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Fourth Coordinated Research Meeting on the
Measurement and Evaluation of Transactinium Isotope Nuclear Data

Vienna, 12-13 October 1981

SUMMARY REPORT

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

December 1981

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

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SUMMARY REPORT

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Abstract

Proceedings of the fourth 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 12-13 October 1981, at IAEA Headquarters in Vienna.

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.

I. SUMMARY OF THE MEETING

Introduction

The fourth 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 12-13 October 1981, at IAEA Headquarters, Vienna. The meeting was chaired by A. Lorenz, IAEA Nuclear Data Section.

The participants in this meeting are listed in Appendix 1.

Main Objectives

The principal objectives of this meeting were to review the status of measurements performed by the participants in this programme, to review and extend the list of proposed half-lives, and to continue the review of the status and accuracy of gamma-ray and alpha emission spectra for the heavy element radionuclides.

The Adopted Agenda is given in Appendix 2.

Conclusions and Results of the Meeting

The meeting reviewed the existing and planned programme for the measurement and evaluation of heavy element radionuclide nuclear decay data of each participating research group. In particular, the meeting

- updated the list of proposed heavy element radionuclide half-lives published in INDC(NDS)-121/NE (December 1980), and decided to release the new version of this list;
- agreed to issue a provisional list of proposed recommended list of alpha radiation spectra (E_{α}/I_{α}) emitted in the decay of heavy element radionuclides
- continued the detailed review of the status and accuracies of the gamma radiation spectra (E_{γ}/I_{γ}) emitted in the decay of heavy element radionuclides.

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

The participants of this CRP agreed on the date of their meeting: it was proposed to be on 1, 2 and 3 September 1982 in Antwerp, Belgium directly preceding the scheduled International Conference on Nuclear Data for Science and Technology, 6-10 September 1982, Antwerp, Belgium.

II. MEETING PROGRAMME

1. Progress Reports

1.1. F. Lagoutine (LMRI/Saclay)

Progress report included as Appendix 4.

1.2. R. Vaninbrouckx (CBNM/Geel)

Progress report included as Appendix 5.

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

Progress report included as Appendix 6.

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

Progress report included as Appendix 7.

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

Progress report included as Appendix 8.

1.6. H. Umezawa (Japan/JAERI)

Progress report included as Appendix 9.

1.7. V.M. Kulakov (USSR/Kurchatov Institute)

Translated progress report included as Appendix 10.

2. Review of recent publications

In addition to the progress reports, 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 11.

3. Review of Recommended List of Half-Lives

The group reviewed the "Proposed Recommended List of Transactinium Isotope Decay Data. Part I. Half-Lives (December 1980 Edition)" published in INDC(NDS)-121/NE, and made the changes listed below, taking into account measurements and evaluations performed since June 1980. The new "Proposed Recommended List of Heavy Element Radionuclide Decay Data. Part I: Half-lives (December 1981 Edition)" will be issued as INDC(NDS)-127/NE.

Changes in the half-lives compilation

- 3.1. A new evaluation of the total half-lives of the uranium isotopes by N.E. Holden (see Appendix 11) was accepted by the group; this resulted in the adoption of the following new values:

232U	$T_{1/2} = (69.8 \pm 1.0)$ years
233U	$T_{1/2} = (1.592 \pm 0.002) \cdot 10^5$ years
234U	$T_{1/2} = (2.454 \pm 0.006) \cdot 10^5$ years
235U	$T_{1/2} = (7.037 \pm 0.011) \cdot 10^8$ years
236U	$T_{1/2} = (2.342 \pm 0.003) \cdot 10^7$ years
238U	$T_{1/2} = (4.468 \pm 0.005) \cdot 10^9$ years

- 3.2. A new measurement of the ^{237}Pu half-life by H. Baba et al at JAERI (see Appendix 9 and 11), has yielded a new value of (45.12 ± 0.03) days. The group recommended that this half-life be evaluated in the light of this new measurement before introducing a new value into the recommended list. CBNM was asked to perform this evaluation (see Action 11).
- 3.3. A new measurement of the ^{239}Pu half-life by D. Brown, et al., at Harwell (see Appendix 11), has yielded a new value of 24088 ± 51 years. The group felt there was no justification to change the accepted recommended value, as the new value falls within the uncertainty of the recommended value.
- 3.4. New measurements of the half-life of ^{241}Pu have been reported in the open literature during 1980/1981^(1,2). These new data, coupled with earlier work indicate reasonable agreement between mass-spectrometric (unweighted mean value of $(14.34 \pm 0.04)\text{y}$) and ^{241}Am in-growth (unweighted mean value of $(14.5 \pm 0.1)\text{y}$) measurements. In view of these developments, the group's earlier recommended value of $(14.7 \pm 0.4)\text{y}$ requires adjustment. Although a number of problems are still unresolved, this group recommends a provisional change to $(14.4 \pm 0.2)\text{y}$.

Among the remaining problems, one particular measurement with a Pu-240, 241, 242 enriched sample continues to show a marked discrepancy from the above value, i.e. $(14.8 \pm 0.1)^{(3,4)}\text{y}$, and the possible effects of chemical bonding on the half-life⁽⁵⁾ still requires investigation.

References to 3.4.

- (1) S.F. Marsh, R.M. Abernathey, R.J. Beckman, J.E. Rein, Int. J. Appl. Rad. Isotopes 31 629 (1980).
- (2) S.K. Aggarwal, S.N. Acharya, A.R. Parab, H.C. Jain, Phys. Rev. 23 C 1748 (1981)
- (3) E.A.C. Crouch, I.C. McKean, UKNDC(78) P38, p. 97(1978)
- (4) A.L. Nichols, IAEA-TECDOC-232, p. 67 (1980)
- (5) V.N. Tikhonov, F.E. Chukreev, INDC(CCP)-151/NF (1980)

- 3.5. A new measurement of the ^{242}Cm half-life by Usuda et al, at JAERI (see Appendix 9) has yielded a value of (161.35 ± 0.30) days. The group recommended that this half-life be evaluated in the light of this new measurement before introducing it in the recommended list. CBNM was asked to perform this evaluation (see Action # 11).
- 3.6. New measurements of the spontaneous fission half-lives for ^{233}U , ^{234}U , ^{235}U and ^{236}U have been reported by H.R. von Gunten et al, at EIR/Wuerenlingen (see Appendix 11). The group accepted these new measurements subject to an assessment by Knitter (CBNM) (see Action # 13). The new values are:

$$\begin{aligned}^{233}\text{U} \quad (T_{1/2})_{\text{SF}} &= 2.7 \cdot 10^{17} \text{ years} \\^{234}\text{U} \quad (T_{1/2})_{\text{SF}} &= (1.42 \pm 0.08) \cdot 10^{16} \text{ years} \\^{235}\text{U} \quad (T_{1/2})_{\text{SF}} &= (9.8 \pm 2.8) \cdot 10^{18} \text{ years} \\^{236}\text{U} \quad (T_{1/2})_{\text{SF}} &= (2.43 \pm 0.13) \cdot 10^{16} \text{ years}\end{aligned}$$

- 3.7. Special consideration was given to the tabulation of spontaneous fission data evaluated and calculated for the UK heavy element decay data library (UKHEDD-1), which is given in the progress report from UK/AEE Winfrith (see Appendix 6). The tabulated data comprises the prompt energy release (Q_{sf}), the branching fraction, and the average number of prompt neutron ($\bar{\nu}_p$) for 26 heavy element nuclides. Of the branching fractions given in this tabulation, most are identical with the proposed recommended values listed in INDC(NDS)-121/NE, or fall within the quoted uncertainties. The only branching fraction (BF) value which is significantly different is that for ^{240}Pu which was recently reevaluated for the UKHEDD-1 library. The following new value was adopted

$$^{240}\text{Pu} \quad (\text{BF})_{\text{SF}} = (5.7 \pm 0.2) \cdot 10^{-8}$$

yielding the revised spontaneous fission half-life of

$$^{240}\text{Pu} \quad (T_{1/2})_{\text{SF}} = (1.15 \pm 0.04) \cdot 10^{11} \text{y}$$

4. Provisional List of Alpha Spectra

The group agreed to issue a provisional list of alpha radiation spectra (E_{α}/I_{α}) emitted in the decay of heavy radionuclides, based on the values published in the Nuclear Data Sheets, supplemented where warranted by values from the Table of Isotopes (7th Edition). The "Provisional List of Alpha Spectra" will be issued together with the December 1981 edition of the proposed recommended half-lives, in the INDC(NDS)-127/NE report. A proposed recommended list of alpha spectra will replace the provisional list after an evaluation of new and ongoing measurements will have been performed by the group.

5. Review of the E_{γ}/I_{γ} data for heavy element radionuclides

The group continued the review of the E_{γ}/I_{γ} data for the set of heavy element radionuclides identified at the May 1979 TND Meeting at Cadarache, and decided to issue a provisional list of these data based on the current information published in the Nuclear Data Sheets.

6. Review of recommended values of decay data for radionuclides used as calibration standards

The group reviewed the tabulation of recommended half-lives and gamma-ray energies and intensities of radionuclides used as calibration standards. This latest revision will be submitted for inclusion in the 1982 INDC/NEANDC Standards File.

7. Conclusion of CRP in 1982 and consideration of follow-up activities

In conclusion of the first phase of this international effort, at the time of the termination of this CRP at the end of 1982, IAEA/NDS plans to publish the decay data of the heavy element radionuclides, which have been reviewed by this group over the past five years, in the form of an IAEA report. This publication will reflect the "state of the art", and will contain the "best values" of half-lives, alpha spectra and gamma spectra which have been reviewed and recommended by the participation in this CRP. In view of the measurements of heavy element radioisotope decay data planned and still in progress, final evaluation of these results will have to be performed after the end of this first phase. It is envisaged that a similar effort, with emphasis on the evaluation of these data, will extend the initial work of this CRP, with the objective of arriving at an internationally accepted evaluated heavy element radionuclide decay data file.

It was the feeling of this group that it would be desirable to incorporate the data recommended by this CRP into existing national and international nuclear data files.

8. Next Meeting

The group agreed to hold the next meeting of this CRP on 1, 2 and 3 September, 1982 in Antwerp, Belgium, directly preceding the scheduled International Conference on Nuclear Data for Science and Technology, 6-10 September, 1982, Antwerp, Belgium.

LIST OF PARTICIPANTS

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

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Appendix 2

Fourth Meeting of the CRP on the

"Measurement and Evaluation of Transactinium Isotope Nuclear Decay Data"

Vienna, Austria, 12-13 October 1981

Agenda

1. Introductory Items
2. Review of Actions from June 1980 meeting
3. Progress reports and activities forecast from CRP members
4. Consideration of the recommended list of half-lives: changes and additions
5. Review of the proposed Tables of E_{α}/I_{α} data
6. Review of the proposed Tables of E_{γ}/I_{γ} data
7. Review of recommended values of decay data for isotopes used as calibration standards
8. Conclusion of CRP in 1982 and consideration of follow-up activities
9. Next meeting

Appendix 3

List of Actions

1. Lorenz Request from US/NNDC for distribution to the CRP group a retrieval from the ENSDF file of the heavy radionuclide decay data (Repeat of 1980 Action # 2)
2. Reich Inform the CRP group of discussion results, regarding systematic entry of horizontal compilations into ENSDF, which resulted from discussions held at the Conference on Nuclear Data Evaluation, 27-30 October 1981, at Asilomar, California
3. Reich Ask Ahmad (ANL) to write NDS regarding the comparison of this Pu-240 I_{α} measurements with those of Baranov (Kurchatov Institute) (Follow-up of 1980 Action # 6)
4. Lorenz Forward information received as a result of Action # 3 to Kulakov (Kurchatov Institute)
5. Lorenz Write S. Pearlstein (NNDC) informing him of IAEA plans to publish results of CRP on actinide decay data at the end of 1982
6. Lorenz Investigate who is currently performing compilations on neutron production data (i.e. (α, n) , $\bar{\nu}$, etc.)
7. Fudge Send to IAEA for distribution to CRP group, report on planned investigation of apparent chemical effects in the half-life measurement of Pu-241
8. Reich Send to IAEA for distribution to CRP group copies of corrected galley proofs on the Pu-239 and Pu-240 E_{γ}/I_{γ} measurements
9. Lorenz Ask actinide data users (through the auspices of INDC) to review the accuracies of the decay values for those nuclides which so far have not been measured to the required accuracies, and attempt to convey this information to the CRP group by March 1982
10. Lorenz Find out from other CRP on actinide cross section evaluation, what accuracies they require for the actinide decay data needed by them
11. Vaninbroukx Request CBNM to evaluate the half-lives of Pu-237 and Cm-242 before next CRP meeting (September 1982)

12. Vaninbroukx Inquire from the US National Bureau of Standards where the Am-241 59-5 KeV gamma-ray intensity value will be published

13. Vaninbroukx Ask Knitter (CBNM) to comment on uranium isotope spontaneous fission half-lives in view of newly published (March 81) measurement by von Gunten, et al

14. Reich Ask US NSDD network if they would consider reviewing/refereeing the results of this CRP, and inform CRP group of results

Appendix 4

Progress Report from LMRI

1) Measurement

Report : Etude du spectre de ^{238}Pu . Mesure des énergies et des intensités absolues des rays γ , Morel et al,. The work is now completed. The report will be available as Note LMRI by the end of 1981.

Report : Etude du spectre de ^{240}Pu . Mesure des énergies et des intensités absolues des rays γ . J. Morel et al,. Detailed analysis of the data is in progress. Will be published as Note LMRI.

2) Evaluation of data decay

- Evaluation of ^{238}Pu , ^{239}Pu , ^{241}Am data decay. Work in progress. The results will be published in "Table de Radionucléides du LMRI" (édition 1982).

- Evaluation of ^{226}Ra decay chain (9 radionuclides) Work in progress. The results will be published in "Table de Radionucléides du LMRI" (édition 1982).

STATUS REPORT

Appendix 5

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

A small group of about 5 people is part time active in the field of the determination of decay data of actinides.

MEASUREMENTS1. Decay Parameters of ^{232}U Daughters

1.1. Alpha-transition probability to the 241 keV level in the decay of ^{224}Ra
G. Bortels, D. Reher, R. Vaninbroukx

Determinations of the α -transition probability to the 241 keV level in the decay of ^{224}Ra have been continued by both α and γ -ray spectrometry. The ^{228}Th sources used were sealed in aluminium containers to prevent possible escape of ^{220}Rn from the source. After establishing secular equilibrium of the decay products (^{224}Ra and daughters) the sources were calibrated against a ^{228}Th solution standardized at The Radiochemical Centre, Amersham. The disintegration rate of the sources was also determined, using a calibrated Ge(Li) detector, by measuring the emission rates of the 583 keV γ ray, the emission probability of which is well known: $P_{\gamma}(583) = (0.3083 \pm 0.0072)^{1,2}$. The γ -ray emission rate of the 241 keV γ transition has been measured with a calibrated intrinsic Ge detector. This yields a preliminary value for

$$P_{\gamma}(241) = 0.0407 \pm 0.0009.$$

-
- (1) M.J. Martin, Nuclear Data Sheets 27, 637 (1979).
(2) W.B. Ewbank, "Transactinium Isotope Nuclear Data-1979"
IAEA-TECDOC-232, IAEA, Vienna (1980) p. 109.

Taking into account the total internal conversion coefficient (0.282 ± 0.007) of this transition from the calculations of Rösler et al. ³⁾ the following preliminary value for the α -transition probability to the 241 keV level can be deduced:

$$P_{\alpha}(241) = 0.0520 \pm 0.0015$$

The quoted uncertainty, corresponding to a 1σ confidence level, takes into account random and systematic influences.

Alpha spectra of ^{224}Ra were measured with a surface barrier detector using sources produced by recoil implantation from a ^{228}Th deposit. These sources were rather weak (about 120 Bq). Therefore a relatively large solid angle of about 5 % of 4π sr was used. A correction of about 1.4 % must be applied to the measured 5.45 MeV α -peak intensity due to simultaneous detection of conversion electrons of the 241 keV transition and 5.45 MeV α particles. Additional measurements with stronger sources of implanted ^{224}Ra (1000 Bq) and a smaller solid angle (1 % of 4π sr) are in progress to reduce the summing correction.

1.2. Gamma-ray emission probabilities in the decay of ^{228}Th and its daughters R. Vaninbrouckx, H.H. Hansen

For the preparation of the ^{228}Th sources special care had to be taken in order to prevent the escape of Rn from the sources; otherwise the equilibrium between all daughter nuclides is not guaranteed. The disintegration rates N_0 of the sources were determined relative to other ^{228}Th sources using a 3" x 3" NaI(Tl) detector. The reference sources were prepared from a standardized ^{228}Th solution obtained from TRC, Amersham. This solution was also standardized at CBNM using a calibrated Ge(Li) detector by observing the count rates under the 583 keV peak, for which the emission probability is rather well known ^{1,2)}. For the determination of the γ -ray emission rates, N_{γ} , two calibrated detectors were used: the Ge(Li) detector mentioned above and a small high purity Ge detector. For the efficiency calibration of the detectors in function of the γ -ray energies, reference sources prepared by deposition of weighed fractions of accurately standardized solutions of suitable radionuclides were used. The γ -ray emission probabilities P_{γ} are obtained by dividing N_{γ} by N_0 . The preliminary results are given in Table 1. The quoted uncertainties, corresponding to a 1σ level, take into account random and systematic errors.

(3) F. Rösler, H.M. Fries, K. Alder, H.C. Pauli, Atomic Data and Nucl. Data Tables 21, 91 (1978).

Table 1. Gamma-ray emission probabilities

Energy E keV	Emission probabilities P_{γ}		
	Ge(Li)	HP-Ge	Mean
238.6	} 0.486 \pm 0.009	0.440 \pm 0.006	0.440 \pm 0.006
241.0		0.0407 \pm 0.0009	0.0407 \pm 0.0009
277.4	0.0224 \pm 0.0006	0.0231 \pm 0.0004	0.0229 \pm 0.0004
300.1	0.0325 \pm 0.0008	0.0323 \pm 0.0006	0.0324 \pm 0.0006
510.8	0.0848 \pm 0.0017	0.0820 \pm 0.0016	0.083 \pm 0.002
583.1	(0.3083 \pm 0.0072)*	0.308 \pm 0.006	0.308 \pm 0.006
727.2	0.0707 \pm 0.0021	-	0.071 \pm 0.002

* Used partly as basis for the determination of the disintegration rates of the sources.

2. Gamma-Ray Emission of ^{235}U and ^{231}Th

R. Vaninbroukx, B. Denecke

The present knowledge of the γ -ray emission probabilities in the decay of ^{235}U and its radioactive daughter ^{231}Th is unsatisfactory. The accuracy required for non-destructive analysis of fuel materials is $\pm 1\%$ while the achieved accuracy is $\pm 10\%$ (4). The emission probabilities for the most prominent γ transitions in the energy range between 25 and 206 keV accompanying the decay of ^{235}U and ^{231}Th , - generally in secular equilibrium with its parent ^{235}U -, were redetermined.

Twenty sources were prepared from three different uranium materials. Some details about the materials used and the definition of the ^{235}U content of the sources are given in Table 2.

For the determination of the ^{235}U disintegration rates of the sources two different methods were applied: mass spectrometric isotope dilution and α counting. The ^{231}Th disintegration rates are equal to the ^{235}U disintegration rates because they are in secular equilibrium.

For the U-Al alloy the number N of ^{235}U atoms for each of the sources was determined by mass spectrometric isotope dilution. For that purpose, the sources, after the measurement of the γ -emission rates, were spiked with a well known amount of ^{233}U and dissolved. The uranium was then separated from

(4) A.L. Nichols, "Transactinium Nuclear Data - 1979", IAEA-TECDOC-232, IAEA, Vienna (1980) p. 67.

Table 2. Materials used and ^{235}U determination

Material	Number of sources	^{235}U content			Activity ratio R	
		atom %	mg per source	method of determination	$\frac{I_a(^{235}\text{U})}{I_a(\text{total})}$	method of determination
U-Al alloy	6	93.22	1.5 - 25	Isotope dilution	-	-
Uranium fluoride	3	99.47	0.1 - 0.6	α counting	0.1690 ± 0.0017	Isotope analysis
Uranyl acetate	11	99.97	0.1 - 1.0	α counting	0.9716 ± 0.0010	α -particle spectrometry

the aluminium and the ratio $^{235}\text{U}/^{233}\text{U}$ was measured. The uncertainty of the ^{235}U determination was estimated as $\pm 0.5\%$. The ^{235}U disintegration rate has been calculated using the recommended half-life value ⁵⁾.

For uranium fluoride and uranyl acetate the ^{235}U disintegration rates were determined by α counting under a well defined low-geometry solid angle.

For the uranium fluoride samples, the corrections for the contribution from other uranium isotopes to the α count rates were calculated from the isotopic composition as determined by mass spectrometry and the decay constants derived from the recommended half-lives. For the uranyl acetate samples these corrections were determined by α -particle spectrometry. The values of these corrections for both materials, expressed as the activity ratio R of the ^{235}U α activity to the total α activity are given in Table 2. The overall uncertainty on the ^{235}U disintegration rates becomes 0.6% for U-Al alloy, $\pm 1.1\%$ for uranium fluoride and $\pm 0.7\%$ for uranyl acetate. All uncertainties correspond to a 1σ confidence level and include random and systematic uncertainties. The γ -ray emission rates of all the sources were determined using three calibrated photon detectors: a Si(Li) detector with an area of 3 cm^2 and a thickness of 3 mm and two high purity Ge detectors, one with an area of 1 cm^2 and a thickness of 8 mm (detector GE-I), and one with an area of 3 cm^2 and a thickness of 10 mm (detector GE-II). The Si(Li) detector was used to measure the γ rays of 25.65 keV and 84.16 keV following the decay of ^{231}Th , while the two Ge detectors were used for the two γ rays mentioned and for those of

(1) A. Lorenz, Ed., INDC(NDS)-121/NE (1980).

143.8, 163.4, 185.7, and 205.3 keV accompanying the decay of ^{235}U . The solid angles of the detector systems were about 0.2 sr for the Si(Li) detector, 0.1 sr for GE-I, and 1 sr for GE-II.

The mean results for the γ -ray emission probabilities, obtained from the measurements on all sources using the three detectors, for the different materials are given in Table 3. The last column of this table gives the weighted mean values; they are our final values.

Table 3. Results of the γ -emission probability determinations

Energy (keV)	Emission probability P_γ			
	U-Al Alloy	Uranium fluoride	Uranium acetate	Final value
25.65	0.1445 \pm 0.0035	0.1444 \pm 0.0030	0.1453 \pm 0.0028	0.145 \pm 0.003
84.16	0.0666 \pm 0.0020	0.0652 \pm 0.0026	0.0626 \pm 0.0034	0.066 \pm 0.003
143.8	0.1090 \pm 0.0021	0.1086 \pm 0.0024	0.1084 \pm 0.0023	0.109 \pm 0.002
163.4	0.0502 \pm 0.0011	0.0488 \pm 0.0015	0.0496 \pm 0.0012	0.050 \pm 0.001
185.7	0.5780 \pm 0.0085	0.5707 \pm 0.0098	0.5755 \pm 0.0088	0.575 \pm 0.009
205.3	0.0494 \pm 0.0015	0.0501 \pm 0.0020	0.0515 \pm 0.0015	0.050 \pm 0.002

Our results for the 185.7 keV ray, the most intense one and often considered as a "reference" γ ray, is 6.5 % higher than the previously adopted value based on a few very old measurements ⁶⁾. The ratios of the emission probabilities of the other γ rays to that of the "reference" ray are in fairly good agreement with recent evaluated values ^{7,8)}.

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- (6) E.K. Hyde, I. Perlman, G.T. Seaborg, "The Nuclear Properties of the Heavy Elements, Vol. II: Detailed Radioactive Properties", Prentice Hall, Inc., Englewood Cliffs, N.J. (1964).
 - (7) M.R. Schmorak, Nucl. Data Sheets 21 (1977) 91.
 - (8) C.M. Lederer, V.S. Shirley, Eds., "Table of Isotopes, 7th edition", John Wiley and Sons, Inc., New York (1978).

HEAVY ELEMENT DECAY DATA:
PROGRESS REPORT FOR THE IAEA CO-ORDINATED RESEARCH PROGRAMME
ON THE MEASUREMENT AND EVALUATION OF TRANSACTINIUM
ISOTOPE NUCLEAR DECAY DATA (OCT 1981)

A L NICHOLS

M F JAMES

Abstract

Spontaneous fission data for 26 specific heavy element nuclides have been evaluated, calculated and inserted into the UK heavy element decay data file (UKHEDD-1). These data include the prompt energy release (Q_{sf}), branching ratio, the average number of prompt neutrons ($\bar{\nu}_p$) and the prompt neutron and gamma spectra. An extensive report is being prepared in which the contents of UKHEDD-1 will be described.

AEE Winfrith
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1 INTRODUCTION

During 1977, work began to produce a computer-based library of heavy element decay data, UKHEDD-1. The format adopted was ENDF/B-V as recommended internationally, permitting the inclusion of spontaneous fission decay data. Progress in the preparation of this data library has been described at previous meetings (1, 2), and UKHEDD-1 is now complete following the addition of spontaneous fission decay data in 1981.

2 SPONTANEOUS FISSION DATA

The spontaneous fission data for 26 heavy element nuclides have been evaluated (Table 1). Much of the required data had to be derived by calculation and extrapolation. The sources of available data and the various calculations are described below.

The prompt energy release from spontaneous fission (Q_{sf}) is defined as:

$$Q_{sf} = Q_{tot} - E_d$$

where Q_{tot} is the total energy released and E_d is the average energy released in the radioactive decay of the fission products. Q_{tot} was estimated by the method described in references (3, 4). If $m(A, Z)$ is the mass excess in MeV of the fissioning nucleus, and y_i and $m(A_i, Z_i)$ are the yield and mass excess of the stable fission product (A_i, Z_i), then

$$Q_{tot} = m(A, Z) - \sum_i y_i m(A_i, Z_i) - \bar{\nu}_p m_n$$

where $\bar{\nu}_p$ is the average number of prompt neutrons emitted per fission, and m_n is the neutron mass excess (8.071431 MeV). However, this equation was only used for 252-Cf, the only spontaneously-fissioning nucleus for which a complete set of yields are available. For other nuclides an interpolative method had to be used (4). The equation for Q_{tot} may be re-written:

$$Q_{tot} = m(A, Z) - \bar{m}_L - \bar{m}_H - \bar{\nu}_p m_n$$

where \bar{m}_L , \bar{m}_H are the mean mass excesses of the light and heavy mass peaks. By using available fission yield sets, the approximate dependences of \bar{m}_L on \bar{A}_L (the mean mass number of the light mass peak) and of \bar{m}_H on \bar{A}_H (the mean mass number of the heavy mass peak) can be found (4). For each spontaneously-fissioning nuclide suitable values of \bar{m}_L and \bar{m}_H can then be estimated by interpolation. Extrapolation formulae (3, 4) were used to estimate E_d , the fission product decay energy: for consistency these were based on a value of $E_d = 23.8$ MeV/f for 235-U (n, f).

Most experimental values for the average number of prompt neutrons ($\bar{\nu}_p$) were obtained from the review of Manero and Konshin (5), but other sources were used when appropriate. For nuclides with no experimental data a systematic approach was adopted:

A was plotted against

$$\bar{\nu}_p(A, Z) - 0.104(Z-90)$$

to give a reasonably smooth curve from which $\bar{\nu}_p$ values were derived.

The prompt neutron energy distribution was assumed to be Maxwellian, with a mean energy B related to $\bar{\nu}_p$ by Terrell's formula (6):

$$B = a + b(\bar{\nu}_p + 1)^{\frac{1}{2}} \text{ MeV}$$

where $a = 0.74 \pm 0.02$ MeV, and $b = 0.653$ MeV.

The prompt gamma distribution for all the spontaneous fissioning nuclides were assumed to be the same as for the neutron-induced fission of ^{235}U .

3 THE UK HEAVY ELEMENT DECAY DATA FILE, UKHEDD-1

The computer-based file is stored in ENDF/B-V format based upon standard 80 column card images. There is a general information data section for each nuclide containing the following information:

- (i) the name of the evaluator(s) and the date of the evaluation (month and year);
- (ii) a list of references used to determine the evaluated data set;
- (iii) detailed comments associated with the evaluation;
- (iv) various decay data not contained in the decay data section, including beta transition data and specific decay energies;
- (v) a consistency check of the evaluated data.

The recommended radioactive decay data are contained within the major data section. The energy data contained within this section are in eV and the intensities are expressed as percentages, calculated from the spectral normalization factor and the listed relative intensities. The data include the following:-

- (i) spin and parity of the decaying nuclide;
- (ii) half-life;
- (iii) average beta, gamma and alpha energies;
- (iv) decay modes, Q-values and branching ratios;
- (v) radiation decay data, including gamma, beta, alpha, neutron, discrete electron and X-ray transitions;
- (vi) spontaneous fission decay data.

At present a Winfrith report is being prepared (7) which will describe the contents of UKHEDD-1. The evaluation procedure and the mean decay data are defined in some detail, and comprehensive listings of the data will also be included.

Our future intention is to compare UKHEDD-1 with equivalent files that are being constructed elsewhere. In this way the file will be maintained and improved at regular publicised intervals. Enquiries concerning this file should be address to:

Mr B S J Davies, Applied Physics Division,
CEGB Berkeley Nuclear Laboratories,
Berkeley, Gloucestershire, England

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- 7 A. L. Nichols, M. F. James. AEEW - R 1407, in preparation.

Table 1: Selected Data for the Spontaneous Fissioning Nuclides in UKHEDD-1

Nuclide	sf branching ratio	Q_{sf} (MeV)	$\bar{\nu}_p$ (a)
230-Th	$(2.5 \pm 2.5) \times 10^{-13}$	169.7 ± 4.2	1.39 ± 0.14 (b)
232-Th	$(5 \pm 5) \times 10^{-12}$	169.4 ± 4.1	1.50 ± 0.30 (c)
232-U	$(1.0 \pm 0.5) \times 10^{-12}$	176.6 ± 4.4	1.71 ± 0.34 (c)
234-U	$(1.2 \pm 0.6) \times 10^{-11}$	176.1 ± 3.3	1.80 ± 0.18 (b)
235-U	$(2 \pm 1) \times 10^{-10}$	176.4 ± 4.6	1.87 ± 0.38 (c)
236-U	$(1.2 \pm 0.6) \times 10^{-9}$	175.6 ± 3.5	1.90 ± 0.06
238-U	$(5.4 \pm 0.2) \times 10^{-7}$	173.6 ± 3.4	2.00 ± 0.04
236-Pu	$(8 \pm 2) \times 10^{-10}$	185.3 ± 3.3	2.12 ± 0.13
238-Pu	$(1.86 \pm 0.06) \times 10^{-9}$	184.5 ± 2.7	2.21 ± 0.08
239-Pu	$(4.4 \pm 0.4) \times 10^{-12}$	184.3 ± 3.6	2.32 ± 0.23 (b)
240-Pu	$(5.7 \pm 0.2) \times 10^{-8}$	184.7 ± 2.5	2.151 ± 0.026
242-Pu	$(5.5 \pm 0.1) \times 10^{-6}$	184.7 ± 3.1	2.141 ± 0.027
244-Pu	$(1.25 \pm 0.06) \times 10^{-3}$	184.2 ± 3.8	2.29 ± 0.19
241-Am	$(3.8 \pm 0.1) \times 10^{-12}$	188.1 ± 4.9	2.50 ± 0.50 (c)
242m-Am	$(1.6 \pm 0.6) \times 10^{-10}$	188.4 ± 3.7	2.56 ± 0.26 (b)
243-Am	$(2.2 \pm 0.3) \times 10^{-10}$	187.8 ± 3.8	2.61 ± 0.26 (b)
242-Cm	$(6.8 \pm 0.7) \times 10^{-8}$	193.2 ± 2.0	2.528 ± 0.033
244-Cm	$(1.347 \pm 0.002) \times 10^{-6}$	191.8 ± 2.4	2.6875 ± 0.0330
246-Cm	$(2.614 \pm 0.005) \times 10^{-4}$	190.4 ± 2.8	2.948 ± 0.037
248-Cm	0.0826 ± 0.0003	189.6 ± 3.4	3.161 ± 0.040
250-Cm	0.7 ± 0.3	189.9 ± 4.1	3.30 ± 0.09
249-Bk	$(4.7 \pm 0.2) \times 10^{-10}$	192.7 ± 3.0	3.400 ± 0.048
249-Cf	$(5.2 \pm 0.2) \times 10^{-9}$	196.0 ± 4.4	3.4 ± 0.4
250-Cf	$(7.7 \pm 0.3) \times 10^{-4}$	194.8 ± 3.2	3.52 ± 0.10
252-Cf	0.03092 ± 0.00008	193.9 ± 2.4	3.756 ± 0.047
253-Es	$(8.7 \pm 0.3) \times 10^{-8}$	198.3 ± 4.4	4.7 ± 0.5

(a) All $\bar{\nu}_p$ values based on $\bar{\nu}_p$ (252-Cf) = 3.756 ± 0.047 ; standard deviation in $\bar{\nu}_p$ includes contributions from this standardisation.

(b) Calculated from reference (5) using equation

$$\bar{\nu}_{th}(A-1, Z) = \bar{\nu}_p + 0.101(BE)_n$$

(c) From systematics.

Transactinium Nuclear Data Evaluation and Measurement (TANDEM)

Project Progress Report

The measurement part of this programme has been severely disrupted during this year by a series of failures to the special gamma ray detectors. This has involved an extensive replacement and re-calibration programme. Measurements are now being started again and it is hoped that results will be available shortly.

Uranium Isotopes. If measurements on these isotopes has been carried out as part of a safeguards programme for the isotopic measurement of uranium non-destructively using gamma rays. For this work most of the NBS uranium isotopic standard reference materials were examined over the range from 0.5% to 93% ^{235}U . This work is continuing.

$^{232}\text{Uranium}$. A source of $^{232}\text{Uranium}$ was purified by an ion-exchange chromatography using 12 M hydrochloric acid. About 14 days elapsed between separation and the first measurements and the gamma ray contribution from daughter products was too great for useful measurements to be made. This work is being continued.

$^{236}\text{Uranium}$. About 8 mg of ^{236}U containing more than 99% of this isotope and ~ 0.7% ^{235}U , was purified by anion exchange chromatography to remove daughter products. The gamma abundances associated with the decay of this nuclide are so low that larger quantities of an even higher enrichment material are required before meaningful measurements can be made. Only gamma rays from ^{235}U can be observed. This work is continuing.

Gamma Standards. The calibration of the gamma ray detector system is of fundamental importance for the accurate measurement of I_γ for transactinium nuclide decay. It has now been established that a range of γ intensity values exists for standard nuclides used for the efficiency calibration of detectors. The values used by different workers have been evaluated and the region particularly below 400 Kev shows disturbing differences. The consequence of this observation is that it is virtually impossible to compare the I_γ values obtained by different experimentalists. Attempts are now in progress to assess the realistic uncertainties of measurements of I_γ and from this to decide if further measurements are likely to give the desired improvements to the decay scheme data.

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

Vienna, October, 1981

C. W. Reich

In this report, we present the current status of the work being carried out in various U. S. laboratories that is specifically oriented toward the objectives of this IAEA CRP. Reported below are the absolute γ -ray intensity measurements at INEL, the half-life measurements carried out 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. ABSOLUTE GAMMA-RAY INTENSITY MEASUREMENTS AT INEL

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

^{240}Pu : The measurement of the absolute intensities of the three prominent γ rays emitted following the decay of ^{240}Pu has been completed. Since interest was expressed at the previous meeting of the CRP in having precise values for the energies of these γ rays, we have also measured the energies of these transitions. A paper describing these measurements and presenting the results has been accepted for publication in the International Journal of Applied Radiation and Isotopes. The measurement results are summarized in Table I.

^{239}Pu : Preliminary values for the absolute intensities of six of the most prominent γ rays from ^{239}Pu were reported at the previous meeting of the CRP participants. Subsequently, the measurements for these six transitions and analysis of the resulting data were completed. At the previous meeting, interest was expressed in having available precise energy values for these γ rays and intensities for a number of

the weaker transitions. These additional measurements have also been completed, and a paper describing them has been submitted to the International Journal of Applied Radiation and Isotopes. The energy and absolute-intensity data for the six prominent γ rays from ^{239}Pu are summarized in Table II, and the data for a number of the weaker ^{239}Pu γ rays are given in Table III. It should be noted that the intensity data in Table III are preliminary values only; a small correction resulting from a time dependence in the efficiency of one of the detectors used in the measurement has not been applied to these data.

^{238}Pu : Preliminary results have been obtained for the absolute intensities of the three most intense γ rays from the ^{238}Pu decay. The source material was ~ 25 μg of ^{238}Pu , consisting of 99.90 ± 0.01 wt. percent ^{238}Pu and $0.10 \pm 0.01\%$ wt. percent ^{239}Pu . This material was obtained by chemically separating Pu from a sample of ^{237}Np which had been irradiated in EBR-II for the purpose of making fast-fission-yield measurements. Preliminary values for the absolute intensities of the three strongest γ rays from ^{238}Pu are summarized in Table IV. The ^{238}Pu sample concentrations on which these values are based were determined from a sample mass analysis. An absolute α emission-rate measurement to confirm this analysis is presently being carried out at NBS.

Because of its potential relevance in some applications, we plan also to measure the absolute intensity of the 766-keV γ ray. Because of its low intensity ($\sim 2.4 \times 10^{-5}\%$), however, we anticipate that the quoted precision of this value will be somewhat worse than those of the other γ rays.

^{233}U : A solution of ^{233}U , with an isotopic purity of 99.99%, has been acquired. This material remained from the SALE interlaboratory comparison program and the analysis had been verified by coulometry as well as by the New Brunswick Laboratory. Measurements on this sample will get underway later on this year.

Absolute Detector-Efficiency Measurements: The measurement of the absolute efficiencies of the two detectors utilized in the γ -ray intensity studies reported here has been completed. The first detector is an $\sim 8\text{-cm}^3$ planar intrinsic Ge detector fabricated at the Lawrence Berkeley Laboratory and has a typical resolution of 0.86 keV (FWHM) at $E_\gamma=88$ keV. The efficiency calibration of this detector was originally carried out in March, 1980 when the first of the ^{240}Pu and ^{239}Pu γ -intensity measurements were made. As reported at the previous meeting of the CRP participants, this calibration involved 19 sources, 18 isotopes and 58 γ rays with energies from 14 to 2754 keV. With this calibration, the uncertainty in the efficiency is quoted as $\sim 2\%$ from 30 to 85 keV, 1% from 85 to 120 keV, 0.75% from 120 to 420 keV and 0.5% from 420-1420 keV. After this original calibration, repeated measurements with a ^{133}Ba source ($T_{1/2}=10.7$ y) indicated that the detector efficiency was decreasing. Consequently, the efficiency-calibration curve was completely remeasured in August, 1981 at the time of the ^{238}Pu γ -intensity measurements. The average rate of change of the efficiency appears to be $\sim 0.5\text{-}1\%$ per year above 300 keV and less at lower energies. For this new efficiency curve, the uncertainties assigned are 1.5% for 50-80 keV, 1% for 80-120 keV, 0.75% for 120-420 keV and 0.5% for 420-1420 keV.

The second detector is an 114-cm^3 closed-end coaxial intrinsic Ge detector. This detector has been calibrated in the same manner as the other detector. The assigned uncertainties in the efficiency calibration are 1.5% for 80-120 keV, 0.75% for 120-420 keV and 0.5% for 420-1420 keV. Efficiency checks, similar to those described above for the smaller detector, have been made over a period of ~ 5 months now and no significant systematic changes ($\leq 0.1\%$) in this efficiency have thus far been observed.

II. ACTIVITIES OF THE HALF-LIFE EVALUATION COMMITTEE

The various participating laboratories have completed their measurements of the ^{240}Pu half-life. Walter Strohm, the chairman of the committee says that they are not yet in a position to recommend

a final value. He feels at the present time that the impact of their final result will be not so much to change the present half-life values as it will be to provide an increased confidence in them. Their final value will probably lie between the recent ANL measurement and the calorimetry-based values. The method of publishing their ^{240}Pu work has not yet been decided but a format similar to that which was done for the ^{239}Pu result is being considered.

The committee does not have any presently planned defined measurement goals beyond ^{240}Pu .

III. MEASUREMENT OF RELATIVE ALPHA INTENSITIES (Irshad Ahmad, Chemistry Division, ANL)

The relative intensities of alpha groups associated with the decay of ^{239}Pu and ^{233}U were measured with a 25-mm² area Au-Si surface-barrier detector. Isotopically pure and essentially massless sources prepared in the Argonne electromagnetic isotope separator were used in these measurements. Spectra were measured at 0.2% geometry in order to reduce distortions in α intensities due to summing of α -particle pulses with the electron and L X-ray pulses. The spectra were accumulated for 3 to 6 days and the gain of the counting system was held constant with a digital-gain stabilizer. Under these conditions a resolution (FWHM) of 13.0 keV was achieved. Peak areas were determined from hand plotted graphs. The relative intensities obtained from the analysis of several spectra are given in Tables V and VI. Additional measurements are in progress.

Table I

Summary of the INEL measurements of the energies and absolute intensities of the three prominent γ rays emitted following the decay of ^{240}Pu . Quantities in parentheses represent the uncertainties in the least significant figure (or figures) of the associated value.

γ -ray energy (keV)	Absolute γ -ray intensity ($\gamma/10^5$ decays)
45.244(2)	43.5(9)
104.234(6)	7.18(7)
150.308(3)	0.402(5)

Table II

Summary of the INEL measurements of the energies and absolute intensities of the six prominent γ rays from the ^{239}Pu decay. Quantities in parentheses represent the uncertainties in the least significant figure (or figures) of the associated value.

γ -ray energy (keV)	Absolute γ -ray intensity ($\gamma/10^5$ decays)
51.624(1)	27.1(5)
129.296(1)	6.41(5)
203.550(5)	0.568(4)
332.845(5)	0.492(4)
375.054(3)	1.547(12)
413.713(5)	1.455(9)

Table III

Energy and intensity values for a number of γ rays from ^{239}Pu decay. The intensity values should be regarded as preliminary only, and small changes in them are anticipated.

Energy (keV)	Relative intensity	Energy (keV)	Relative intensity
30.251(10)	15.4(4)	307.81(10)	0.08(2)
38.660(2)	159.(4)	311.78(4)	0.40(3)
41.848(10)	4.13(10)	316.41(4)	0.20(4)
46.218(10) ^a	16.1(7) ^a	320.862(20)	0.86(3)
51.624(1)	435.(10)	323.841(29)	0.84(2)
54.039(8)	3.10(7)	332.845(5)	7.70(11)
56.825(3)	18.1(4)	336.120(12)	1.73(4)
65.675(20)	0.75(6)	341.502(19)	1.00(4)
67.674(12)	2.80(11)	345.013(4)	8.72(13)
68.696(6)	8.17(18)	361.90(6)	0.22(2)
77.592(14)	7.0(2)	367.096(26)	1.44(3)
78.44(3)	2.1(2)	368.557(27)	1.37(3)
89.73(4)	0.47(8)	375.054(3)	24.3(3)
UK _{α}	198.	380.191(6)	4.78(7)
103.086(14)	3.81(10)	382.698(16)	4.03(6)
UK _{β}	39.1	392.914(14) ^a	8.61(13) ^a
119.73(3)	0.53(2)	399.54(9)	0.09(1)
124.729(20) ^a	2.18(10) ^a	406.77(25)	0.046(11)
129.296(1)	\cong 100.0	413.713(5)	23.0(3)
141.62(4)	0.46(8)	422.598(19)	1.88(4)
144.201(3)	4.83(9)	445.81(10) ^a	0.18(2) ^a
146.094(6)	2.02(10)	451.481(10)	2.96(4)
161.482(12)	1.96(4)	481.78(12)	0.11(1)
171.393(6)	1.74(4)	617.96(15)	0.09(1)
179.220(12)	1.04(3)	639.99(10)	0.16(2)
189.360(10)	1.30(2)	645.98(3)	0.21(2)
195.679(8)	1.68(3)	651.79(10)	0.125(15)
203.550(5)	8.90(13)	658.63(15)	0.125(14)
225.42(4)	0.23(2)	718.0(5)	0.73(15)
243.39(6)	0.38(3)		
249.06(9)	0.11(1)		
255.384(15)	1.27(3)		
263.97(3)	0.42(4)		
297.46(3)	0.78(2)		
302.88(10)	0.075(10)		

^aThe data of Gunnink et al. [UCRL-52139] indicate that this peak is a multiplet.

Table IV

Summary of γ -ray intensity measurements for ^{238}Pu . The listed absolute-intensity values should be regarded as preliminary.

E_γ (keV)	Sample	Uncorrected peak area (counts/mg-sec)	Absolute γ -ray intensity values	
			(γ 's/mg-sec)	($\gamma/10^5$ decays)
43.48	^{238}Pu -4	0.0300	3.92	
	-3	0.0304	3.98	
	-2	0.0302	3.95	
	-1	0.02995	4.02	
			Avg.	3.97
99.86	^{238}Pu -4	0.00547	0.764	
	-3	0.00547	0.763	
	-2	0.00552	0.771	
	-1	0.00542	0.776	
			Avg.	0.768
152.68	^{238}Pu -4	----	----	
	-3	0.000464(15)	0.0998	
	-2	0.000441(16)	0.0949	
	-1	0.000441(3)	0.0972	
			Avg.	0.0973

Table V
 ^{239}Pu α Intensities

Energy (MeV)	Excited State Energy (keV)	Relative Intensity (%)
5.155	0	88.0 \pm 0.3
	13	
5.105	51	12.0 \pm 0.2
	82	

Table VI
 ^{233}U α Intensities

Energy (MeV)	Excited State Energy (keV)	Relative Intensity (%)
4.824	0	82.7 \pm 0.3
4.783	42	14.9 \pm 0.2
4.729	97	1.85 \pm 0.05

Progress Report for the IAEA Coordinated Research Programme
on the Measurement of Transactinium Isotope Nuclear Decay Data

October 1981

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With respect to the measurement of transactinium isotope nuclear decay data, following studies have been performed in Japan. Half-life measurements have been made on ^{242}Cm by Usuda et al.[1] and on ^{237}Pu by Baba et al.[2]. Intensity of gamma rays of ^{239}Pu has been precisely measured by Yoshizawa et al.[3]. Progress in the works is described.

1. Half-life of ^{242}Cm

The measurement of alpha-decay half-life of ^{242}Cm was completed. The best value obtained from the measurements was 161.35 ± 0.30 day that is significantly shorter than the other data appeared in literature. Cause for the deviation has not been identified yet. A draft of the work was accepted for publication in the Journal of inorganic and nuclear Chemistry and its preprint was already distributed to the CRP members as JAERI-memo 9512.

[1] H. Umezawa, Progress Report to IAEA on the Measurement of Nuclear Decay Data of ^{242}Cm under the Research Agreement No. 2170/R2/CF (June 1981).

[2] H. Baba, T. Suzuki and K. Hata, J. inorg. nucl. Chem., 43, 1059(1981).

[3] Y. Yoshizawa, Report to JAERI under a Resarch Contract with the Hiroshima University (March 1981).

2. Partial half-life of ^{242}Cm for spontaneous fission decay

Mica detector techniques which have been tested by using ^{242}Pu as reported previously [1,4], were applied to the ^{242}Cm samples. For the purposes of determining the partial half-life of ^{242}Cm for spontaneous fission, it is important to determine the ratio of the spontaneous fission to alpha decay so accurately that the alpha activity of each sample was measured prior to the spontaneous fission measurement with a mica detector.

A piece of mica foil that was fully etched with hydrofluoric acid beforehand to erase background fission tracks, was placed in face-to-face to the ^{242}Cm sample electroplated on a platinum disk. The stack of the detector and sample was sandwiched between a couple of plain glass plates, pressed and allowed to stand for an appropriate period to make exposure the detector to the fission fragments of the sample. Then the detector was taken out and chemically treated for etching fission tracks: Immersed in concentrated hydrofluoric acid for several hours at room temperature, washed with nitric acid, water, ethylalcohol and acetone, in turn, and dried. After etching the mica piece was fixed on an optical glass plate sectioned with lines in 0.2mm x 0.3mm squares having address numbers. Microphotographs were taken in series covering all the sections over the sample area, and the number of fission tracks were totally counted on the photographs. Figure 1 shows the distribution of tracks counted on a typical sample of ^{242}Cm and Table 1 gives current results of the measurements.

[4] H. Umezawa, Progress Report to IAEA on the Measurement of Nuclear Decay Data of ^{242}Cm under the Research Agreement No. 2170/R1/CF, JAERI-memo 8963(1980).

3. Half-life of ^{237}Pu

The work has been published in the Journal of inorganic and nuclear Chemistry[2].

Sample material was prepared by $^{238}\text{U}(^3\text{He},4n)^{237}\text{Pu}$ reaction using a cyclotron of the Institute of Physical and Chemical Research followed by adequate chemical treatment. Details of the procedures are described elsewhere[5].

The half-life of ^{237}Pu was determined by periodic measurements of low-energy-photon spectra with a planar germanium detector. A sample for measurement was prepared by depositing an aliquot of the purified sample solution on Mylar film supported with aluminum ring and covering it with Scotch tape. Among impurities involved in the ^{237}Pu sample, ^{236}Pu and ^{238}Pu were the most interfering for the decay measurement, amounting to about 1% in activity. The measurements were carried out 43 times for about 600 days. Variation of the detection efficiency during the period was monitored by use of a reference source of ^{241}Am . Shaping time of the electronic circuit used was set to 2 μ sec and total count rate was as low as 600 cps from the beginning, so that the distortion of spectra due to pile-up effect was negligible.

Four single-gamma-ray peaks and two complex peaks which were not affected by the activity of ^{236}Pu and ^{238}Pu , were chosen to make decay curve of ^{237}Pu . The half-life was deduced from each of the decay curves by the least squares fit. As weighted average of the six values, the half-life of ^{237}Pu was found to be 45.12 ± 0.03 day.

[5] K. Hata, H. Baba, H. Umezawa, T. Suzuki and T. Nozaki,
Intern. J. appl. Radiat. Isotopes, 27, 713(1976).

4. Gamma-ray intensity of ^{239}Pu

The work has been performed by Yoshizawa et al. of the Hiroshima University under a research contract with JAERI. A method making accurate determination of gamma-ray intensity with a Ge(Li) detector has been developed by Yoshizawa et al.[6] and applied to the case. The sample material was of 97% ^{239}Pu , 3% ^{240}Pu and 0.0% ^{241}Pu . After purified from americium, it was prepared in 4M hydrochloric acid solution. 1.5ml of the solution that was accurately weighed, was taken in a capsule made of acrylic resin and used for relative intensity measurement. Another capsule was contained the same amount of the plutonium solution mixed with a known amount of a standardized ^{133}Ba solution and used to determine absolute intensity of gamma rays of ^{239}Pu in reference with the intensity of 356-keV gamma-ray peak of ^{133}Ba . For making relative detection efficiency curve of the detector, prepared were ^{152}Eu and ^{154}Eu samples in which the physical conditions of the solution were adjusted to obtain the same self-absorption characteristics to that of the plutonium samples by adding uranium of natural isotope abundance. The measurements were carried out in JAERI with a Ge(Li) detector in an arrangement shown in Fig. 2.

The plutonium sample was measured 48 times and each measurement took 2 hours. The detection efficiency for the 356-keV gamma ray was determined to be $(4.66 \pm 0.03) \times 10^{-4}$.

Results of the relative intensity of gamma rays from ^{239}Pu are shown in Table 2 in comparison with those measured by Cline[7] and Gunnink et al.[8]. The present results are rather close to those of Gunnink et al. but deviated systematically below 200 keV.

The amounts of plutonium contained in the sample capsules are going to be analysed by isotope dilution mass spectrometry, and absolute intensity of the gamma rays is to be obtained.

[6] Y. Yoshizawa, Y. Iwata, T. Kaku, T. Katoh, J. Ruan, T. Kojima and Y. Kawada, Nucl. Instrum. Meth., 174, 109(1980).

[7] J. E. Cline, Nucl. Phys., A106, 481(1968).

[8] R. Gunnink, J. E. Evans and A. L. Prindle, UCRL-52139(1976).

Table 1.

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

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

Table 2. Relative gamma-ray intensities for ^{239}Pu .

Gamma-ray energy (keV)	Relative intensities (%)					
	Present		Cline ²⁾		Gunnink et al. ⁴⁾	
129.3	445	± 7	373	± 47	420	± 9
141.7	2.6	± 0.8	6.5	± 0.6	2.15	± 0.06
143.4	} 22.3	± 0.6	1.80	± 0.27	1.16	± 0.05
144.2					19.0	± 0.4
146.1	8.5	± 0.4	6.5	± 0.7	79.9	± 1.7
160.2	} 13.4	± 0.6	0.33	± 0.13	0.42	± 0.08
161.5			6.7	± 1.3	8.06	± 0.13
171.3	7.7	± 0.4	6.5	± 0.6	7.42	± 0.16
179.2	4.33	± 0.19	1.93	± 0.20	4.42	± 0.10
184.3	0.09	± 0.07				
188.2	0.61	± 0.06	0.28	± 0.04	0.73	± 0.07
189.3	5.69	± 0.10	4.9	± 0.6	5.57	± 0.15
195.7	7.43	± 0.16	5.9	± 0.7	7.15	± 0.15
198.0	0.08	± 0.07				
203.5	39.0	± 0.5	32.0	± 2.7	37.6	± 0.7
218.0	0.07	± 0.06				
225.4	0.91	± 0.08	0.73	± 0.13	1.05	± 0.05
237.4	1.27	± 0.08	0.93	± 0.13	0.97	± 0.04
242.1	} 2.41	± 0.14	0.62	± 0.06	0.49	± 0.03
243.4			1.3	± 0.3	1.70	± 0.05
244.9			0.33	± 0.05	0.34	± 0.03
249.0			0.50	± 0.08	0.50	± 0.06
255.4	5.30	± 0.10	4.4	± 0.5	5.41	± 0.15
263.9	1.64	± 0.11	1.27	± 0.20	1.75	± 0.06
265.6	0.06	± 0.07				
281.2	0.11	± 0.06	2.7	± 0.4	0.148	± 0.020
285.3	0.08	± 0.06				
297.5	3.32	± 0.08	2.7	± 0.4	3.37	± 0.09
302.9	0.32	± 0.06	0.48	± 0.06	0.343	± 0.027
307.9	0.34	± 0.06	0.26	± 0.05	0.369	± 0.027
311.7	3.46	± 0.07	1.33	± 0.13	1.73	± 0.05
316.4	0.86	± 0.06	0.66	± 0.08	0.91	± 0.03
320.9	3.69	± 0.08	3.1	± 0.3	3.60	± 0.08
323.8	3.51	± 0.07	2.9	± 0.3	3.64	± 0.08
332.8	35.1	± 0.3	28.7	± 2.7	34.0	± 0.7
336.1	7.69	± 0.16	6.3	± 0.7	7.62	± 0.15
341.5	4.58	± 0.13	4.0	± 0.5	4.45	± 0.09
345.0	37.35	± 0.28	33	± 3	37.6	± 0.7
361.9	0.71	± 0.04	0.73	± 0.13	0.82	± 0.05
367.1	5.59	± 0.08	5.5	± 0.6	5.81	± 0.11
368.6	5.89	± 0.08	5.4	± 0.6	6.06	± 0.12
375.0	104.4	± 0.7	100	± 7	105.4	± 2.0
380.2	20.38	± 0.15	20.0	± 2.0	20.5	± 0.4

Table 2. (continued)

Gamma-ray energy (keV)	Relative intensities (%)					
	Present		Cline ²⁾		Gunnink et al. ⁴⁾	
382.8	17.40	± 0.13	16.7	± 2.0	17.4	± 0.3
392.5	8.3	± 1.1	18	± 3		
393.1	28.7	± 1.1	18	± 3	37.1	± 0.7
399.5	0.45	± 0.06	0.60	± 0.10	0.410	± 0.020
411.2			0.93	± 0.20	0.47	± 0.20
413.7	100.0	± 0.7	100	± 7	100.0	± 2.0
422.6	8.27	± 0.08			8.01	± 0.16
426.7	1.76	± 0.04				
430.1	0.481	± 0.027			0.289	± 0.011
445.7	0.57	± 0.05	0.50	± 0.07	0.584	± 0.017
451.5	12.74	± 0.11	12.0	± 1.3	12.69	± 0.27
457.6	0.101	± 0.026	0.080	± 0.027	0.100	± 0.002
474.4	0.003	± 0.002				
481.5	0.299	± 0.019	0.34	± 0.07	0.309	± 0.006
493.1	0.054	± 0.014				
550.6	0.034	± 0.013				
598.0	0.128	± 0.015			0.112	± 0.004
612.8	0.068	± 0.015			0.064	± 0.003
617.1	} 0.300	± 0.020	0.140	± 0.027	0.090	± 0.005
618.3			0.140	± 0.027	0.137	± 0.005
633.2	0.157	± 0.014	0.080	± 0.020	0.170	± 0.004
637.8	0.202	± 0.018			0.172	± 0.004
640.1	0.613	± 0.021	0.38	± 0.06	0.551	± 0.011
646.0	1.022	± 0.026	0.87	± 0.20	1.000	± 0.020
649.3	0.067	± 0.021			0.048	± 0.003
652.1	0.443	± 0.021	0.34	± 0.07	0.440	± 0.009
654.9	0.097	± 0.020	0.100	± 0.020	0.151	± 0.003
658.9	0.645	± 0.021	0.64	± 0.08	0.651	± 0.014
664.6	0.113	± 0.015			0.111	± 0.003
674.1	0.035	± 0.014			0.0346	± 0.0012
686.0	0.100	± 0.015			0.0586	± 0.0019
690.8	0.060	± 0.014			0.0374	± 0.0020
701.1	0.046	± 0.015			0.0344	± 0.0011
703.7	0.302	± 0.017	0.32	± 0.06	0.265	± 0.005
717.7	0.183	± 0.015	0.120	± 0.027	0.184	± 0.004
727.9	0.109	± 0.015			0.0083	± 0.0005
756.4	0.235	± 0.016	0.26	± 0.05	0.233	± 0.005
769.4	0.806	± 0.021	0.64	± 0.08	0.752	± 0.015

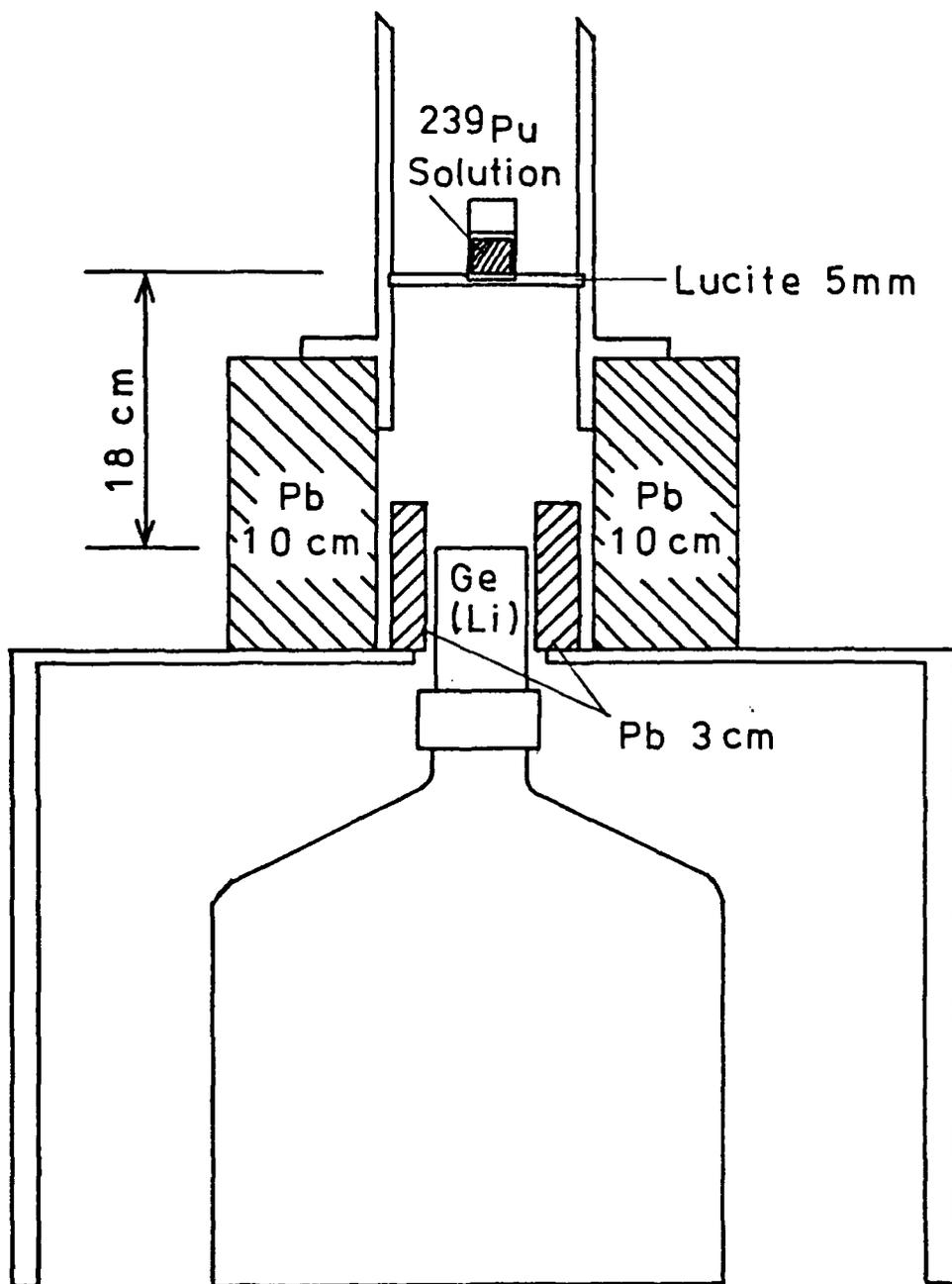


Fig. 2. Geometrical arrangement of Ge(Li) detector to the Pu sample.

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Translated from Russian

REPORT TO THE FOURTH MEETING OF THE CO-ORDINATED RESEARCH PROGRAMME
ON THE MEASUREMENT AND EVALUATION OF TRANSACTINIUM NUCLEAR DATA

The work of Soviet scientists on measurement and evaluation of
actinide nuclear data in 1980-81

V.M. Kulakov

In 1980-81, a considerable amount of work was done in the Soviet Union on the evaluation of non-neutron (decay) nuclear data for actinides.

The group led by Yu.V. Khol'nov (Radium Institute of the USSR Academy of Sciences, Leningrad) published tables [1] of evaluated nuclear physics data obtained from an analysis of original experimental work and by processing data found in the literature with application of the rules of mathematical statistics. The tables include the following information: half-lives; decay energy; energies of α , β , X and γ radiation and conversion electrons; absolute intensities of X-rays, γ radiation and conversion electrons; mean energies of α and β particles and electromagnetic radiation; conversion coefficients, ionization gamma constants and certain other characteristics of specific nuclides (for example, partial half-lives, fluorescence yields, etc.). All papers, including unpublished ones, mentioned up to January 1979 in abstracting journals and other periodicals were taken into account. A procedure was worked out for finding a "best" or recommended value with a confidence level of 95%.

Altogether, the authors analysed 100 radionuclides, 13 of which were actinides (^{234}U , ^{237}Np , ^{239}Np , ^{236}Pu , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am , ^{243}Am , ^{242}Cm and ^{244}Cm).

At the XXXI All-Union Meeting on Nuclear Spectroscopy and the Structure of the Atomic Nucleus (Samarkand, April 1981) the same authors presented a paper on the evaluation of the half-lives of some 200 radionuclides [2]. Table 1 below shows evaluated half-lives for actinides with their errors (shown in brackets) for a confidence level $P = 0.68$.

Isotope	$T_{I/2}$	Isotope	$T_{I/2}$
Th - 228	1,9131(9) a	Pu -240	6,56(1) · 10 ³ a
Th - 232	1,40(1) · 10 ¹⁰ a	Pu -241	14,61(10) a
U - 232	68,9(4) a	Pu -242	3,742(14) · 10 ⁵ a
U - 233	1,5927(14) · 10 ⁵ a	Am -241	432,1(3) a
U - 236	2,3415(14) · 10 ⁷ a	Am - 242	16,02(2) h
U - 237	6,752(2) d	Am - 242m	152(7) a
U - 238	4,4683(24) · 10 ⁹ a	Am - 243	7,37(2) · 10 ³ a
Np -237	2,14(1) · 10 ⁶ a	Cm -242	163,0(2) d
Np -239	2,347(3) d	Cm -244	18,11(3) a
Pu -236	2,85(1) a	Cf -249	350,7(20) a
Pu -238	87,74(5) a	Cf -252	2,638(6) a
Pu -239	24113(11) a		

At the Centre for Data on the Structure of the Atomic Nucleus and Nuclear Reactions (TsAYaD) an evaluation of non-neutron (decay) data was performed for isotopes of atomic mass 238 and 242. Under the bilateral agreement a magnetic tape with evaluated data was sent to the United States national centre (Brookhaven National Laboratory). Table 2 shows half-lives of isotopes with atomic mass 238 and 242.

Table 2

Isotope	$T_{I/2}$	Isotope	$T_{I/2}$
U -238	4,468(3) · 10 ⁹ a	Pu -242	3,763(20) · 10 ⁵ a
Pa-238	2,3(1) min	Cm-242	162,8(4) d
Am-238	98(2) min	Cf-242	3,68(44) min
Np -238	2,117(2) d	Am-242	16,02(2) h
Cm -238	2,4(1) h	Am -242m	141(2) a

In addition to the half-lives, the following were evaluated: energies and relative intensities of α , β and γ radiation; energies of levels of daughter nuclei; multipolarity of radiation; and conversion coefficients and coefficients for forbidden α decay.

Apart from the evaluations of decay data, work continued in the Soviet Union on measurements of the nuclear physics characteristics of actinides.

At the I.V. Kurchatov Institute of Atomic Energy the group led by Professor L.A. Mikaehlyan measured the times of spontaneous fission (T_{SF}) and the first moments of distribution of the total number of prompt neutrons ($\bar{\nu}$ and σ_{ν}^2) per fission event for uranium isotopes. The measurements were made using a neutron detector based on ^3He counters in a moderator containing hydrogen, the source studied being placed in the centre. The work was performed in an underground laboratory at a depth of 80 metres of water equivalent with a considerably reduced external background and induced fission of sample nuclei. In addition, the detector was protected with a layer of borated polyethylene and anti-coincidence plates made of scintillation plastic which suppressed the meson background of the device (by a factor of over 100).

The efficiency of the detector in respect of neutrons of the ^{252}Cf spontaneous fission spectrum ($\bar{\nu} = 3.735$) was 0.49. With the change to spectra of uranium isotopes a correction of $\sim 2\%$ for the variation in efficiency was made; the correction was obtained by numerical modelling of the neutron diffusion processes in the detector.

For the measurements use was made of samples of uranium highly enriched in certain isotopes (^{238}U , ^{236}U and ^{235}U) weighing between 20 and 50 g. Estimates and experimental measurements were made of the contribution of self-multiplication and induced fission by neutrons of the (α, n) reaction to the counting speed for spontaneous fission events in samples. For the processing of the measurement results the shape of the multiplicity distribution of prompt neutrons was taken to be Gaussian.

The following results were obtained (see Table 3).

Table 3

Isotope	T_{SF}	$\bar{\nu}$	σ_{SF}
$U-238$	$(8,3 \pm 0,4) \cdot 10^5$ a	$1,97 \pm 0,10$	$1,03 \pm 0,08$
$U-236$	$(2,7 \pm 0,4) \cdot 10^{16}$ a	$1,79 \pm 0,18$	$1,02 \pm 0,12$
$U-235$	$> 3 \cdot 10^{18}$ a	-	-

The results for ^{238}U agree well with the data recommended in the literature. The spontaneous fission half-life and mean number of prompt neutrons per ^{236}U fission event confirm the only measurements performed, which are in Refs [3] ($T_{SF} = (2.42 \pm 0.17) \cdot 10^{16}$ a) and [4] ($\bar{\nu} = 1.89 \pm 0.05$). The spread of the multiplicity distribution of ^{236}U neutrons is given for the first time. The figure for spontaneous fission of ^{235}U is in agreement with recent measurements ($T_{SF} = (9.8 \pm 2.8) \cdot 10^{18}$ a) [5].

A group of workers at the Institute of Atomic Energy (S.A. Baranov et al.) attempted to verify the existence of a relatively long-lived isomer of ^{241}Pu [6]. The authors analysed the situation possibly occurring during detection of α radiation on the assumption that there was an isomer of ^{241}Pu with a half-life of about one third of a year. Comparisons were then made between the intensity of fine structure lines of α radiation for the "old" product of ^{241}Pu more than three years old and that for the product obtained by irradiation in a reactor of a target enriched in ^{240}Pu (the irradiation time was 280 d, and the fluence $5 \cdot 10^{21}$ n/cm²). Within the limits of statistical error (~20%) the measured ratios of the intensities of weak α transitions (0.2-1.3%) were the same. The conclusion was reached from the experimental data obtained that with a lifetime of $1/3$ a the relative probability of the isomer being formed in the reactor does not exceed 0.01 if the isomer has a spin $J = \frac{1}{2}$ and 0.3 if $J \geq \frac{9}{2}$.

With a view to checking the correctness of reactor programs and to predicting transuranium isotope breeding, the group of A.G. Zelenkov measured the fuel composition of a fuel-element sample from an assembly in the fourth reactor unit of the Novovoronezh nuclear power station [7]. By means of radiochemical, mass-spectrometric and radiometric techniques developed by them, the authors determined the contents of 20 heavy nuclides from ^{232}U to ^{246}Cm . The relative contents in the sample of

practically all the nuclides (with the exception of the long-lived uranium isotopes) were determined by detection of the α radiation with a semiconductor alpha spectrometer meter and a precision magnetic alpha spectrometer with a position-sensitive detector. The relative content of ^{243}Cm was determined with a semiconductor detector from the intensity of γ radiation with energies of 210, 229 and 278 keV. The authors point out a contradiction in the content of ^{245}Cm in the samples determined from α and γ radiation. Analysis of the data has shown that the accuracy with which quantum yields of γ rays during the decay of ^{243}Cm and ^{245}Cm are determined must be improved.

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