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ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

DECAY DATA FOR RADIONUCLIDES USED FOR THE CALIBRATION OF X- AND γ-RAY SPECTROMETERS

Données relatives à la décroissance des radionucléides servant au calibrage des spectromètres à rayons X et γ

A.R. RUTLEDGE, L.V. SMITH and J.S. MERRITT

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Chalk River, Ontario

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Résumé

Les périodes radioactives et les probabilités d'émission de rayons γ ont été résumées à partir de résultats obtenus par le groupe d'étalonnage des radio-éléments au cours de la dernière dizaine d'années. Les périodes radioactives sont données pour trente-trois radionucléides; à savoir ^{7}Be, ^{18}F, ^{22}Na, ^{24}Na, ^{42}K, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{56}Mn, ^{60}Co, ^{64}Cu, ^{65}Ni, ^{82}Br, ^{85}Sr, ^{95}Nb, $^{99}Tc^{m}$, ^{109}Pd, $^{113}In^{m}$, ^{113}Sn, $^{115}In^{m}$, ^{133}Xe, ^{133}Ba, ^{134}Cs, $^{134}Cs^{m}$, $^{137}Ba^{m}$, ^{137}Cs, ^{139}Ce, ^{141}Ce, ^{152}Eu, ^{169}Yb, ^{198}Au, ^{203}Hg et ^{233}Pa. Les probabilités d'émission de rayons gamma sont données pour les onze radionucléides suivants: ^{7}Be, ^{42}K, ^{65}Ni, ^{75}Se, ^{85}Kr, $^{99}Tc^{m}$, $^{113}In^{m}$, $^{115}In^{m}$, ^{137}Cs, ^{139}Ce et ^{141}Ce. Le matériel de comptage comprenait la chambre d'ionisation $4\pi\gamma$, le système de coincidence $4\pi\beta$-γ et un compteur Ge(Li). Chaque méthode de mesure fait l'objet de commentaires. Tout au long de cette étude on a employé une incertitude statistique tenant compte d'une déviation normale. Des précisions sont soupçonnées dans ving-et-un spécimens de radionucléide, parmi ceux dont il est question dans ce rapport, font l'objet de commentaires dans une annexe.

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ABSTRACT

Half-life values and γ-ray emission probabilities are summarized from results determined by the Radioisotope Standardization Group over the past decade or so. Half-life values are given for thirty-three radionuclides: these are ^{7}Be, ^{18}F, ^{22}Na, ^{24}Na, ^{42}K, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{56}Mn, ^{60}Co, ^{64}Cu, ^{65}Ni, ^{82}Br, ^{85}Sr, ^{95}Nb, $^{99}Tc^{m}$, ^{109}Pd, $^{113}In^{m}$, ^{113}Sn, $^{115}In^{m}$, ^{133}Xe, ^{133}Ba, ^{134}Cs, $^{134}Cs^{m}$, $^{137}Ba^{m}$, ^{137}Cs, ^{139}Ce, ^{141}Ce, ^{152}Eu, ^{169}Yb, ^{198}Au, ^{203}Hg, and ^{233}Pa. Gamma-ray emission probabilities are given for the eleven radionuclides ^{7}Be, ^{42}K, ^{65}Ni, ^{75}Se, ^{85}Kr, $^{99}Tc^{m}$, $^{113}In^{m}$, $^{115}In^{m}$, ^{137}Cs, ^{139}Ce, and ^{141}Ce. Counting equipment included the $4\pi\gamma$ ionization chamber, the $4\pi\beta$-γ coincidence system, and a Ge(Li) counter. Each method of measurement is discussed. A statistical uncertainty of one standard deviation has been used throughout and detailed information is given about the assessment of other uncertainties. The impurities found or suspected in twenty-one of the reported samples of radionuclides are discussed in an appendix.

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1. INTRODUCTION

The α-, β-, and γ-ray Spectrometry Working Group (SWG) of the International Committee for Radionuclide Metrology (ICRM) has requested information about measurements of γ-ray emission probabilities and half-lives for radionuclides that are issued as standards for calibration purposes. Rather detailed information was requested, especially about the assessment of uncertainties in the final values. The SWG plans to examine critically the collected data to draw attention to discrepancies, and to encourage further and more accurate measurements to resolve these discrepancies. Ultimately this should allow γ-ray spectrometers to be calibrated more accurately.

This report is the contribution from the Radioisotope Standardization Group of Atomic Energy of Canada Limited (AECL) to the SWG, and it summarizes and reviews such measurements made in this laboratory during the past decade or more.

2. GENERAL REMARKS AND LISTS OF DATA

The attempt by the AECL Radioisotope Standardization Group (RSG) to summarize its data in a consistent manner has been complicated by the fact that while the results of some measurements have been fully documented others have appeared only as short paragraphs in Physics Division Progress Reports. In the past the RSG has generally used a confidence interval of 3σ for the statistical uncertainty and estimates of other sources of uncertainty usually were added linearly. To conform with the request of the SWG for consistent statements about accuracy at a confidence level of 68%, in order to facilitate recognition of discrepancies and a uniform evaluation of the data[1,2], considerable research and recalculation were required. As a result, in most cases the uncertainties in the values tabulated in this report differ from those in the referenced publications or reports.

The radionuclides for which half-life data are submitted are listed in Table I. Column 2 gives the number of half-lives over which observations have been made, followed by the number of observations in brackets. The

Table I

Radionuclides Issued as Standards for which

Half-Life Data are Submitted to SWG of ICRM

Radionuclide	No. of Half-Lives followed (No. of Points)	Counting Equipment[a]	Completion Status	Half-Life	Reference[b]
^{7}Be	9.3(43)	I.C.	Yes	53.284 ± 0.004 d[c]	4 (PRP)
^{18}F	6 to 20 (~30)	4πPC	Yes	109.73[d] ± 0.04 min	5 (PRP)
^{18}F	10 to 11 (25)	I.C.	Yes	109.71[d] ± 0.02 min	5 (PRP)
^{22}Na	3.9 (92)	I.C.	No	950.30[d] ± 0.27 d	6 (PRP)
^{24}Na	10 (33)	4πPC	Yes	14.965 ± 0.010 h	7 (PRP)
^{24}Na	3 to 8	4πPC	Yes	14.959 ± 0.010 h	8
^{24}Na	10 to 14 (397)	I.C.	Yes	14.965 ± 0.001 h	none
^{42}K	~10	4πPC	Yes	12.358 ± 0.007 h	9
^{46}Sc	12.4 (41)	I.C.	Yes	83.752 ± 0.015 d	10 (PRP)
^{51}Cr	13.6 (40)	I.C.	Yes	27.704 ± 0.003 d	4 (PRP)
^{54}Mn	11 (83)	I.C.	Yes	312.21 ± 0.03 d	11 (PRP)
^{56}Mn	3.2 (32)	I.C.	Yes	2.5764± 0.0008 h	12 (PRP)
^{60}Co	0.8	4πPC-γ	No	1923.78 ± 0.94 d	13 (PRP)
^{60}Co	0.5 to 3.5	I.C.	No	1924.33[d] ± 1.36 d	13 (PRP)
^{64}Cu	~10 (32)	I.C.	Yes	12.701 ± 0.003 h[c]	14 (PRP)
^{65}Ni	6 to 9	4πPC	Yes	2.520 ± 0.001 h	15

Table I (Continued)

Radionuclide	No. of Half-Lives followed (No. of Points)	Counting Equipment[a]	Completion Status	Half-Life	Reference[b]
^{82}Br	~10	4πPC	Yes	35.344 ± 0.016 h	9
^{85}Sr	~15 (26)	I.C.	Yes	64.845 ± 0.009 d	16 (PRP)
^{95}Nb	12 (37)	I.C.	Yes	34.98[d] ± 0.02 d	4 (PRP)
$^{99}Tc^{m}$	15 (89)	I.C.	Yes	6.008 ± 0.004 h[c]	17 (PRP)
^{109}Pd	5.3 (36)	4πPC	Yes	13.402 ± 0.006 h	none
$^{113}In^{m}$	4 to 5 (19)	4πPC	Yes	99.49 ± 0.06 min	18 (PRP)
^{113}Sn	~ 9 (29)	I.C.	Yes	115.12 ± 0.13 d	16 (PRP)
$^{115}In^{m}$	5 to 11 (10)	I.C.	Yes	4.49 ± 0.01 h	19 (PRP)
^{133}Xe	14 (57)	I.C.	Yes	5.243 ± 0.001 d	20 (PRP)
^{133}Ba	0.17 (22)	I.C.	No	3785.17[d] ± 27.17 d	6 (PRP)
^{134}Cs	1.36 (34)	I.C.	No	753.78[d] ± 0.30 d	6 (PRP)
$^{134}Cs^{m}$	8.5 (22)	4πPC	Yes	2.914 ± 0.002 h	21 (PRP)
$^{137}Ba^{m}$	10 to 20	4πPC	Yes	2.554 ± 0.002 min	22
^{137}Cs	0.06 (22)	I.C.	No	10677.5[d] ± 140.3 d	6 (PRP)
^{139}Ce	12.7 (50)	I.C.	Yes	137.65 ± 0.03 d	23 (PRP)
^{141}Ce	9.0 (9)	I.C.	Yes	32.50 ± 0.03 d	none
^{152}Eu	0.31 (50)	I.C.	No	4892.3[d] ± 8.2 d	6 (PRP)
^{169}Yb	13.3 (32)	I.C.	Yes	32.015 ± 0.009 d	24 (PRP)

Table I (Continued)

Radionuclide	No. of Half-Lives followed (No. of Points)	Counting Equipment[a]	Completion Status	Half-Life	Reference[b]
^{198}Au	10 to 14 (252)	I.C.	Yes	2.6935[d] ± 0.0004 d	25 (PRP)
^{203}Hg	9 (13)	I.C.	Yes	46.60 ± 0.01 d	21 (PRP)
^{233}Pa	11 (112)	I.C.	Yes	26.967 ± 0.002 d	26 (PRP)

a) 4πPC indicates 4π gas flow proportional counter; I.C. indicates ionization chamber; 4πPC-γ indicates 4πβ-γ coincidence system.

b) 'none' indicates work done in this laboratory but never published; PRP indicates Physics Division Progress Report.

c) Form of source for ^{7}Be was dilute HCl solution; for ^{64}Cu was metal, and solutions of HNO_3, H_2SO_4, and NH_3; for $^{99}Tc^{m}$ was $NaTcO_4$ in solution.

d) These values have been updated since the reference given.

counting equipment used is listed next. Then the completion status is given: 'No' indicates that the experiment is ongoing, 'Yes' indicates the converse. Most of the entries indicated as ongoing are for rather long half-lives ($\geq$2 a); it is planned to continue these measurements for several more years and to update the values at appropriate intervals.

The half-life values are given in the fifth column. The stated uncertainties contain statistical and systematic components; Table IV gives a breakdown of the contribution from various sources of uncertainties to this overall uncertainty. For most of the measurements, the decay of the radionuclide was followed for one or more samples prepared from the same batch of the radionuclide; the statistical uncertainty used here is one standard deviation in the least squares fit to the counting data. In other cases half-life values from more than one preparation or supply of the nuclide were obtained. These are identifiable where Table I, column 2, shows a range in the number of half-lives followed. For these cases the external error in the weighted mean value is used as the statistical uncertainty.

Details about our estimates of sources of systematic uncertainty are given in sections 3 and 4 of this report. Column 5 of Table I lists the overall uncertainties for each nuclide. Where there is only one known source of systematic uncertainty its estimate is added to the statistical uncertainty to get the value shown in the table. Where more than one source of a systematic nature is estimated, the individual estimates are combined in quadrature and then added to the statistical uncertainty. Because the number of known sources of systematic uncertainty is very small, other published methods of combining statistical and systematic uncertainties[2,3] seem less appropriate here; they would tend to give smaller overall uncertainties.

In the reference column the letters PRP indicate that the only available account is in an AECL Physics Division Progress Report.

Table II lists the radionuclides for which we have determined gamma-ray emission probabilities (P_γ). The energy of the gamma ray and the γ-counting equipment are also listed. P_γ is given in percent

Table II

Radionuclides Issued as Standards for which P_γ Data are Submitted to SWG of ICRM

Radionuclide	E_γ(keV)	γ-counting equipment	P_γ(%)	Reference
^{7}Be	477	IC	10.32 ± 0.04	27, 28 (PRP)
^{42}K	1530	IC	19.1 ± 0.6	29 (PRP)
^{65}Ni	1482	IC	23.5 ± 0.4	15
^{75}Se	400.5	Ge(Li)	12.5 ± 0.3	30 (PRP)
^{85}Kr	510	scintillation spectrometer	0.46 ± 0.03	31
$^{99}Tc^{m}$	140 and 142	4πPC-γ	88.75 ± 0.14	32 (PRP)
$^{113}In^{m}$	392	IC	64.9 ± 0.2	18 (PRP)
$^{115}In^{m}$	336	IC	45.9 ± 0.3	19 (PRP)
^{137}Cs	662	IC	84.7 ± 0.6	33
^{139}Ce	165	4πPC-γ	79.95 ± 0.06	34
^{141}Ce	145.44	IC	48.5 ± 0.4	35

and the uncertainties are stated on the same basis as for Table I. Again, PRP in the Reference column indicates that the only available account is in an AECL Physics Division Progress Report.

3. HALF-LIFE MEASUREMENTS WITH A $4\pi\gamma$ IONIZATION CHAMBER

A $4\pi\gamma$ ionization chamber (IC) is the instrument that has been used for most half-life measurements in this laboratory. Decay data are taken relative to a ^{226}Ra reference source in order to correct for short-term fluctuations in the IC response. The decay usually is followed for about ten half-lives. In computing the data, corrections are made for ^{226}Ra decay using a half-life value of 1600 ± 7 years[36].

The ionization chamber is a TPA MkII[37,38] reentrant model filled with twenty atmospheres of argon. A diagram of the chamber[39] and a discussion about the use of ionization chambers for high-precision measurements have been given in recent review articles[39,40].

Table IV gives information about the contribution from various sources of uncertainty toward the overall uncertainty given in Table I.

The experimental setup, described in an earlier report[41], allows the charge built up on an integrating capacitor mounted across a vibrating reed electrometer to be read out with a digital voltmeter. The standard deviation for a single observation is typically ±0.02 to ±0.04% for conditions of source strength and counting interval that are representative during the first few half-lives of a half-life measurement.

Systematic error from long-term instability of the response of the ionization chamber has been investigated and found trivial. The same ionization chamber was used in an earlier study[41], which showed no evidence of gas leakage during the ten-year period prior to 1967; our calibration data since that time confirm this finding and indicate that any decrease in efficiency has been <0.1%. An example is the set of response factors shown in Table III for ^{198}Au calibrations versus a ^{226}Ra reference source. The ^{198}Au activity was determined independently by the $4\pi\beta$-γ coincidence method. The statistical uncertainty since 1969 has been ≈0.03%, but earlier measurements were less precise.

Table III

Ionization Chamber Response Factor
for ^{198}Au over a Period of Time

Date	IC Response Factor
1978 - May	3.1940
1977 - Jan	3.1950
1974 - May	3.1947
1969 - Sept	3.1924
1966 - Sept	3.196
1966 - July	3.192
1965	3.188
1963	3.194

The source was a sample of solution sealed in an ampoule. For most of the radionuclides studied these were glass ampoules, but for hydrofluoric acid solutions (e.g. ^{95}Nb and ^{233}Pa) polyethylene ampoules were used. The carrier solution is selected for stability and usually is the same as that used for standards of radionuclides[42]. No evidence of solution instability has been observed and therefore no contribution to systematic error from it has been included. Non-reproducibility of the source position in the chamber contributes to the statistical uncertainty in the half-life measurements. Tests with ^{60}Co have revealed that this effect is $\leq 0.01\%$[41] and arises largely from anisotropy in the chamber response[41]. The effect is greater for lower energy γ rays and correlates with the somewhat larger statistical uncertainties in the half-life values of nuclides that emit only low energy γ rays.

The ^{226}Ra reference sources were obtained from Amersham Corporation between 1964 and 1967. They are doubly encapsulated in iridium-platinum alloy. In 1972 the purity was investigated by γ-ray spectrometry[14] to assess the content of 6-year ^{228}Ra. None was detected and a limit of <0.01% was estimated at that time. Therefore, no correction was made for ^{228}Ra impurity when computing any of the half-lives reported here, but the possibility of its presence contributes slightly to the systematic uncertainty in some of the half-life values, as shown in Table IV. The accuracy of the ^{226}Ra half-life (1600 ± 7 a) is sufficient to allow us to neglect the effect of its uncertainty at this time, but if the decay of some of the longer-lived samples is followed for many more years, this might become significant. For example, it would contribute ≈ ±0.02 d to the uncertainty in the ^{60}Co half-life value.

Saturation of the ionization chamber is another source of error that has been investigated[40]. Tests with a series of ^{60}Co sources of various activities have been done to demonstrate the source strength at which saturation of the ion-current level gives a noticeable loss. Source strengths have been selected to stay below this level and thus avoid the difficulty.

Other systematic uncertainties in the measurements arise from radioactive impurities in the source samples. For some radionuclides chemical separations were performed prior to commencement of the measurement period, and these are indicated by CP in the first column of Table IV. The presence of impurities was investigated by two methods. One method was the identification of impurities by γ-ray spectrometry. If an impurity was found, corrections were made to the decay data. The difference between half-life values computed with and without these corrections was used to deduce the systematic uncertainty caused by impurities. The other method used to test for impurities was to divide the decay data into two or more sections, compute the half-life for each section separately, and examine the results for a significant difference. The magnitude of the difference found, if any, gave an indication of systematic uncertainty. Details about the actual impurities found or suspected in specific radionuclides are given in Appendix 1.

Table IV

Uncertainties in Half-Life Values Determined with the $4\pi\gamma$ Ionization Chamber

Radionuclide	Uncertainty in Half-Life Value from (a) Statistical (1 σ)	(b) Purity of ^{226}Ra Reference Source	(c) Impurities in Sample
^{7}Be CP*	0.004 d	nil	nil
^{18}F CP	0.013 min	nil	< 0.01 min
^{22}Na CP	0.12 d	0.1 d	0.11 d
^{24}Na CP	0.0005 h	nil	nil
^{46}Sc	0.011 d	0.004 d	nil
^{51}Cr CP	0.003 d	nil	nil
^{54}Mn CP	0.017 d	0.01 d	nil
^{56}Mn	0.0004 h	nil	≤ 0.0004 h
^{60}Co CP	0.77 d	0.2 d	0.56 d
^{64}Cu	0.001 h	nil	0.002 h
^{85}Sr	0.007 d	0.002 d	0.0005 d
^{95}Nb CP	0.002 d	nil	0.018 d
$^{99}Tc^{m}$	0.0007 h	nil	≤ 0.003 h
^{113}Sn	0.032 d	0.005 d	0.10 d
$^{115}In^{m}$ CP	0.005 h	nil	0.005 h
^{133}Xe SI*	0.0007 d	nil	0.0003 d
^{133}Ba CP	27.1 d	0.1 d	nil
^{134}Cs CP	0.26 d	0.04 d	nil

Table IV (Continued)

Radionuclide	Uncertainty in Half-Life Value from (a) Statistical (1 σ)	(b) Purity of ^{226}Ra Reference Source	(c) Impurities in Sample
^{137}Cs CP	140.2 d	0.1 d	nil
^{139}Ce	0.024 d	0.005 d	nil
^{141}Ce	0.02 d	nil	0.01 d
^{152}Eu	7.2 d	0.1 d	1.0 d
^{169}Yb	0.007 d	nil	0.002 d
^{198}Au	0.0002 d	nil	0.0002 d
^{203}Hg	0.005 d	nil	0.002 d
^{233}Pa	0.002 d	nil	nil

*CP indicates chemically purified; SI indicates separated isotope was irradiated to produce the radionuclide.

4. HALF-LIFE MEASUREMENTS WITH A 4π PROPORTIONAL COUNTER

A 4π proportional counter was used for most other half-life measurements reported here. This instrument is less suitable than an ionization chamber because it is subject to many more sources of systematic uncertainty. Chief among these are: 1) the dead time of the counting system; 2) instability of the radioactive source over a prolonged period of time; and 3) change in the voltage plateau.

Table V shows the contribution from individual sources of uncertainty toward the overall uncertainty given in Table I.

The dead time of the counting system was measured with the two-source method or the source-pulser method, both of which have been discussed by Taylor in a recent review[43]. Typically, the dead time was 2 μs with a standard deviation of ±0.12 μs for a single observation. For the half-lives reported here the dead-time correction to the first data point was $\leq$3%. To study the magnitude of uncertainty in the half-

life value from an erroneous dead-time value, the data from a run were corrected for a dead-time value different by one standard deviation; the half-life was recomputed, and the difference in the half-life value was taken as the systematic uncertainty contributed by dead time. This approach gives an uncertainty estimate that is much lower than the maximum conceivable limit of error, and seems consistent with the 68% confidence level adopted here.

It is well-known that the thin sources required for 4π counting are susceptible to small gradual changes which affect the overall 4π counting efficiency. One such change is in the source material itself. An example is the sorption of water vapour, which frequently is encountered with halides of rare earths, alkaline earths and alkali metals, and depending upon circumstances, can change the self-absorption by ± 1% over a period of several months.

Another type of change can occur in the metallic coating of the film used as the source mount. A decrease in the electrical conductivity of the film may alter the voltage plateau and hence the counting rates observed at the selected counting voltages. For a typical plateau, with a slope of $\approx$ 0.2% per 100 V, an effect of $\geq$ 0.1% has been observed for sources of long-lived nuclides over a period of a few months. It was impracticable to make quantitative observations of this effect for the actual sources used in the half-life determinations reported here because the counting statistics were inadequate after the decay of several half-lives. Our only estimate of the magnitude of this source of uncertainty is based upon our experience with longer-lived nuclides. The effect is trivial for short-lived nuclides.

Another source of systematic uncertainty has been considered and found to be small. It is the effect from change in the counter response that is not associated with the source material or source mount. We have called this "instability of the counter" in Table V. It arises from changes in the gain of the detector, such as those associated with source

Table V

% Uncertainty from Various Sources in Half-life Values
Determined with a 4π Proportional Counter

Radionuclide	Statistical	Impurities	Dead-time	Instability of Source	Instability of Counter
^{18}F CP[a]	0.01 min	0.003 min	0.03 min	negligible	<0.01 min
^{24}Na CP	0.003 h	nil	0.007 h	0.001 h	0.001 h
^{42}K CP	0.003 h	nil	0.004 h	0.001 h	0.001 h
^{60}Co[b] CP	0.56 d	nil	0.38 d[b]	N/A[b]	N/A[b]
^{65}Ni SI[a]	0.0004 h	0.0001 h	0.0004 h	negligible	0.0003 h
^{82}Br CP	0.004 h	nil	0.009 h	0.006 h	0.004 h
^{109}Pd SI	0.0019 h	nil	0.004 h	0.001 h	0.0007 h
$^{113}In^{m}$ CP	0.02 min	0.002 min	0.04 min	negligible	0.005 min
$^{134}Cs^{m}$	0.0003 h	0.001 h	0.001 h	0.0003 h	0.0002 h
$^{137}Ba^{m}$ CP	0.001 min	0.001 min	0.001 min	negligible	0.0002 min

a) CP indicates chemically purified; SI indicates separated isotope was irradiated to produce the radionuclide.

b) By 4πβ-γ coincidence method which compensates for instabilities in the source and 4π proportional counter, and for which the methods of assessment of uncertainties in timing corrections have been described[44].

rate and the accumulation of a deposit on the counter wires over a period of time; these changes alter the voltage-plateau characteristics. Although our normal practice in recording an individual half-life datum is to average three observations taken at different voltages on the plateau, a small shift in either the slope or the position of the plateau might not be noticed and would bias the half-life value. In our judgment the limit of this shift gives ≈ 0.2% in the relative counting rate over the activity range and period usually followed. Assuming that one-third of this limit is consistent with a confidence interval of one, and taking into account the number of half-lives over which the decay was followed, the estimates listed in the last column of Table V were deduced.

5. MEASUREMENT OF GAMMA-RAY EMISSION PROBABILITIES (P_γ)

Most of the P_γ values listed in Table II were direct observations of the ratio of γ-ray emission rate to activity. The counting methods and instruments used were essentially the same as discussed in earlier sections of this report. The accuracy, when applied to individual P_γ measurements, is outlined in sections 6 and 7.

Determination of internal conversion data allowed P_γ values to be deduced for other radionuclides, namely $^{99}Tc^m$, ^{139}Ce, $^{113}In^m$ and $^{115}In^m$. The data for $^{99}Tc^m$ and ^{139}Ce were determined with a method patterned on that published by Taylor for ^{203}Hg[45]. The effect of variation in efficiency of the 4π(PC) upon 4πPC-γ coincidence results is analyzed to derive the fraction of the γ-ray transitions that are internally converted. The usual techniques for efficiency variation (variation of self- and film-absorption) were used here instead of the suspended foils described by Taylor[45]. Observations of the ratio, electron emission rate to γ-ray emission rate, gave internal conversion coefficients for $^{113}In^m$ and $^{115}In^m$.

6. UNCERTAINTIES IN GAMMA-RAY MEASUREMENTS FOR P_γ VALUES

Gamma-ray emission rates for ^{7}Be, ^{42}K, ^{65}Ni, $^{113}In^{m}$, $^{115}In^{m}$, ^{137}Cs and ^{141}Ce were determined with the calibrated $4\pi\gamma$ ionization chamber (see section 3). Response versus energy curves, relative to the response of the same ^{226}Ra reference sources described in section 3, were determined for three sample holders made from (1) 0.05-mm thick aluminum, (2) 0.4-mm cadmium, and (3) 1.2-mm cadmium inside 0.12-mm tantalum. The $4\pi\beta$-γ coincidence method was used to calibrate samples of suitable radionuclides. For data taken in recent years, the uncertainty in an individual calibration point (the combined uncertainty in the coincidence counting and ion-chamber measurements) was typically ± 0.3%[44]. These data were for 140-keV $^{99}Tc^{m}$ γ rays, 165-keV ^{139}Ce, 279-keV ^{203}Hg, 411-keV ^{198}Au, annihilation radiation from ^{18}F, 514-keV ^{85}Sr, 766-keV ^{95}Nb, 835-keV ^{54}Mn, 889- and 1120-keV ^{46}Sc, and 1173- and 1332-keV ^{60}Co. The calibration data for the energy region 279 to 1332 keV were fitted to a second-order polynomial, and for the entire energy range, to a third-order polynomial. To deduce the uncertainties in the fitted calibration curves for the three sample holders, response data were compared for test γ-ray energies, and the standard deviation among the three results was taken as the uncertainty. Uncertainties in the calibration are listed in Table VI. They are greatest for the low energy region and for energies above 1.33 MeV. For example, the uncertainties in instrument calibration in the cases of ^{42}K and ^{65}Ni are larger because the response curves had to be extrapolated beyond the highest energy calibration point. A further contribution to the uncertainty for ^{42}K is that at the time of this P_γ measurement, high-precision instrumentation and stable ^{226}Ra references were not yet available.

Gamma-ray emission rates for ^{75}Se and ^{85}Kr were obtained by gamma-ray spectrometry with Ge(Li) and NaI(Tℓ) detectors respectively. For the 401-keV ^{75}Se γ ray, the most relevant energy calibration points were ^{198}Au at 411 keV and ^{69}Zn at 439 keV; the ^{198}Au data were obtained from a $4\pi\beta$-γ coincidence standardization and the ^{69}Zn from the calibrated ion chamber. ^{22}Na standardized by $4\pi\beta$-γ coincidence counting was used to calibrate the NaI(Tℓ) detector in the energy region of ^{85}Kr; most of the uncertainty shown for this calibration comes from uncertainty in the source geometry used to simulate the Kr gas sample[31].

Table VI

Breakdown of Uncertainty (%) in P_γ Values

Radionuclide	Statistical	Impurities	γ-counting Calibration Curve	Other*
^{7}Be	0.07	nil	0.16	0.18, 0.2
^{42}K	0.2	nil	2.5	0.5
^{65}Ni	<0.1	nil	1.0	1.3, 0.2
^{75}Se	0.4	nil	1.0	1.0, 1.0
^{85}Kr	2.4	nil	2.5	0.3, 0.3, 1.5
^{137}Cs	0.15	0.01	0.5	0.083
^{141}Ce	0.05	0.1	1.6	0.1, 0.2

* See text, page 16 - 17.

7. UNCERTAINTIES IN ACTIVITY MEASUREMENTS FOR P_γ VALUES

The 4π(PC)-γ coincidence method was used to determine the activity of ^{7}Be[27], ^{65}Ni[15], ^{75}Se[30], ^{137}Cs[33] and ^{141}Ce[35]; ^{42}K[29] and ^{85}Kr[31] activities were determined by 4π proportional counting and internal gas counting, respectively. For ^{7}Be, the difference between results from two different γ-channel gates was 0.18%, and uncertainty from spurious pulses was estimated as ± 0.2%. These are given in the last column of Table VI.

For ^{42}K, the other uncertainty listed is the uncertainty in self-absorption and source-mount absorption corrections.

The systematic uncertainty in $4\pi\beta$-γ coincidence counting ^{65}Ni was estimated as ± 0.2%. The other major source of uncertainty arises from the correction to the ion chamber data for the response of other ^{65}Ni γ rays, for which published values for relative gamma intensities[46] were used.

The determination of the activity of ^{75}Se by $4\pi\beta$-γ coincidence counting was complicated by uncertainties in the decay scheme. The uncertainty in the correction for the decay to $^{75}As^{m}$ was taken to be ± 1.0%. In addition, the complexity of the decay scheme necessitated higher-order polynomial fits to the efficiency data. Uncertainty here was judged to contribute another ± 1.0%, by examining the differences among results from two different γ-channel gates and second- and third-order fits.

For ^{85}Kr, the systematic uncertainties in internal gas counting were from wall effect (± 0.3%), uncertainty in counter volume (± 0.3%) and slope of the differential voltage plateau (± 1.5%). The slope was 1% per 100 V for a 300 V long plateau, and the average counting rate along the plateau was used as the result.

Sources of systematic uncertainty in an accurate measurement of the activity of ^{137}Cs determined by the 4π(PC)-γ efficiency-tracing method with ^{134}Cs as tracer have been reported in detail[33]. They are from dead-time and resolving-time corrections (± 0.022%), decay-scheme corrections (± 0.067%), sensitivity of the 4π(PC) to γ rays (± 0.024%), ^{134}Cs impurity (0.010%), and ^{134}Cs decay corrections (± 0.034%). Their combination in quadrature gives ± 0.083%.

The activity measurement of ^{141}Ce was done with less attention to minute details but has also been reported[35]. Here the main sources of systematic uncertainty were from dead-time and resolving-time corrections (± 0.1%), efficiency-dependent correction (± 0.2%), and impurities of ^{46}Sc and ^{139}Ce (± 0.1%).

8. UNCERTAINTIES IN P_γ VALUES FOR $^{99}Tc^{m}$, ^{139}Ce, $^{113}In^{m}$ and $^{115}In^{m}$

We have not been able to break down the overall uncertainty value given for $^{99}Tc^{m}$ [32] into its components. It was stated that the uncertainty consisted principally of allowances for various sources of possible systematic error. The final result was the mean value from five runs,

which came from three different preparations of $^{99}Tc^{m}$; thus it seems unlikely that impurities contributed significantly to the overall uncertainty.

For ^{139}Ce, other measurements[47] have been made from time to time since the first determination in 1962[34], and these have been in good agreement. The statistical uncertainty, though not always the same, has been unimportant compared with other sources of uncertainty. Typically these were from non-detection of conversion electrons in the 4π(PC) (± 0.1%), uncertainty in the correction for the sensitivity of the 4π(PC) to γ rays (± 0.1%), uncertainty in other instrumental corrections for dead time and resolving time (± 0.15%), and uncertainty in extrapolation of the efficiency function (± 0.2%). They are listed in Table VII.

Table VII

Breakdown of Uncertainty (%) in α_T* Values

Radionuclide	Statistical	Impurities	Other†
^{139}Ce	0.09	nil	0.1, 0.1, 0.15, 0.2
$^{113}In^{m}$	0.08	nil	0.15, 0.1, 0.4, 0.1, 0.1
$^{115}In^{m}$	0.23	.03	0.15, 0.1, 1.6, 0.6, 0.1

* $P_\gamma = 1/(1+\alpha_T)$, where α_T is total internal conversion coefficient.

† See text, page 18 - 19.

The electron emission rate from sources of $^{113}In^{m}$ was determined by 4π(PC) counting. Uncertainties in it were from the slope of the plateau (± 0.15%), and from the correction for sensitivity of the 4π(PC) to γ rays, $\varepsilon_{\beta,\gamma}$ (± 0.1%). Systematic uncertainty in the correction for dead-time was considered negligible because results for different counting rates were consistent. The γ-ray emission rate was determined for other samples of $^{113}In^{m}$ with the 4π ionization chamber, by using the three sample

holders. The uncertainty in calibration of the ionization chamber was taken as the standard deviation in the results from the three holders (± 0.4%). The uncertainty in the γ-ray energy was estimated to affect the results by ± 0.1%. Corrections for Bremsstrahlung contributed another ± 0.1% uncertainty.

Similar measurements were made to get electron and γ-ray emission rates for $^{115}In^{m}$. The result of the experiment was a set of conversion-coefficient data. The same uncertainties were assigned for the 4π(PC) counting (± 0.15% for plateau and ± 0.1% for $\varepsilon_{\beta,\gamma}$). A correction was necessary for the contribution to the 4π(PC) rate from $^{115}In^{m}$ β rays. At the time of the measurement this was estimated as 6 ± 1% but it seems likely this was in error. A recent evaluation gives 3.7 ± 0.8% [48], and its uncertainty contributes ± 1.6% to α_T. (This branching ratio gives α_T = 1.096.) However, it can be shown that P_γ is unaffected by the β-branching ratio. The experimental data revealed that the 4π(PC) counting rate, $N_{4\pi}$, was equal to 1.1768 times the γ-ray emission rate, N_γ. Thus we have

$$N_{4\pi} = 1.1768\ N_\gamma$$

But $$N_{4\pi} = N_\beta + N_e + N_\gamma \varepsilon_{4\pi,\gamma}$$

and $$N_o = N_{4\pi} + N_\gamma$$

where N_β is the counting rate from the β-branch,
N_e is the counting rate from conversion electrons, and
$\varepsilon_{4\pi,\gamma}$ is the very small efficiency of the 4π(PC) counter for 336-keV γ rays (≈0.1%). Since

$$P_\gamma = N_\gamma / N_o$$

it is unaffected by the relative contributions from β particles and conversion electrons to the 4π counting rate.

The uncertainty in calibration of the ionization chamber for $^{115}In^{m}$ γ rays, by the same criterion as for $^{113}In^{m}$, was deduced to be ± 0.6%. Bremsstrahlung was estimated as ± 0.1% and uncertainty in the γ-ray energy was considered negligible.

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APPENDIX 1

IMPURITIES FOUND OR SUSPECTED IN SAMPLES OF RADIONUCLIDES

^{18}F - Chemical separation removed ^{3}H and traces of ^{24}Na, ^{38}Cl and ^{198}Au. The only residual activity was ^{187}W which was detected in all 7 preparations. The impurity varied between 0.002 and 0.015% at the start of a run. Its effect on the half-life value from the 4π(PC) results was 0.015 min but was negligible for the ionization chamber results.

^{24}Na - Chemical separation effectively removed ^{42}K.

^{42}K - Chemical separation effectively removed ^{24}Na.

^{56}Mn - Possibly a small impurity of 2.5-h ^{65}Ni. This would have a negligible effect on the result because the half-lives are nearly equal.

^{64}Cu - Gamma-ray spectrometry detected no impurities. ^{56}Mn is the most suspected impurity; 0.2% ^{56}Mn would cause an error of 0.004 h in the ^{64}Cu half-life.

^{65}Ni - An impurity of 0.03% ^{24}Na was typical at the start of a run.

^{85}Sr - Impurities of ^{57}Co (0.61%), ^{65}Zn (0.12%) and ^{133}Ba (0.24%) were estimated at the end of the measurement period. Corrections for these amounted to 3.7×10^{-4} % at the beginning and 0.9% at the end of the measurement period. If the impurities are not taken into account, the half-life value would be in error by 0.001 day.

^{95}Nb - An impurity of ^{95}Zr was detected. The correction for it was 0.1% at the start and 10% at the end of the period followed. If the ^{95}Nb half-life is computed without making a correction for the presence of ^{95}Zr, the result is longer by 0.09 d. It is assumed that the accuracy of the correction is ± 20% of this 0.09-d difference.

$^{99}Tc^{m}$ - For the sample used for the half-life measurement, an impurity of ^{99}Mo was detected. This was 0.1% at the beginning and 90% at the end of the measurement period.
Different samples were used for the α_T measurement. Specific information about their purity is not available.

$^{113}In^{m}$ - Impurities of ^{113}Sn were present in most samples, and the amounts varied between 0.001 and 0.003% at the start of the measurement period.

^{113}Sn - Impurities of ^{125}Sb, ^{60}Co and $^{114}In^{m}$ were detected, with ^{125}Sb predominant. The total correction at the start of the run was 0.76% and at the end it was 50%.

$^{115}In^{m}$ - $^{115}In^{m}$ samples were prepared by neutron irradiation of ^{114}Cd, followed by a 1-day waiting period, before chemical separation. The 1-day waiting period reduces the content of ^{117}In impurity. No long-lived impurities were detected, and it is estimated that short-lived impurities were $\leq$ 0.005% at the start of a run.

^{133}Xe - An impurity of $^{131}Xe^{m}$ was detected. This was 0.009% at the start and 2% at the end of the run. If the correction for this impurity is not made, the half-life value would be longer by 0.0006 day.

$^{134}Cs^{m}$ - Impurities of ^{24}Na and ^{134}Cs were detected; these were $\simeq$ 0.5% at the start of a run.

^{137}Cs - An impurity of 0.03 ± 0.01 % ^{134}Cs was present.

$^{137}Ba^{m}$ - An impurity of ^{137}Cs was present (0.003 % at the start).

^{141}Ce - Impurities of ^{139}Ce, ^{152}Eu and ^{154}Eu were present in the sample used for the half-life measurement. The correction for these was 0.04% at the beginning and 0.23% at the end of the measurement period.
Impurities of ^{139}Ce (0.11%) and ^{46}Sc (0.01%) were present in the sample used for P_{γ} determination.

^{152}Eu - An impurity of ^{154}Eu ($\approx$ 0.5%) was detected.

^{169}Yb - An impurity of ^{170}Tm was detected. This was 0.01% at the start of the run and 7.3% at the end. If the correction is neglected, the half-life value would be 0.010 day longer.

^{198}Au - It was calculated that the irradiation which produced this material should have produced 0.02% ^{199}Au. The effect of this on the ^{198}Au half-life value is 0.0001 day. Two separate irradiations gave values that differed by 0.0004 day.

^{203}Hg - Impurities of ^{60}Co and $^{110}Ag^{m}$ were detected. These were < 0.01% at the beginning and 3.6% at the end of the run.