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INTERNATIONAL NUCLEAR DATA COMMITTEE

First Coordinated Research Meeting on the
Measurement of Transactinium Isotope Nuclear Data

Vienna, 20-21 April 1978

SUMMARY REPORT

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Nuclear Data Section
International Atomic Energy Agency

August 1978

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Abstract

The first meeting of the participants in the IAEA coordinated research programme to measure and evaluate required nuclear decay data of transactinium isotopes, was convened by the IAEA Nuclear Data Section on 20-21 April 1978, at IAEA Headquarters in Vienna.

The meeting participants discussed the current status of decay half-lives of transactinium isotopes, compiled preliminary lists of proposed half-life values, and discussed ways and means of collaboration between the participating research groups.

Table of Content

	Page
Abstract	i
Table of Content	ii
List of Appendices	iii
Foreword	iv
1. <u>Summary of the Meeting</u>	1
2. <u>Status Reports from Participating Groups</u>	2
3. <u>Nature and Scope of the Coordinated Research Programme</u>	2
4. <u>Collaboration between Participating Research Groups</u>	3
a) Exchange and distribution of information	3
b) Procurement of source samples and their exchange between participating groups	4
c) Participation in the Coordinated Research Programme	6
5. <u>Current Status of Transactinium Isotope Half-Life Data</u>	7

List of Appendices

1. List of Meeting Participants.
2. Adopted Agenda.
3. List of Actions.
4. List of Participants in the Programme.
5. Report from the Central Bureau for Nuclear Measurements, Geel, Belgium. R. Vaninbrouckx.
6. JAERI's Programme for Source Preparation and Decay Measurement of ^{242}Cm . Presented by T. Fuketa.
7. Status Report on the U.S. Programme. C.W. Reich.
8. Report of the Laboratoire de Metrologie des Rayonnements Ionisants. J. Legrand and G. Malet.
9. Report to 1st Coordinated Research Meeting on Trans-actinium Nuclear Decay Data. Presented by A.J. Fudge.

FOREWORD

The presently available accuracies of half-lives, and alpha and gamma-ray intensities of a number of transactinium isotopes are not adequate to meet the requirements set for fuel analysis, safeguards applications, mass determination and the preparation of standards.

In view of these discrepancies the IAEA Advisory Group Meeting on Transactinium Isotope Nuclear Data (TND), November 1975, recommended the organization of an international cooperative research programme to measure and evaluate needed decay data of transactinium isotopes which would be coordinated by the IAEA Nuclear Data Section (see INDC(NDS)-74/L+, March 1976).

The Coordinated Research Programme which was approved by the IAEA on 8 December 1977, 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 related to their nuclear characteristics required in many applications in science and industry.

It was proposed that the programme consist of extensions of on-going and initiation of measurements and evaluations, the co-ordination and intercomparison of results between laboratories concerned, and the exchange of samples and techniques. In particular, the participants would perform measurements of alpha and β decay, and spontaneous fission half-lives of transactinium isotopes, and of the relative intensities and energies of emitted alpha particles and associated gamma-rays.

The objective of the programme is to arrive at a consistent set of required decay data which satisfies the required accuracies.

All institutes and laboratories participating in this coordinated research programme are signatories to IAEA Research Agreements, and are as such subject to the rules governing these agreements.

1. SUMMARY OF THE MEETING

Introduction

The first meeting of the participants in the IAEA Coordinated Research Programme to measure and evaluate nuclear decay data of transactinium isotopes, was convened by the IAEA Nuclear Data Section on 20-21 April 1978, at IAEA Headquarters in Vienna. The attendees of this meeting are listed in Appendix 1. The meeting was chaired by Dr. A. J. Fudge, of AERE Harwell.

Meeting Objectives

The principal objective of this meeting was to initiate the collaboration between the research groups participating in this cooperative project, review the accuracy of existing data so as to agree on the priority of required measurements, and to discuss areas of possible cooperation between the participating research groups.

The Adopted Agenda is given in Appendix 2.

Conclusions and Results of the Meeting

The meeting reviewed the existing and planned programmes for the measurement and evaluation of transactinium isotope nuclear decay data of each participating research group; in particular, the meeting

- discussed the nature and scope of the coordinated research programme.
- agreed on ways and means to exchange and disseminate information pertinent to this programme.
- considered the possibility to exchange source samples among the participants of this programme.
- recommended the participation of scientists from developing countries in the programme.
- reviewed the status of transactinium isotope half-life data, compiled a list of proposed half-life values, and recommended the future review of the spontaneous fission data status.

The participants agreed that the next meeting of this coordinated research programme be convened in conjunction with the Advisory Group Meeting on Transactinium Isotope Nuclear Data planned to be held at a location in Europe in May 1979.

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

2. REPORTS FROM PARTICIPATING GROUPS

The reports presented by the members of this coordinated research programme are reproduced in this report as Appendices. Summary of these presentations and the reference to the relevant Appendix are given below.

1. Report from the Central Bureau for Nuclear Measurements, Geel, Belgium, R. Vaninbrouckx. Appendix 5.
2. JAERI's Programme for Source Preparation and Decay Measurement of ^{242}Cm . Presented by T. Fuketa. Appendix 6.
3. Status report on the U.S. programme, C.W. Reich. Appendix 7.
4. Report of the Laboratoire de Metrologie des Rayonnements Ionisants. J. Legrand and G. Malet. Appendix 8.
5. Report to the First Coordinated Research Meeting on Transactinium Nuclear Decay Data. A.J. Fudge. Appendix 9.

3. NATURE AND SCOPE OF THE COORDINATED RESEARCH PROGRAMME (CRP)

The coordinated research programme for the measurement and evaluation of transactinium isotope nuclear decay data consists at present of a cooperative effort of five research groups coordinated by the International Atomic Energy Agency. The name and laboratory address of the participants in this programme are listed in Appendix 4. Under this arrangement, each participating laboratory represented by a Chief Investigator, is party to a Research Agreement and is responsible to submit a yearly progress report of the measurements undertaken in context of the programme.

The IAEA, in addition to its coordinating function, assists in the distribution and exchange of information pertinent to the programme, and invites the chief investigators from each participating laboratory to attend Research Coordination Meetings at appropriate intervals at IAEA expense. At the conclusion of a coordinated research programme, the participants, in cooperation with the IAEA, produce a final report for publication by the Agency in its Technical Report Series. In the case of this programme, the final report is proposed to consist primarily of a comprehensive compilation of recommended nuclear decay data (including half-lives, and partial α - and γ - yields with their uncertainties) for all transactinium isotopes.

In addition to their individual concern regarding the existence and accuracy of measured and evaluated transactinium isotope decay data, the participants noted that most of the data considered in the scope of this programme are of basic importance to the development and applications of techniques used for nuclear materials safeguards purposes. It was also felt by the participants that an important function of this group is to identify discrepant transactinium isotope decay data (particularly half-lives of isotopes which are of prominence in the nuclear fuel cycle), and to alert the user community of these discrepancies.

4. COLLABORATION BETWEEN PARTICIPATING RESEARCH GROUPS

a) Exchange and Distribution of Information

In the discussion on the exchange and distribution of information, the group distinguished two categories of information: (a) information generated by this group (and associated measurement groups) of interest to data users, and (b) information on current measurements of interest to data producers.

It was agreed that information of both categories, (a) and (b), pertinent to this project be sent in 10 copies, to IAEA/NDS by each of the participating teams for distribution to the group. (See Action # 6)

Of the category (a) information, (i.e. measurement or evaluation results) of interest to data users (such as the first list of proposed half-lives reproduced in this report as Table 3), IAEA/NDS is prepared to distribute this information to the scientific community (e.g. members of the International Nuclear Data Committee (INDC), members of National Nuclear Data Committees, etc.) if it is so requested by members of this group.

Regarding category (b) information, on current measurements of interest to data producers (i.e. measurers and evaluators), the group made the following recommendation:

That authors of reports containing newly measured nuclear decay data (e.g. $T_{1/2}$, I_{γ} , I_{α}) send these data to IAEA/NDS either in their published form (e.g. laboratory report or preprint) or in summary form which could be reproduced, and that IAEA/NDS distribute these to the members of the group.

It was suggested by the group that this recommendation be implemented with the help of the INDC and members of the National Nuclear Data Committees.

The group also suggested that IAEA/NDS look into the possibility of creating a file of information on current nuclear decay measurements, and of distributing this information to decay data measurers, possibly in the form of an informal "information bulletin" (see Action # 8).

To aid in the creation of this information file, Reich was asked to investigate whether this information could be extracted from the "Secondary Sources" file kept by the Nuclear Data Project at the Oak Ridge National Laboratory (USA) (see Action # 7), and IAEA/NDS was asked to inquire whether a similar source of information existed in the Soviet Union. (See Action # 9).

b) Procurement of Source Samples and their Exchange between Participating Groups

The group discussed the availability of isotope samples required for decay data measurements. While it requested each member of the group to investigate the existence of isotope samples at their home laboratories (Action # 10), the group compiled a preliminary list of available sample material; this list is included in this report as Table 1. It was suggested that IAEA/NDS complement this table of available samples with the information received from the other members of the group and distribute it to all members of the group.

It was pointed out at the meeting, that in a number of cases, larger quantities of these isotopes were being used by the measurers of cross-sections.

The meeting took note of the requirements of the BARC/Trombay group for their decay data measurements (see Table 2 below). It was suggested that some of this material could perhaps be procured through the IAEA/NDS Targets and Sample Programme, or through direct bilateral arrangements with a supplier country.

Table 2

^{241}Pu (enrichment > 93%), 20 mg
^{242}Pu (enrichment > 99%), 10 mg
^{240}Pu (enrichment > 98%), 20 mg
^{239}Pu (enrichment > 99.99%), 20 mg
^{243}Am , 10 mg
^{241}Am , 500 mg
Plutonium chemical assay standard SRM 949e, 250 mg
Plutonium isotopic standard SRM 946, 250 mg

In particular, the group recognized the importance of having the origin and history of samples used in decay data measurements documented as accurately as possible.

The group also discussed existing measurement projects in which the same isotope sample was used for the same measurement. Information was given on one such project "the AS-76 Interlaboratory Alpha Spectrum Comparison", for the α - spectrometric measurements of the activity ratio of mixtures of Pu isotopes. Vaninbrouckx was asked to send more information about this project to the members of the group (see Action # 12). In general the group endorsed the exchange of source samples between members of the group but pointed to existing restriction in the transport of radioactive materials, particularly in liquid form.

TABLE 1

AVAILABILITY OF ISOTOPE SAMPLES FOR DECAY DATA MEASUREMENTS

PRELIMINARY LIST *

^{228}Th	Available at Harwell.
^{229}Th	Available at Harwell.
^{230}Th	13% enriched material available at Harwell, very small quantity of > 90% enriched material also available at Harwell.
^{232}Th	Available at Harwell.
^{231}Pa	60-100 gm available at Harwell, 10 gm available in the US.
^{233}Pa	Can be obtained by separation.
^{232}U	Available in the US, same at Harwell.
^{233}U	Some quantity of high purity (> 99%) available at Harwell. (Difficulty to obtain pure material.)
^{234}U	20 mg of 99% pure material available at Harwell.
^{235}U	Material of 99.996% purity available at ORNL.
^{236}U	9 mg available at Harwell (gm - quantities of this material are used by the Cross-Section Measurement Group at Harwell).
^{237}U	Milked from ^{241}Pu .
^{238}U	99.996% pure material available at Harwell and ORNL.
^{237}Np	Stock of 100% pure material available at Harwell and France.
^{238}Pu	Material is available.
^{239}Pu	gm - quantities of > 99% pure material available at Harwell.
^{240}Pu	gm - quantities of > 99% pure material available at Harwell.
^{241}Pu	Many - gm stock of 93% purity available at Oak Ridge. INEL plans to produce some by fast reactor irradiation.
^{242}Pu	95% pure material available in France.
^{241}Am	Large quantities available at the Karlsruhe Kernforschungszentrum (purity uncertain).
^{243}Am	Available at Harwell (?)
^{242}Cm	(?)
^{243}Cm	(?)
^{244}Cm	Available at Oak Ridge (?)

* This is not a complete list.

c) Participation in the Coordinated Research Programme

In the discussions regarding the composition of this coordinated research programme, the group suggested that Dr. Thiele from the Bundesanstalt für Materialprüfung in Berlin, and Dr. Ramthun from the Physikalische Technische Bundesanstalt in Braunschweig, be asked to join this coordinated effort. (See Action # 11).

The group also discussed the desirability to invite the participation of scientists from developing countries (e.g. Egypt, Iran, Nigeria, Pakistan). It was suggested that the training of such scientists could be done at the home laboratories of some of the members of this group. In view of the precedent cases at BCMN/Geel and LMRI/Saclay, the representatives from these two laboratories indicated that the proposed training could be done at their laboratories as part of their own programmes in context of this coordinated effort. The IAEA/NDS was asked to investigate if it were possible to obtain IAEA fellowship support for such training of scientists from developing countries. (See Action # 14).

5. CURRENT STATUS OF TRANSACTINIUM ISOTOPE HALF-LIFE DATA

In context of the overall objective of this coordinated research programme, that is to arrive at a consistent set of transactinium isotope decay data which satisfies the required accuracies, the group reviewed the current status of half-lives and their assigned uncertainties. The thirty-five considered isotopes were singled out at the 1975 Karlsruhe TND meeting as those requiring a half-life accuracy of 1% or better, and supplemented by isotopes identified by this group to be of importance in specific fields of applications, such as safeguards.

The resultant list, included in this report as Table 3, represents a critical appraisal of the current status of transactinium isotope half-lives by the members of this group, and is presented as a proposed list of recommended values.

The group concluded that only two of the nuclides of importance listed in Table 3 justified additional measurement of their half-life: (1) the α half-life of Pu-241, because it is based on the discrepant values of the Pu-241 β half-life, and (2) the half-life of Np-237 because its value is based on a single measurement.

In ascribing uncertainties to the recommended half-life values, the group agreed to adopt the criterion of

1 sigma random error plus 1/3 the linear sum of the systematic errors

based on a statistical confidence level of 68.3%. The group felt, however, that in no case should the ascribed total uncertainty be lower than 0.1%.

Another area which the group felt was in need of a critical appraisal, was the status of the spontaneous fission half-lives and their uncertainties. The group proposed to have IAEA/NDS generate a tabulation of spontaneous fission half-lives on the basis of the current INEL actinide file, and include this table in this report (see Action # 16). This tabulation, presented in Table 4, is proposed to serve as a "first draft" to an eventual list of recommended values, and should stimulate the measurement and evaluation of these quantities.

The list of transactinium isotopes included in these two tabulations is not assumed to be complete. It is the intent of this group to complete these lists at subsequent meetings. In particular, as it is anticipated that some tons of the minor actinides (e.g. Np, Am and Cm isotopes) will be generated in the next 20 years, their potential as fuels and strategic materials will make it imperative to know their decay characteristics to a large degree of accuracy.

TABLE 3

PROPOSED HALF-LIFE VALUES AND THEIR ACCURACIES

<u>Isotope</u>	<u>Decay Type</u>	$T_{1/2} \pm \Delta T_{1/2}$		<u>% Error</u>	<u>Comments</u>
^{228}Th	α	$(1.913 \pm .003)$	y	0.16	(b)
^{230}Th	α	$(7.7 \pm .3) \cdot 10^4$	y	3.9	(b)
^{232}Th	α	$(1.405 \pm .006) \cdot 10^{10}$	y	0.42	(b)
^{231}Pa	α	$(3.276 \pm .011) \cdot 10^4$	y	0.34	(b)
^{232}Pa	β^-	$(1.31 \pm .02)$	d	1.53	(b)
^{233}Pa	β^-	$(2.70 \pm .01) \cdot 10^1$	d	0.37	(b)
^{232}U	α	$(7.2 \pm .1) \cdot 10^1$	y	1.39	(b)
^{233}U	α	$(1.592 \pm .002) \cdot 10^5$	y	0.13	(b)
^{234}U	α	$(2.446 \pm .007) \cdot 10^5$	y	0.29	(b)
^{235}U	α	$(7.038 \pm .007) \cdot 10^8$	y	0.10	(a) 1
^{236}U	α	$(2.342 \pm .004) \cdot 10^7$	y	0.17	(a) 2
^{237}U	β^-	$(6.75 \pm .01)$	d	0.15	(b)
^{238}U	α	$(4.468 \pm .004) \cdot 10^9$	y	0.09	(b)
^{239}U	β^-	$(2.350 \pm .005) \cdot 10^1$	m	0.21	(b)
^{236}Np	β^-/EC	$(1.15 \pm .12) \cdot 10^5$	y	10.4	(b)
$^{236\text{m}}\text{Np}$	β^-/EC	$(2.25 \pm .04) \cdot 10^1$	h	1.78	(b)
^{237}Np	α	$(2.14 \pm .01) \cdot 10^6$	y	0.47	(b)
^{238}Np	β^-	$(2.117 \pm .002)$	d	0.09	(b)
^{239}Np	β^-	$(2.354 \pm .006)$	d	0.25	(b)
^{236}Pu	α	$(2.851 \pm .008)$	y	0.28	(b)
^{237}Pu	EC/α	$(4.56 \pm .02) \cdot 10^1$	d	0.44	(b)
^{238}Pu	α	$(8.774 \pm .009) \cdot 10^1$	y	0.10	(a) 3
^{239}Pu	α	$(2.411 \pm .003) \cdot 10^4$	y	0.12	(a)
^{240}Pu	α	$(6.553 \pm .008) \cdot 10^3$	y	0.12	(a) 4
^{241}Pu	β^-	$(1.47 \pm .04) \cdot 10^1$	y	2.72	(a) 5
^{241}Pu	α	$(6.0 \pm .1) \cdot 10^5$	y	1.67	(b) 6
^{242}Pu	α/SF	$(3.76 \pm .02) \cdot 10^5$	y	0.53	(a) 7
^{244}Pu	α/SF	$(8.2 \pm .1) \cdot 10^7$	y	1.22	(b)
^{241}Am	α	$(4.326 \pm .006) \cdot 10^2$	y	0.14	(a)
^{242}Am	β^-/EC	$(1.601 \pm .002) \cdot 10^1$	h	0.12	(b)
$^{242\text{m}}\text{Am}$	IT/α	$(1.52 \pm .07) \cdot 10^2$	y	4.6	(b)
^{243}Am	α	$(7.38 \pm .04) \cdot 10^3$	y	0.54	(b)
^{242}Cm	α	$(1.628 \pm .004) \cdot 10^2$	d	0.25	(a) 8
^{244}Cm	α/SF	$(1.811 \pm .002) \cdot 10^1$	y	0.11	(a)
^{252}Cf	α/SF	$(2.64 \pm .01)$	y	0.38	(a)

Comments to Table 3

- (a) Isotopes singled out at the 1975 Karlsruhe TND Meeting as those requiring a half-life accuracy of 1% or better.
- (b) Isotopes added to the (a) list by this group.

- - - - -

1. Used ANL value adding a realistic uncertainty.
2. Only few measurements available.
3. Adopted Ellis' evaluated value published in NDS 21, 590(8/71).
4. Adopted arithmetic mean of Jaffey's and Oetting's measured values.
5. The problem with the Pu 241 half-life is not one of inaccurate measurements but lies in the spread of measured values. The spread of measured values seems to reflect different sources of the material. The error quoted only indicates the span of measured values.
6. Value is based on the accepted Pu 241 β^- half-life.
7. The quoted value does not include the recently measured value of Meadows (ANS/NDM-38, Aug. 77) because it is based on the earlier used Pu 239 half-life of 24290 ± 70 years, and not on the one recommended in this set.
8. Adopted Ellis & Haese value published in NDS 21, 4(8/77).

TABLE 4

PROPOSED SPONTANEOUS FISSION HALF-LIVES DERIVED FROM THE INEL DATA BASE

Isotope	$[T_{1/2} \pm \Delta T_{1/2}]_{\text{Total}}$ (in years)*	Branching fraction	$[T_{1/2} \pm \Delta T_{1/2}]_{\text{SF}}$ (in years)	% Error
^{232}U	$(7.17 \pm 0.09) \cdot 10^1$	$(0.9 \pm 0.7) \cdot 10^{-12}$	$(7.97 \pm 6.22) \cdot 10^{13}$	78.0
^{233}U	$(1.5918 \pm 0.0015) \cdot 10^5$	$(1.3 \pm 0.3) \cdot 10^{-12}$	$(1.22 \pm 0.28) \cdot 10^{17}$	23.0
^{234}U	$(2.446 \pm 0.007) \cdot 10^5$	$(1.2 \pm 0.6) \cdot 10^{-11}$	$(2.04 \pm 1.02) \cdot 10^{16}$	50.0
^{236}U	$(2.3415 \pm 0.0014) \cdot 10^7$	$(1.2) \cdot 10^{-9}$	$(1.95) \cdot 10^{16}$	
^{238}U	$(4.4683 \pm 0.0024) \cdot 10^9$	$(5.45 \pm 0.06) \cdot 10^{-7}$	$(8.20 \pm 0.09) \cdot 10^{15}$	1.1
^{236}Pu	$(2.851 \pm 0.008) \cdot 10^0$	$(8.1 \pm 2.3) \cdot 10^{-10}$	$(3.52 \pm 1.00) \cdot 10^9$	28.4
^{238}Pu	$(8.775 \pm 0.005) \cdot 10^1$	$(1.84 \pm 0.05) \cdot 10^{-9}$	$(4.77 \pm 0.13) \cdot 10^{10}$	2.7
^{239}Pu	$(2.411 \pm 0.010) \cdot 10^4$	$(4.4) \cdot 10^{-12}$	$(5.48) \cdot 10^{15}$	
^{240}Pu	$(6.55 \pm 0.07) \cdot 10^3$	$(5.0 \pm 0.2) \cdot 10^{-8}$	$(1.31 \pm 0.05) \cdot 10^{11}$	3.8
^{242}Pu	$(3.763 \pm 0.020) \cdot 10^5$	$(5.50 \pm 0.06) \cdot 10^{-6}$	$(6.842 \pm 0.075) \cdot 10^{10}$	1.09
^{244}Pu	$(8.2 \pm 0.1) \cdot 10^7$	$(1.25 \pm 0.06) \cdot 10^{-3}$	$(6.56 \pm 0.32) \cdot 10^{10}$	4.95
^{241}Am	$(4.322 \pm 0.002) \cdot 10^2$	$(4.1 \pm 0.1) \cdot 10^{-12}$	$(1.05 \pm 0.03) \cdot 10^{14}$	2.9
$^{242\text{m}}\text{Am}$	$(1.52 \pm 0.07) \cdot 10^2$	$(1.6 \pm 0.6) \cdot 10^{-10}$	$(9.5 \pm 3.6) \cdot 10^{11}$	3.78
^{243}Am	$(7.38 \pm 0.04) \cdot 10^3$	$(2.2 \pm 0.2) \cdot 10^{-10}$	$(3.35 \pm 0.31) \cdot 10^{13}$	9.1
^{242}Cm	$(4.460 \pm 0.008) \cdot 10^{-1}$	$(6.8 \pm 0.6) \cdot 10^{-6}$	$(6.56 \pm 0.58) \cdot 10^6$	8.8
^{244}Cm	$(1.811 \pm 0.001) \cdot 10^1$	$(1.347 \pm 0.002) \cdot 10^{-6}$	$(1.344 \pm 0.002) \cdot 10^7$	0.15
^{246}Cm	$(4.73 \pm 0.10) \cdot 10^3$	$(2.614 \pm 0.005) \cdot 10^{-4}$	$(1.809 \pm 0.038) \cdot 10^7$	2.12
^{248}Cm	$(3.397 \pm 0.032) \cdot 10^5$	$(8.26 \pm 0.03) \cdot 10^{-2}$	$(4.112 \pm 0.041) \cdot 10^6$	1.01
^{249}Bk	$(8.761 \pm 0.164) \cdot 10^{-1}$	$(4.60 \pm 0.25) \cdot 10^{-10}$	$(1.91 \pm 0.11) \cdot 10^9$	5.8
^{249}Cf	$(3.506 \pm 0.021) \cdot 10^2$	$(5.02 \pm 0.10) \cdot 10^{-9}$	$(6.98 \pm 0.14) \cdot 10^{10}$	2.0
^{250}Cf	$(1.308 \pm 0.009) \cdot 10^1$	$(7.7 \pm 0.3) \cdot 10^{-4}$	$(1.70 \pm 0.07) \cdot 10^4$	4.1
^{252}Cf	$(2.638 \pm 0.010) \cdot 10^0$	$(3.092 \pm 0.008) \cdot 10^{-2}$	$(8.532 \pm 0.039) \cdot 10^1$	0.46
^{253}Es	$(5.604 \pm 0.005) \cdot 10^{-2}$	$(8.7 \pm 0.3) \cdot 10^{-8}$	$(6.44 \pm 0.22) \cdot 10^5$	3.4

* Conversions made are based on 365.24 days/year.

LIST OF MEETING PARTICIPANTS

A. J. Fudge Chemistry Division, Building 220 AERE Harwell, Didcot Oxfordshire OX11 0RA United Kingdom	Chief investigator (AERE, Harwell).
T. Fuketa Nuclear Data Center Japan Atomic Energy Research Institute Tokai-Mura, Naka-Gun Ibaraki-Ken 319-11 Japan	Proxy for chief investigator H. Umezawa (JAERI).
J. Legrand Laboratoire de Metrologie des Rayonnement Ionisants Boite Postale No. 2 F-91190 Gif-sur-Yvette France	Chief investigator (LMRI, Saclay).
A. Lorenz IAEA Nuclear Data Section	Project officer (IAEA/NDS).
G. Malet Centre d'Etudes Nucleaires de Saclay B.P. No. 2 F-91190 Gif-sur-Yvette France	Member of LMRI Group.
M. K. Mehta Head, Nuclear Physics Division Bhabha Atomic Research Centre Trombay, Bombay 400 085 India.	Proxy for chief investigator H. C. Jain (BARC, Trombay).
Charles W. Reich Nuclear Physics Branch E.G. & G. Idaho Inc. P.O. Box 1625 Idaho Falls, Idaho 83401 U.S.A.	Chief investigator (EG&G, Idaho).
J. J. Schmidt Head, IAEA Nuclear Data Section	Head, IAEA Nuclear Data Section.
R. Vaninbrouckx Central Bureau for Nuclear Measurements Steenweg naar Retie B-2440 Geel Belgium	Chief investigator (CBNM, Geel).

First Coordinated Research Meeting on the
Measurement of Transactinium Isotope Nuclear Data

Vienna, 20-21 April 1978

Adopted Agenda

1. Introduction of members of the coordinated research programme (CRP) on the measurement and evaluation of transactinium isotope nuclear decay data.
2. Current status and plans of TND decay data measurements.
3. Progress reports and forecasts for this year's activities by each group.
4. Role of IAEA/NDS: coordination and information dissemination. (Also questions of participants regarding procedural matters).
5. Coordination with the international nuclear structure and decay data effort (IAEA/NDS ↔ US/NDP), and publication of final report containing tabulation of the recommended TND decay data.
6. Collaboration between participating research groups:
 - a) Communication of all new measurement results of all members of the CRP
 - b) Procurement of isotope samples, and their exchange between CRP members
 - c) Exchange of personnel
 - d) Exchange of all other pertinent information
7. Discussion of the current status and updating of the "Current Status Table" (Revision of summary table and inclusion of all currently accepted data).
8. Preliminary consideration of the Agenda for the 1979 Advisory Group Meeting on TND.
9. Next meeting of this coordinated research group.

10. All participants
Send to NDS, as soon after the meeting as possible, a list including the quantity and purity of existing isotope samples for decay data measurements available at their own and other laboratories.
11. IAEA/NDS
Inquire whether the research groups at BAM(Berlin) and PTB(Braunschweig) in the Federal Republic of Germany would be interested in joining this coordinated effort.
12. Vaninbrouckx (BCM/N/Geel)
Find out about European interlab comparison of Pu-238 (or mixture of Pu isotopes) and communicate this information to the group through IAEA/NDS.
13. Fudge (AERE/Harwell)
Provide IAEA/NDS with the names of groups in developing countries which could eventually participate in this measurement programme.
14. IAEA/NDS
Alert developing countries to the possibility of their participation in this measurement programme, and investigate the possibility of IAEA fellowship support for training scientists from developing countries in laboratories of developed countries.
15. All participants
Send in to IAEA/NDS lists of measurements of I_y and I_x considered important, before the next meeting.
16. IAEA/NDS
Include table of branching ratios and spontaneous fission half-lives into the Minutes of this meeting.
17. Reich (EG&G/Idaho Falls)
Investigate in the US the best way to identify and channel nuclear data requests for safeguards purposes.

Coordinated Research Programme for the
Measurement of Transactinium Isotope Nuclear Decay Data

List of Participants in the Programme

CEC/Geel	Dr. R. Vaninbrouckx Central Bureau for Nuclear Measurements Steenweg naar Retie B-2440 Geel, Belgium
LMRI/France	Dr. J. Legrand Laboratoire de Metrologie des Rayonnement Ionisants Boite Postale No. 2 F-91190 Gif-sur-Yvette, France
BARC/India	Dr. H. C. Jain Radiochemistry Division Bhabha Atomic Research Centre Trombay, Bombay 400 085, India
JAERI/Japan	Dr. H. Umezawa Division of Chemistry JAERI Taokai-mura, Naka-gun Ibaraki-ken 319-11 Japan
Harwell/UK	Dr. A. J. Fudge Chemistry Division, V.E.C. Group Building 220 AERE Harwell, Didcot Oxfordshire OX11 0RA United Kingdom
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REPORT FROM THE CENTRAL BUREAU FOR NUCLEAR MEASUREMENTS, GEEL, BELGIUM

R. Vaninbrouck

1. Introduction

At CBNM only a small group of 4-5 people is generally part-time active in this field: about 2 man-year of real effort.

The activities cover:

a. Measurement of Data

Half-lives: ^{239}Pu , ^{241}Pu , ^{238}Pu

I_{IX} and I_{γ} (40-50 KeV band) and I_{α} (feeding the ~ 40 KeV level); I_{γ} and I_{α} combined will yield the total conversion coefficient α_{T} , when only few data are available; these experimental α_{T} values can then be compared with theoretical values.

I_{γ} of the 60 KeV transition in the decay of ^{241}Am .

b. Compilation/Evaluation of Data

A new EUR-Report, like the 1974 Report on half-lives of long lived actinides, is being prepared, but only for Pu isotopes.

Since 1975, a Status File on the ^{239}Pu is updated for every NEANDC meeting. The recommended value for ^{239}Pu since 1976 has been $(2.411 \pm 0.010) \cdot 10^4$ years.

2. Current Status, Progress and Plans

a. Measurement of the ^{239}Pu half-life

The measurements were performed using an NBS material (used for the fabrication of targets for NBS) - origin is ORNL - containing 99.99% ^{239}Pu . The Pu content of the samples was determined by mass spectrometric isotope dilution. The isotopic composition by mass spectrometric isotope analysis and by α - particle spectrometry for the ^{238}Pu (0.02% in activity). The α emission rate was determined by low geometry α counting and by liquid scintillation α counting. All measurements are finished. Only the "best" mean value from the results, lying between 2.105×10^5 years and 2.112×10^5 years, has still to be computed. The final accuracy (sum of random \pm SE + linear sum of systematic uncertainties) at the 99.73% confidence level will probably be of the order of $\pm 0.3\%$. The results will be published.

b. Measurement of the ^{238}Pu half-life

Measurements were planned at an earlier stage, but not performed because very good values became available in the meantime, e.g.

the ANL measurements, based on the in-growth of ^{238}Pu in pure ^{242}Cm samples. However, in view of the Russian result [Polyukhov et al, *Atomnaya Energiya*, 40, 61 (1976) and translation in *Sov. J. Atom. Energy*, 40, 66 (1976)] of (86.98 ± 0.39) years, based on specific α activity measurements, it could be of interest still to do one measurement by determining the specific activity. Such a measurement could be done at CBNM in the framework of the I_{CX} , $I_{\gamma 43}$, 48 and I_{α} determinations, without appreciable supplementary effort.

c. Measurement of the ^{241}Pu half-lives

The material used - originated from ORNL in 1968, (but probably irradiated in 1965 or 1966) contained 92.7 atom % ^{241}Pu beginning 1976 at the start of the CBNM measurements. The half-life was measured using the following methods:

- (1) Measurement of the ^{241}Pu decay by following the change in time, using mass spectrometers, the ratio $(^{241}\text{Pu}/^{240}\text{Pu})$ and the ratio of ratios $(^{241}\text{Pu}/^{240}\text{Pu})/(^{240}\text{Pu}/^{239}\text{Pu})$.

The mass spectrometric measurements are still in progress; the current preliminary result is:

$$(14.60 \pm 0.14)\text{years}$$

- (2) Measurement of the ^{241}Am in-growth by: 2.1 α low geometry, where corrections for the contribution to the count rate by all Pu isotopes, including the α branch of ^{241}Pu , have to be applied.

Measuring the in-growth via the 60 KEV $_{\gamma}$ line using SiLi-detectors, calibrated with ^{241}Am reference sources; here a correction has to be applied for the 60 KEV $_{\gamma}$ from the decay of ^{237}U .

The in-growth measurements are finished; the final result is:

$$(14.60 \pm 0.10)\text{years}$$

Also measured was the partial α half-life using the following methods:

- (1) α - particle spectrometry during the first days after separation, where the correction for tailing effects due to the in-growing ^{241}Am was still small,
- (2) via the 208 KEV $_{\gamma}$ in the decay of ^{237}U using calibrated Ge-detectors and taking for $I_{\gamma-208}$ a weighted mean from Cline (1972 and Gunnink (1976), and
- (3) via the 60 KEV $_{\gamma}$ line during the first days after the separation, where the contribution of the in-growing ^{241}Am can be calculated with sufficient accuracy.

The value obtained for the partial α half-life of ^{241}Pu is:

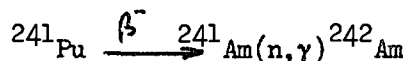
$$(6.04 \pm 0.06) \cdot 10^5 \text{ years}$$

JAERI'S PROGRAMME FOR SOURCE PREPARATION AND DECAY MEASUREMENT OF ^{242}Cm

Presented by T. Fuketa

1. Material

JFDR-I spent fuel specimens are used. The fuel had been irradiated for a long time; October 1963 - August 1969, and considerable accumulation of ^{242}Am can be expected.

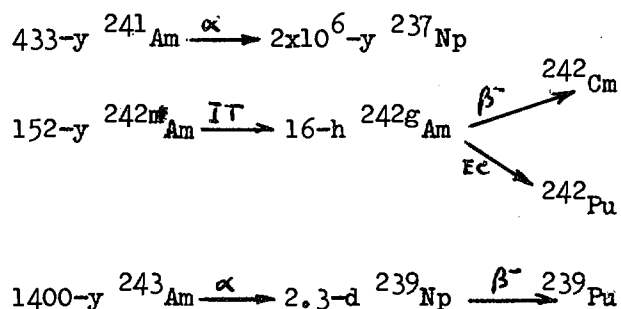


Analytical results are

$$^{152}\text{-y } ^{242}\text{Am}/\text{U} = 5 \times 10^{-6}$$

2. Separation of pure ^{242}Cm

After separating americium which is consisted of three isotopes, ^{241}Am , ^{242}Am and ^{243}Am , from the spent fuel, it is allowed to stand for 2 - 3 months. Then curium is separated from the americium which contains only ^{242}Cm as curium isotope.



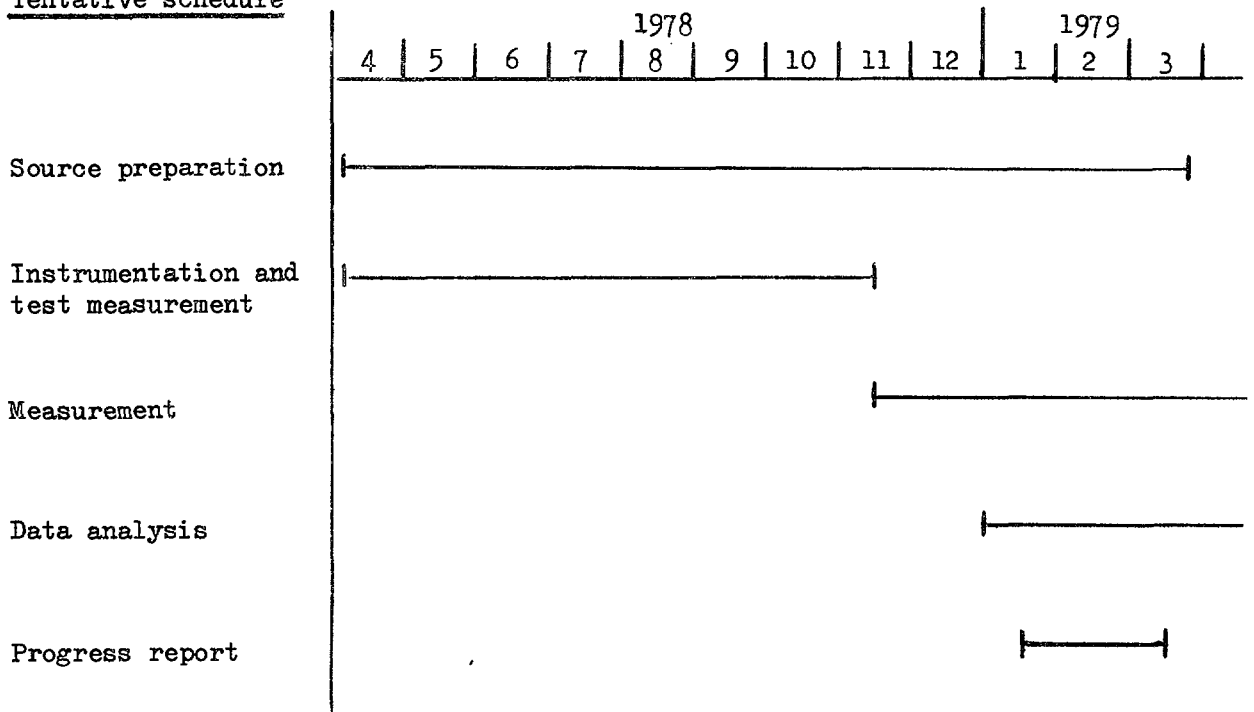
3. Source preparation

The separated pure ^{242}Cm is electrodeposited on a platinum plate with dimethyl sulphoxide as electrolyte. The techniques have been established for plutonium samples. Performance can be seen in an attached spectrum of a plutonium sample.

4. Measurement instruments

A new proportional counter has been ordered and is going to be equipped soon.

5. Tentative schedule



References: 1) H. Natsume, et al.: Jour. Nucl. Sci. Technol.
14 (1977) 745

"Gamma-Ray Spectrometry and Chemical Analysis
Data of JPDR-1 Spent Fuel"

2) S. Baba, et al.: JAERI 1211 (1971)

"Decay Analysis of Some Fission Product Nuclides
with Medium Half-lives"

STATUS REPORT ON THE U.S. PROGRAMME

C. W. Reich

UNITED STATES REPRESENTATION IN THE COORDINATED RESEARCH PROGRAMME

In the case of the United States participation in this CRP, Dr. C. W. Reich, of the Nuclear Physics Branch of EG & G Idaho Inc. at the Idaho National Engineering Laboratory, has been officially appointed by the US Department of Energy to serve as the representative of the combined US effort in this field. The US research groups which are active in this field and which are indirectly represented on this CRP are:

<u>Group</u>	<u>Measurement Area</u>
<u>Half-Life Evaluation Committee</u>	Half-lives
W. Strohm, Chairman (Mound Lab.)	"
A. Jaffey (Argonne National Lab.)	"
J. Rein (Los Alamos Scientific Lab.)	"
A. Prindle (Lawrence Livermore Lab.)	"
L. Lucas (National Bureau of Standards)	"
R. Carpenter (Rocky Flats)	"

Independent Groups .

I. Ahmad (Argonne National Lab.)	α-intensities
Nuclear Physics Branch (Idaho National Engineering Lab.)	γ-ray intensities
D. Hoppes (National Bureau of Standards)	(being considered)

Scope and Status

The Half-Life Evaluation Committee was formed several years ago to address the poor status of half-life data on the more important Pu isotopes. Their present plans call for the precise measurement of the half-lives of ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, using a variety of techniques. The work on ²³⁹Pu is now completed and will soon appear in the International Journal of Applied Radiation and Isotopes. In this work, a value of

$$T_{1/2}({}^{239}\text{Pu}) = 24,119 \pm 26 \text{ y}$$

is recommended. Work on the half-lives of ²⁴⁰Pu and ²⁴¹Pu is underway. Beyond these nuclides, the plans of the Committee are not definite. An interest has been expressed in measurements on other isotopes on the IAEA importance list, and the existence of the CRP has served to heighten this, but the Committee is not in a position to make definite commitments at present. The possibility of extending their efforts to include additional nuclides is under discussion, however.

The work at ANL will involve measurement of the α intensities identified in the IAEA list. The source material, in few- μg amounts, will be obtained from the ANL mass separator. Solid-state detectors will be used to measure the α spectra. It is expected that, for the more intense transitions, the accuracy requirements can be met.

At INEL, it is planned to measure absolute γ -ray intensities for the isotopes $^{238-242}\text{Pu}$, ^{233}Pa , ^{233}U , ^{235}U and ^{232}U . In addition, attention is being given to the problem of the ^{241}Pu half-life. Plans are underway to produce samples of ^{241}Pu from both thermal and fast-neutron irradiation of ^{240}Pu . Measurements on both of these samples, produced under known irradiation conditions, will be carried out in order to help resolve the presently puzzling half-life situation, particularly as regards the interesting possibility that there may be two distinct activities involved in the decay of " ^{241}Pu ". A measurement of the absolute intensity of the prominent 312-keV γ ray from the decay of ^{233}Pa has recently been completed. The value obtained is

$$I_{\gamma}(312 \text{ keV}) = 38.6 \pm 0.4 \text{ photons/100 decays,}$$

which is significantly larger than the values quoted in several recent evaluations. (This datum is required in the analysis of measurements of the ^{232}Th neutron capture cross section which employ activation techniques.)

An interest in the CRP has been expressed by personnel at NBS. They are currently investigating the possibility of such an involvement, as well as its extent and form.

Related Decay-Data Evaluation Activities

Several years ago, the scope of ENDF/B was expanded to include detailed decay data. INEL was assigned the responsibility for preparing evaluated sets of decay data for this file. As a part of this responsibility, we have prepared evaluated decay-data sets for all the nuclides included in the ENDF/B-V Actinide File, scheduled to be released in the near future. The nuclides to be included in this file are as follows:

Th - 228*, 230, 231, 232, 233
Pa - 231, 232, 233
U - 232, 233, 234, 235, 236, 237, 238, 239
Np - 236, 236m, 237, 238, 239
Pu - 236, 237, 238, 239, 240, 241, 242, 243, 244
Am - 240, 241, 242, 242m, 243, 244, 244m
Cm - 241, 242, 243, 244, 245, 246, 247, 248, 249
Bk - 249, 250
Cf - 249, 250, 251, 252, 253
Es - 253

*includes the members of this decay chain.

It is anticipated that our involvement in both the measurement and the evaluation of transactinium-isotope decay data, through the CRP and ENDF/B, respectively, will be beneficial to both programmes.

REPORT OF THE LABORATOIRE DE METROLOGIE DES RAYONNEMENTS IONIZANTS

J. Legrand and G. Malet

1. Measurement of the ^{241}Am Gamma-Ray Spectrum

The energies and intensities of the ^{241}Am gamma-rays have been determined in the energy region between 98.93 keV and 962 keV. The energy errors are equal to ± 0.2 keV for the gamma-rays having intensities greater than 10^{-8} , and to ± 0.5 keV for the gamma-rays of lesser intensities. The evaluation of errors of the absolute intensities were performed using the following two methods:

- 1) The uncertainty is assumed to be equal to the sum of the systematic and random errors combined quadratically for a statistical confidence level of 0.997.
- 2) The uncertainty is assumed to be equal to the quadratic sum of systematic and random errors for a statistical confidence level of 0.68.

The obtained results have been compared with those of R. Gunnink and J.E. Cline.

2. Measurement of the ^{239}Pu Gamma-Ray Spectrum

The energies and relative intensities of gamma-rays from a ^{239}Pu source, having an isotopic purity of 99.83%, have been measured in the energy region between 94.70 keV and 786.61 keV. Similarly to the ^{241}Am measurement, the energies were determined to accuracies of ± 0.2 and ± 0.5 respectively, and the relative intensities were compared to the R. Gunnink data by normalizing to the 129.29 keV peak.

The values of the absolute intensities are in the process of being analyzed.

3. Isotope Sources Available at LMRI

- ^{239}Pu - 145 mCi of 99.98% pure isotope in stainless steel container.
- ^{241}Am - 18 mCi of 97.21% pure isotope in stainless steel container.

REPORT TO FIRST COORDINATED RESEARCH MEETING ON

TRANSACTINIUM NUCLEAR DECAY DATA

A. J. Fudge

Introduction

Decay scheme work on Actinide Nuclides at Harwell has been fragmented and sporadic in time. The I.A.E.A. suggestion of a coordinated programme of research served to catalyse the moves towards an established coordinated programme. A project, called TANDEM (Trans Actinium Nuclear Decay, Evaluation and Measurement), has been set up to coordinate I_{α} , I_{γ} and $T_{1/2}$ measurements of actinide nuclides. Dr. A.J. Fudge (Chemistry Division) has been appointed coordinator.

Measurements

In order to make the measurement programme as efficient as possible simultaneous I_{α} , I_{γ} and $T_{1/2}$ measurements will be made on each nuclide. The following sequence of operation is proposed.

- (a) Procurement and purification
- (b) Characterisation of mass, isotopic composition, and impurities
- (c) Source preparation
- (d) Measurement
- (e) Data evaluation
- (f) Decay scheme evaluation.

The equipment available includes surface ionisation mass spectrometers, constant current and controlled potential coulometers, emission spectrographs, X-ray fluorescence spectrometers, low and medium fixed geometry alpha counters, α spectrometers (solid state and gridded ion chambers), Ge(Li) and Si(Li) gamma and X-ray spectrometers.

Material Availability

A wide range of high isotopic purity of many actinide nuclides exists at Harwell as well as expertise in purification, and characterisation of these nuclides. The order of measurement will depend on local allocations of priority for manpower and equipment.

Time Scales

The individual measurements would continue until completed. The coordinated programme would start in mid - 78 on a number of Np and U nuclides as newer equipment and facilities became available.

Evaluation

The measurements on $T_{1/2}$ for ^{241}Pu have failed to produce a consistent result from the different measurement techniques and source materials. This work will continue with freshly prepared source material from known irradiation facilities.

118 nuclides are being evaluated by Dr. A. Nichols (now at A.E.E. Winfrith) in ENDF/B5 format as part of the U.K. Chemical Nuclear Data File. About half will be completed by August 1978. M. James (also of A.E.E. Winfrith) is evaluating spontaneous fission data for actinide nuclides but on a longer time scale. The coordinator of the Data File sub-committee of the U.K. Chemical Nuclear Data File is B.S.J. Davies of C.E.G.B., Berkeley Nuclear Laboratory and the Chairman of the U.K. Chemical Nuclear Data Committee is J.G. Cuninghame, Chemistry Division, Harwell.

