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REPORT ON THE IAEA COORDINATED RESEARCH PROGRAMME ON THE MEASUREMENT AND EVALUATION OF TRANSACTINIUM ISOTOPE NUCLEAR DECAY DATA†

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ABSTRACT

As one result of the First IAEA Advisory Group Meeting on Transactinium Isotope Nuclear Data, held in November 1975 at Karlsruhe, an IAEA Coordinated Research Program was set up to address certain identified actinide-isotope decay-data needs in reactor technology. At present, laboratories from five nations are involved in this effort. In this paper, we give an overview of this program, including its origin and the present status of the measurements being carried out. The current status of the actinide-nuclide half-life, spontaneous-fission branching-ratio, α-intensity and γ-intensity data of concern to the Coordinated Research Program is presented and briefly discussed.

1. INTRODUCTION

At the first IAEA Advisory Group Meeting on Transactinium Isotope Nuclear Data [1], held in November, 1975, at Karlsruhe, one of the problem areas addressed was the status of the decay data (half-lives, α and γ

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intensities) for the transactinium ($Z \geq 90$) nuclides. It was pointed out that the accuracy of many of these data was not adequate to satisfy a number of needs in such areas of reactor technology as safeguards, fuel assay, sample-mass determination and standards preparation. At that meeting, a list of these important transactinium isotopes and the accuracy requirements for their decay data was drawn up. Further, it was recommended that an internationally coordinated research program of decay-data measurement and evaluation be initiated to meet these identified data needs. Subsequently, the IAEA Nuclear Data Section set up a Coordinated Research Program (CRP) on the measurement and evaluation of transactinium-isotope nuclear decay data, with groups from six nations agreeing to participate. The first meeting of the national representatives for this program was held in Vienna, April 20-21, 1978; and a summary report of this meeting was subsequently issued [2]. The second meeting of this group was held on the two days (April 30-May 1, 1979) immediately preceding this conference (the Second IAEA Advisory Group Meeting on TND).

In this paper, the present status and future plans of the Coordinated Research Program are discussed. In addition, a brief summary is given of the current status of those decay data with which this program is primarily concerned. Finally, attention is called to a recent precise measurement of the absolute intensities of the γ rays from the ^{233}Pa decay and to the implications which this result carries for certain aspects of nuclear decay-data evaluation.

2. THE COORDINATED RESEARCH PROGRAM: PARTICIPANTS AND PLANS

2.1 Participating Research Groups

At the first meeting of the national representatives for the Coordinated Research Program, the following participating laboratories were represented.

Laboratory (Nation)	National Representative
Central Bureau for Nuclear Measurements (Belgium)	R. Vaninbroukx
Laboratoire de Métrologie des Rayonnements Ionisants (France)	J. Legrand
Bhabha Atomic Research Center (India)	H. C. Jain
Japan Atomic Energy Research Institute (Japan)	H. Umezawa
Atomic Energy Research Establishment Harwell (United Kingdom)	A. J. Fudge
Idaho National Engineering Laboratory (United States of America)	C. W. Reich

Within the U.S., work at a number of laboratories is relevant to the objectives of the CRP. Absolute α-particle intensity measurements are being carried out at Argonne National Laboratory by I. Ahmad. At INEL, we are involved in the carrying out of absolute γ-ray intensity measurements. The Half-Life Evaluation Committee is involved in measurement and evaluation of half-life values for selected Pu isotopes. This latter group, representing individuals from six laboratories, was formed several years ago to address the then-current poor status of half-life data on the more common Pu isotopes. The following laboratories (and representatives) make up this Committee: Mound Laboratory (W. Strohm, Chairman); Argonne National Laboratory (A. Jaffey); Los Alamos Scientific Laboratory (J. E. Rein); Lawrence Livermore Laboratory (A. Prindle); National Bureau of Standards (L. Lucas); and Rocky Flats Laboratory (R. Carpenter).

2.2 Measurement Goals of the CRP

Prior to the first meeting of the participants in the Coordinated Research Program, the status of the measurement activities and the plans for future measurements at the participating laboratories were assessed. A summary of this information was prepared [3] and formed a basis for some of the discussions at that meeting. This summary, slightly modified, is presented in Table I. It reflects the presently defined overall measurement goals of the CRP and, as such, forms the basis for much of our subsequent discussion.

TABLE I. STATUS AND PLANS OF TND DECAY DATA MEASUREMENT.
(Adapted from A. Lorenz, private communication, April 1978)

Required Accuracy: listed are those supplied by Harwell, values recommended at the 1975 Karlsruhe meeting are given in parentheses if different.

Participating Laboratory: Measurement Status:
Plan = measurement is planned to be performed; parentheses imply that plans are not firm; (U) indicates urgency.
Prog = measurement in progress
Finished = measurement completed; parentheses mean that data are being processed.

Priorities: priorities given by each participating group are given as a number in parentheses to the right of measurement status.

Isotope and Quantity		Required Accuracy	Participating Laboratory (or organization)						
			INEL	$T_{\frac{1}{2}}$ Comm.	ANL	Harwell	CBNM	LMRI	JAERI
^{233}Pa	I_γ		Plan(1)						
^{233}U	I_γ		Plan(2)						
^{234}U	I_α	1%			Plan	Plan(1)			(3)
	I_γ	5%				Plan(2)			(3)
^{235}U	$T_{\frac{1}{2}}(\alpha)$	1%				Plan(2)			(3)
	I_α	1%			(Plan)	Plan(2)			(3)
	I_γ	1%	Plan(1)			Plan(1)			(3)
^{236}U	$T_{\frac{1}{2}}(\alpha)$	1%				Plan(2)			(3)
^{237}U	I_γ						Plan		
^{238}U	I_α	1%			(Plan)	Plan(2)			(3)
^{237}Np	I_α	1%			Plan	Plan(2)			(3)
^{238}Pu	$T_{\frac{1}{2}}(\alpha)$	0.5%[a]				Plan(1)	Prog.		(2)
	I_α	0.5%(0.1%)			Plan	Plan(1)			(2)
	I_γ	1%	Plan(1)			Plan(1)		Plan	(1)
	I_{LX}						Plan		
^{239}Pu	$T_{\frac{1}{2}}(\alpha)$	0.5%[a]		Prog.		Plan(1)	(finished)		(2)
	I_α	1%[a]			Plan	Plan(1)			
	I_γ	1%	Plan(1)			Plan(1)		finished	(1)
^{240}Pu	$T_{\frac{1}{2}}(\alpha)$	1%		Plan		Plan(1)	Prog.		(2)
	I_α	1%			Plan	Plan(1)			
	I_γ	1%(0.2%)	Plan(1)			Plan(1)		Plan	(1)

TABLE I. STATUS AND PLANS OF TND DECAY DATA MEASUREMENT (Cont.)

Isotope and Quantity	Required Accuracy	Participating Laboratory (or organization)						
		INEL	$T_{1/2}$ Comm.	ANL	Harwell	CBNM	LMRI	JAERI
^{241}Pu $T_{1/2}(\alpha)$	1%		Plan		Plan(U)	(finished)		(1)
I_γ	1%	Plan(1)			Plan(1)	Plan	Plan	(1)
^{242}Pu $T_{1/2}(\alpha)$	1%[a]		(Plan)		Plan(2)			(3)
I_α	4%			Plan	Plan(2)			(2)
^{241}Am $T_{1/2}(\alpha)$	1%				Plan(2)			(2)
I_γ	1%				Plan(1)	Plan	Finished	(2)
^{242}Cm $T_{1/2}(\alpha)$	0.5%(0.1%)[a]				Plan(1)			Plan(1)
$T_{1/2}$(S.F.)	1%(3%)				Plan(1)			Plan(1)
^{244}Cm $T_{1/2}$(S.F.)	2%(0.3%)[a]				Plan(1)			(1)
^{252}Cf $T_{1/2}(\alpha)$	0.5%(0.2%)				Plan(2)			(3)

a) Required accuracy achieved by a known recent measurement.

3. CURRENT STATUS OF DECAY DATA RELEVANT TO THE CRP

Before discussing the progress to date of the measurement activities related to the goals of the Coordinated Research Program, it is helpful to consider the present status of the decay data. Since the 1975 Karlsruhe meeting, several data evaluations relevant to the transactinium isotopes have appeared. The most comprehensive of these are the Nuclear Data Sheets [4] and the Seventh Edition of the Table of Isotopes [5]. In addition, an Evaluated Nuclear Structure Data File, ENSDF, based on the data evaluations contained in the Nuclear Data Sheets, is being produced by the Nuclear Data Project at ORNL and updated on a periodic basis. A description of this file, in the context of Transactinium Isotope Nuclear Data, is being given as a Review Paper [6] presented at this meeting.

3.1 Half-life data

In Table II, we summarize the status of the half-life data, as contained in several recent evaluations, on those nuclides that were discussed at the first meeting of the participants in the Coordinated Research Program. The data in the third column of Table II are those in the most recent version of ENSDF, dated 1 October 1978, as summarized in Ref. [7]. The values associated with the Table of Isotopes are those "adopted" for the Seventh Edition [5]. No uncertainties have been associated with these values since the authors of Ref. [5] have chosen not to quote "adopted" uncertainties for them. Rather, it is intended that the precision quoted for a given value convey an estimate of its associated uncertainty [9]. For example, the listed half-life for ^{238}Np, 2.117 d, indicates that the associated uncertainty is to be taken to be < 0.005 d; if the uncertainty to be associated with this value were > 0.005 d (but less than 0.05 d), then the value would have been written as 2.12 d. The ENDF/B values, column 5, are those prepared at INEL [10] for inclusion in the Actinide File of ENDF/B-V. These data were, in general, prepared prior to the appearance the other two evaluations and in some cases (e.g., ^{237}Pu and ^{240}Pu) do not incorporate measured values which were published subsequently. The values listed in the sixth column of Table II

TABLE II. SUMMARY OF HALF-LIFE VALUES OF SELECTED TRANSACTINIUM ISOTOPES. Quantities in parentheses represent uncertainties in the least significant figure (or figures) of the associated value.

Nuclide	Half-Life Units	ENSDF[a]	Table of Isotopes	ENDF/B	CRP-I
Th-228	y	1.9131(9)	1.9131	1.91313(88)	1.913(3)
230	10^4y	7.7(3)	8.0	7.7(3)	7.7(3)
232	10^{10}y	1.405(6)	1.41	1.405(6)	1.405(6)
Pa-231	10^4y	3.276(11)	3.28	3.276(11)	3.276(11)
232	d	1.31(2)	1.31	1.31(2)	1.31(2)
233	d	27.0(1)	27.0	27.0(1)	27.0(1)
U-232	y	72.(2)	72.	71.7(9)	72.(1)
233	10^5y	1.592(2)[b]	1.592	1.5918(15)[c]	1.592(2)
234	10^5y	2.445(10)	2.45	2.446(7)[c]	2.446(7)
235	10^8y	7.038(5)	7.038	7.038(5)	7.038(7)
236	10^7y	2.3416(39)	2.342	2.3415(14)	2.342(4)
237	d	6.75(1)	6.75	6.75 (1)	6.75(1)
238	10^9y	4.468(3)	4.468	4.4683(24)	4.468(4)
239	m	23.50(5)	23.5	23.50(5)	23.50(5)
Np-236[d]	10^5y	1.15(12)	1.1	1.15(12)	1.15(12)
236m[e]	h	22.5(4)	22.5	22.5(4)	22.5(4)
237[e]	10^6y	2.14(1)	2.14	2.14(1)	2.14(1)
238[e]	d	2.117(2)	2.117	2.117(2)	2.117(2)
239	d	2.355(4)	2.35	2.354(6)	2.354(6)
Pu-236[e]	y	2.851(8)	2.85	2.851(8)	2.851(8)
237	d	45.3(2)	45.4	45.63(20)	45.6(2)
238	y	87.74(4)	87.74	87.75(5)	87.74(9)
239	10^4y	2.411(10)	2.41	2.411(10)	2.411(3)
240	10^3y	6.537(10)	6.57	6.55(7)[c]	6.553(8)
241	y	14.4(2)	14.4	14.7(4)	14.7(4)
242	10^5y	3.763(20)	3.76	3.763(20)	3.76(2)
244	10^7y	8.26(9)	8.1	8.2(1)[c]	8.2(1)
Am-241	y	432.2(5)	433.	432.2(2)	432.6(6)
242	h	16.02(2)	16.01	16.01(2)	16.01(2)
242m[e]	y	152.(7)	152.	152.(7)	152.(7)
243	10^3y	7.380(40)	7.37	7.380(40)	7.38(4)
Cm-242	d	162.8(4)	162.8	162.9(3)	162.8(4)
244	y	18.11(2)	18.11	18.11(1)	18.11(2)
Cf-252	y	2.638(10)	2.64	2.638(10)	2.64(1)

a) Data as summarized in Ref. [7].

b) Uncertainty given as 0.020 x 10^5y in Ref. [7].

c) Value taken from R. Vaninbroukx (Ref. [8]).

d) Only one measured value reported.

e) Listed value based essentially on only one measurement.

are taken from a proposed list of recommended values drawn up during the first meeting of the participants in the CRP [2]. They were derived largely from the INEL data base, represented by the ENDF/B values. However, in some cases the uncertainties have been increased, reflecting the consensus of the group that, in consideration of the experimental techniques involved, the total uncertainty ascribed should in no case be smaller than $\sim$ 0.1%.

Generally, the status of the half-life data is reasonably good. In most instances, the accuracy criteria established at the first TND meeting appear to be met. The outstanding exception at present is the important Pu isotope, ^{241}Pu. However, it is to be anticipated that the results of measurement programs currently underway (see below) will help to clarify this situation, as well as to provide increased accuracy and precision for the half-life values of ^{239}Pu and ^{240}Pu. As indicated in Table II, the half-life values for several nuclides are based on one, or essentially only one, measurement. For those applications where these data are of importance, it might be useful to re-measure these values to provide a check on their accuracy.

3.2 Spontaneous-fission branching ratios

The spontaneous-fission branching-ratio data for those transactinium isotopes considered at the first meeting of the participants in the CRP [2] are summarized in Table III. The spontaneous-fission branching ratios are not listed explicitly in the Table of Isotopes for most of these nuclides; only the S.-F. half-lives are given. Since no "adopted" values for these latter quantities are listed in Ref. [5], we have chosen not to derive branching ratios from these data. Since the data base for the three evaluations in Table III is essentially the same, values deduced from the Table of Isotopes information should not differ strikingly from those listed. A number of the S.-F. branching ratios are based on only one measurement and several result from two rather discrepant measurements. The spontaneous-fission half-life of ^{238}U has been extensively investigated, with roughly 40 measurements reported. This situation has

TABLE III. SUMMARY OF SPONTANEOUS-FISSION BRANCHING RATIOS FOR SELECTED TRANSACTINIUM ISOTOPES.
Quantities in parentheses represent uncertainties in the least significant figure (or figures) of the associated value.

Nuclide	S.-F. Branching Ratio (in %)[a]					
	ENSDF[b]		Table of Isotopes[c]		ENDF/B	
U-232[d]	1.	$\times 10^{-10}$			0.9(7)	$\times 10^{-10}$
-233[d]	1.3(4)	$\times 10^{-10}$			1.3(3)	$\times 10^{-10}$
-234[d]	1.2(6)	$\times 10^{-9}$			1.2(6)	$\times 10^{-9}$
-236[d]	≈1.2	$\times 10^{-7}$			1.2	$\times 10^{-7}$
-238	5.4(8)	$\times 10^{-5}$			5.45(6)	$\times 10^{-5}$
Pu-236[d]	8.1(23)	$\times 10^{-8}$			8.1(23)	$\times 10^{-8}$
-238	1.84(6)	$\times 10^{-7}$			1.84(5)	$\times 10^{-7}$
-239[d]	4.4	$\times 10^{-10}$			4.4	$\times 10^{-10}$
-240	4.95(20)	$\times 10^{-6}$			5.0(2)	$\times 10^{-6}$
-242	5.50(6)	$\times 10^{-4}$			5.50(6)	$\times 10^{-4}$
-244[e]	0.125(6)				0.125(6)	
Am-241[e]	3.77(8)	$\times 10^{-10}$			4.1(1)	$\times 10^{-10}$
-242m[d]	1.6(6)	$\times 10^{-8}$			1.6(6)	$\times 10^{-8}$
-243[e]	2.2(2)	$\times 10^{-8}$			2.2(2)	$\times 10^{-8}$
Cm-242	6.8(7)	$\times 10^{-6}$			6.8(6)	$\times 10^{-6}$
-244	1.347(2)	$\times 10^{-4}$			1.347(2)	$\times 10^{-4}$
-246	0.02614(5)				0.02614(5)	
-248	8.26(3)		8.26(3)		8.26(3)	
Bk-249	4.7(2)	$\times 10^{-8}$			4.60(25)	$\times 10^{-8}$
Cf-249	5.2(2)	$\times 10^{-7}$			5.02(10)	$\times 10^{-7}$
-250	0.077(3)				0.077(3)	
-252	3.092(8)		3.092(8)		3.092(8)	
Es-253	8.7(3)	$\times 10^{-6}$			8.7(3)	$\times 10^{-6}$

a) These values have generally been computed from the measured spontaneous-fission half-lives and total half-life values.

b) Values summarized in Ref. [7].

c) With the exceptions listed, the spontaneous-fission branching ratios are not explicitly given in this reference. See the discussion in the text.

d) Only one measured value is reported.

e) Two discrepant values are reported. For ^{241}Am, several measurements are reported.

been carefully evaluated by Apt [11]; and he adopts the value $\lambda_{S.F.} = 8.46 \times 10^{-17} y^{-1}$, with an estimated uncertainty of $\sim$ 1%. This is the value from which the ^{238}U S.-F. branching ratio included in ENDF/B was derived. (Incidentally, Apt's adopted value is essentially identical to that measured by Galliker et al. [12].) The two sets of evaluated S.-F. branching-ratio data listed in Table III are in generally good agreement, except possibly for ^{241}Am, where different evaluation criteria were adopted.

3.3 Absolute α-transition intensities

The absolute α-transition intensity data for selected "important" transactinium isotopes, as summarized in a number of data compilations, are given in Table IV. We have followed the usual convention, labelling the individual α transitions by the energy of the daughter-nucleus state which is directly fed by the transition. We have also, for convenience, given the initial- and final-state spin-parity (J^{π}) values. Generally, only the more intense α transitions, those subject to direct experimental measurement, are included.

Since the appearance in 1973 of the major evaluation of α-transition data by Rytz[13], a number of α-intensity measurements have been published. Consequently, in a number of cases the α-intensity values in this reference have been superseded. It still remains authoritative, however, as regards α-transition energies. The intensity values of Baranov et al. [15] are those given in their review paper, presented at the Karlsruhe meeting on TND [1].

The agreement among all the evaluations (except that of Ref. [15]) for the α-intensity data for ^{238}U and ^{237}Np results from the fact that they are all based on the results of a single measurement for each nuclide. A number of differences in the listed I_{α} values for other isotopes is apparent from inspection of Table IV. Of some interest, although not perhaps of overriding importance from a practical point of view, are the differences in the reported intensities of the α transitions feeding the 4^{+} members of

TABLE IV. ABSOLUTE-INTENSITY DATA FOR SELECTED α-TRANSITIONS FROM TRANSACTINIUM ISOTOPES OF INTEREST TO THE CRP. Quantities in parentheses represent uncertainties in the least significant figure (or figures) of the associated values.

Nuclide and J^π	α-group[a], and final-state J^π	I_α (% per decay)					
		Rytz [13]	Nuclear Data Sheets	Table of Isotopes	ENDF/B	Rogers [14]	Baranov et al. [15]
^{234}U 0^+	α_0 , 0^+	72.(2)	72.5(20)	72.	72.5(30)	72.(2)	
	α_{53} , 2^+	28.(2)	27.5(15)	28.	27.5(15)	28.(2)	
	α_{174}, 4^+	-	0.24(3)[b]	0.3	0.3	0.3	
^{235}U $7/2^-$	α_0 , $5/2^+$	4.	5.0(5)	4.6	5.4(5)	1.2	
	α_{42} , $7/2^+$	-	4.2(3)	3.7	4.5(5)	1.7	
	α_{205}, $7/2^-$	56.	55.(3)	57.	56.(3)	53.	
	α_{237}, $9/2^-$	18.	~17. [c]	18.	17.(2)	12.3	
	α_{278}, $11/2^-$	-	4.6(5)	3.	4.7(5)	3.5	
	α_{388}, $7/2^-$	6.	5.7(6)	5.7	5.7(6)	6.2	
^{238}U 0^+	α_0 , 0^+	77.(4)	77.(4)	77.(4)	77.(4)	77.(4)	
	α_{50} , 2^+	23.(4)	23.(4)	23.(4)	23.(4)	23.(4)	
	α_{143}, 4^+	-	0.23(7)	0.23(7)	0.23(7)	0.23(7)	
^{237}Np $5/2^+$	α_0 , $3/2^-$	2.6(2)	2.6(2)	2.6(2)	2.6(2)	2.6(2)	-
	α_{57} , $7/2^-$	-	2.5(4)	2.5(4)	2.5(4)	2.5(4)	-
	α_{86} , $5/2^+$	47.(9)	47.(9)	47.	47.(9)	47.(9)	51.3(8)
	α_{104}, $7/2^+$	25.(6)	25.(6)	25.	25.(6)	25.(6)	19.4(4)
	α_{109}, $9/2^+$	8.(3)	8.(3)	8.	8.(3)	8.(3)	16.8(4)
	α_{238}, $5/2^+$	6.2(1)	6.18(12)	6.18(12)	6.18(12)	6.18(12)	-
^{238}Pu 0^+	α_0 , 0^+	71.1(12)	71.6(6)	72.	71.1(12)	71.1(12)	72.13(6)
	α_{43} , 2^+	28.7(12)	28.3(6)	28.	28.7(12)	28.7(12)	27.87(3)
	α_{143}, 4^+	-	0.10(3)	0.11	0.13(1)	0.068(5)	-
^{239}Pu $1/2^+$	$\alpha_{0.07}$, $1/2^+$	73.3	73.3(7)	73.	73.3(7)	73.3	73.3(7)
	α_{13} , $3/2^+$	15.1	15.1(2)	15.1	15.1(2)	15.1	15.1(2)
	α_{52} , $5/2^+$	11.5	11.5(2)	11.5	11.5(2)	11.5	11.5(2)

TABLE IV. ABSOLUTE-INTENSITY DATA FOR SELECTED α-TRANSITIONS FROM TRANSACTINIUM ISOTOPES OF INTEREST TO THE CRP. (cont'd)

Nuclide and J^π	α-group[a], and final-state J^π	I_α (% per decay) Rytz [13]	Nuclear Data Sheets	Table of Isotopes	ENDF/B	Rogers [14]	Baranov et al. [15]
^{240}Pu 0^+	α_0 , 0^+	76.	73.4(8)	73.3	73.4(8)	76.	73.4(8)
	α_{45} , 2^+	24.	26.5(4)	26.6	26.5(4)	23.	26.5(4)
	α_{149}, 4^+	-	0.091(6)	0.084	0.091(6)	0.09	-
^{242}Pu 0^+	α_0 , 0^+	77.	77.5(30)	74.	77.5(30)	77.	79.7(27)
	α_{45} , 2^+	23.	22.4(20)	26.	22.4(20)	23.	20.2(11)
	α_{148}, 4^+	-	0.098(17)	0.11	0.098(17)	-	-

a) The subscript gives the energy (in keV) of the state in the daughter nucleus to which the α transition proceeds.

b) Intensity value inferred from level scheme.

c) Two α groups are presumed to lie within this peak. The listed value is the sum of the two intensities.

the ground-state rotational bands in the doubly even nuclei. In some cases (e.g., ^{234}U) these differences result from the fact that the intensity values for these transitions deduced from level-scheme intensity-balance considerations are not the same as those determined experimentally. Since these former values are based on γ-ray intensity measurements (together with the associated internal-conversion coefficients) and since both absolute I_α and I_γ measurements will be carried out under the CRP for several of these nuclides, it is to be hoped that one of the results of this Program will be a resolution of these differences.

3.4 Absolute photon-intensity values

Table V presents a summary of absolute-intensity values for selected prominent γ-ray transitions from the decay of transactinium isotopes to be investigated in the CRP. The evaluation of Kocher [16] is derived largely from data contained within the Nuclear Data Sheets and ENSDF; and because of intensity-limit considerations adopted in the computer programs employed to produce the data listings in Ref. [16], no intensity values are given for the γ-ray transitions for a number of these isotopes. The values of Gunnink et al. [17] represent an upgrading of data presented in an earlier report [18]. Where available, the data of Ref. [17] are those incorporated into the decay data in ENDF/B. The errors given in Ref. [17] are generally those associated with the γ-ray peak fitting alone and, for this reason, are not reproduced in Table V. The errors given for the "Gunnink et al." intensity values in ENDF/B (see Column 7, Table V) represent estimates by the compilers of that data file, based on the listed "peak-fitting" errors [17] and estimates of the other errors, in consultation with Gunnink.

In several instances, the absolute γ-ray intensities are based on intensity-balance considerations in the adopted decay schemes. This procedure is generally rather sensitive to the details of these schemes and, as is evident, can lead to significant differences in the inferred intensity values. The I_γ value listed in ENDF/B for the 311.9-keV γ ray

TABLE V. ABSOLUTE-INTENSITY VALUES OF SELECTED γ-RAY TRANSITIONS FROM TRANSACTINIUM ISOTOPES OF INTEREST TO THE CRP. Quantities in parentheses represent uncertainties in the least significant figure (or figures) of the associated values.

Nuclide	E_γ(keV)	I_γ (photons/100 decays)				
		Nuclear Data Sheets	Table of Isotopes	Kocher [16]	Gunnink [17]	ENDF/B
^{233}Pa	311.9	36.(2)[a]	37.(2)[a]	33.7[a]		38.6(5)
^{233}U	42.4	0.062(9)	0.062(9)			0.062(8)
	54.6	0.015(2)	0.015(2)			0.015(2)
	97.1	0.022(3)	0.022(3)			0.022(3)
	317.2	0.008(1)	0.008(1)			0.008(1)
^{234}U	53.2[a]	0.119(10)	0.119(10)	0.118(10)		0.12(1)
	120.9[b]	0.041(4)	0.041(4)			0.041(6)
^{235}U	185.7	54.	54.	54.		54.1(19)
^{237}U	208.0	21.7(23)[a]	23.[a]	23.3[a]	21.7	21.7(3)
^{239}Np	228.2	10.7(7)	10.7(7)	10.7(7)		11.3(2)
	277.6	14.1(4)	14.1(4)	14.1(4)		14.3(2)
^{238}Pu	43.5	0.0390(5)	0.0394(11)[a]		0.0393	0.0393(5)
	99.8	0.00724(20)	[c]		0.00724	0.00724(10)
	152.7	0.00101(20)	[c]		0.000956	0.000956(15
^{239}Pu	51.6	0.0208(6)	0.0208(2)		0.0270	0.0270(4)
	129.3	0.00620(20)	0.00620(6)		0.00626	0.00626(9)
	413.7	0.00151(5)	0.00151(2)		0.00149	0.00149(2)
^{240}Pu	45.2	0.0450(5)	0.0450(5)		0.0453	0.0453(6)
	104.2	0.0070(1)	0.00700(7)		0.00698	0.00698(10)
	160.3	0.000420(4)	0.000420(4)		0.000402	0.000402(7)
^{241}Pu[d]	103.7	1.01(1)	1.03(6)		1.01	1.01(2)
	148.6	1.86(2)	1.9(1)		1.87	1.87(1)
^{241}Am	26.3	2.4(1)	2.4(1)	2.58(22)	2.45	2.45(3)
	59.5	35.9(6)	35.7(5)	36.3(4)	35.9	35.9(4)

a) Intensity values deduced from level scheme

b) Intensity deduced from measured $I_\gamma(120)/I_\gamma(53)$ ratio.

c) Only relative I_γ values listed. These are consistent with the other listed I_γ values

d) These I_γ values are based on a value of 0.00245% for the intensity of the α-decay branch of ^{241}Pu. The listed I_γ values must all be multiplied by a factor of 10^4 in order to represent photons/100 decays.

from ^{233}Pa is the result of a recent measurement at INEL [19] and will be discussed later (see Section 5 below). Similarly, the ENDF/B value for the intensity of the 185.7-keV γ ray from ^{235}U decay is that supplied by Bemis [20]. Previously, the only reported value for this quantity was 54.%, reported in 1957 with no quoted uncertainty [21].

The determination of the isotopic composition of Pu samples has important applications for fuel reprocessing and for the safeguards and waste-management aspects of the nuclear fuel cycle, as well as for the physics of reactors operating under different spectrum and fuel-burnup conditions. Since Ge(Li)-based γ-ray spectrometry provides a non-destructive, relatively convenient and flexible means of measuring these isotopic concentrations, considerable attention has been focused on obtaining values for the absolute intensities of the γ rays emitted from the important Pu isotopes ($238 \leq A \leq 241$). A recent summary of these data, as used by various groups, has been given by Banham [22]. In Table VI we present a somewhat modified version of these data as listed in Ref. [22]. The latest I_γ data of Gunnink et al. [17] were not available to Banham; we have included them in Table VI partly to trace the evolution of these values from 1971 [18] to 1976 [17] and partly to illustrate how they compare with those used by the other groups. Significant differences in the three data sets ([17], [22] and [23]), particularly for the even-A Pu isotopes, are observed.

Interest in L-x-ray intensity data was voiced at the first meeting of the participants in the CRP [2]. In Table VII, we summarize the results of a recent measurement [24] of the L-x-ray spectra for a number of trans-actinium isotopes. Also included in the table are the results of a determination [25] of the intensities of the prominent L-x-ray lines from the decay of ^{238}Pu and those for the ^{241}Am decay, measured several years ago by Campbell and McNelles [26]. The measured values reported in [24] and [25] represent absolute determinations. However, the intensity standards utilized to determine the absolute detection efficiencies in these two studies are those of Campbell and McNelles and, consequently, the reported intensity data are not completely independent of the ^{241}Am L-x-ray intensity data measured by these authors [26].

TABLE VI. COMPARISON OF ABSOLUTE INTENSITIES OF GAMMA RAYS FROM PU ISOTOPES, AS LISTED IN VARIOUS REFERENCES. The table is adapted from that given in Ref. [22].

Isotope	E_γ(keV)	Absolute Intensity (photons/10^6 decays)			
		Parker [23]	Gunnink [18]	Gunnink [17]	Banham [22]
238	152.8	10.1	10.1	9.56	9.5
239	129.3	62.	62.	62.6	62.
	144.2	3.25	2.86	2.83	2.96
	161.5	1.25	1.3	1.20	1.24
	171.4	1.13	1.09	1.105	1.11
	195.7	1.07	1.07	1.064	1.09
	203.5	5.6	5.6	5.60	5.70
	345.0	5.61	5.61	5.592	5.96
	375.0	15.8	15.8	15.70	16.4
	413.0	15.	15.1	14.89	15.1
240	160.3	4.2	4.2	4.02	4.2
241	148.6	1.9	1.9	1.87	1.88
	164.6	0.45	0.45	0.453	0.46
	208.0	5.12	5.12	5.34	5.6
	267.5	0.177	0.177	0.182	0.19
	332.4	0.280	0.280	0.298	0.30
^{241}Am	125.3	39.5	39.5	40.8	39.5
	146.6	4.58	4.58	4.61	4.58
	208.0	7.6	7.6	7.91	7.6
	332.3	1.45	1.45	1.490	1.45
	335.4	4.7	4.7	4.960	4.7

TABLE VII. MEASURED INTENSITIES OF L-X-RAYS FROM TRANSACTINIUM ISOTOPES. Unless otherwise indicated, the data are those of Bemis and Tubbs (Ref. [24]). Quantities in parentheses represent uncertainties in the least significant figure (or figures) of the associated value.

Nuclide	L-X-ray intensity (in photons/100 decays).				
	L_ℓ	L_α	L_β	L_γ	Total L
^{234}U	0.22(1)	3.66(8)	4.87(10)	1.05(4)	9.81(13)
^{235}U [a]	2.01(10)	45.4(20)	94.3(42)	15.2(7)	157.(4)
^{237}Pu	1.06(5)	16.3(4)	17.0(5)	3.83(20)	38.2(7)
^{238}Pu	0.26(1)	4.15(7)	5.61(7)	1.36(2)	11.38(10)
^{238}Pu [b]	-	5.05(6)	7.41(9)	1.48(2)	-
^{239}Pu	0.113(5)	1.82(4)	2.16(4)	0.53(1)	4.63(6)
^{240}Pu	0.24(1)	3.78(6)	4.84(7)	1.20(3)	10.06(10)
^{242}Pu	0.21(2)	3.10(8)	4.15(10)	1.08(4)	8.54(14)
^{241}Am [c]	0.86(3)	13.20(35)	19.25(60)	4.85(20)	38.2(7)
^{243}Am [d]	1.8(1)	27.5(6)	30.6(8)	7.69(21)	67.6(10)
^{244}Cm	0.25(1)	3.86(7)	4.30(7)	1.03(2)	9.44(10)
^{245}Cm	3.2(2)	49.8(22)	47.6(21)	12.5(6)	113.1(31)
^{246}Cm	0.21(1)	3.33(7)	3.71(7)	0.86(2)	8.11(10)
^{250}Cf	0.21(1)	3.27(8)	3.85(8)	0.85(3)	8.18(12)
^{252}Cf	0.23(2)	3.09(28)	3.80(34)	0.93(8)	8.05(45)

a) Includes Pa L-x-rays from the decay of the ^{231}Th daughter.

b) Values reported by Vasilik and Martin, Ref. [25].

c) Values reported by Campbell and McNelles, Ref. [26].

d) Includes Pu L-x-rays from the decay of the ^{239}Np daughter.

3.5 Recent measurements

Several recent measurements, not yet generally incorporated into the evaluations, of interest to the work of the CRP should be noted. The first formal publication of the work of the Half-Life Evaluation Committee has recently appeared [27]. It describes the comprehensive effort undertaken by that group to provide an accurate value for the half-life of ^{239}Pu. Their recommended value for this quantity is

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

A measurement of the half-life of ^{240}Pu has recently been published [28]. The value quoted by the authors for this quantity is

$$T_{\frac{1}{2}}\ (^{240}\text{Pu}) = 6569 \pm 6\ \text{y}.$$

The α-particle spectra of ^{246}Cf, ^{248}Cm and ^{240}Pu have been remeasured [29]. For ^{240}Pu, these authors report for the intensities of the α transitions to the 0-, 45- and 149-keV levels in ^{236}U the values

$$I(\alpha_0) = 73.51(36)\%$$
$$I(\alpha_{45}) = 26.39(21)\%$$
and $$I(\alpha_{149}) = 0.071(1)\%$$

The absolute-intensity values for the prominent γ rays from the decays of ^{233}Pa and ^{235}U have been mentioned in Sect. 3.4 above and listed in Table V.

4. STATUS OF CURRENT MEASUREMENTS BEING PERFORMED IN THE CONTEXT OF THE CRP

In this section, we present brief summaries of the measurement activities being conducted at the participating laboratories which are related to the objectives of the CRP. These summaries have been taken from material kindly supplied by the representatives of these participating laboratories.

4.1 CBNM

The present effort largely involves measurements of the half-lives of ^{239}Pu and ^{241}Pu.

The determination of the half-life of ^{239}Pu has been finished. During the reporting period the disintegration rates of five samples of a Pu material containing 99.98 atom % ^{239}Pu have been determined by counting α particles in a defined solid angle of low geometry. Corrections for the contribution of ^{238}Pu and ^{240}Pu have been allowed for. The first was determined by α-particle spectrometry and the latter calculated from the isotopic composition of the sample which was deduced from mass-spectrometric measurements. Finally, the specific α-emission rate of ^{239}Pu and its half-life have been calculated.

In a following step the α-emission rate of samples of the same Pu material, but after spiking with ^{242}Pu for the determination of the Pu content by mass-spectrometric isotope dilution techniques, were determined by liquid scintillation counting. Corrections for the contribution of ^{238}Pu, ^{240}Pu, and the spike material ^{242}Pu to the count rates were applied and again the specific α-emission rate and the half-life of ^{239}Pu were deduced.

The results of both series of measurements are given in Table VIII in which the uncertainties quoted are at the 1σ level taking into account random and systematic effects.

TABLE VIII

SPECIFIC α-EMISSION RATE AND HALF-LIFE OF ^{239}Pu

Method	Specific α-emission $s^{-1}/\mu g$ ^{239}Pu	Half-life years
Low geometry	2298 ± 3	$(2.4085 \pm 0.0030)10^4$
Liquid Scintillation	2295 ± 3	$(2.4114 \pm 0.0030)10^4$
Mean	2296 ± 3	$(2.4100 \pm 0.0030)10^4$

In an attempt to resolve the existing discrepancy of several percent between the values reported for the half-life of ^{241}Pu, a new determination was performed. The half-life was determined by following the change in time of the Pu isotopic composition by mass spectrometry and by measuring the ^{241}Am ingrowth using α- and γ-counting techniques. As a sideproduct the partial α half-life was determined. The following results were obtained: $T_{\frac{1}{2}}$ = (14.30 ± 0.14)y and (14.60 ± 0.10)y for the mass spectometric and ingrowth method, respectively, and $T_{\frac{1}{2}}$ (α) = (6.04 ± 0.06)10^5y.

4.2 Harwell

Measurements of the half-life and absolute α- and γ-ray intensities for ^{237}Np have been initiated. These measurements will be followed by studies of the uranium isotopes.

4.3 LMRI

The measurement of the energies and absolute intensities of the γ rays in the energy range from 20 keV to 60 keV from the decay of ^{241}Am has been completed. In addition,the energies and absolute intensities of the γ rays from the ^{239}Pu decay have been measured.

Measurements of the energies and absolute intensities of γ rays from the decay of ^{238}Pu and ^{240}Pu are in progress and are expected to be completed later on this year.

4.4 JAERI

The measurement program has had as its emphasis an accurate half-life determination for ^{242}Cm. The ^{242}Cm can be very purely prepared by means of milking decay products from 152-y ^{242m}Am. In the present work, americium was extracted from a plutonium bearing fuel specimen and purified for curium and other actinides. Curium-242 was separated from the americium after allowing it to stand for a several-month period. Several samples of ^{242}Cm were prepared for measurement, and alpha activities have been

measured with a proportional counter and a silicon surface barrier detector. Measurements of spontaneous fissions are also being studied at present.

Although ^{241}Am and ^{243}Am coexisted with ^{242m}Am, which is the ancestor of questioned nuclide, ^{242}Cm has grown in as the only curium nuclide in the americium and could be extracted pure by performing the same chemical treatment as the purification of the americium, after allowing it to stand for an appropriate period for the growth of ^{242}Cm. Six samples were prepared for the half-life measurement by depositing a drop of the hydrochloric solution of purified ^{242}Cm on a platinum plate of 24mm diameter and 0.2mm thick. Alpha activities of those samples are being measured with a windowless proportional counter. Decay of the alpha activity has been followed for 4 months. The efficiency of the counter has slowly changed within a one or two percent range through the whole period of the measurements. The deviation was estimated by measuring a reference sample of ^{238}Pu.

The results obtained so far from the decay measurements are summarized in Table IX. The measurements will be continued further to obtain more accurate results.

TABLE IX
RESULTS OF HALF-LIFE MEASUREMENTS ON ^{242}Cm SAMPLES

Sample No	Half-life (days)
1	164.79
2	163.41
3	163.94
4	160.66
5	164.74
6	162.12
Mean	163.28
Standard deviation	1.62

4.5 Measurement within the U.S.

4.5.1 Half-Life Evaluation Committee.

The objectives of this committee, consisting of participants from six laboratories (see Sect. 2.1 above), are the measurement of accurate half-life values for ^{239}Pu, ^{240}Pu and ^{241}Pu. The first phase of this work, the measurement of the ^{239}Pu half-life, has now been completed. The results have been published as a collection of papers in the August, 1978 issue of The International Journal of Applied Radiation and Isotopes [27]. The value of the ^{239}Pu half-life recommended from this work is 24,119 ± 26 y (as indicated in Sect. 3.5 above).

The members of the committee are currently measuring the half-lives of ^{240}Pu and ^{241}Pu. The measurement procedures for ^{240}Pu are similar to those employed for ^{239}Pu [27]. The sample material has been acquired and has been distributed to the participating laboratories for characterization, and measurements have gotten under way. The ^{241}Pu half-life measurement is of a more limited scope, involving only a mass-spectrometric technique. This investigation was undertaken earlier than the ^{240}Pu study but, because of the nature of the measurement, will not be completed until some time after the ^{240}Pu work is finished. At the present time, the early results from this study suggest a value of $\sim$14.4 y for the ^{241}Pu half-life, in reasonable agreement with independent measurements at the U.S. National Bureau of Standards and at the CBNM in Geel, Belgium (See Sect. 4.1 above).

4.5.2 ANL.

Absolute α-intensity measurements will be getting under way this Spring. The first isotopes to be studied will be $^{238,239,240,242}Pu$. Samples, containing a nominal few-μg amounts of Pu, will be prepared by isotope separation. A small (25mm^2 area) Si surface-barrier counter, with an energy resolution of $\sim$12 keV, will be used to count the α particles. The counting geometry will be at 1% or lower; and measurements will be taken at several geometries, to assess the effects of electron-α summing.

4.5.3 INEL

The INEL measurement [19] of the absolute γ-ray intensities from the β^- decay of ^{233}Pa has been completed and the results accepted for publication. 4π β-γ coincidence techniques were utilized to determine the absolute disintegration rates of the ^{233}Pa sources (obtained from milking an 0.5-g sample of ^{237}Np). The value obtained for the absolute intensity of the prominent 312-keV γ ray was 38.6 ± 0.5 photons/100 decays (see Table V and Sects. 3.4 and 5.).

The next nuclides for which absolute I_γ measurements will be made are ^{239}Pu and ^{240}Pu. High-purity samples of ^{239}Pu (99.995% in mass 239) and ^{240}Pu (≤1% ^{238}Pu by α activity) have been acquired for these measurments.

To permit absolute-intensity measurements of γ rays from actinide samples to be made with precisions of 1% or better, careful attention must be given to all aspects of the measurement process. The improvement of our techniques of precision γ-ray spectrometry to make possible measurements with this required precision has gotten underway this year. An early emphasis of this activity is a careful study of the shapes of full-energy γ-ray peaks observed in spectra measured using Ge-based spectrometers. This has as its object the development of a method of reliably and consistently determining the number of events contained in these peaks. To do this requires a means of treating the effects of "tailing" in the peaks and accounting for the spectral distribution underlying the peaks in a reproducible manner. Typical peak shapes observed in spectra acquired using two different Ge-semiconductor spectrometers are shown in Figs. 1 and 2. Inspection of these figures reveals that the contribution of tailing (as defined in the Figures) can be 1-2% of the peak area and extends over a large number of channels. If peak-area determinations to a precision of 1% or so are desired, these effects can be accounted for fairly simply. However, if the overall intensity data are desired to a precision of <1%, the contribution of the uncertainty

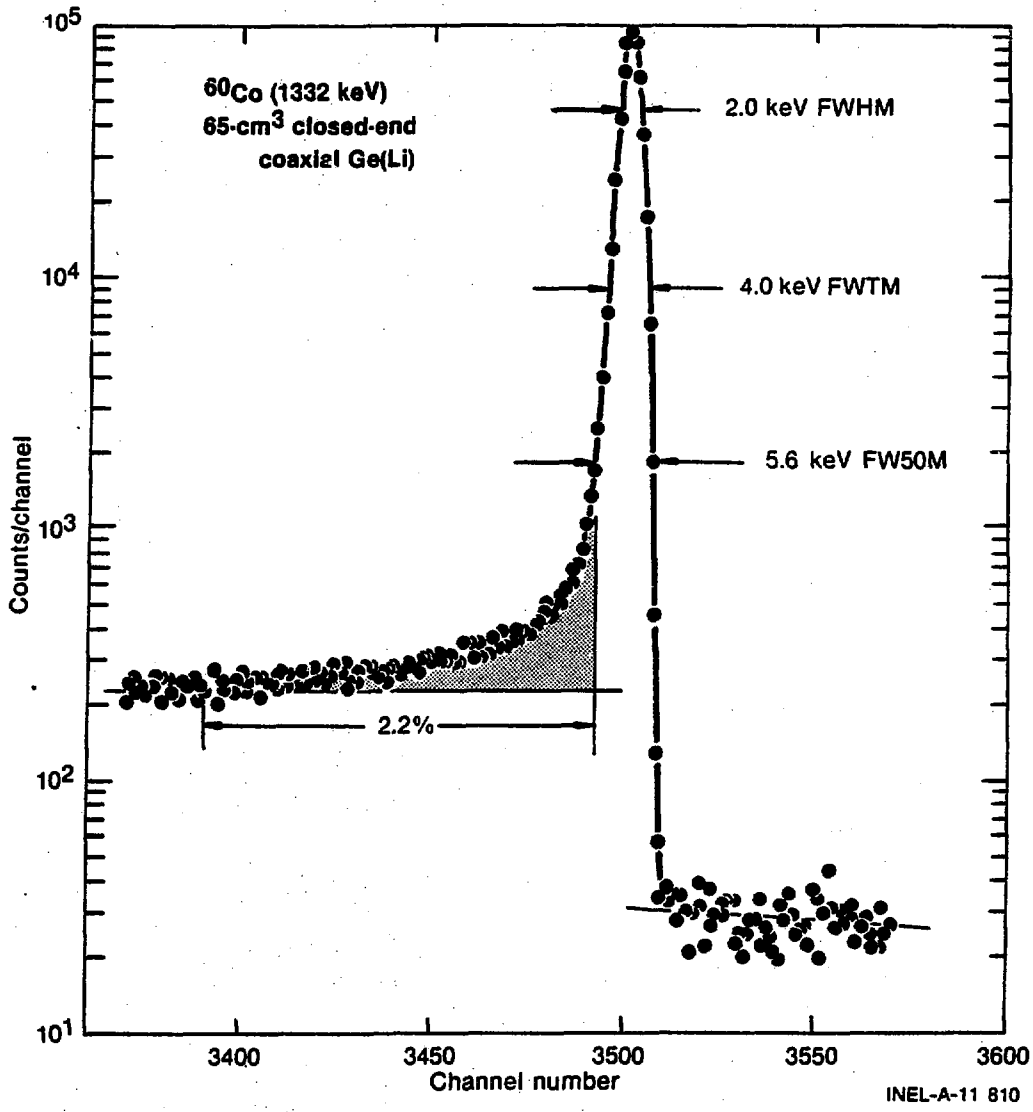

Fig. 1 Spectrum of the full-energy peak of the 1332-keV γ ray from ^{60}Co, measured using a 65-cm^3 closed-end coaxial Ge(Li) detector. The shaded area represents the contribution of the low energy "tail" to the peak area. This figure was provided by R. G. Helmer.

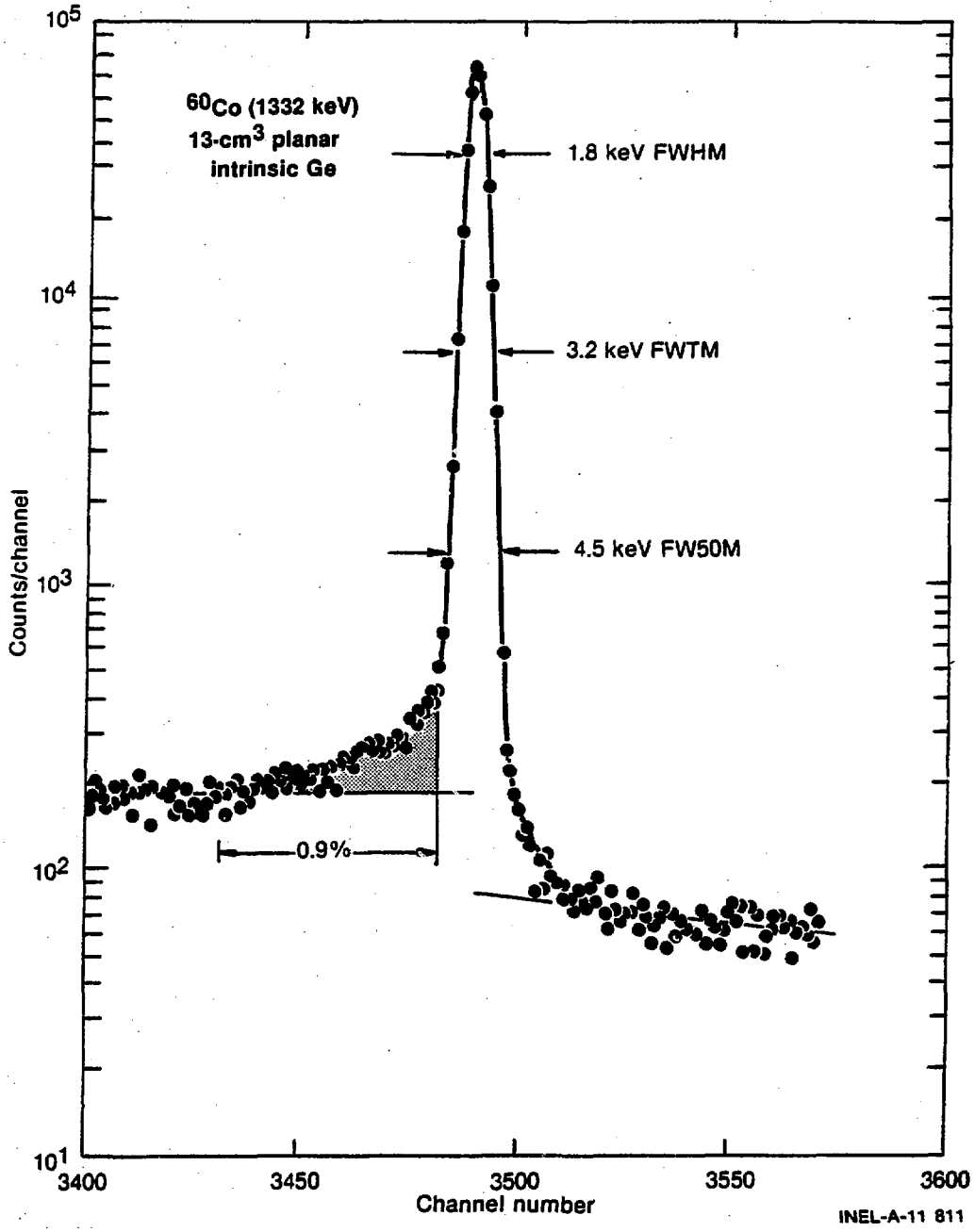

Fig. 2. Spectrum of the full-energy peak of the 1332-keV γ ray from ^{60}Co, measured using a 13-cm^3 planar intrinsic Ge detector. The shaded area represents the contribution of the low-energy "tail" to the peak area. This figure was provided by R. G. Helmer.

in peak area must be significantly reduced, say to the order of a few tenths of a %. To achieve this precision, a careful investigation of all facets of γ-ray peak-shape analysis is required.

5. INTENSITY-BALANCE CONSIDERATIONS IN THE ^{233}PA DECAY SCHEME[†]

The essential features of the ^{233}Pa decay scheme are illustrated in Fig. 3. From the measured value, 38.6 ± 0.5 photons/100 decays, of the absolute intensity of the 311.9-keV γ ray and the relative intensities of the γ rays, the sum of the transition intensities (γ-ray plus conversion-electron) of the γ rays which feed into the ground-state rotational band of ^{233}U is calculated to be (101.4 ± 1.0)%, assuming zero uncertainty in

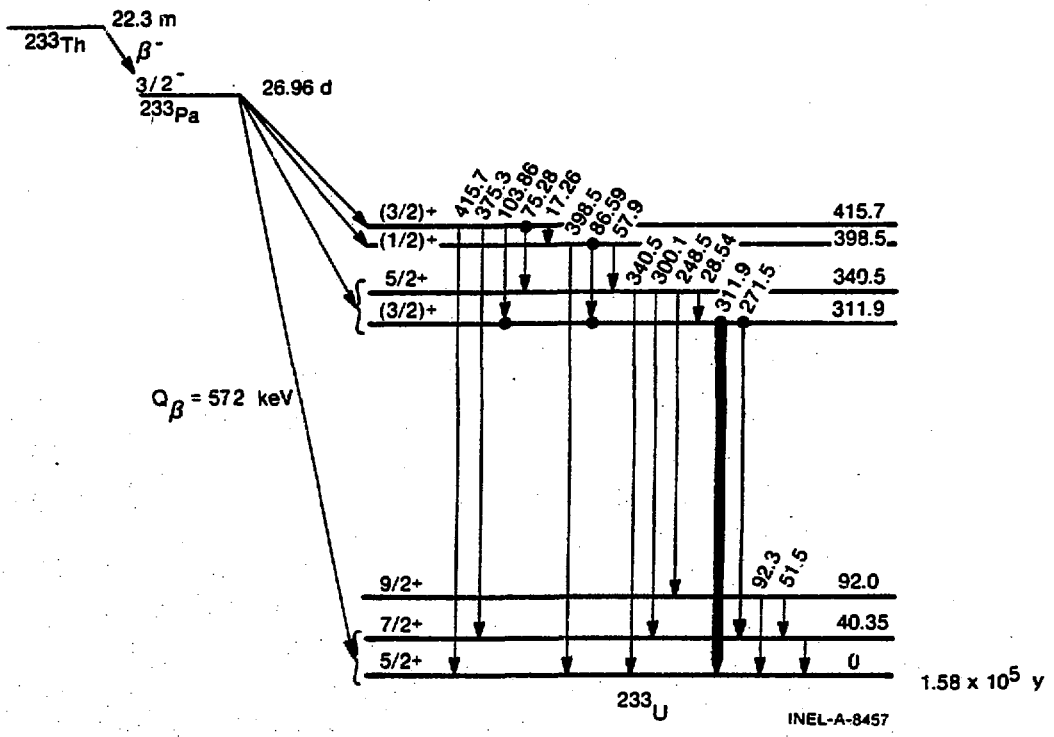

Fig. 3 Partial decay schemes of ^{233}Th and ^{233}Pa.

[†] The discussion in this section is drawn from that given in Ref. [19].

in the multipolarities and internal-conversion coefficients (ICC). This calculation was performed with the reported [4] multipolarities (including M1 for 300 and 312 keV and 90% M1 + 10% E2 for 340 keV), the K, L and M ICC tables of Hager and Seltzer [30] and N tables of Dragoun et al. [31]. (Higher shells will contribute an additional ∿ 0.3% conversion and were omitted.)

It might be argued that this summed γ-transition intensity is sufficiently close to the 100% upper limit that no significant intensity balance problems exist; but to do so would require the assumption of essentially zero direct β^- feeding of the ground and 40-keV states in ^{233}U. It seems clear, however, that there is appreciable direct feeding of these two states. The results of Refs. [32] and [33] give a value of 5% for this feeding intensity, while those of Ref. [34] yield a value of 12%. Unfortunately, no uncertainties are reported for these values so that it is not possible to assess the extent of their agreement (or disagreement). Nonetheless, a definite excess of feeding intensity beta + gamma + conversion-electron) at the ^{233}U ground state is implied; and this amount probably lies in the range of 6-13%.

At present, the origin of this problem cannot be established with certainty. It is conceivable that this imbalance results from errors in the ^{233}Pa decay scheme, our absolute γ-intensity values or the γ-ray multipolarities but, while such a situation is always possible, we do not regard it as a likely explanation here. It seems to us more likely that the problem is associated with the internal-conversion coefficients.

Since the ICC values, denoted here by α, of the γ rays from ^{233}Pa decay are large (e.g., α∿ 1.0 for an M1 transition of energy 300 keV and ∿ 0.2 for an E2 transition), the calculated transition intensities will depend significantly on the values employed for these coefficients. There are three pieces of evidence that indicate that the theoretical M1 ICC values of Ref. [30] may be too large. First, our measured K-x-ray intensities are ∿ 16% lower than the values calculated using theoretical [30] α_K values

and our γ-ray intensities. Since the major contribution to the K x rays is from the M1 transitions, this suggests smaller α_K (M1) values. Second Bisgård et al.[34] report a measured $\alpha_K(312) = 0.69 \pm 0.07$ which is consistent with a smaller α_K value (although it is also consistent with the theoretical value of 0.76). Third, in the electron data of Albridge et al. [35] the K-line intensities of the M1 transitions are weaker relative to the K lines of the E2 transitions than expected from the theoretical values. Interestingly, recent calculations by Band et al. [36] give K- and L-shell ICC values for M1 transitions in this region of Z that are ∿ 6% lower [37] than those of Ref. [30]. We have consequently recalculated the above summed transition intensity of the eight γ rays >200 keV feeding the ground and the 40-keV states, assuming the maximum E2 content reasonably consistent with the experimental measurements for mixed M1/E2 transitions and assuming α(M1) values 6% lower than those of Ref. [30]. This yields a value of ∿ 98% for the summed γ-ray transition intensities and would allow some feeding of the ground and 40-keV levels of ^{233}U, but still appears to conflict with the reported 5-12% beta feeding. Thus if all the errors are in the same direction, the "conflict" may be within the experimental uncertainties.

The question of the intensity balance in the ^{233}Pa decay, thus, remains open at present. To assess the extent to which it results from inaccuracies, resulting either from errors in the theoretical calculations or from other effects such as penetration, in the presently used values of the internal-conversion coefficients would require a number of careful experimental ICC-value measurements. However, intensity-balance considerations, based in part on theoretical ICC values, are quite generally employed by evaluators of nuclear decay data in deducing β- and α-feeding intensities in the decay schemes of transactinium nuclei and, where absolute γ-ray intensities are not experimentally determined, such considerations are frequently used to deduce values for them (see,e.g., Table V). Consequently, the possibility that this procedure may produce erroneous results, at least for the transactinium nuclei, underscores the need for additional studies to resolve this question.

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N.D.S. 19, No. 1 (1976); A=243,245,247
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