International Atomic Energy Agency

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INDC INTERNATIONAL NUCLEAR DATA COMMITTEE

Fifth Research Coordination Meeting on the Measurement and Evaluation of Transactinium Isotope Nuclear Data

Central Bureau for Nuclear Measurements
Geel, Belgium, 1-3 September 1982

SUMMARY REPORT

Prepared by A. Lorenz
Nuclear Data Section
International Atomic Energy Agency

December 1982

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SUMMARY REPORT

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Abstract

Proceedings of the fifth meeting of the participants in the IAEA Coordinated Research Programme to measure and evaluate the required nuclear decay data of heavy element radionuclides, convened by the IAEA Nuclear Data Section on 1-3 September 1982 at CBNM, Geel, Belgium.

The meeting participants reviewed the data requirements, updated and extended the recommended list of half-lives, and continued to review the status of alpha and gamma radiation spectra emitted in the decay of transactinium isotopes.

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I. SUMMARY OF THE MEETING

Introduction

The fifth meeting of the participants in the IAEA Coordinated Research Programme (CRP) on the Measurement and Evaluation of Transactinium Isotope Nuclear Decay Data was convened by the IAEA Nuclear Data Section on 1-3 September 1982, at the Bureau Central de Mesures Nucléaires (CBNM), Geel, Belgium. The meeting was chaired by A. Lorenz, IAEA Nuclear Data Section.

The participants in this meeting are listed in Appendix 1.

Objectives

This coordinated research programme aims specifically at improving the quality and accuracy of nuclear decay data required to calculate the effects of transactinium isotopes on the fuel cycle of both thermal and fast reactors, to assess their impact on nuclear waste management, to improve the accuracy of safeguard techniques, and to improve the knowledge of their nuclear characteristics required in many applications in science and industry.

The principal objectives of this meeting were to review the status of measurements performed by the participants in this programme, to review the list of proposed half-lives, and to continue the review of the status and accuracy of gamma-ray emission spectra for the heavy element radionuclides. Another objective of this meeting was to define the format and content of the final report of this project.

The Adopted Agenda is given in Appendix 2.

Conclusions and Results of the Meeting

The meeting reviewed the current and projected programmes for the measurement and evaluation of heavy element radionuclide nuclear decay data of each research group participating in the CRP.

In particular, the meeting

- updated the list of proposed heavy element radionuclide half-lives published in INDC(NDS)-127/NE (December 1981), and agreed on the release of the new update of this list;

- recommended to issue the list of gamma-ray energies and intensities for isotopes used by the CRP as calibration standards in their measurements of heavy element radionuclide nuclear decay data;

- performed a comprehensive review of the measurement and evaluation status of all decay data for the radionuclides identified at the IAEA Meetings on Transactinium Nuclear Data; and

- decided on the format and content of the final report of this project.

Specific Actions which resulted from this meeting are listed in Appendix 3.

The participants of this CRP agreed to have their next meeting in Idaho Falls, Idaho, USA, during the second half of June 1983.

II. MEETING PROGRAMME

1. Progress Reports

1.1. R. Vaninbroukx (CBNM/Geel)

Progress report included as Appendix 6.

1.2. F. Lagoutine (LMRI/France)

Progress report included as Appendix 7.

1.3. C.W. Reich (US/INEL)

Progress report included as Appendix 8.

1.4. H. Umezawa (Japan/JAERI)

Progress report included as Appendix 9.

1.5. A.L. Nichols (UK/AEE Winfrith)

Progress report included as Appendix 10.

1.6. A.J. Fudge (UK/AERE Harwell)

Progress report included as Appendix 11.

2. Cooperation with ICRM

Bambynek (CBNM), who serves as Secretary to the International Committee for Radionuclide Metrology (ICRM), described the function and activities of the ICRM (see Appendix 5). Of direct interest to this CRP is ICRM's effort to evaluate decay data used as calibration standards in nuclear data measurements. The CRP endorsed the ICRM evaluation of calibration standards and Vaninbroukx (CBNM) was asked to inform the CRP on the progress of this ICRM activity (see Action # 5).

3. Review of recent publications

In addition to the progress report, the group took into consideration recently published measurements and evaluations which it considered in its review of the proposed recommended lists of data. The list of these references is given in Appendix 4.

4. Review of Recommended List of Half-Lives

The group reviewed the "Proposed Recommended List of Transactinium Isotope Decay Data. Part I. Half-Lives (December 1981 Edition)" published in INDC(NDS)-127/NE, and made the changes listed below, taking into account measurements and evaluations performed since October 1981. The new "Proposed Recommended List of Heavy Element Radionuclide Decay Data. Part I: Half-lives (December 1982 Edition)" will be issued as INDC(NDS)-139/NE.

Changes in the half-life compilation

4.1. Two new measurements of the ^{238}U spontaneous fission half-life have been reported in the literature:

$(8.16 \pm 0.13) \times 10^{15}$ y	H.G. De Carvalho, et al. Nucl. Instr. and Methods 197(1982)
$(1.05 \pm 0.03) \times 10^{16}$ y	Z.N.R. Baptista, et al. An.Acad.Brasil.Cienc 53(1981)

The group recommended that these results be evaluated before changing the currently accepted value. (See Action # 8).

4.2. A new decay measurement by M. Lindner, et al. (J.inorg.nucl. Chem. 43 12 (1981) 3071-3080) of the ^{236}Np nuclide has yielded the following values:

Total half-life :	(1.55×10^{5})y	($\sim \pm 0.5$ %)	
Electron capture:	(1.29×10^{6})y	($\pm$ 0.5 %)	BF=.88
Beta decay :	(1.76×10^{5})y	($\mp$ 0.2 %)	BF=.12
Alpha decay :	(9.5×10^{7})y	($\mp$ 2.6 %)	BF=.002

The group decided to accept these new values.

4.3. The new half-life value for ^{237}Pu, as evaluated by R. Vaninbroukx (see Appendix 13), of (45.17 ± 0.06)d was accepted by the group.

4.4. A new ^{238}Pu half-life measurement by V.D. Sevastianov and V.P. Yanin in the USSR was reported in the literature (Nuclear Constants 5(44) (1981)). The group recommended that this new measurement, yielding an evaluated value of 86.96 $\pm$ 0.55 years, be evaluated before changing the currently accepted value. (See Action # 9).

4.5. With regard to the recent ^{242}Cm half-life evaluation by R. Vaninbroukx (see Appendix 13), yielding a value of (162.94 ± 0.06)d, the group decided to wait for the results of the ongoing Harwell measurement, so as to resolve the low value

obtained in the recent Japanese measurement (see the Progress Report by H. Umezawa Appendix 9 this report). It was proposed to have this measurement repeated by CBNM and Harwell, using the sample used in the Japanese measurement.

4.6. On the basis of the assessment by H.H. Knitter (CBNM) (See Appendix 6 of this report) of the spontaneous fission half-lives measured by H.R. von Gunten, et al., (Phys.Rev. C23,1110(1981)) (see INDC(NDS)-126/NE, page 4, and Action # 13 from the October 1981 CRP meeting), the spontaneous fission half-life for ^{235}U of $(9.8 \pm 2.8).10^{18}y$ supersedes the earlier value of $(3.5 \pm 0.9).10^{17}y$.

5. Review of the status of the half-lives for which the required accuracies had not yet been achieved

Of the 41 radionuclides, identified at the IAEA TND Meetings in 1975 (Karlsruhe) and 1979 (Cadarache), whose decay constants did not have the accuracy required by the user community, 30 are now considered to satisfy the prescribed requirement (see Table I). The accuracy of the decay constants of the following 11 radionuclides had still not reached the prescribed requirements as of August 1982. (Mode of decay is given in parentheses for alpha, beta and spontaneous fission (SF), the half-life values quoted are taken from the Proposed Recommended List of Heavy Element Radionuclide Decay Data, INDC(NDS)-127/NE).

Th-229 — The currently accepted value of this half-life is $(7.34 \pm 0.16).10^{3}y$ (uncertainty = 2.2 %). The requested 1 % accuracy is difficult to realize, and is questionable for the specified application. Required accuracy to be checked (Action # 11).

U-232 — The currently accepted value of this half-life is (69.8 ± 1.0) years (uncertainty = 1.4 %). The required accuracy is 0.5 %. Measurement is foreseen by AERE/Harwell.

Pu-238 (SF) — The currently accepted value of this half-life is $(4.77 \pm 0.13).10^{10}y$ (uncertainty = 2.7 %). The requested accuracy of 1 % is considered to be justified. A new measurement of this half-life is recommended.

Pu-240 (SF) — The currently accepted value of this half-life is $(1.15 \pm 0.04).10^{11}y$ (uncertainty = 3.5 %). The required accuracy is 2 %. A new measurement of this half-life is recommended.

Pu-241 (β^-) — The currently assigned value of this half-life is (14.4 ± 0.2) years (uncertainty = 1.4 %). The required accuracy is 0.5 %. Work is in progress at Harwell, CBNM and at the NBS in the USA. In addition, the possible effect of chemical bonding on the half-life is to be investigated experimentally at Harwell (see Appendix 12), and theoretically by Bambynek (CBNM) and Behrens (KFK).

Pu-241 (α) The currently accepted value of this half-life is $(6.00 \pm 0.25).10^5$ years (uncertainty = 4.2 %). The required accuracy of 1 % is difficult to realize and is a questionable accuracy requirement. Required accuracy to be checked (Action # 11).

Am-242m The currently accepted value of this half-life is $(3.13 \pm 0.15).10^4$ years (uncertainty = 4.8 %). The required accuracy of 1 % is difficult to achieve. Required accuracy to be checked (Action # 11).

Am-242m (SF) The currently accepted value of this half-life is $(8.8 \pm 3.3).10^{11}$ years (uncertainty = 38 %). The required accuracy is 2 %, although 5 % should satisfy the specified application. A new measurement of this half-life is recommended.

Cm-242 (SF) The currently accepted value of this half-life is $(6.5 \pm 0.6).10^6$ years (uncertainty = 9.2 %). The required accuracy of 3 % is difficult to achieve. Required accuracy to be checked (Action # 11). A new measurement of this half-life is recommended.

Cm-246 The currently accepted value of this half-life is $(4.73 \pm 0.10).10^3$ years (uncertainty = 2.1 %). Questionable accuracy requirement is 1 %. Measurement may be foreseen at Harwell.

Cf-252 (α) The currently accepted value of this half-life is (2.72 ± 0.01) years (uncertainty = 0.4 %). The required accuracy is 0.2 %. It is unlikely that the required accuracy can be achieved. Required accuracy to be checked (Action # 11).

6. Review of the Status of the gamma-ray emission probabilities (E_γ / I_γ) for which the required accuracies have not yet been achieved

Of the 28 radionuclides, identified at the IAEA TND Meetings (Karlsruhe 1975 and Cadarache 1979), whose gamma-ray emission probabilities (E_γ / I_γ) had not been measured to the required accuracies, 18 are planned to be evaluated by the CRP using the most recent absolute measurements. The remaining 10, because of the lack of accurate data and/or the practical impossibility to achieve the requested accuracy, will not be evaluated. The Summary status of the (E_γ / I_γ) data for the priority nuclides is given in Table II.

7. Review of the Status of the Alpha-decay data (E_α / I_α) for which the required accuracies have not yet been achieved

Of the 16 radionuclides, identified at the IAEA TND Meetings (Karlsruhe 1975 and Cadarache 1979), whose alpha emission probabilities (E_α / I_α) had not been measured to the required accuracies, 5 are now considered to be known to the required accuracy. Of the remaining 11, 3 are now in the process of being measured or evaluated; the remaining 8, whose currently accepted uncertainties range from 5 % to 20 %, are very difficult to measure to the required accuracy, and are not planned to be measured by the CRP. The summary status of the (E_α / I_α) data for which the required accuracies have not yet been achieved is given in Table III.

Table I

NUCLIDES FOR WHICH THE ACCURACIES OF THE HALF-LIVES HAVE BEEN SATISFIED

Nuclides	% Accuracy Required	% Accuracy Achieved	Source of Adopted Values
Th-228	1	0.1	ENSDF A-chain Evaluation (Horen, 1976)
Th-230	1	0.4	Measurement (Meadows, et al, 1980)
Th-233	1	0.5	UK Evaluated Decay Data File
Pa-231	1	0.3	ENSDF, A-chain evaluation (Schmorak, 1977)
Pa-233	0.5	0.4	ENSDF, A-chain evaluation (Ellis, 1978)
U -233	0.5	0.13	ENSDF, A-chain evaluation (Ellis, 1978)
U -234	0.3	0.24	1981 Evaluation (N. Holden)
U -235	0.5	0.16	1981 Evaluation (N. Holden)
U -236	0.5	0.13	1981 Evaluation (N. Holden)
U -238	0.5	0.11	1981 Evaluation (N. Holden)
U -238 (SF)	2	1	1978 ENDF/B Evaluation (C.W. Reich)
U -239	1	0.2	ENSDF, A-chain evaluation (Schmorak, 1977)
Np-236	5	0.5	Measurement (Lindner et al, 1981)
Np-236m	5	2	ENSDF, A-chain evaluation (Schmorak, 1977)
Np-237	0.5	0.5	ENSDF, A-chain evaluation (Ellis, 1978)
Np-238	1	0.1	ENSDF, A-chain evaluation (Ellis, 1977)
Np-239	1	0.3	ENSDF, A-chain evaluation (Schmorak, 1977)
Pu-236	1	0.3	ENSDF, A-chain evaluation (Schmorak, 1977)
Pu-238	0.5	0.1	ENSDF, A-chain evaluation (Ellis, 1977)
Pu-239	0.5	0.1	1978 US Half-life Evaluation Committee
Pu-240	0.5	0.3	ENSDF, A-chain evaluation (Schmorak, 1977)
Pu-242	1	0.6	ENSDF, A-chain evaluation (Ellis + Haese, 1977)
Pu-242 (SF)	1	1	1978 ENDF/B Evaluation (C.W. Reich)
Am-241	0.2	0.1	ENSDF, A-chain evaluation (Ellis, 1978)
Am-242	1	0.1	Table of Isotopes (1978)
Cm-243	1	0.7	ENSDF, A-chain evaluation (Ellis, 1981)
Cm-244	1	0.1	ENSDF, A-chain evaluation (Chukreev, et al., 1981)
Cm-244 (SF)	3	0.2	1978 ENDF/B Evaluation (C.W. Reich)
Cm-245	1	1	ENSDF, A-chain evaluation (Ellis, 1981)
Cf-252 (SF)	1	0.5	1978 ENDF/B Evaluation (C.W. Reich)

Table II

NUCLIDES FOR WHICH THE ACCURACIES OF I HAVE NOT BEEN ACHIEVED AS OF AUGUST 1982

Nuclide	% Accuracy Required	% Accuracy Achieved	New CRP Measurements	Evaluation to be performed by	Comments
Th-228*	2	2	CBNM, Harwell, INEL	Vaninbroukx (CBNM)	
Th-229*	2	-	(none)	Reich (INEL)	D
Th-233	2	10-20	Harwell (83-84?)	Reich (INEL)	
Pa-231	2	5-10	Harwell (end 1982)	Nichols (AEE, Winfrith)	
Pa-233	0.5	1	Harwell, INEL, (CBNM, 1983)	Reich (INEL)	A
U -232	2	5-10	INEL (end 1982), Harwell (83)	Fudge (Harwell), Reich (INEL)	
U -233	1	10	INEL, Harwell (1983)	Reich (INEL)	
U -234	2	10	Harwell and JAERI (1983)	Nichols (AEE, Winfrith)	
U -235	1	10	Harwell, CBNM, INEL	Vaninbroukx (CBNM)	
U -236	1	5-10	(none)	(none)	A B D
U -237			(Data from Pu-241 meas.)	Fudge (Harwell)	C
U -238	1	15	Harwell (83?)	(none)	A B
U -239	2	10	(none)	(none)	A B D
Np-237	0.5	10	Harwell (81), CBNM, INEL (83?)	Reich (INEL)	A
Np-238	2	10	(none)	(none)	A B D
Np-239	1	2	(none)	(none)	A B D
Pu-236	2	30	(none)	(none)	A B
Pu-238	1	2- 5	CBNM, INEL, LMRI	Lagoutine (LMRI)	
Pu-239	1	2- 5	Harwell, INEL, LMRI, JAERI/HU[1]	Umezawa (JAERI)	
Pu-240	1	2- 5	INEL, LMRI	Lagoutine (LMRI)+(CBNM)	
Pu-241	1	2- 5	Harwell, INEL (83)	Lagoutine (LMRI)	D
Pu-242	5	10	(none)	(none)	A B D
Am-241	1	2- 3	CBNM (82-83), LMRI	Bambynek (CBNM)	
Cm-242			(none)	Umezawa (JAERI)	C
Cm-243	1	5-10	(none)	(none)	A B D
Cm-244	1	10	(none)	Lagoutine (LMRI)	A B D
Cm-245	2		(none)	(none)	A B D
Cm-246	2		(none)	(none)	A B D

* with daughters in equilibrium
A. Questionable if required accuracy can be achieved
B. Not enough data for adequate evaluation
C. No accuracy requirement for this nuclide
D. No plans for more measurements
1 Measurement performed by JAERI together with Hiroshima University

Table III

NUCLIDES FOR WHICH THE ACCURACIES OF I_α HAVE NOT BEEN ACHIEVED AS OF AUGUST 1982

Nuclide	% Accuracy Required	% Accuracy Achieved	Comment	Status
Pa-231	2	2- 5	D	Half-life measurement performed at Harwell (Glover, et al). Publication includes best I values. No further work planned at Harwell.
U -234	1	4	A	No work in progress, material is of not sufficient purity. Measurement planned at Harwell.
U -235	1	5-10	A D	No work planned anywhere.
U -236	3	5-10	A D	Measurement at Harwell discontinued.
U -238	1	5-20	A D	No work planned anywhere.
Np-237	1	20	A	Could never achieve 1 % accuracy. Bortels (CBNM) plans to do measurement in 1983.
Pu-239	1	1- 2		Half-life measurement at Harwell yielded best I_α values (Fudge to send values to IAEA). Recently measured values by Ahmad (ANL) have <1 % uncertainties. Work planned at LMRI.
Pu-240	1	3	D	Ahmad (ANL) recently measured values have < 1 % uncertainties.
Cm-243	2	2-10	A D	
Cm-245	2	0.5- 5	A D	
Cm-246	2	1- 5	A	

A. Questionable if required accuracy can be achieved
B. Not enough data for adequate evaluation
C. No accuracy requirement for this nuclide
D. No plans for more measurements

Appendix 1

LIST OF PARTICIPANTS

Participants in the Coordinated Research Programme are indicated by an asterisk.

	Bambynek, W. (part-time observer)	Central Bureau for Nuclear Measurements Steenweg naar Retie B-2440 Geel Belgium
*	Fudge, A.J.	Actinide Analysis Group Chemistry Division Building 220 AERE Harwell, Didcot OX11 ORA U.K.
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*	Umezawa, H.	Division of Chemistry JAERI Tokai-Mura, Naka-Gun Ibaraki Ken Japan
*	Vaninbroukx, R.	Central Bureau for Nuclear Measurements Steenweg naar Retie B-2440 Geel Belgium

Appendix 2

Fifth Meeting of the CRP on the

"Measurement and Evaluation of Transactinium

Isotope Nuclear Decay Data"

CBNM, Geel, 1-3 September 1982

Adopted Agenda

1. Introductory items

2. Review of actions from October 1981 meeting

3. Cooperation with the International Committee for Radionuclide Metrology (ICRM)

4. Progress reports and activity forecasts

5. Review of new measurements and update of half-life data compilation

6. Summary of current (E_γ / I_γ) and (E_α / I_α) data status

7. Extension of CRP by 2 years (1983, 1984)

   - remaining and on-going measurements
   - coordination of final meeting with IAEA Standards or TND meeting in 1984

8. Discussion of final report

   - outline and content
   - international acceptance
   - coordination with other efforts

9. Discussion of decay data evaluation guidelines

10. Next meeting

Appendix 3

List of Actions

1.	Lorenz	Investigate who is currently performing compilations of neutron production data (i.e., (α,n), $\bar{\nu}$, etc...)
2.	Vaninbroukx	Inquire with Liskien (n,2n) and Axton ($\bar{\nu}$) about the status of their compilation, and inform Lorenz
3.	Fudge	Inquire about the status of (α,n) measurements, compilations and evaluations at Harwell and inform Lorenz
4.	Vaninbroukx	Distribute to CRP results of the review which is to be performed by Knitter (CBNM) on uranium isotope spontaneous fission half-lives (see 1981 Action # 13)
5.	Vaninbroukx	Inform CRP on any ICRM evaluation of calibration standards
6.	Lagoutine	Send to IAEA LMRI report on the Ra226 evaluation for distribution to the CRP
7.	Reich	Inquire about the status of the Pu240 half-life summary report, and send report when available, to IAEA for distribution to CRP
8.	Vaninbroukx	Review with Knitter (CBNM) the new spontaneous fission data for U238 by Carvalho et al. and by Baptista et al
9.	Reich	Ask Holden (BNL) if he is contemplating the evaluation of plutonium half-life data (including the recent USSR data of Sevastianov and Yarina)
10.	Reich	Inform CRP about the state of publication of the Pu241 half-life measured by the US NBS
11.	Lagoutine	Ask Robin (CAD) to review the required half-life accuracies assigned at the Cadarache meeting (see Table I of this report)
12.	Fudge	Check the availability of the I_γ values which have resulted from the Harwell Pu239 half-life measurements

13. Vaninbroukx	Inquire with Bortels (CBNM) if any measurements are planned on Cm243, Cm245 and Cm246
14. Lorenz	Re-issue the corrected calibration standards listings as a separate report
15. Lorenz	Inquire with the Baranov (Kurchatov) Group the status of their I_{α} data evaluations
16. Fudge	Inquire whether any measurements are foreseen at Harwell on the U232, Pu240 and Cm246 half-lives
17. Nichols	Send IAEA 10 copies of his final heavy element data compilation report, for distribution to the CRP
18. Fudge	Send IAEA recent measurement results of the Pa231 half-life by Glover et al

Appendix 4

Actinide Decay Data References Distributed and Reviewed by the CRP

- R.G. Helmer and C.W. Reich. Emission Probabilities and Energies of the Prominent γ-Ray Transitions from the ^{240}Pu Decay. Int. J. Appl. Radiat. Isot. Vol. 32.pp.829 to 831, 1981.

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- M. Lindner, et al. Lifetime of the long-lived Isomer of ^{236}Np from α -,β- and Electron-capture Decay Measurements. J. Inorg. Nucl. Chem.43 12 (1981) 3071-3080.

- R. Vaninbroukx. Review of the Half-lives of ^{237}Pu and ^{242}Cm. Private communication. August 1982. (Submitted at the meeting).

- R. Vaninbroukx and B. Denecke. Determination of Gamma-ray Emission Probabilities in the Decay of ^{235}U and ^{231}Th. Nucl. Instr. and Methods 193 (1982) 191-196. (Submitted at the meeting).

- P. De Bievre, M. Gallet and R. Werz. A Determination of the ^{241}Pu Half-life on Highly Enriched ^{241}Pu by Isotope Mass Spectroscopy. Private Communication. August 1982. (Submitted at the meeting). To be published.

- F. Lagoutine. Schema de Desintegration du ^{239}Pu. (Preliminary results). Private Communication. September 1982. (Submitted at the meeting).

- A.J. Fudge. Note on the Proposed Investigation of the Effect of the State of Chemical Combination on the Decay Constant for Beta Decay, with Special Reference to the Half-life of ^{241}Pu. August 1982. (Submitted at the meeting).

Appendix 5

International Committee for Radionuclide Metrology

I C R M

An International Committee for Radionuclide Metrology (ICRM) was set up during 1974. The ICRM is an informal association of scientists representing national laboratories engaged in the field of radioactivity standards, and also international organizations such as the Bureau International des Poids et Mesures (BIPM), the International Atomic Energy Agency (IAEA), and the Central Bureau of Nuclear Measurements (CBNM) of the European Communities.

The objects of the ICRM are to promote the advancement of radionuclide metrology, to study measurement problems that are associated with applied radioactivity and nuclear energy, to engage in such other activities as are conducive to the applications of radionuclide metrology and to the dissemination of pertinent knowledge. ICRM endeavours to cooperate with other organizations that are concerned with radionuclide metrology such as the Bureau International des Poids et Mesures.

With these objectives in view, the ICRM currently operates through five working groups concerned with the dissemination of information and the organization of intercomparative measurements in the following fields:

- Non-neutron nuclear data
- α-, β-, γ-ray spectrometry
- low level activity measurement techniques
- radionuclide metrology needs related to nuclear energy, and to
- life sciences.

Scientists who wish to cooperate with the committee (or to discuss their activity-measurement problems) are invited to contact either a member of the Executive Board or the coordinator of the group who is concerned with their field of interest (see below).

The ICRM comprises two classes of membership:

- Regular members, i.e., persons who represent an institute, agency or laboratory that actively pursues the objects of ICRM
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Appendix 6

STATUS REPORT 1982

J.R.C. - C.B.N.M. Participation in the I.A.E.A. Coordinated Research Programme on the Measurement and Evaluation of Transactinium Nuclear Decay Data

R. Vaninbroukx

Central Bureau for Nuclear Measurements, Geel, Belgium

INTRODUCTION

The status of the work carried out at CBNM during the period September 1981 - September 1982 is presented. Reported are the measurements on the half life of ^{241}Pu and the alpha-and photon emission probabilities in the decay of ^{232}U daughters and the decay of ^{238}Pu - ^{234}U, and the evaluations of the half lives of ^{237}Pu and ^{242}Cm. Some comments on recent determinations of the spontaneous fission half life of ^{235}U are given too.

MEASUREMENTS

1. Half life of ^{241}Pu

P. De Bièvre, M. Gallet, R. Werz

The determination of the half life of ^{241}Pu by direct decay measurements using mass-spectrometric methods has continued.
Samples of Pu material (ORNL 210A, Harwell 94/241/10 and "Wilkins") were followed by measurements of the ratios $(^{241}Pu/^{240}Pu)/(^{240}Pu/^{239}Pu)$ or $(^{241}Pu/^{240}Pu)/(^{242}Pu/^{241}Pu)$ over a period of 6 years. During that period about 29 % of the ^{241}Pu atoms decayed. Values for the half life were derived from least-squares fits. The deviation of the data points from the fitted values is less than 0.06 %.

A detailed statistical analysis, based upon iterative weighted-least-squares fits, has been performed of the half-life measurements. Very small but unavoidable contributions of ingrown ^{241}Am to the mass-241 signals were evaluated for their effect on the half-life value. Table 1 shows the results for the three samples measured of the ^{241}Pu half-life determinations. The quoted uncertainties are the standard deviations. Details about the measurements and the statistical analysis are described in the internal CBNM report GE/NI/MS/41/82.

TABLE 1. Results of the ^{241}Pu half-life determinations

Mass spectrometric Determination	Samples	Number of measured points	Half life (a)
$\frac{^{241}Pu/^{240}Pu}{^{240}Pu/^{239}Pu}$	CBNM (ORNL 210A)	9	14.331 ± 0.021
$\frac{^{241}Pu/^{240}Pu}{^{242}Pu/^{241}Pu}$	Harwell ("Wilkins")	3	14.352 ± 0.007
	Harwell (94/241/10)	3	14.330 ± 0.006

2. Decay Parameters of ^{232}U Daughters

2.1. Emission probabilities for the 5.449 MeV α particles and 241 keV γ rays in the ^{224}Ra - ^{220}Rn decay.

G. Bortels, D. Reher, R. Vaninbroukx

The measurements are finished and the results have been accepted for publication in the International Journal of Applied Radiation and Isotopes with following abstract :

" The emission probability $P_{\alpha 1}$ for the 5.449 MeV α particles, which in the decay of ^{224}Ra populate the 241 keV level in ^{220}Rn, was measured using ^{224}Ra sources produced by recoil implantation from ^{228}Th and Si-surface-barrier detectors. Additionally, the 241 keV γ-emission probability, $P_{\gamma(241)}$ was measured using ^{228}Th sources and a calibrated intrinsic Ge detector. A pair of deduced values, $P_{\gamma(241)}$, $P_{\alpha 1}$, was obtained from the measured data of $P_{\alpha 1}$ and $P_{\gamma(241)}$, respectively. The weighted mean of the measured and deduced data yielded the results of $P_{\alpha 1} = 0.0507 \pm 0.0005$ and $P_{\gamma(241)} = 0.0400 \pm 0.0007$. The quoted overall uncertainties of the mean correspond to a 68 % confidence level."

2.2. Determination of γ-ray emission probabilities in the decay of the ^{228}Th daughters.

R. Vaninbroukx, H.H. Hansen

The measurements are finished and the results will be submitted for publication with following abstract :

" Gamma-ray emission probabilities useful for the assessment of ^{232}Th and ^{232}U by γ-ray spectrometry have been determined very accurately. The results were deduced from γ-ray spectra measured with a Ge(Li) and a high purity Ge detector using sources of ^{228}Th, being part of the decay chains of ^{232}Th and ^{232}U."

3. Alpha- and photon emission probabilities in the ^{238}Pu - ^{234}U decay.

G. Bortels, B. Denecke, R. Vaninbroukx

Alpha emission probabilities were determined from several series of measurements using ^{238}Pu sources evaporated in vacuum and Si-surface barrier detectors of 25 and 50 mm^2 active area. The resolution obtained was 12.5 keV FWHM for source-to-detector geometries down to 0.04 % (0.005 sr).
Similar sources were used for the determination of photon-emission probabilities (LX-and γ rays) in the energy range 10 - 160 keV. These measurements were performed with calibrated high purity Ge and Si(Li) detectors. The disintegration rates of the sources were determined by α counting using detectors with well defined low-geometry solid angles.
The results are summarized in Table 2. Quoted are the overall uncertainties of the mean corresponding to a 68.3 % confidence level.

TABLE 2. Alpha- and photon emission probabilities in the decay of ^{238}Pu.

Level in ^{234}U	Corresponding E_α (MeV)	Corresponding E_γ (keV)	Emission probabilities P_α	Emission probabilities P_{photon}
296 keV	5.206	152.7	0.000030 ± 0.000001(*)	$(9.3 \pm 0.1)10^{-6}$
143 keV	5.358	99.9	0.00105 ± 0.00004	$(7.3 \pm 0.1)10^{-5}$
43 keV	5.457	43.5	0.2898 ± 0.0010	$(3.96 \pm 0.10)10^{-4}$
ground state	5.499	—	0.7091 ± 0.0010	—
		U - LX rays		
		$\bar{E}$ = 11.6 keV Lℓ		0.0026 ± 0.0001
		$\bar{E}$ = 13.6 keV Lα		0.0406 ± 0.0006
		$\bar{E}$ = 17.2 keV Lβ		0.0585 ± 0.0009
		$\bar{E}$ = 20.2 keV Lγ		0.0138 ± 0.0002

(*) Value deduced from the γ-ray measurements and taking into account the theoretical conversion coefficient.

EVALUATIONS (COMMENTS)

1. Review of the half lives of ^{237}Pu and ^{242}Cm

R. Vaninbroukx

This review prepared as part of the CBNM contribution to the IAEA Coordinated Research Programme on the Measurement and Evaluation of Transactinium Isotope Nuclear Data evaluates the experimental data and recommends values for the total half lives of ^{237}Pu and ^{242}Cm.

The recommended values and their uncertainties on a 1σ confidence level are :

^{237}Pu : T1/2 = (45.17 ± 0.06)d
^{242}Cm : T1/2 = (162.94 ± 0.06)d

For detailed information see **Appendix 13 of this report.**

2. Comment on the spontaneous fission half life of ^{235}U as given by H.R. von Gunten et al. (1)

H.-H. Knitter

The authors[1] measured the spontaneous fission half lives of ^{233}U, ^{234}U, ^{235}U and ^{236}U using the spinner detector technique. Their results agree for the isotopes ^{233}U, ^{234}U and ^{236}U with most of the values found in the literature. Only in the case of ^{235}U they get a spontaneous fission half life of $(9.8 \pm 2.8).10^{18}$ a, which is a factor 54 and 28 larger than the existing measurements of ref. (2) and (3), respectively.

The advantages of the present measurement of von Gunten et al.[1] compared to those of ref. (2) and (3) are :

- the very high enrichment of the ^{235}U sample of 99.76 %;
- the well known isotopic composition of the sample material;
- advantageous detection technique (4π geometry, 100 % detection efficiency, no pile-up effects for α particles, no absorption corrections).

The work seems to be done with great care, also what concerns the estimation of corrections and background.

Due to these reasons it is advisable to take the value of von Gunten et al. as a recommended value of the spontaneous fission half life of ^{235}U. Since one cannot exclude systematic errors, it should be recommended to verify in a second, independent experiment the spontaneous fission half life of ^{235}U.

References

1. H.R. von Gunten, A. Grütter, H.W. Reist and M. Baggenstos, Phys. Rev. C23, 1110 (1981)

2. E. Segrè, Phys. Rev. 86, 21 (1951)

3. B.M. Aleksandrov, L.S. Krivokhatskii, L.Z. Malkin and K.A. Petrzhak At. Energ. 20, 315 (1965), Sov. J. At. En. 20, 352 (1966)

Appendix 7

MEETING OF THE CRP ON THE MEASUREMENT AND EVALUATION OF

TRANSACTINIUM ISOTOPE NUCLEAR DECAY DATA

1 - 3 September 1982 Antwerp, Belgium

Summary of progress report from LMRI

1) Evaluation of data decay

Evaluation of decay data was performed for ^{239}Pu and ^{226}Ra decay chaim. The results will be published in "Table de Radionucléides du LMRI (édition 1982)".

The half-lives were taken from the list of proposed values by the groupe CRP INDC (NDS)-126/NE (1981)

Comments -

- Because of recent interest in low energie X-rays this work has been extended to provide new data of the X_L-rays.

- In this work the recent paper of Helmer Reich et al (1982) was taken into account.

- Our values of the strong α: groups are rather close to those of Irshad Ahmad*

^{238}Pu, ^{241}Am - work in progress

* preliminary results received four months back

2) Review of recommended list of half-lives of isotopes used as gamma ray standards

The LMRI reviewed the recommended list of half-lives published in INDC (NDS)-126/NE, and made the changes taking into account measurements performed since 1980.

Appendix 8

Report on the Participation of U. S. Laboratories in the Work of the IAEA Coordinated Research Program on the Measurement of Transactinium Isotope Nuclear Decay Data

Geel, September, 1982

C. W. Reich

In this report, we present the current status of the work being carried out in various U. S. laboratories specifically oriented toward the objectives of this IAEA CRP. Reported below are the gamma-ray emission-probability measurements, and related studies, at INEL, the half-life measurements being done by the participants in the U. S. Half-Life Evaluation Committee (ANL, LANL, LLNL, Mound Lab., NBS and Rocky Flats), and the absolute α-intensity measurements at ANL.

I. GAMMA-RAY EMISSION-PROBABILITY MEASUREMENTS FOR SELECTED ISOTOPES OF Pu AND U

(R. G. Helmer, C. W. Reich)

The measurement of precise values for the emission probabilities of the prominent γ rays from $^{238,239,240}Pu$ and $^{233,235}U$ is nearing completion. The results for ^{239}Pu and ^{240}Pu have now been published (see, Refs. [1] and [2], respectively). Preliminary values are now available for the remaining three nuclides. Final results for these nuclides must await the completion of an extensive series of measurements on the variation with time of the efficiencies of the Ge detectors used in these studies and the measurement of the correction to the detector efficiencies due to the finite diameter of the sources.

The ^{235}U measurements were done using a calibration solution obtained from NBS. This solution was calibrated in μmoles/gm of solution with

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[1] R. G. Helmer, C. W. Reich, R. J. Gehrke and J. D. Baker, Int. J. Appl. Radiat. Isotopes 33, 23 (1982).

[2] R. G. Helmer and C. W. Reich, Int. J. Appl. Radiat. Isotopes 32, 829 (1981).

an uncertainty (1σ level) of 0.06%. This sample contained 99.8% atom-percent ^{235}U. The ^{233}U material has been described previously [3]. In addition to its original calibration, this material was also calibrated by measurement of the isotopic abundance ratio by mass spectrometry for samples mixed with the above ^{235}U material.

The ^{238}Pu source material has also been described previously [3].

Preliminary results for these three nuclides are given in Tables I-III. The changes in these values resulting from including the effects of the time dependence of the detector efficiency calibration and the finite diameter of the sources are expected to be relatively small (<1%).

II. Ge DETECTOR EFFICIENCIES

(R. G. Helmer)

In our measurements of the γ-ray emission probabilities for ^{238}Pu, ^{233}U and ^{235}U, two Ge detector spectrometers, known locally as LBL-P and OR-12, have been used. As discussed at the previous meeting of the CRP participants, the efficiency of the LBL-P detector had changed with time. Since that time it has been found that the OR-12 detector efficiency has also changed. Although these changes have generally been less than 1%, they are significant for precise measurements such as these. As a result of the existence of this time dependence, the efficiencies of both detectors have been remeasured. The measurements have been as follows:

detector	LBL-P	OR-12
volume (cm^3)	~8	114
source-detector distance (cm)	10	15
type	Ge, planar	Ge, coaxial
data of original efficiency calibration	March, 1980	April, 1981 (?)
date of recalibration	August, 1981	April, 1982

[3] See the previous report to the CRP, contained in the IAEA Report INDC(NDS)-126/NE, December, 1981, pp. 25-33.

source used to monitor efficiency drift	^{133}Ba	^{152}Eu

The ^{133}Ba and ^{152}Eu sources were measured every few weeks to maintain a monitor on the changes in the efficiency at various energies (53-383 keV for ^{133}Ba and 121-1408 keV for ^{152}Eu). The spectra for the above Pu and U nuclides were measured from July 1981 through January 1982.

The composite of all this information will be used to obtain the best efficiency values at the time of each set of measurements.

III. EFFICIENCY CORRECTIONS FOR DISK SOURCES
(R. G. Helmer)

Our basic γ-ray efficiency calibrations for Ge detectors are done with "point" sources. However, the low specific activity for the Pu and U sources used in the γ-emission probability measurements reported here required the use of disk sources with diameters of 1.0-2.2 cm. Therefore, it is necessary to correct the original efficiency curves for the change in efficiency with source diameter.

In our earlier papers on the γ-emission probabilities, this correction was generated from a "calculated" correction and a measurement at one γ-ray energy. In an effort to improve our knowledge of this correction, we have made a series of measurements of this correction. These measurements were done with a set of calibrated sources obtained from PTB. The set includes a point source and disks of 2, 3, 4 and 5 cm diameters. Each source contains known amounts of ^{54}Mn, ^{57}Co, ^{65}Zn, ^{85}Sr, ^{88}Y, ^{133}Ba, ^{137}Cs and ^{139}Ce. This allowed measurements of the correction from 53 to 1115 keV (the 1836-keV line from ^{88}Y was not used).

These measurements were made at our standard source-detector distances -10 cm on the LBL-P detector and 15 cm on the OR-12 detector. Measurements were also made at 3 cm on LBL-P and 1 and 5 cm on OR-12. Some results of these measurements are shown in Table IV.

II. ACTIVITIES OF THE HALF-LIFE EVALUATION COMMITTEE

All of the experimental papers on the ^{240}Pu half-life measurements from the various participating laboratories have been submitted for publication in Journal of Applied Radiation and Isotopes. Walter Strohm, the chairman of this committee, said that a recommended value for the ^{240}Pu half-life is not available yet. The committee's recommendation will be included in a summary paper, which is presently being written. He indicated that this would be available within approximately a month.

The committee does not have plans for any further half-life measurements at this time.

III. MEASUREMENT OF RELATIVE ALPHA INTENSITIES

(Irshad Ahmad, Chemistry Division, ANL)

No further α-intensity measurements have been carried out since the last meeting of the CRP (in October, 1981). Isotopes thus far measured are ^{233}U and $^{238,239,240}Pu$. These measured α-intensity values were submitted earlier this year to A. Lorenz so that other interested investigators could be informed of them.

Table I

Preliminary values of energies and emission probabilities for the prominent γ-ray transitions from ^{238}Pu.

γ-ray energy (keV)	γ-ray emission probability (γ/10^5 dis)	
	present values	values from previous report [3]
43.498±0.001	38.6 ±0.8	38.0 ±0.8
99.854±0.003	7.48 ±0.07	7.35 ±0.08
152.720±0.002	0.944±0.012	0.931±0.009

Table II

Preliminary values for the energies and emission probabilities of selected γ-ray transitions from ^{235}U.

γ-ray energy (keV)	γ-emission probability (γ/10^2dis)
25.509±0.009	12.
84.221±0.001	6.6
143.768±0.001	10.9
163.357±0.003	5.1
185.722±0.004	56.
205.318±0.004	4.9

Table III

Preliminary γ-ray energies and emission probabilities for the decay of ^{233}U.

γ-ray energy (keV)	γ-emission probability ($\gamma/10^5$ dis)
29.19	12.0
42.47	87.
54.70	18.1
118.96	4.07
120.82	3.34
135.	2.33
146.34	6.59
164.52	6.26
208.17	2.30
245.34	3.64
291.35	5.39
317.16	7.79
320.54	2.91

Table IV

Corrections of a point source efficiency for finite source diameter.

Detector	Source-detector distance (cm)	γ-ray energy (keV)	Source diameter (cm)	Correction (%)
LBL-P	10	165-1115	2	0.7
			3	1.4
			5	3.1
		81-122	2	1.1
			3	2.0
			5	4.3
		53	2	~1.7
			3	~2.9
			5	~6.
OR-12	15	165-1115	2	0.15
			4	0.4
		81-122	2	0.5
			4	1.1

Appendix 9

PROGRESS REPORT TO THE INTERNATIONAL ATOMIC ENERGY AGENCY ON THE MEASUREMENT OF NUCLEAR DECAY DATA OF CURIUM-242 UNDER THE RESEARCH AGREEMENT NO. 2170/R3/CF

September 1982

H. Umezawa
Japan Atomic Energy Research Institute

During the period of the Research Agreement No. 2170/R3/CF between the Agency and our Institute, measurements of the spontaneous fission half-life of ^{242}Cm have been carried out.

Four new samples of ^{242}Cm were prepared for the measurements. Methods of preparation of the samples and examination of those for impurities are the same to that reported previously[1].

On each of the samples prepared, spontaneous fissions were counted by means of solid state track detectors. In order to determine the ratio of the spontaneous fission rate to the alpha decay rate, each sample was measured by alpha counters for which a windowless 2π-proportional counter and a silicon surface barrier detector were used, before and after the fission track counting that took several tens days. For calibration of the fission tracks to alpha counts ratio, sources of ^{242}Pu were prepared as the same physical demention to the samples of ^{242}Cm and measured under the same conditions. The principles and procedures involved in the measurements have also been described in our previous reports[1,2].

Table 1 shows results of the ratios of alpha counting rate to fission tracks counting rate and spontaneous fission half-lives obtained from the results referring to the half-life data of ^{242}Pu: 3.763×10^{6}yr. for alpha decay and 6.75×10^{10}yr. for spontaneous fission decay. Considering uncertainties associated with the reference half-life data of ^{242}Pu and that associated with the half-life of ^{242}Cm for that we took 161.35±0.1(1σ)[3], our best value of the spontaneous fission half-life of ^{242}Cm is $(6.89\pm0.17)\times10^{6}$yr. In Table 2, the result is compared with others reported. Our measurements on some other samples are still continued so that elaboration of the result is expected.

References:

[1] H. Umezawa, Progress Report to IAEA on the Research Agreement No. 2170/CF(1979).

[2] H. Umezawa, Progress Report to IAEA on the Research Agreement No. 2170/R1/CF(1980).

[3] H. Umezawa, Progress Report to IAEA on the Research Agreement No. 2170/R2/CF(1981),
S. Usuda, H. Umezawa, J. Inorg. Nucl. Chem., 43[12], 3081(1981).

Table 1.

Spontaneous Fission Half-life of ^{242}Cm obtained from the Present Measurements

Cm-242 Source	$[\alpha(2\pi)]_{cm}/[\text{Track}]_{cm}$	$T_{1/2}$(S.F.)	$[\alpha(\text{Si})]_{cm}/[\text{Track}]_{cm}$	$T_{1/2}$(S.F.)
No.6(0.491μCi)	8.091×10^{6}	6.847×10^{6}y	6.743×10^{4}	6.807×10^{6}y
8(0.420μCi)	8.266	6.996	6.948	7.013
9(0.827μCi)	7.937	6.717	6.661	6.724
10(0.351μCi)	8.269	6.998	6.959	7.024
Mean		6.890×10^{6}y		6.892×10^{6}y
Standard Deviation		±0.135		±0.150

Table 2.

Comparison of Data Reported for the Spontaneous Fissin Half-life of ^{242}Cm

Reference	$T_{1/2}$(S.F.)	Method
G.C.Hanna, 1951	$(7.2 \pm 0.2) \times 10^{6}$ y	Fission Chamber
R.J.Armani & R.Gold, 1966	(6.09±0.18)	Measurment of S.F. Neutron
C.Huan-qiao et.al., 1979	(7.46±0.06)	Mica Detector
Present work	(6.89±0.17)	Mica Detector

Appendix 10

PROGRESS REPORT FOR THE IAEA CO-ORDINATED RESEARCH PROGRAMME ON THE MEASUREMENT AND EVALUATION OF TRANSACTINIUM ISOTOPE NUCLEAR DECAY DATA (AUGUST 1982)

A.L. NICHOLS, M.F. JAMES
AEE Winfrith
Dorchester
Dorset, UK

Effort has concentrated upon the production of a Winfrith report (AEEW - R 1407) describing the contents of UKHEDD-1, a library of evaluated heavy element decay data for the UK Chemical Nuclear Data Committee (UKCNDC). The development and form of this library have been described in some detail at previous CRP meetings[1]. These computer based data files have been produced in ENDF/B-V format, listing the data references used to formulate the proposed decay schemes and identifying their inadequacies. Evaluated data include half-life, total decay energies, branching ratios, alpha, beta and gamma energies and intensities, spontaneous fission decay data, average energies, internal conversion coefficients and associated uncertainties. Extensive tabulations of the data will be presented in the report by means of a variety of computer codes developed at the Brookhaven National Laboratory Centre[2] and the CEGB Berkeley Nuclear Laboratories[3]. A final draft of the report has undergone peer review and is in the process of declassification at AEE Winfrith.

Our future evaluation efforts will concentrate on the development of an activation products decay data library for reactor applications. This work constitutes an updating of a UKCNDC data library (UKPADD-1) and will include the evaluation of decay data for approximately 410 nuclides. Enquiries concerning all UKCNDC libraries should be addressed to:

Mr B.S.J. Davies,
CEGB Berkeley Nuclear Laboratories,
Berkeley, Gloucestershire, UK

References

(1) A.L. Nichols, M.F. James in INDC(NDS)-126/NE, Editor: A. Lorenz.

(2) CINDU-11, March 1976, IAEA Vienna: the listing program used is a modified version of LISTFC described on page 93.

(3) A. Tobias, CEGB Report RD/B/5170N81, 1981.

Appendix 11

Transactinium Nuclear Data Evaluation and Measurement (TANDEM) Project Progress Report

A.J. Fudge

Chemistry Division, A.E.R.E., Harwell, Oxfordshire. OX11 ORA

1. Status of Programme

Work on the project has progressed with increased interest during this year. Some new measurements have been initiated, but much of the available effort has been spent on applications of the data and this in turn has identified new or more exacting requirements of previously measured data. Progress then will be reviewed under the headings of measurements, applications, and investigations.

2. Measurements

2.1 231-Protactinium M.F. Banham, R. Jones, D. Brown and B. Whittaker

Weighable quantities of $PaCl_5$ were prepared and purified by vacuum sublimation. This material was dissolved in concentrated nitric acid and its purity and mass checked by alpha spectrometry. Small quantities of daughter decay nuclides were found and allowed for in subsequent measurements. Measurements of the γ-rays and x-rays emitted from liquid and thin solid sources have been measured on calibrated Ge(Li), H.P.Ge, and Si(Li) detectors. 90 emissions have identified and measured over the range 10 to 600 KeV. Absolute intensities have been assigned to all of these emissions and comparisons made with other measurements and evaluations. The emissions from daughter nuclides have been measured by repeated measurements after known time intervals. The results will be published shortly when the decay scheme has been re-evaluated with A. Nichols of A.E.E. Winfrith.

2.2 235-Uranium

As a result of the marked differences noted by Vaninbroukx and Denecke for some of the principle gamma emissions from 235-uranium, measurements were made to check these and investigate all the emissions from this nuclide. Very high purity uranium metal enriched to 93.1% in the 235 isotope was used initially for this work. This was dissolved in 10M nitric acid and measured on a Ge(Li) detector against liquid standards in similar geometry. A resolution of 670 eV F.W.H.M. was obtained for energies around 180 KeV. The preliminary data on the major emission probabilities quoted by Vaninbroukx et al.

are shown in table 1.

Table 1. Comparison of γ-Emission Probabilities for 235-Uranium

Energy in KeV	A.E.R.E. Values	C.B.N.M. Values
84.1	0.068	0.066
140.7	0.002	-
143.7	0.103	0.109
145.8	0.002	-
163.3	0.049	0.050
164.1	0.001	-
182.6	0.004	-
185.7	0.561	0.575
186.6	0.009	-
202.2	0.011	-
205.4	0.050	0.050

It can be seen from these values that we confirm and agree very closely with the C.B.N.M. results if the smaller peaks are included as indicated. These results are preliminary and further work is in progress.

3. Applications

Use has been made during this period of γ-emission probability data for the non-destructive assay of a number of transactinium nuclides and hence, elements, in the presence of one another. These measurements would normally require extensive chemical separations and complex chemical measurements. Using gamma emissions of very closely related energies it is possible to obtain these abundances and hence ratios of masses and concentrations fairly simply if nuclear decay data of γ-emission probabilities are avaiable. The uncertainty of the measurement will then depend to a large extent on the uncertainties of these probabilities. This has high-lighted the need for improvements in the uncertainty associated with abundances of some of the abundant emissions.

3.1 Americium in Plutonium

The method developed at present uses the following γ photo-peaks

^{241}Am

125.29 KeV 122.99 KeV

^{239}Pu

129.29 KeV 125.21 KeV

124.51 KeV 123.62 KeV

Present estimates indicate that the abundances of these photopeaks are known to ±5%. It is desirable that they be known to ±2%. Other photopeaks,

some of which are used in the non-destructive measurements of isotopic composition of plutonium, are being examined. These are at 146.56, 169.56, 208.0, 332.35, 335.41 and 376.60 KeV.

3.2 Neptunium in Plutonium

The most convenient photopeak for the assay of neptunium is at 312 KeV and is due to the ^{233}Pa daughter, so measurements can only be made when this nuclide is in equilibrium with its parent. The 316.4 and 311.7 KeV peaks for ^{239}Pu are the corresponding peaks used for the ratio measurements.

3.3 232-Uranium in Uranium

Again a photopeak from the ^{228}Th daughter is used as a measure of the ^{232}U content. So it has to be assumed that it is equilibrium with its parent nuclide. Peaks at 238.6 KeV and 241.0 KeV are measured in relation to the 240.9 KeV peak for ^{235}U (the isotopic composition of the uranium must be known from mass spectrometry measurements).

4. 241-Plutonium Half-life

Progress in this problem area has been limited to the initiation of a study of the possible influence of chemical composition on the half-life. This work is reported separately. Assessments of previous mass spectrometric measurements have not lead to any significant changes to previously reported data. It is proposed to carry out further measurements in the near future on existing isotopic mixtures by mass spectrometry.

Appendix 12

Note on the Proposed Investigation of the Effect of the State of Chemical Combination on the Decay Constant for Beta Decay, With Special Reference to the Half-life of ^{241}Pu

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It has been pointed out, several years ago, that β transitions occurring with the emission of very low energy electrons, might take place by two processes:

(i) Normal β decay, releasing an electron to the continuum with the expected spectrum of energies dependent on the partition of energy between the electron and the anti-neutrino.

(ii) Decay leading to the orbitally bound state of the electron in the new atom. For this, it is assumed that the anti-neutrino carries off practically all of the available energy.

The latter process, if constituting an appreciable proportion of the decay events, could be expected to be affected by the state of chemical combination of the parent species. This will be influenced particularly by the oxidation state, since this affects the wave function of the newly occupied orbital. Consequently, two decay process producing found electrons will only be important if the energy released in the transition, and in particular the energy of the emitted electrons, is small. Direct detection of such decay would be difficult. Differences in the measurement of the half-life of such a nuclide might be expected between measurements made of the specific activity and measurements made by measurement of the amount of the daughter product arising from a known period of decay of a known weight of parent. The specific activity measurement would be expected to indicate a shorter half-life than other methods. Further, the discrepancy should vary with the chemical state of the material used while decay is taking place in the care of daughter growth measurement.

This predicted effect has been suggested to account for the differing values obtained for the half-life of ^{241}Pu and also to increase the uncertainty on the half-life of tritium, but without any experimental evidence. Experimental evidence seems to have been found for two such modes of decay from one nucleus in the case of ^{187}Re. Here the bound electron mode of

decay may amount to 30% or even more of the total decay and this could well be the reason for the great variability of half-life quoted for this nuclide.

In the particular case of ^{241}Pu which proceeds via β^- decay to ^{241}Am (99.998%) with $E_{\beta max}$ of 20.8 KeV, it has been calculated that the fraction of decay by emission of the electron into $7S_{\frac{1}{2}}$ orbital of the daughter could be as high as 50%. Clearly, if the fraction of decay by this mode is as large as this, the chemical valency state of the plutonium will show the effect by a variation in half-life with composition very clearly and could account for the observed differences in $t_{\frac{1}{2}}$ measurements for ^{241}Pu.

To investigate the magnitude of this effect compounds of differing valency states prepared from the same nuclide or isotopic mixture must be prepared and measured. It is important to be able to characterise the material produced for its stoichiometry at the beginning of the measurements and when the final measurements have been made. Changes in valency state may occur due to radiation effects during the decay period being measured. These must be avoided if the influence of valency on the half-life is to be proven beyond reasonable doubt by this technique. Plutonium compounds are thermally stable in three valency state +3, +4, and +6. Compounds in these states, together with plutonium in the metallic form, are to be prepared and characterised prior to measurements of the half-life being made. The choice of the compounds used will be made when preliminary work on radiation stability has been completed.

Because of the radiological hazards involved with the use of plutonium compounds for this investigation, a complementary investigation using ^{210}Pb has been initiated. This nuclide has very similar decay characteristics to ^{241}Pu and since lead also exhibits two well defined valency stages, as well as a metallic state, it should be possible to study this possible effect using this nuclide. Two other nuclides, ^{227}Ac and ^{228}Ra, which have low energy β^- branches have been suggested as possible nuclides for study because they also have associated γ-ray emissions which can be measured. Both these elements have only one common valency state so this and the metallic state would have to be used in this study.

Appendix 13

REVIEW OF THE HALF LIVES OF ^{237}Pu AND ^{242}Cm

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Abstract

This review prepared as part of the CBNM contribution to the IAEA Coordinated Research Programme on the Measurement and Evaluation of Transactinium Isotope Nuclear Data evaluates the experimental data and recommends values for the total half lives of ^{237}Pu and ^{242}Cm.

The recommended values and their uncertainties on a 1σ confidence level are :

^{237}Pu : T1/2 = (45.17 ± 0.06)d
^{242}Cm : T1/2 = (162.94 ± 0.06)d

1. Introduction

In this review the measurements of the half lives of ^{237}Pu and ^{242}Cm are described summarily and assessed. Results and some details are given in the Tables 1 and 2 for ^{237}Pu and ^{242}Cm, respectively. If for one publication various individual significant results are available, e.g. by measuring samples of different origin, or by applying independent methods or by observing different radiations, they are given too. All uncertainties are listed here at the 1σ confidence level (68.3%). They were determined from the author's quoted uncertainties. In some cases, when only random uncertainties were quoted, estimated systematic uncertainties have been added. Random and systematic uncertainties are always combined linearly, which may slightly change, in a very few cases, the overall uncetainties quoted by the authors . From the listed individual results weighted mean and recommended values have been deduced, by taking the reciprocals of the squared overall uncertainties as weights. The consistency of the whole set of data, individual values with their weights, is checked by comparing, according to Topping [1], the external and internal standard errors.

2. Half life of ^{237}Pu

Thomas et al. [2] followed the decay of ^{237}Pu by counting the Auger electrons

accompanying the electron capture decay using a windowless proportional counter. These measurements yield only an approximative value and are only given here for information.

Hoffman [3] measured the decay of two ^{237}Pu samples from different origins, over respectively 207 and 146 days, by periodic photon counting with a NaI(Tl) scintillation detector. Both samples give half-life values with a small standard deviation of about 0.1%. However, they deviate from each other by about 0.6%. No information is given on the energy range of the observed photons. Therefore, it is difficult to estimate the possible influence of other Pu isotopes, especially ^{236}Pu and ^{238}Pu, which may be present, on the photon count rates. Such influences could explain the deviation between both results and also the higher half-life value obtained for the longest period of observation. The overall uncertainty of 0.2d is probably underestimated. To the quoted random uncertainties we added an estimated systematic uncertainty of 0.27d, being the difference between the results obtained for the two samples.

Smith et al. [4] determined the half life by periodic counting of the Np KX rays with a NaI(Tl) detector. Two samples were measured over a period of about one year. They yield individual results with standard deviations of about 0.2%. Again the two results differ by about 0.8%. Furthermore, it should be noted that for both samples the half-life values calculated from the data measured during the first half year are about 4% lower than those obtained for the second half year.Therefore, it can be concluded that the overall uncertainty of 0.2 d is underestimated. To the quoted random uncertainties we added 0.35d, being the difference between the two results.

Baba et al.[5] determined the half life by periodic measurement of the photon spectrum in the energy range 25 to 120 keV over a period of about 600 days. Due to the high resolution of solid state detectors, as used for these measurements, the contribution by other Pu isotopes can be well estimated. Furthermore, peaks which were not affected by ^{236}Pu and ^{238}Pu activities were selected for the determinations. From this and also from the fact that any change in the detection efficiency was observed always with a ^{241}Am reference source, it may be deduced that the systematic uncertainty is certainly not higher then 0.04 d. This estimated systematic uncertainty was added to the quoted random uncertainties.

The ^{237}Pu samples used in Ref.2 to 4 were produced by the reaction $^{235}U(\alpha,2n)^{237}Pu$ on enriched ^{235}U. The sample used in Ref.5 was produced by the reaction $^{238}U(^{3}He,4n)^{237}Pu$.

Conclusions : The individual results are given in Table 1. The weighted mean value is 45.17d. The external standard error $[\Sigma(x_i - \bar{x})^2 . w_i/(n-1)\Sigma w_i]^{-1/2}$ is 0.055 d, and the internal standard error $(1/\Sigma w_i)^{-1/2}$ is 0.059 d. Here, x_i and w_i are, as usual, the individual results and their weights. The ratio of the external to the internal standard error is 0.93, from which we may conclude that the individual values with their attributed weights are consistent, since for 10 values this ratio should be 1.00 ± 0.24 .[1] The recommended value is (45.17 ± 0.06)d.

3. Half life of ^{242}Cm

All ^{242}Cm samples used in the various experiments were produced by

$$^{241}Am(n,\gamma)^{242}Am \xrightarrow[83\%]{\beta^-} {}^{242}Cm$$

In all cases, except for Hanna et al. [6], Cm/Am separations by ion exchange were performed. The correction for ^{238}Pu growth was always applied, as well as corrections, if necessary, for radionuclidic impurities especially those for remaining ^{241}Am.

Hanna et al. [6] followed by α-particle spectrometry with a proportional counter the decay of three samples with $^{242}Cm/^{241}Am$ activity ratios at the beginning of the measurements of respectively 2, 119 and 144. The samples were measured respectively 3, 2 and 4 times over periods of up to one year.

Glover and Milsted [7] followed the decay of an exhaustively purified ^{242}Cm sample with a low-geometry proportional counter. During a period of about 7 months, 14 measurements were made, each to a statistical accuracy of ± 0.1%. To the quoted uncertainty, being only statistical, an estimated systematic uncertainty of 0.05 d was added.

Hutchinson and White [8] measured over a period of 12 months the change of the heat output of a 2 μg ^{242}Cm sample using a microcalorimeter. The small size of the sample seriously limited the obtainable accuracy.

Flynn et al. [9] used a 2π proportional counter and the measurements were carried out for about 3 years. Part of the uncertainty arises from the need for estimating the amount of ^{241}Am not removed in the Cm/Am separation. The data given here are not those in the cited reference, but have been derived by A.H. Jaffey by re-estimation from the original data (see Ref. 11).

Kerrigan and Banick [10] followed over a period of 287 days the change in heat output of two 5 mg ^{242}Cm samples with a microcalorimeter. Corrections for the contribution to the heat output by other nuclides varied between 1.1% initially and 4.6% at the end of the period of observation. These corrections were deduced from mass-spectrometric isotope analysis and from γ and β counting. The uncertainty quoted by the authors is on the 2σ confidence level and has been reduced here to the 1σ level.

In the framework of a larger programme on the determination of the ^{238}Pu half life by measuring the ^{238}Pu growth in ^{242}Cm, Diamond et al.[11] followed also the decay of ^{242}Cm by α-counting techniques using an intermediate-geometry proportional counter. The ^{242}Cm half-life determination rests on only three points in each of five samples taken over a 262 day interval. The extraordinary self-consistency of the data, due to the good stability of the detection efficiency and the high purity of the samples allowed, in spite of the few measurements, the estimation of the half life with a good accuracy. The quoted random uncertainty is $\pm$ 0.014 d and the sum of the systematic uncertainties is $\pm$ 0.065 d. Combining both uncertainties linearly an overall uncertainty of $\pm$ 0.08 d is obtained, which is a factor of two higher than the overall uncertainty quoted by the authors, who combined all individual uncertainties quadratically.

Huan-Qiao et al. [12] followed the decay of a ^{242}Cm sample for 308 days using α spectrometry with a low-geometry proportional counter. The uncertainty quoted takes only into account random effects. From the difference between the results obtained for respectively the first and second half of the observation period we estimated a systematic uncertainty of 0.07 d, yielding an overall uncertainty of $\pm$ 0.18 d.

Jadhav et al. [13] followed the decay of four samples for 237 days using α spectrometry with a solid state detector. The result is rather inaccurate, but much more accurate results were obtained later at the same laboratory (see Ref. 15).

Usuda and Umezawa [14] followed the decay of five samples for 540 days. Two 2π proportional counters were used. For each of the samples about 90, respectively 60, individual points were recorded with the two detectors. The quoted uncertainty is only random but on a 3σ level. The obtained result is significantly smaller than the other results. No obvious explanation could be found for this lower value, which was not confirmed by more recent measurements (Ref. 15 and 16), although the authors of these measurements were aware of the existing low value. In view of the significant deviation of the result from the other ones, and taking into account that no systematic uncertainties were considered, the quoted 3σ uncertainty was maintained and regarded as a kind of overall uncertainty.

Aggarwal et al. [15] determined the ^{242}Cm half life by two rather independent methods : (1) by following the decay of 20 sources with a proportional counter, recording for each of the sources seven points over a period of 250 days ; (2) by measuring the changes in the $^{242}Cm/^{244}Cm$ activity ratios of synthetic mixtures of ^{242}Cm and ^{244}Cm by α-spectrometry using a solid state detector. This method has the advantage that any change in the detection efficiency, either by changes in the intrinsic detector efficiency or in the counting geometry or by changes in the quality of the sources, will not affect the activity-ratio measurement. For each of the nine sources six measurements were carried out over a period of 220 days. The quoted uncertainties are only statistical ones. Adding $\pm$ 0.05 d as an estimate for the systematic uncertainty the overall uncertainties become $\pm$ 0.11 d and $\pm$ 0.26 d for the two methods, respectively.

Wiltshire et al. [16] counted four sources on each of two low geometry counters for a period of about 490 days. The ^{238}Pu content of each source was determined by α spectrometry for all data points. This reduces the uncertainty on the correction for ^{238}Pu growth, since some ^{238}Pu could be lost from the samples by recoil effects.

Conclusions : The individual results are given in Table 2. The weighted mean of all results is 162.90 d with an external standard error of $\pm$ 0.10 d and an internal one of $\pm$ 0.05 d. The ratio of the external to the internal standard error is 2.0, from which we may conclude that the individual values with their attributed weights are not consistent, since for 11 values the ratio should be

1.0 ± 0.2. The non-consistency is only due to the significantly different value from Ref. 14. The weighted mean of the other results is 162.94 d with an external standard error of ± 0.063 d and an internal one of ± 0.054 d,giving a consistent set of data.

The recommended value is (162.94 ± 0.06) d.

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Table 1. ^{237}Pu half-life determinations

Author	Year	Reference	Method	Sample or photon energy	Period of observation (days)	Half life (days)
Thomas et al.	1957	2	electron counting with proportional counter	-	-	(44±2)
Hoffman	1957	3	photon counting with NaI(Tl) detector	I	207	45.81±0.31 (0.04)
				II	146	45.54±0.30 (0.03)
				Average and overall uncertainty estimated by author		45.63±0.20
Smith et al.	1977	4	photon counting with NaI(Tl) detector	I	390	45.50±0.46 (0.11)
				II	330	45.15±0.44 (0.09)
				Average and overall uncertainty estimated by author		45.3 ±0.2
Baba et al.	1981	5	photon counting with solid state detector	26.3 keV γ ray	500	45.16±0.22 (0.18)
				59.5 keV γ ray	580	45.23±0.17 (0.13)
				Np $K\alpha_2$ X ray	580	45.05±0.13 (0.09)
				Np $K\alpha_1$ X ray	580	45.15±0.14 (0.10)
				Np Kβ X ray I	580	45.13±0.14 (0.10)
				Np Kβ X ray II	580	45.06±0.15 (0.11)
				Average and overall uncertainty estimated by author		45.12±0.03
In parentheses the random uncertainties quoted by the authors are given				Weighted mean and recommended value		45.17±0.06

Table 2. ^{242}Cm half-life determinations

Author	Year	Reference	Method	Period of observation (days)	Half life (days)
Hanna et al.	1950	6	α spectrometry with low-geometry proportional counter	300	162.5 ±2. (2.)
Glover and Milsted	1954	7	α counting with low-geometry proportional counter	210	162.46±0.32(0.27)
Hutchinson and White	1954	8	heat output by calorimetry	365	163.0 ±1.8 (1.8)
Flynn et al.	1965	9	α counting with 2π proportional counter	1187	163.1 ±0.4 (0.4)
Kerrigan and Banick	1975	10	heat output by calorimetry	287	163.2 ±0.2 (0.4)
Diamond et al.	1977	11	α counting with intermediate-geometry proportional counter	260	162.76±0.08(0.04)
Huan-Qiao et al.	1979	12	α counting with low-geometry proportional counter	308	163.02±0.18(0.11)
Jadhav et al.	1980	13	α spectrometry with solid-state detector	237	162.13±2.25(2.25)
Usuda and Umezawa	1981	14	α counting with 2π proportional counter	540	161.35±0.30(0.30)
Aggarwal et al.	1982	15	1) α counting with proportional counter	250	163.17±0.11(0.06)
			2) α spectrometry on synthetic ^{242}Cm - ^{244}Cm mixtures with solid-state detector	208	162.82±0.26(0.21)
Wiltshine et al.	1982	16	α counting with low-geometry proportional counter	490	163.03±0.16(0.16)
Weighted mean and recommended value					162.94±0.06

In parentheses the uncertainties quoted by the authors are given.