)0.008 (1)0.0109 (5)201.640.024 (3)0.020 (2)0.020 (1)0.020 (1)204.140.223 (3)0.23 (3)0.200 (1)0.200 (17)204.980.16 (3)0.326 (16)0.262 (7)0.256 (11)0.206 (1)204.980.53 (5)0.253 (16)0.262 (7)0.256 (11)0.268 (13)204.980.39 (5)0.257 (1)0.256 (11)0.268 (13)0.267 (1)0.256 (11)205.080.25 (3)0.288 (14)1.25 (6)1.20 (1)0.256 (11)0.268 (13)210.621.25 (14)0.218 (1)1.25 (6)1.20 (1)0.265 (11)0.268 (13)218.90.110 (14)0.03 (5)0.097 (15)1.21 (5)1.21 (5)218.90.110 (14)0.013 (5)0.097 (15)1.30 (5)246.120.111 (14)0.021 (1)'0.117 (2)0.25 (1)252.50.111 (18)0.093 (5)0.140 (15)0.121 (1)254.630.71 (14)0.71 (4)0.120 (1)1.26 (5)272.910.51 (4)0.45 (5)1.46 (1)0.45 (1)0.45 (1)262.870.051 (4)0.46 (2)0.131 (8)0.33 (18)272.910.51 (4)0.59 (3)0.35 (1)0.35 (1)286.091.	141.42	0.119 (23)	0.123 (6)			0.110 (14)	0.118 (8)
162.190.008 (3)0.017 (2)0.017 (2)168.360.015 (3)0.012 (1)0.012 (1)0.012 (1)169.950.005 (22)0.008 (1)0.009 (1)0.000 (1)197.560.013 (4)0.02 (2)0.109 (4)0.020 (17)201.640.23 (3)0.202 (2)0.020 (1)0.200 (17)204.140.23 (3)0.36 (16)5.200 (17)0.200 (17)204.140.23 (3)0.36 (16)5.200 (17)0.200 (17)204.142.23 (3)0.36 (16)5.200 (17)0.200 (17)204.042.23 (3)0.36 (16)5.200 (12)0.256 (11)0.266 (13)204.142.43 (2)0.25 (3)0.288 (14)1.25 (5)0.262 (1)0.256 (13)210.621.25 (1)1.21 (5)1.21 (5)1.21 (5)1.21 (5)1.21 (5)212.7 + 212.70.968 (13)0.97 (5)1.28 (4)1.26 (4)1.21 (5)1.21 (5)218.91.10 (14)0.103 (5)1.28 (4)1.28 (4)1.30 (5)1.33 (5)1.33 (5)1.30 (5)235.961.20 (14)0.201 (2)1.24 (6)1.21 (5)1.30 (6)0.77 (7)1.25 (5)0.140 (8)0.77 (7)255.230.101 (12)0.124 (6)0.21 (2)0.24 (2)0.21 (2)0.23 (1)0.23 (2)0.23 (2)256.230.051 (14)0.54 (25)1.80 (8)1.85 (5)1.86 (6)0.53 (2)0.53 (2)279.80.054 (14)0.054 (25)1.80 (8)1.85 (5)1.86 (6)0.53 (2) </td <td>150.14</td> <td>0.011 (3)</td> <td>0.017 (2)</td> <td></td> <td></td> <td></td> <td>0.017 (2)</td>	150.14	0.011 (3)	0.017 (2)				0.017 (2)
168.360.015 (3)0.012 (1)0.012 (1)169.950.0055 (22)0.008 (1)0.008 (1)0.008 (1)201.640.024 (3)0.202 (2)0.020 (1)0.020 (1)204.142.023 (3)0.202 (1)0.200 (17)0.200 (17)204.980.16 (3)0.336 (16)0.256 (7)0.256 (11)0.336 (16)204.14 + 204.980.39 (5)0.256 (7)0.256 (11)0.368 (13)204.14 + 204.980.39 (5)0.262 (7)0.256 (11)0.368 (13)204.021.25 (4)1.21 (6)1.25 (6)1.20 (4)1.18 (5)1.21 (5)210.621.25 (1)0.99 (13)0.97 (5)0.97 (5)0.097 (5)0.097 (5)1.007 (5)218.90.110 (14)0.103 (5)0.97 (15)0.101 (6)0.97 (7)0.12 (2)0.11 (2)252.50.111 (18)0.93 (5)1.86 (8)7.04 (2)7.0 (3)0.12 (2)254.630.71 (14)0.71 (4)7.6 (4)6.88 (18)7.04 (22)7.0 (3)266.270.017 (12)0.514 (2)1.80 (8)8.8 (18)7.04 (22)7.0 (3)262.570.117 (8)0.514 (2)1.60 (6)0.533 (21)0.033 (18)0.04 (3)272.910.51 (4)0.046 (2)1.80 (8)1.85 (5)1.86 (6)0.533 (21)272.910.51 (4)0.46 (2)1.71 (8)0.77 (7)0.338 (18)0.533 (21)0.733 (18)284.420.040 (13)0.338 (18)1.80 (8)1.85 (5)1.86 (6)1.87 (7) <td>162.19</td> <td>0.008 (3)</td> <td>0.017 (2)</td> <td></td> <td></td> <td></td> <td>0.017 (2)</td>	162.19	0.008 (3)	0.017 (2)				0.017 (2)
169.950.0055 (22)0.008 (1)0.009 (1)0.009 (3)197.560.013 (4)0.010 (4)0.010 (4)0.010 (5)201.640.23 (3)0.20 (2)0.020 (1)0.200 (17)204.142.33 (3)0.336 (16)0.201 (1)0.201 (1)204.14 + 204.980.39 (5)0.151 (12)0.151 (12)0.151 (12)206.080.25 (3)0.288 (14)0.262 (7)0.256 (11)0.268 (13)210.621.25 (14)1.21 (6)1.25 (6)1.20 (4)1.85 (6)1.21 (5)212.7 + 212.70.98 (13)0.097 (5)0.097 (15)0.010 (6)1.33 (6)1.28 (6)0.017 (2)235.960.110 (14)0.103 (5)1.28 (6)1.28 (6)0.13 (5)1.30 (5)246.120.0123 (14)0.093 (5)1.28 (8)1.86 (8)1.61 (8) (8)0.117 (24)252.550.111 (18)0.93 (5)1.21 (5)0.33 (3)0.77 (7)254.630.107 (12)0.124 (6)7.6 (4)6.88 (18)7.04 (22)7.03 (13)256.2377.0 (6)6.84 (12)7.6 (4)6.88 (18)7.04 (22)0.23 (21)279.910.51 (14)0.90 (33) (8)1.20 (11) (5)0.124 (8)0.338 (18)0.373 (13)0.338 (13)279.920.56 (14)0.303 (18)1.80 (8)1.85 (5)1.86 (6)1.77 (8)0.338 (13)280.091.74 (21)1.90 (93) (3)1.80 (8)1.85 (5)1.86 (6)1.57 (8)299.94 * 300.52.22 (20)2.16 (10)2.2	168.36	0.015 (3)	0.012 (1)				0.012 (1)
197.560.013 (4)0.020 (2)0.0109(A)0.0109 (5)201.640.024 (3)0.020 (2)0.020 (1)0.200 (17)204.980.16 (3)0.336 (16)1.200.200 (17)0.151 (18)204.14 + 204.980.39 (5)0.262 (7)0.256 (11)0.366 (13)206.080.253 (3)0.288 (14)0.262 (7)0.256 (13)0.366 (13)210.621.25 (14)1.21 (6)1.25 (6)1.20 (4)1.18 (5)1.21 (5)212.7 + 212.70.098 (13)0.097 (5)0.097 (15)0.010 (6)1.31 (5)1.30 (5)235.961.29 (11)1.28 (6)1.33 (6)1.28 (4)1.31 (5)1.30 (5)252.50.111 (18)0.093 (5)1.40 (8)0.117 (24)254.630.17 (14)0.012 (2)*0.140 (8)0.117 (24)254.540.101 (14)0.093 (5)1.40 (8)0.117 (24)254.530.107 (12)0.124 (6)0.88 (18)0.410 (8)0.177 (7)254.640.107 (12)0.124 (6)1.80 (8)1.81 (8)0.413 (18)0.426 (1)272.910.51 (4)0.534 (25)1.51 (10)0.533 (18)0.533 (18)0.533 (18)0.533 (18)272.910.51 (4)0.038 (18)1.85 (5)1.86 (6)0.533 (18)0.338 (18)0.465 (12)0.338 (18)286.091.74 (21)1.09 (9)1.80 (8)1.85 (5)1.86 (6)0.359 (13)0.358 (18)0.56 (15)0.358 (18)299.94 + 300.52.26 (0)0.593 (1)1.65	169.95	0.0055 (22)	0.008 (1)				0.008 (1)
201.64 0.024 (3) 0.020 (2) $0.020(1)$ $0.020(1)$ 204.14 0.23 (3) 0.23 (16) 0.336 (16) 0.151 (17) 0.151 (18)204.98 0.39 (5) 0.39 (5) 0.526 (17) 0.256 (17) 0.256 (17) 0.256 (17) 0.256 (17)206.08 0.25 (3) 0.288 (14) $0.262(7)$ 0.256 (17) 0.997 (15) 0.997	197.56	0.013 (4)			0.0109(4)		0.0109 (5)
204.140.23 (3) $0.200 (17)$ 0.200 (17)204.980.16 (3)0.36 (16) $0.151 (18)$ 0.151 (17)0.151 (18)204.14 + 204.980.25 (3)0.288 (14)0.262(7)0.256 (11)0.268 (13)210.621.25 (14)1.21 (6)1.25 (6)1.20 (4)1.28 (5)0.200 (17)0.268 (13)212.7 + 212.70.098 (13)0.097 (5)0.097 (15)0.010 (6)235.9612.9 (11)12.8 (6)13.3 (6)12.8 (4)13.1 (5)13.0 (5)246.120.102 (14)0.201 (2°0.104 (8)0.117 (24)252.50.111 (18)0.093 (5) $$	201.64	0.024 (3)	0.020 (2)		0.020(1)		0.020 (1)
204.980.16 (3)0.336 (16)0.151 (17)0.151 (18)204.14 + 204.980.39 (5)0.396 (16)0.336 (16)0.336 (16)0.336 (16)206.080.25 (3)0.288 (14)0.262(7)0.256 (11)0.636 (13)210.621.25 (14)1.21 (6)1.25 (6)1.20 (4)1.18 (5)1.21 (5)212.7 + 212.70.098 (13)0.097 (5)	204.14	0.23 (3)				0.200 (17)	0.200 (17)
204.14 + 204.98 0.39 (5) 0.336 (16) 0.336 (16)206.08 0.25 (3) 0.25 (3) 0.268 (13) 0.268 (13) 0.268 (13)210.62 1.25 (14) 1.21 (6) 1.25 (6) 1.20 (13) 0.268 (13)212.7 + 212.7 0.098 (13) 0.097 (5) 0.097 (5) 0.097 (5)218.9 0.101 (14) 0.103 (5) 0.097 (15) 0.097 (15) 0.097 (15)235.96 12.9 (11) 12.8 (6) 13.3 (6) 12.8 (4) 3.1 (5) 0.097 (15)246.12 0.012 (2) 0.021 (2) 0.021 (2) 0.021 (2) 0.117 (2)252.5 0.111 (18) 0.093 (5) 0.140 (8) 0.140 (8) 0.177 (7)254.63 0.71 (14) 0.71 (4) 0.129 (15) 0.129 (15) 0.126 (6)256.23 7.0 (6) 6.8 (4) 7.6 (4) 6.88 (18) 0.129 (15) 0.126 (6)272.91 0.51 (4) 0.044 (2) 1.26 (6) 0.024 (2) 7.6 (3) 0.174 (8) 0.174 (8) 0.038 (18)279.8 0.054 (14) 0.046 (2) 1.87 (7) 0.174 (8) 0.0338 (18) 0.174 (8) 0.038 (18)284.24 0.040 (13) 0.0338 (18) 1.85 (5) 1.86 (6) 1.87 (7)292.41 0.066 (10) 0.059 (3) 0.457 (12) 0.455 (15) 0.455 (15)299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8)308.4 0.49 (6) 0.49 (20) 1	204.98	0.16 (3)	0.336 (16)			0.151 (17)	0.151 (18)
206.08 $0.25 (3)$ $0.288 (14)$ $0.262(7)$ $0.256 (11)$ $0.268 (13)$ 210.62 $1.25 (14)$ $1.21 (6)$ $1.25 (6)$ $1.20(4)$ $1.18 (5)$ $1.21 (5)$ 212.7 + 212.7 $0.098 (13)$ $0.097 (5)$ $0.097 (5)$ $0.097 (15)$ $0.097 (15)$ 218.9 $0.110 (14)$ $0.013 (5)$ $0.097 (15)$ $0.097 (15)$ $0.010 (16)$ 235.96 $12.9 (11)$ $12.8 (6)$ $13.3 (6)$ $12.8 (4)$ $13.1 (5)$ $13.0 (5)$ 246.12 $0.012 (14)$ $0.021 (2)^c$ $0.140 (8)$ $0.140 (8)$ $0.17 (24)$ 254.63 $0.171 (14)$ $0.71 (4)$ $8.8 (18)$ $7.04 (22)$ $7.0 (3)$ 256.23 $7.0 (6)$ $6.8 (4)$ $7.6 (4)$ $6.88 (18)$ $7.04 (22)$ $7.0 (3)$ 256.24 $0.107 (12)$ $0.124 (6)$ $0.415 (5)$ $0.126 (6)$ 272.91 $0.51 (4)$ $0.534 (25)$ $0.17 (18)$ $0.232 (21)$ 279.8 $0.056 (11)$ $0.338 (18)$ $0.017 (8)$ $0.0338 (18)$ $0.017 (8)$ $0.0338 (18)$ 284.24 $0.040 (13)$ $0.0338 (18)$ $1.86 (5)$ $1.86 (6)$ $1.87 (7)$ 292.5 $0.44 (5)$ $0.436 (21)$ $0.467 (12)$ $0.455 (15)$ $0.454 (17)$ 299.98 + 300.5 $2.22 (20)$ $2.18 (10)$ $2.26 (10)$ $2.26 (6)$ $2.25 (8)$ 304.5 $1.15 (16)$ $1.08 (5)$ $1.01 (5)$ $1.11 (3)$ $1.08 (4)$ $1.00 (4)$ 308.4 $0.49 (6)$ $0.499 (23)$ $0.493 (22)$ $1.03 (18)$ $0.503 ($	204.14 + 204.98	0.39 (5)					0.336 (16)
210.621.25 (14)1.21 (6)1.25(6)1.20(4)1.18 (5)1.21 (5)212.7 + 212.70.098 (13)0.097 (5)0.097 (5)0.097 (5)218.90.101 (14)0.103 (5)0.097 (5)0.097 (5)235.9612.9 (11)12.8 (6)13.3 (6)12.8 (4)13.1 (5)13.0 (5)246.120.0123 (14)0.021 (2)°12.8 (4)13.1 (5)10.01 (2)252.50.111 (18)0.93 (5)0.404 (2)0.117 (24)254.630.71 (14)0.71 (4)8.88 (18)7.04 (2.2)7.0 (3)256.270.107 (12)0.124 (6)0.129 (15)0.126 (6)272.910.51 (4)0.534 (25)0.613 (18)0.523 (21)279.80.55 (14)0.406 (2)0.117 (8)0.124 (8)281.420.404 (13)0.338 (18)0.031 (18)286.091.74 (21)1.90 (9)1.80 (8)1.85 (5)1.86 (6)0.455 (15)292.410.66 (10)0.659 (3)0.455 (15)0.45 (17)295.50.44 (5)0.436 (21)0.467 (12)0.455 (15)0.45 (17)299.98 + 300.52.22 (20)2.18 (10)2.26 (10)2.26 (6)2.25 (8)308.40.07 (3)0.020 (1)1.11 (3)0.86 (1)0.020 (1)308.40.49 (6)0.489 (23)0.493 (22)5.05 (18)0.455 (15)0.42 (1)	206.08	0.25 (3)	0.288 (14)		0.262(7)	0.256 (11)	0.268 (13)
212.7 + 212.70.098 (13)0.097 (5)0.097 (5)218.90.101 (14)0.103 (5)0.097 (15)0.101 (6)235.9612.9 (11)12.8 (6)13.3 (6)12.8 (4)0.097 (15)0.021 (2)246.120.0123 (14)0.021 (2)c0.110 (8)0.117 (24)252.50.111 (18)0.093 (5)0.140 (8)0.117 (24)254.630.71 (14)0.71 (4)6.88 (18)7.04 (22)7.0 (3)266.237.0 (6)6.8 (4)7.6 (4)6.88 (18)7.04 (22)7.0 (3)262.870.107 (12)0.124 (6)0.129 (15)0.123 (12)0.123 (12)27.9 10.51 (14)0.046 (2) \cdot 0.041 (5)0.124 (6)0.127 (8)27.9 80.054 (14)0.046 (2) \cdot 0.041 (5)0.124 (8)0.172 (8)284.420.040 (13)0.038 (18) \cdot 0.017 (8)0.172 (8)0.038 (18)0.172 (8)286.091.74 (21)1.90 (9)1.80 (8)1.85 (5)1.86 (6)1.87 (7)290.50.444 (5)0.436 (21)0.467 (12)0.455 (15)0.454 (17)299.8 $+$ 300.52.22 (20)2.18 (10)2.26 (10)2.26 (6)2.25 (8)304.450.173 (8)0.202 (1)1.11 (3)1.00 (21)0.002 (1)314.850.49 (6)0.499 (23)0.493 (22)1.50 (18)0.405 (18)	210.62	1.25 (14)	1.21 (6)	1.25(6)	1.20(4)	1.18 (5)	1.21 (5)
218.9 0.110 (14) 0.103 (5) 0.097 (15) 0.101 (6) 235.96 12.9 (1) 12.8 (6) 13.3 (6) 12.8 (4) 13.1 (5) 13.0 (5) 246.12 0.012 (314) 0.021 (2° . . 0.140 (8) 0.117 (24) 252.5 0.111 (18) 0.993 (5) . . 0.404 (8) 0.117 (24) 254.63 0.71 (14) 0.71 (4) . <td>212.7 + 212.7</td> <td>0.098 (13)</td> <td>0.097 (5)</td> <td></td> <td></td> <td></td> <td>0.097 (5)</td>	212.7 + 212.7	0.098 (13)	0.097 (5)				0.097 (5)
235.96 12.9 (11) 12.8 (6) 13.3 (6) 12.8 (4) 13.1 (5) 13.0 (5) 246.12 0.0123 (14) 0.021 (2)° . 0.021 (2) 252.5 0.111 (18) 0.093 (5) . 0.83 (3) 0.17 (24) 254.63 0.71 (14) 0.71 (4) . 0.83 (3) 0.77 (7) 256.23 7.0 (6) 6.8 (4) 7.6 (4) 6.88 (18) 7.04 (22) 7.0 (3) 262.87 0.107 (12) 0.124 (6) . 0.129 (15) 0.126 (6) 272.91 0.51 (4) 0.534 (25) . 0.611 (5) 0.174 (8) 0.533 (21) 279.8 0.554 (14) 0.046 (2) . 0.041 (5) 0.044 (3) 281.42 0.404 (13) 0.171 (8) . . 0.172 (8) 284.24 0.404 (13) 0.038 (18) 296.5 0.444 (5) 0.436 (21) 	218.9	0.110 (14)	0.103 (5)			0.097 (15)	0.101 (6)
246.12 0.0123 (14) 0.021 (2) ^e 0.021 (2) 252.5 0.111 (18) 0.093 (5) 0.140 (8) 0.117 (24) 254.63 0.71 (14) 0.71 (4) 0.83 (3) 0.77 (7) 256.23 7.0 (6) 6.8 (4) 7.6 (4) 6.88 (18) 7.04 (22) 7.0 (3) 262.87 0.107 (12) 0.124 (6) 0.129 (15) 0.126 (6) 272.91 0.51 (4) 0.534 (25) 0.041 (5) 0.044 (3) 279.8 0.054 (14) 0.46 (2) 0.174 (8) 0.042 (3) 281.42 0.178 (19) 0.171 (8) 0.172 (8) 0.172 (8) 284.24 0.040 (13) 0.0338 (18) 0.174 (8) 0.172 (8) 286.09 1.74 (21) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 0.467 (12) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 <td>235.96</td> <td>12.9 (11)</td> <td>12.8 (6)</td> <td>13.3(6)</td> <td>12.8(4)</td> <td>13.1 (5)</td> <td>13.0 (5)</td>	235.96	12.9 (11)	12.8 (6)	13.3(6)	12.8(4)	13.1 (5)	13.0 (5)
252.5 0.111 (18) 0.093 (5) 0.140 (8) 0.117 (24) 254.63 0.71 (14) 0.71 (4) 0.83 (3) 0.77 (7) 256.23 7.0 (6) 6.8 (4) 7.6 (4) 6.88 (18) 7.04 (22) 7.0 (3) 262.87 0.107 (12) 0.124 (6) 1.12 (24) 0.129 (15) 0.126 (6) 272.91 0.51 (4) 0.534 (25) 0.513 (18) 0.523 (21) 279.8 0.054 (14) 0.46 (2) 0.174 (8) 0.172 (8) 281.42 0.178 (19) 0.171 (8) 0.172 (8) 0.172 (8) 284.24 0.040 (13) 0.0338 (18) 0.174 (8) 0.187 (7) 292.41 0.066 (10) 0.059 (3) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 1.80 (8) 1.85 (5) 1.86 (6) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.00 (4) 308.4 0.049 (6) 0.489 (23)	246.12	0.0123 (14)	0.021 (2) ^c				0.021 (2)
254.63 0.71 (14) 0.71 (4) 0.83 (3) 0.77 (7) 256.23 7.0 (6) 6.8 (4) 7.6 (4) 6.88 (18) 7.04 (22) 7.0 (3) 262.87 0.107 (12) 0.124 (6) 0.129 (15) 0.126 (6) 272.91 0.51 (4) 0.534 (25) 5.13 (18) 0.523 (21) 279.8 0.054 (14) 0.046 (2) 0.041 (5) 0.041 (5) 281.42 0.178 (19) 0.171 (8) 0.174 (8) 0.172 (8) 286.09 1.74 (21) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 0.467 (12) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.02 (1) 308.4 0.017 (3) 0.202 (1) 0.203 (18) 0.203 (18)	252.5	0.111 (18)	0.093 (5)			0.140 (8)	0.117 (24)
256.23 7.0 (6) 6.8 (4) 7.6 (4) 6.88 (18) 7.0 (2) 7.0 (3) 262.87 0.107 (12) 0.124 (6) 0.129 (15) 0.126 (6) 272.91 0.51 (4) 0.534 (25) 0.513 (18) 0.523 (21) 279.8 0.054 (14) 0.046 (2) 0.041 (5) 0.044 (3) 281.42 0.78 (19) 0.171 (8) 0.174 (8) 0.174 (8) 284.24 0.040 (13) 0.038 (18) 0.038 (18) 0.038 (18) 286.09 1.74 (21) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 0.445 (17) 0.059 (3) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.00 (20 (1) 308.4 0.049 (6) 0.489 (23) 0.493 (22) 503 (18) 0.495 (18)	254.63	0.71 (14)	0.71 (4)			0.83 (3)	0.77 (7)
262.87 0.107 (12) 0.124 (6) 0.129 (15) 0.126 (6) 272.91 0.51 (4) 0.534 (25) 0.513 (18) 0.523 (21) 279.8 0.054 (14) 0.046 (2) 0.041 (5) 0.044 (3) 281.42 0.178 (19) 0.171 (8) 0.174 (8) 0.172 (8) 284.24 0.040 (13) 0.338 (18) 0.046 (10) 0.059 (3) 292.41 0.066 (10) 0.059 (3) 1.88 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 0.454 (17) 296.5 0.44 (5) 0.436 (21) 0.467 (12) 0.455 (15) 0.454 (17) 299.8 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (1) 308.4 0.017 (3) 0.020 (1) 0.503 (18) 0.495 (18) 0.495 (18)	256.23	7.0 (6)	6.8 (4)	7.6 (4)	6.88 (18)	7.04 (22)	7.0 (3)
272.91 0.51 (4) 0.534 (25) 0.513 (18) 0.523 (21) 279.8 0.054 (14) 0.046 (2) 0.041 (5) 0.044 (3) 281.42 0.178 (19) 0.171 (8) 0.172 (8) 0.172 (8) 284.24 0.040 (13) 0.0338 (18) 0.0338 (18) 0.0338 (18) 286.09 1.74 (21) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 0.445 (12) 0.456 (12) 0.455 (15) 0.454 (17) 296.5 0.44 (5) 0.436 (21) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.01 (3) 1.08 (4) 1.10 (4) 304.5 0.017 (3) 0.020 (1) 0.020 (1) 0.020 (1) 0.020 (1) 314.85 0.49 (6) 0.489 (23) 0.493 (22) 50.33 (18) 0.495 (18)	262.87	0.107 (12)	0.124 (6)			0.129 (15)	0.126 (6)
279.8 0.054 (14) 0.046 (2) 0.041 (5) 0.044 (3) 281.42 0.178 (19) 0.171 (8) 0.172 (8) 0.172 (8) 284.24 0.040 (13) 0.0338 (18)	272.91	0.51 (4)	0.534 (25)			0.513 (18)	0.523 (21)
281.42 0.178 (19) 0.171 (8) 0.172 (8) 284.24 0.040 (13) 0.0338 (18) 0.0338 (18) 286.09 1.74 (21) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 0.0467 (12) 0.455 (15) 0.454 (17) 296.5 0.44 (5) 0.436 (21) 0.467 (12) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (15) 0.454 (17) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 0.017 (3) 0.020 (1) 0.503 (18) 0.495 (18) 314.85 0.49 (6) 0.489 (23) 0.493 (22) 5.05 (18) 0.495 (18)	279.8	0.054 (14)	0.046 (2)			0.041 (5)	0.044 (3)
284.24 0.040 (13) 0.0338 (18) 0.0338 (18) 286.09 1.74 (21) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) 0.059 (3) 0.059 (3) 0.059 (3) 296.5 0.44 (5) 0.436 (21) 0.467 (12) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 0.017 (3) 0.020 (1) 0.493 (22) 0.503 (18) 0.495 (18)	281.42	0.178 (19)	0.171 (8)			0.174 (8)	0.172 (8)
286.09 1.74 (21) 1.90 (9) 1.80 (8) 1.85 (5) 1.86 (6) 1.87 (7) 292.41 0.066 (10) 0.059 (3) . . 0.059 (3) 296.5 0.44 (5) 0.436 (21) 0.467 (12) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 0.479 (6) 0.489 (23) 0.493 (22) 5.03 (18) 0.495 (18)	284.24	0.040 (13)	0.0338 (18)				0.0338 (18)
292.41 0.066 (10) 0.059 (3) 0.059 (3) 296.5 0.44 (5) 0.436 (21) 0.467 (12) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 0.017 (3) 0.020 (1) 0.020 (1) 314.85 0.49 (6) 0.489 (23) 0.493 (22) 0.503 (18) 0.495 (18)	286.09	1.74 (21)	1.90 (9)	1.80 (8)	1.85 (5)	1.86 (6)	1.87 (7)
296.5 0.44 (5) 0.436 (21) 0.467 (12) 0.455 (15) 0.454 (17) 299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 0.017 (3) 0.020 (1) 0.493 (22) 0.503 (18) 0.495 (18)	292.41	0.066 (10)	0.059 (3)				0.059 (3)
299.98 + 300.5 2.22 (20) 2.18 (10) 2.26 (10) 2.26 (6) 2.25 (8) 304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 0.017 (3) 0.020 (1) 0.020 (1) 0.020 (1) 0.020 (18) 0.495 (18)	296.5	0.44 (5)	0.436 (21)		0.467 (12)	0.455 (15)	0.454 (17)
304.5 1.15 (16) 1.08 (5) 1.10 (5) 1.11 (3) 1.08 (4) 1.10 (4) 308.4 0.017 (3) 0.020 (1) 0.020 (1) 0.020 (1) 0.020 (1) 314.85 0.49 (6) 0.489 (23) 0.493 (22) 0.503 (18) 0.495 (18)	299.98 + 300.5	2.22 (20)	2.18 (10)	2.26 (10)	2.26 (6)		2.25 (8)
308.4 0.017 (3) 0.020 (1) 0.020 (1) 314.85 0.49 (6) 0.489 (23) 0.493 (22) 0.503 (18) 0.495 (18)	304.5	1.15 (16)	1.08 (5)	1.10 (5)	1.11 (3)	1.08 (4)	1.10 (4)
314.85 0.49 (6) 0.489 (23) 0.493 (22) 0.503 (18) 0.495 (18)	308.4	0.017 (3)	0.020 (1)				0.020 (1)
	314.85	0.49 (6)	0.489 (23)	0.493 (22)		0.503 (18)	0.495 (18)
346.45 0.0120 (16) 0.0096 (11) 0.0096 (11)	346.45	0.0120 (16)	0.0096 (11)				0.0096 (11)
466.8 0.00049 (5) 0.0014 (4) 0.0014 (4)	466.8	0.00049 (5)	0.0014 (4)				0.0014 (4)

^a Negligible interferences from the ²²⁷Ac 60.6 keV γ ray with P_{γ} = 0.000041 % and the ²²³Fr 61.43 keV γ ray with P_{γ} = 0.000049 %.

 $^{b}\,$ Interference from the ^{227}Th 112.6 keV γ ray with P_{γ} = 0.009 % has been subtracted.

 $^{\rm c}\,$ Interference from the $^{227}\text{Th}\,245.6\,\text{keV}\,\gamma$ ray with $P_{\gamma}\,$ = 0.00027 is considered negligible.

Table 7

Absolute γ -ray emission probabilities (%) measured in the 223 Ra \longrightarrow 219 Rn decay.

Energy (keV)	DDEP	JRC	ENEA	CEA	СМІ	This work
122.319	1.238 (19)	1.37 (7)	1.39 (6)		1.42 (6)	1.39 (6)
144.27	3.36 (8)	3.67 (17)			3.76 (16)	3.71 (15)
154.208	5.84 (13)	$6.4(3)^{d}$	6.40 (28) ^d		6.52 (21) ^d	6.43 (23)
158.635	0.713 (16)	0.78 (4) ^e	0.80 (4) ^e		0.80 (3) ^e	0.795 (30)
165.8	0.0054 (28)	0.0037 (15)				0.0037 (15)
177.3	0.047 (4)	0.055 (3)				0.055 (3)
179.54	0.154 (14)	0.186 (9)	0.168 (9)	0.155 (5)	0.133 (15)	0.161 (12)
221.32	0.036 (6)	0.030 (2)				0.0304 (19)
269.46	14.23 (32)	13.7 (7)	14.2 (6)		14.4 (5)	14.1 (5)
288.18	0.161 (5)	0.126 (6)				0.126 (6)
323.87	4.06 (9)	3.74 (18)			3.96 (13)	3.85 (17)
338.282	2.85 (6)	2.66 (13)	2.78 (12)		2.84 (9)	2.76 (10)
355.5	0.0043 (14)	0.0085 (7)				0.0085 (7)
371.68	0.499 (11)	0.445 (21)			0.468 (17)	0.457 (18)
445.03	1.28 (4)	$1.25~(6)^{f}$	$1.28(6)^{f}$		1.29 (4) ^f	1.28 (5)
487.5	0.011 (2)	0.0064 (6)				0.0064 (6)
527.61	0.073 (4)	0.068 (3)			0.075 (15)	0.070 (4)
537.6	0.0021 (7)	0.0033 (5) ^g				0.0033 (5)
541.99	0.0014 (6)	0.0012 (5)				0.0012 (5)
598.721	0.092 (4)	0.087 (4)			0.083 (18)	0.086 (4)
623.68	0.009 (4)	0.007 (2) ^h				0.007 (2)
711.3	0.0037 (1)	0.0030 (4)				0.0030 (4)

 d Interference from the ^{223}Fr 155.5 keV γ ray with P_{γ} = 0.000038% is considered negligible. e Interference from the ^{227}Ac 159.2 keV γ ray with P_{γ} = 0.00055% is considered negligible.

^f Interference from the ²²³Fr 444.5 keV γ ray with $P_{\gamma} = 0.000015\%$ is considered negligible.

 g Interference from the ^{223}Fr 537.2 keV γ ray with P_{γ} = 0.00007% is present.

^h Interference from the ²²⁷Th 623.8 keV γ ray with P_{γ} = 0.00016% is considered negligible.

Table 8

Absolute γ -ray emission probabilities (%) measured in the ²¹⁹Rn \rightarrow ²¹⁵Po decay.

Energy (keV)	DDEP	JRC	ENEA	CEA	СМІ	This work
271.228	11.07 (22)	11.1 (6)	11.4 (5)		11.5 (4)	11.3 (4)
293.56	0.075 (3)	0.065 (3)				0.065 (3)
401.81	6.75 (22)	6.8 (4)	6.98 (30)		7.14 (23)	6.98 (26)
517.6	0.043 (3)	0.046 (3) ⁱ		$0.046(2)^{i}$	$0.047 (14)^{i}$	0.046 (2)
676.66	0.018 (2)	0.0208 (12)				0.0208 (12)
891.1	0.0009 (2)	0.0005 (2) ^j				0.0005 (2)

ⁱ Interference from the ²²⁷Th 516.6 keV γ ray with P_{γ} = 0.00028% is considered negligible.

^j Interference from the ²²⁷Th 891 keV γ ray with P_{γ} = 0.000019% is considered negligible.

Table 9

Absolute γ-emission	probabilities ((%) measured	in the	²¹⁵ Po—	\rightarrow^{211} Pb c	lecay.

Energy (keV)	DDEP	JRC	ENEA	CEA	СМІ	This work
438.9	0.058 (19)	0.0544 (21) ^k		$0.0565 (16)^{k}$	0.073 (13) ^k	0.058 (4)

^k Interferences from the ²²⁷Ac 439.6 keV γ ray with P $_{\gamma}$ = 0.000034% and the ²²³Fr 439.6 keV with P γ = 0.0000042% are considered negligible.

2.3. Source covering

The ²²⁷Ac activity of each source can be calculated from the total alpha activity assuming that the ²²⁷Ac and its decay products nuclides are in equilibrium during measurement (denoted as $^{227}\mathrm{Ac}_\mathrm{eq}\text{)}.$ It was apparent from the first measurements that the initial covering with $2 \times 35 \,\mu\text{g/cm}^2$ VYNS foils did not suffice to contain all recoil atoms and the radon gas, thus invalidating the equilibrium assumption. The background count rate increased a hundred times after the measurement but decreased and levelled off after approximately five hours. The Absolute γ -ray emission probabilities (%) measured in the ²¹¹Pb \rightarrow ²¹¹Bi decay.

Energy (keV)	DDEP	JRC	ENEA	CEA	СМІ	This work
404.83	3.83 (6)	4.07 (19) ¹	4.22 (18) ¹		4.31 (14) ¹	4.20 (15)
427.15	1.81 (4)	1.91 (9)	1.99 (9)		1.99 (8)	1.96 (7)
429.65	0.008 (3)	0.004 (1)				0.004 (1)
504.07	0.0059 (8)	0.0021 (4)				0.0021 (4)
704.675	0.47 (1)	0.510 (24) ^m	0.527 (24) ^m		0.530 (28) ^m	0.522 (19)
766.68	0.62 (4)				0.781 (26) ⁿ	0.78 (4)
831.984	3.50 (5)	3.64 (17)	3.75 (16)		3.76 (12)	3.72 (14)
865.92	0.0046 (2)	0.0060 (4)				0.0060 (4)
1014.38	0.0173 (5)	0.012 (5)°				0.012 (5)
1080.64	0.0121 (5)	0.0130 (7)				0.0130 (7)
1103.52	0.0047 (7)	0.0036 (3)				0.0036 (3)
1109.509	0.116 (3)	0.119 (6)	0.129 (7)		0.16 (6)	0.125 (8)
1196.33	0.0103 (4)	0.0101 (6)				0.0101 (6)
1234.3	0.0009 (3)	0.00064 (22)				0.00064 (22)
1270.75	0.0068 (12)	0.0066 (4)		0.0071 (3)		0.0069 (3)

 1 Interferences from the ^{219}Rn 405.4 keV γ ray with P_{γ} = 0.00025% is considered negligible.

 m Interferences from the $^{227} Th$ 704.3 keV γ ray with P_{γ} = 0.00008% is considered negligible.

ⁿ Interferences from the ²²⁷Th 439.6 keV γ ray with $P_{\gamma} = 0.0003\%$ and the ²²³Fr 766.64 keV with $P_{\gamma} = 0.00031\%$ are considered negligible.

^o Interference from the ²²⁷Th 1015.2 keV γ ray with P_{γ} = 0.0000148% is considered negligible.

Table 11

Absolute γ -ray emission probabilities (%) measured in the ²¹¹Bi \rightarrow ²⁰⁷Tl decay.

Energy (keV)	DDEP	JRC	ENEA	CEA	CMI	This work
351.03	13.00 (19)	13.5 (5) ^p	14.0 (7) ^p		14.1 (5) ^p	13.9 (5)

 $^p~$ Interference from the ^{227}Th (350.54 + 352.61) keV γ rays with P_{γ} = 0.12% have been subtracted mathematically.

Table 12

Absolute $\gamma\text{-ray}$ emission probabilities (%) measured in the $^{207}\text{Tl}\longrightarrow{}^{207}\text{Pb}$ decay.

Energy (keV)	DDEP	JRC	ENEA	CEA	CMI	This work
897.77	0.263 (9)	0.27 (1) ^q	0.28 (2) ^q			0.28 (2)

 $^q\,$ Interferences from the ^{223}Fr (896.7 keV) and ^{211}Po (897.8 keV) γ rays with P_γ = 0.016% have been subtracted mathematically.

increase of the background count rate was mainly caused by the ²¹¹Bi ($T_{\frac{1}{2}} = 2.14 \text{ min}$) fed by the ²¹¹Pb ($T_{\frac{1}{2}} = 36.1 \text{ min}$) (Fig. 3). The sources were later covered with additional VYNS foils, attaching them to the

source disc using acetone vapours, but escape of recoil atoms and Rn was still evident.

Several foil covering strategies were tested to trap the ²¹⁹Rn and the recoil atoms in the source, until eventually a practical and easy method was developed. The sources were covered gradually with $35 \,\mu g/cm^2$ VYNS foils, each applied on the source using a cyanoacrylate glue, one foil at a time, until the background count rate remained constant after the source measurement. Each foil was afterwards heated gently with a hot air blower to attach it completely onto the glass disc. In most of the cases, three foils were adequate to retain the ²¹⁹Rn and the recoil atoms. This method proved generally effective, but in few cases the VYNS leaked after a few hours, meaning that the seal was broken. This test verifies the short-term validity of the method only, since radiation damage to the VYNS is expected to make it friable on the long run.

Diffusion of ²¹⁹Rn through the VYNS foils was considered insignificant. The diffusion coefficient of Rn in VYNS is less than 10^{-11} m²/s. In conjuction with its short half-life, the diffusion length is less than 8 µm (Khan, 2015; Keller and Hoffmann, 2000; Pressyanov et al., 2011). A total uncertainty of 0.15 % on the geometrical efficiency (see Table 2) buffers safely any errors due to diffusion. The thickness of VYNS foils were of the order of 60 µm each.

Table 13

Typical relative standard uncertainties (in %, k = 1) for the emission probabilities of the ²²⁷Th \rightarrow ²²³Ra 235.96 keV and 286.09 keV γ rays, as estimated by the individual laboratories.

		235.96 keV				286.09 keV		
Component (%) [*]	JRC	ENEA	CMI	CEA	JRC	ENEA	CMI	CEA
Counting statistics	0.04	0.2	0.1	0.04	0.12	0.54	0.8	0.12
Efficiency	3.6	3	3	1.5	3.6	3	3	1.5
Coincidence summing	3	3	2	0.1	3	3	2	0.2
Source activity	0.55	0.6	0.8	0.9	0.55	0.6	0.8	0.9
Dead Time/pile-up	0.12	< 0.1	0.1	< 0.1	0.12	< 0.1	0.1	< 0.1
Total	4.7	4.3	3.7	2.6	4.7	4.3	3.2	2.6

* Decay correction during measurement is negligible.



Fig. 6. The PMM values (solid lines) of the measured ²²⁷Th absolute emission probabilities for γ rays measured by at least three out of the four laboratories. The standard uncertainties (red dotted lines) of the mean values (blue dashed lines) include a systematic 1 % uncertainty on the CCFs. The evaluated P_{γ} values from the NDS database (upright black triangles) are included in the graphs for easy comparison. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

2.4. Sources for γ -ray spectrometry

After execution of the activity standardisation measurements, the sources were covered more robustly to facilitate the handling for γ -ray spectrometry measurements and to further prevent material loss from the source. Discs were cut out of polyethylene film of 0.1 *mm* thickness at the same diameter as the source substrates. They were attached to the top face of the sources, using the same technique as for VYNS foils; they were first glued by applying cyanoacrylate glue at the periphery of the disc and then heated with a hot air blower.

3. Activity standardisation

3.1. Alpha counting at defined solid angle

The primary standardisation (Pommé, 2007) of the sources' activities was carried out using a defined solid angle (DSA) alpha counting setup (Pommé and Sibbens, 2008; Pommé, 2015). The detector used was a passivated implanted planar silicon PIPS^{*} detector with a 900 mm² active area. The sources were placed at 5 cm distance from the detector. Two baffles (thin metal plates with 29 mm and 64 mm diameter central cut-out) were placed inside the counting chamber to minimise detection of alpha-particles scattered from the inner walls of the counter chamber. The sources were mostly measured at a 0.7 % geometrical efficiency (= solid angle/4 π). The activity distribution was obtained by digital autoradiography and the source to the detector distance by an optical focusing distance measuring device. Such low efficiency was chosen to minimise the chance for coincident signals from successive decays, in particular to avoid alpha decays being obscured by the system dead time due to decays from a short-lived successor in the decay chain.

The energy window selected for DSA counting was such as to include all the alpha decays of 227 Ac and its daughters, from 2.5 MeV up to 10 MeV. The 227 Ac is considered to be in secular equilibrium with its daughters. The counts below the threshold are accounted for by extrapolation of the low-energy spectrum towards zero energy (Fig. 4).



Fig. 7. Comparison of absolute γ -ray emission probabilities from this work with recommended data from the DDEP and the NDS databases using the ζ -score. The circled data points indicate emission probabilities for which formerly no uncertainties were assigned.

The standardised activities of the sources used in the project to determine the γ -ray emission probabilities and their associated combined standard uncertainties are summarized in Table 1. A typical uncertainty budget is presented in Table 2.

Drying the sources in open air instead of using a hot plate had as an outcome the formation of comparably large crystals and an increased self-absorption of the alpha particles. Consequently, the number of events was increased by a factor of 1.006–1.013, depending on the source quality, to compensate for the sub-threshold events due to self-absorption. This correction is the main contributor to the total uncertainty of the activity of the sources (see extrapolation uncertainty in Table 2). Additional uncertainty was introduced for non-equilibrium of ²²⁷Ac with its decay products, to account for some recoil atoms escaping from the sources before the adequate covering was applied and for hypothetical losses of Rn which may not show up in the background measurements,.

3.2. Independent check

The activity of one of the sources (Ac227G1509) was checked by γ -ray spectrometry with high-purity germanium (HPGe) detectors, one above ground and one underground in the Hades facility (Andreotti et al., 2011). The measurements lasted a month each. Emission



Fig. 8. Comparison of absolute γ -ray emission probabilities from this work with those measured by Pibida (2015), Kossert et al. (2015) and Collins et al. (2015b) using the ζ -score.

probabilities from the DDEP and NDS databases were used for the calculation of the activities of the ²²⁷Ac decay products ²²³Ra, ²¹⁹Rn, ²¹¹Pb and ²¹¹Bi and the selected peaks are identified in the ²²⁷Ac γ -ray spectrum of Fig. 5. Activities determined from both γ -ray spectrometry measurements agreed within one standard uncertainty with results from the alpha-counting measurement (Table 3). This can be used only as a rough check, due to the 5 % relative uncertainty on the measurements of γ -ray spectrometry.

3.3. Impurities check

Via the production of ^{227}Ac from the irradiation of ^{226}Ra , minor traces of ^{226}Ra could be present in the obtained solution. One of the produced sources was measured using γ -ray spectrometry with a HPGe detector to identify possible traces of ^{226}Ra . Due to the vast number of γ -ray lines in the spectrum, the 242 keV γ -ray line following the decay of ^{214}Pb was selected to quantify the amount of ^{226}Ra . The activity of ^{226}Ra in the produced sources was less than 0.03 % of their total activity.

4. γ-ray spectrometry

4.1. Method

The participating laboratories received one ²²⁷Ac_{eq} source each. All



Fig. 9. Absolute γ -ray emission probabilities from this work and those measured by Pibida (2015), Kossert et al. (2015) and Collins et al. (2015b) for the 223 Ra \rightarrow ²¹⁹Rn decay.

sources were measured with HPGe detectors calibrated for the full energy peak (FEP) detection efficiency $\varepsilon(E_{\gamma})$ as a function of the γ -ray energy. Most of the laboratories calculated a transfer efficiency correction factor by Monte Carlo simulations, thus adjusting the measured $\varepsilon(E_{\gamma})$ curve to account for the differences in geometry and matrix between the calibration sources and the ²²⁷Ac_{eq} sources. True coincidence summing correction factors (CCFs) were calculated by all participants. Due to the low branching ratio of ²²⁷Ac \rightarrow ²²³Fr, γ rays emitted in the decays of ²²³Fr \rightarrow ²¹⁹At \rightarrow ²¹⁵Bi \rightarrow ²¹⁵Po were neglected from the data analysis. Details on the experimental methods are presented in Table 4. Typical uncertainties associated with the γ -ray spectrometry technique have been discussed by Lépy et al. (2015).

4.2. Results

Gamma rays reported in the DDEP database for ²²⁷Ac, ²²³Ra, ²¹⁹Rn, ²¹⁵Po, ²¹¹Pb, ²¹¹Bi and ²⁰⁷Tl, and those included in the decay data evaluation published in NDS for ²²⁷Th (Browne, 2011) were considered in the analysis of the spectra.

An overview of the reported γ -ray emission probabilities from this work is presented in Tables 5–12 and a typical uncertainty budget in

Table 13. For γ -ray lines on which more than one laboratory reported a result, the γ -ray emission probabilities were combined into a final value by calculating the power-moderated mean or PMM (Pommé and Keighley 2015). Emission probabilities for peaks suffering from spectral interferences not resolved by the fitting software were not reported, unless explicitly stated in the table. For the rest of the y-ray lines interferences are considered negligible. As seen in Table 13, each laboratory estimated the uncertainty on the CCFs in a different way. To avoid that differences in the uncertainty assessments would having a large impact on the relative weighting factors, this CCF uncertainty component was initially not taken into account for the calculation of the PMM. Thus, a more stable mean value is obtained and this systematic error was taken into account by adding a 3 % uncertainty in quadrature to the PMM uncertainty. The results are generally intermediate between arithmetic and weighted mean, depending on the level of mutual consistency. A coverage factor of k = 2 was selected to identify extreme data and a default value was used for the power of the weighting factors ($\alpha = 2-3/N$). Fig. 6 shows the PMM for the ²²⁷Th γ -ray lines for which at least three out of the four laboratories reported an emission probability.



Fig. 10. Absolute γ-ray emission probabilities from this work and those measured by Pibida (2015), Kossert et al. (2015) and Collins et al. (2015b) for the ²¹⁹Rn down to ²⁰⁷Pb decay.

5. Discussion

When excluding the reported sum peaks from the evaluation, 64 out of the 84 measured P_{γ} values reported in this work (Tables 5–13) agree with the evaluated data from the DDEP and NDS. In addition, uncertainties on the emission probabilities for 45 γ rays have been reduced. To compare the P_{γ} from this work with the evaluated data, the ζ score is calculated:

$$\zeta = \frac{P_y^m - P_y^{ev}}{\sqrt{u_m^2 + u_{ev}^2}}$$

The index *m* refers to values measured in this work and *ev* to evaluated data. P_{γ} and *u* are the γ -emission probabilities and their standard uncertainties. Assuming a normal distribution, a value of $|\zeta| < 2$ corresponds to a 95 % confidence interval.

A graphical representation of the above criterion is shown in Fig. 7. It is evident that most measured values are in agreement with the evaluated data, however exceptions do exist. For example, the P_{γ} for the ²²⁷Th \rightarrow ²²³Ra 40.2 keV γ ray from NDS (0.015(4) %) and this work (0.066(5) %) do not match ($\zeta = 11$). It should be noted though that the NDS value is extracted from only two studies held in the sixties lacking uncertainty assignment (Briançon et al., 1968; Davidson, Connor, 1970). Disagreement exists as well for the ²²⁷Th \rightarrow ²²³Ra 113.11 keV

 γ ray (ζ = 3). Although there is a discrepancy with the value from (Briançon and Walen, 1971), there is good agreement with the more recent measurement by Abdul-Hadi et al. (1993).

A comparison has also been made with the most recently measured γ -ray emission probabilities in the decay of $^{223}\text{Ra} \rightarrow ^{219}\text{Rn}$, $^{219}\text{Rn} \rightarrow ^{215}\text{Po}$, $^{211}\text{Pb} \rightarrow ^{211}\text{Bi}$, and $^{211}\text{Bi} \rightarrow ^{207}\text{Tl}$ by Pibida (2015), Kossert et al. (2015) and Collins et al. (2015b). These are not included yet in the evaluations by DDEP or NDS. Applying the $|\zeta| < 2$ criterion, the absolute γ -ray emission probabilities are in most of the cases in agreement with the values published in this work (Fig. 8). A direct comparison of absolute emission probabilities is shown for various γ -ray energies in Fig. 9 and Fig. 10. The estimated uncertainties in this work are significantly higher than in the three recent works, where uncertainties of 1 % or less are quoted.

It is evident that the absolute emission probabilities reported in this work are consistently higher than the most recently published ones. An explanation could be a possible underestimation of the activity standardisation of the sources. The most probable reason could be either the loss of Rn gas or the underestimation of the self-absorption correction.

The exclusion of the first was realised in two ways. Firstly, the background has been checked before and after each activity measurement and it remained constant proving the containment of Rn on the source substrate within the measuring time. Secondly, the experimental



Fig. 11. Relative γ -ray emission probabilities from this work and those measured by Pibida (2015), Kossert et al. (2015) and Collins et al. (2015b) for the 223 Ra \rightarrow 219 Rn decay.

ratio of counts of the ²²⁷Ac-²²⁷Th-²²³Ra to the rest of the decay products, ²¹⁹Rn down to ²⁰⁷Pb, was investigated. If ²¹⁹Rn was escaping, it would be manifested by a decrease in the integral of the measured counts from the decay products ²¹¹Bi, ²¹⁹Rn and ²¹⁵Po compared to theory assuming equilibrium. The alpha spectra obtained for each source were fitted with the deconvolution software 'BEST' (Pommé and Marroyo, 2015). The experimental and theoretical ratios of the integral counts of ²²⁷Ac, ²²⁷Th and ²²³Ra to ²¹¹Bi, ²¹¹Po, ²¹⁹Rn and ²¹⁵Po were calculated and compared. There was no significant difference between the two ratios within a 2 % absolute uncertainty.

On the other hand, the formation of comparably large crystals after drying the sources freely in open air increased the self-absorption of the alpha particles. It affected also the correction for the counts lost below the threshold which is done by extrapolating the spectrum to zero energy (Fig. 4). Because of the bad quality of the alpha sources for primary standardisation one of the two aforementioned corrections might not have been fully accounted. For that reason, and to exclude the activity as a parameter in the determination of the absolute emission probabilities (Fig. 9, Fig. 10), the relative emission probabilities ($P_{\gamma(rel)}$) to the ²²³Ra 269.3 keV γ -ray emission probability for each laboratory are shown in Fig. 11 and Fig. 12. The relative emission probabilities of this work are in a very good agreement with those by Pibida et al., Kossert et al. and Collins et al.. This is clearly depicted by comparing Fig. 8 with its corresponding graph of ζ -scores for the relative emission probabilities (Fig. 13).

6. Conclusions

The γ -ray emission probabilities of ²²⁷Ac and its decay products were measured by γ -ray spectrometry with HPGe detectors. Emission probabilities from 87 γ -ray lines are reported in this work; the majority agree within uncertainties with the DDEP/NDS recommended values and with the most recently published values in the literature. The primary standardisation of activity of the measured sources and the completeness of the uncertainty budget provided by the participating laboratories contribute to the SI-traceability of the provided reference decay data. However, there is doubt on the accuracy of the absolute standardisation due to self-absorption problems. Nevertheless, it was clear that the relative γ -ray emission probabilities of this work were in better agreement with the most recent published ones compared to the absolute emission probabilities.



Fig. 12. Relative γ -ray emission probabilities values from this work and those measured by Pibida (2015), Kossert et al. (2015) and Collins et al. (2015b) for the ²¹⁹Rn down to ²⁰⁷Pb decay.



Fig. 13. Comparison of the relative γ -ray emission probabilities values from this work with those measured by Pibida (2015), Kossert et al. (2015) and Collins et al. (2015b) for the ²¹⁹Rn down to ²⁰⁷Pb decay using the ζ -score.

Acknowledgments

This work has been supported by the European Metrology Research Programme (EMRP), JRP-Contract IND57 MetroNORM (www. emrponline.eu). The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union.

The authors would like to thank Gerd Marissens, Jan Paepen and Heiko Stroh for their technical support.

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