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EVALUATION OF THE ACCURACY OF DECAY SCHEME CONSTANTS AND CALIBRATION STANDARDS FOR CESIUM-137

JULY, 1965

BATTELLE-NORTHWEST
RICHLAND, WASHINGTON

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EVALUATION OF THE ACCURACY OF DECAY SCHEME CONSTANTS AND CALIBRATION STANDARDS FOR CESIUM-137

By
W. Y. Matsumoto

Analytical Laboratories Section
Chemistry Department

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PACIFIC NORTHWEST LABORATORY
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INTRODUCTION

Accurate assay of Cs^{137} radioactivity is important because of its wide use as a γ-ray standard and its use as a burnup monitor for nuclear reactor fuels. Assay accuracy by absolute counting is limited by uncertainties in decay scheme data. Reported values for the three key constants cover the following ranges: half-life, 26 to 33 years; beta branching ratio, 0.92 to 0.97; and total internal conversion coefficient, 0.08 to 0.12.

Cs^{137} decays by beta emission, ~5% to stable Ba^{137} and ~95% to metastable 2.6 min Ba^{137m} which decays by emission of a 662 keV γ-ray (~ 90%) and by internal conversion (~10%). Since there are no suitable prompt coincidence events, standardization by high accuracy coincidence counting methods is not possible. Measurements of the total electron emission rate by $4\pi\beta$ counting must be corrected for the contribution from conversion electrons. Disintegration values computed from absolute "beta" counting rates using the range of values reported for the internal conversion coefficient and the beta branching ratio have a spread of about 3% relative. Disintegration values calculated on the same basis from total γ-ray emission rates have a spread of about 9% relative.

Accurate knowledge of decay scheme constants would greatly reduce the area of uncertainty associated with the standardization of Cs^{137} by absolute counting methods. Recently several means became available to narrow that area of doubt. Decay scheme data determined by especially accurate experimental methods were recently published. Also, a Cs^{137} standard that has been standardized in terms of Cs^{137} atoms/volume is available for cross-checking absolute counting methods. That standard, the Cs^{137}-ASTM standard issued by the Nuclear Chicago Corporation, was standardized by the isotopic dilution method using mass spectrometry.[1,2] It was also standardized in terms of γ-ray emission by the National Bureau of Standards.

An experimental cross-check program and a detailed study of the literature appeared to offer the soundest basis for choosing the best decay scheme values for Cs^{137}. This paper presents the data obtained in that work and the constants chosen for Cs^{137} calibration work done at Battelle-Northwest.

SUMMARY

A detailed review of published values for the decay scheme constants of Cs^{137} was made. The total internal conversion coefficient of 0.110 reported by Merritt and Taylor[3] was chosen as the most reliable value published for that constant. Values reported for the beta branching ratio since 1958 have a relative spread of about 4%. Half-life values since 1960 have a relative spread of about 10%.

An experimental program of absolute beta counting and γ-ray spectrometry was conducted on certified standards obtained from the National Bureau of Standards (NBS), the International Atomic Energy Agency (IAEA) and the Nuclear Chicago Corporation (NC). Measured values by this laboratory are consistent with a half-life of 31.1 years, a beta branching ratio of 0.954, and a total internal conversion coefficient of 0.110.

A weighted estimate, with all of the interrelated parameters considered, was made to assign "best estimates" to the Cs^{137} decay scheme constants and to the ASTM-Cs^{137} standard. The values were chosen to be internally consistent and to minimize the conflict within the published data and with experimental results. The "best estimates" together with the closest published values for the Cs^{137} decay scheme and the certified values for the ASTM-Cs^{137} standard are:

	"Best Estimate"	Published Value
Half-life years	30.76	30.72 ± 0.12[52]
β Branching Ratio, 518 keV β	0.954	0.952 ± 0.01[3]
Internal Conversion Coefficient	0.110	0.110 ± 0.0011[3]
γ/sec/ml, ASTM-Cs^{137} Standard	1.05×10^5	1.05×10^5
Atoms/ml, ASTM-Cs^{137} Standard	1.71×10^{14}	1.72×10^{14}

The difference between the value of 31.12 yr calculated for the half-life later in the "Cs^{137} Half-Life" section of this paper and the "best estimate" of 30.76 yr given above, reflects our attempt to minimize the conflict between published values and our experimental data. The best estimate of 1.71×10^{14} atoms/ml for the ASTM-Cs^{137} standard differs from the 1.72×10^{14} atoms/ml certified value for the same reason.

EXPERIMENTAL AND DISCUSSION

Cs^{137} Decay Scheme

Although the decay scheme for Cs^{137} is known to be relatively simple (Figure 1), it has resisted accurate definition for several reasons.

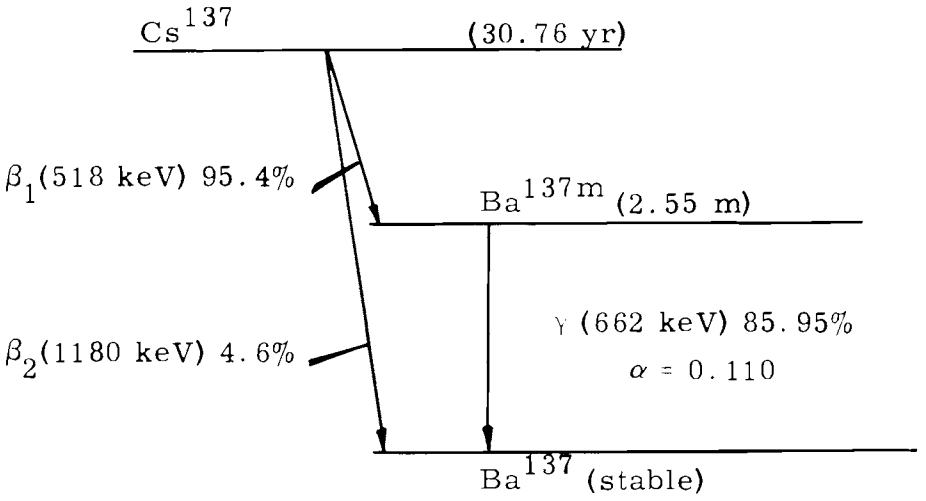

FIGURE 1
Cs^{137} Decay Scheme

Until recently, Cs^{137} free from Cs^{134} contamination was not available. The absence of suitable prompt coincidence events in the Cs^{137} decay scheme limited the accuracy with which absolute counting measurements could be made. Beta spectrometer measurements to determine the beta branching ratio and the internal conversion coefficient with precisions and accuracies of much better than ± 4% are apparently not feasible.

The estimates given in the decay scheme shown in Figure 1 are based on weighed evaluations of published data and experimental measurements on several Cs^{137} standards calibrated at other laboratories. Extra weight was given to measurements made on the ASTM-Cs^{137} standard.

Conversion of Measured γ-Ray and Electron Emission Rates to Disintegrations

The equations for computing disintegration values from counting measurements are:

(1) Cs^{137} disintegrations/min/ml = $\frac{(\gamma/\text{min/ml})(1+\alpha)}{P_1}$

(2) Cs^{137} disintegrations/min/ml = $\frac{\text{Total Electron Emissions/min/ml}}{1+\frac{P_1\alpha}{1+\alpha}}$

where: α = the internal conversion coefficient for 662 keV transition from Ba^{137m} (2.55 min) to stable Ba^{137}. $\alpha = e^-/\gamma$ or the ratio of conversion electron emission (e^-) to the rate of γ-ray emission.

P_1 = the beta branching ratio to Ba^{137m}.

The effect of uncertainties in the values chosen for the decay constants on the calculation of disintegration rates can be deduced from examination of the previous equations. Disintegration rates computed from γ-ray emission rates are quite sensitive to uncertainties in the value chosen for P_1, but relatively insensitive to uncertainties in the value used for α when $\alpha << 1$. By contrast, disintegration rates based on total electron emission rates (beta particles plus internal conversion electrons) are relatively insensitive to uncertainties in the values chosen for α and P_1 for values in the range of interest.

Internal Conversion Coefficient

Values which have been reported for the internal conversion coefficient, α, for the 662 keV transistion of Ba^{137m} to the ground state are given in Table I.

TABLE I

INTERNAL CONVERSION COEFFICIENTS REPORTED FOR THE 662 keV TRANSITION OF Ba^{137m}

α	α_K	Reference	
0.12		Townsend[5]	1948
0.118		Mitchell[6]	1949
0.081		Osaba[7]	1949
0.119[a]	0.097 ± 0.003	Waggoner[8]	1950
0.117[a]	0.095 ± 0.005	Heath[9]	1952
0.135[a]	0.11 ± 0.01	Dolishnyk[10]	1953
0.118[a]	0.096 ± 0.005	Azuma[11]	1954
0.113[a]	0.092 ± 0.006	Wapstra[12]	1954
0.117[a]	0.095 ± 0.008	McGowan[13]	1957
0.114 ± 0.002		Ricci[14]	1957
0.120[a]	0.0976 ± 0.0055	Yoshizawa[15]	1958
0.114[a]	0.093 ± 0.006	Hultberg[16]	1959
0.114[a]	0.093 ± 0.005	deVries[17]	1960
0.117[a]	0.095 ± 0.004	Hultberg[18]	1961
0.114 ± 0.022	0.093 ± 0.003	Daniel[19]	1962
0.110 ± 0.0011	0.0894 ± 0.0011	Merritt[3]	1965

(a) Calculated from α_K reported using K/L and L/MN from Reference (15).

Merritt and Taylor[3] have determined α with the smallest uncertainty reported to date. They separated 2.6 min Ba^{137m} from the Cs^{137} by rapid ion-exchange method and made direct measurements of the conversion electron emission rate and γ-ray emission rate on the same Ba^{137m} source. Those measurements were used to calculate, $\alpha = e^-/\gamma$, directly, free from uncertainties inherent in the methods used for previously reported values. Their value of $\alpha = 0.110 \pm 0.0011$ was adopted for the decay scheme presented in this work.

Beta Branching Ratio

The beta branching ratio of direct concern is P_1, which is equal to the probability for the 518 keV (β_1) decay of Cs^{137} to Ba^{137m}. Values reported in the literature to date are listed in Table II.

TABLE II

REPORTED BRANCHING RATIOS FOR THE 518 keV Cs^{137} BETA

>0.95, <0.98	Osaba[7]	1949
0.95	Peacock[20]	1949
0.92	Langer[21]	1951
0.97	Wapstra[12]	1954
0.952 ± 0.003	Ricci[14]	1957
0.924 ± 0.008	Yoshizawa[15]	1958
0.935 ± 0.002	Daniel[19]	1962
0.965 ± 0.01	Merritt[22]	1963
0.952 ± 0.01	Merritt[3]	1965
0.954	This work	

The value of 0.954 given in the decay scheme shown in Figure 1 is weighted average of that reported by Merritt and Taylor[3] and measurements made at Battelle-Northwest.

Cs^{137} Half-Life

Reported values for the half-life of Cs^{137} are given in Table III. Values reported since 1960 range from 27 to 32.6 yr. Limits of uncertainty reported on the individual values range from ± 0.05 to ± 1.6 yr. In general, the limits are much too small to account for the wide spread of the values.

TABLE III

Cs^{137} HALF-LIFE MEASUREMENTS REPORTED

Half-Life, yr	Method of Measurement	Reference
33 ± 3	Specific activity measured in three monitored irradiations. 6.0% used for the fission yield.	L. E. Glendenin[23] pre-1951
33 ± 2	Observed change in the Cs^{137}/Cs^{133} ratio over 5 yr by mass spectrometry.	D. R. Wiles[24] 1953
26 ± 1	Observed change in the Cs^{137}/Cs^{133} ratio over 8 yr by mass spectrometry.	Melaika[25] 1954
26.6 ± 0.4	Specific activity ($4\pi\beta$ counting); Cs^{137} determined by isotopic dilution-mass spectrometry.	D. M. Wiles[26] 1955
30.0 ± 0.4	Specific activity ($4\pi\beta$ counting); Cs^{137} determined by isotopic dilution-mass spectrometry.	F. Brown[27] 1955
28.6^{+2}_{-1}	Specific activity ($4\pi\beta$ counting); Cs^{137} determined by isotopic dilution-mass spectrometry.	A. J. Moses[28] 1958
28.4 ± 1.4	Specific activity (γ-ray spectrometer, Nuclear Chicago Corp. Cs^{137} standard); Cs^{137} from ANL irradiation—6.14% F. Y.	B. F. Rider[29] 1960
32.6 ± 1.6	Specific activity ($4\pi\beta$ counting); Cs^{137} determined by isotopic dilution-mass spectrometry.	L. E. Glendenin[30] 1960
30.4 ± 0.4	Observed rate of Ba^{137} formation in a measured amount of Cs^{137} by isotopic dilution-mass spectrometry. (50 days)	H. Farrar[31] 1960
27	Utilized large quantities (~50 g) of purified fission product CsCl analyzed mass spectrographically.	T. A. Butler[32] 1960

TABLE III (Contd.)

Cs^{137} HALF-LIFE MEASUREMENTS REPORTED

Half-Life, yr	Method of Measurement	Reference
29 ± 1	Radiometric	M. P. Glazunov[33] 1961
29.15 ± 0.25	Observed rate of Ba^{137} formation in a measured amount of Cs^{137} by isotopic dilution-mass spectrometry, 100 days.	B. F. Rider[1] 1962
30.0 ± 0.7	Specific activity (liquid scintillation absolute β-counting); Cs^{137} determined by isotopic dilution-mass spectrometry.	D. G. Fleishman[34] 1962
30.1 ± 0.7	Mass spectrometry	I. V. Burovina[35] 1963
29.2 ± 0.3	Observed rate of Ba^{137} formation from a measured amount of Cs^{137} by isotopic dilution-mass spectrometry; 100 days.	B. F. Rider[2] 1963
30.35 ± 0.38	Observed change in the Cs^{137}/Cs^{135} ratio over ~1 1/2 yr, using the Cs^{135}/Cs^{133} ratio as an internal standard (mass spectrometry).	L. A. Dietz[4] 1963
29.68 ± 0.05	Observed radioactive decay rate with a well-type ionization chamber, ~ 3 yr. Corrected for Cs^{134}, 0.5% ± 0.05%.	S. G. Gorbics[36] 1963
30.7 ± 0.6	Observed radioactive decay rate with a well-type ionization chamber and a γ-ray spectrometer, ~ 4 yr.	S. A. Reynolds[37] 1963
30.6 ± 0.7	Observed radioactive decay rate with a well-type ionization chamber and a γ-ray spectrometer, ~5 yr.	S. A. Reynolds[48] 1964
30.8 ± 0.3	Specific activity.	R. E. Lewis[49] 1964

TABLE III (Contd.)

Cs^{137} HALF-LIFE MEASUREMENTS REPORTED

Half-Life, yr	Method of Measurement	Reference
29.4 ± 0.18	Difference in 133/135/137 ratios measured by mass spectrometry 5 yr apart.	H. D. Cook[50] 1964
29.3 ± 0.6	Observed radioactive decay with a $2\pi\beta$ proportional counter, 10.2 yrs.	K. F. Flynn[51] 1964
31.4 ± 0.9	Observed radioactive decay with a $2\pi\beta$ proportional counter, 6.6 yrs.	K. F. Flynn[51] 1964
30.9 ± 0.7	Specific activity. Average of 10 independent fission product sources.	K. F. Flynn[51] 1964
30.55 ± 1.55	Observed change in 137/135 and 137/133 ratios by mass spectrometry. Average of 6 measurements.	J. H. Gillette[52] 1965
29.78 ± 0.14	Determination of Cs^{137} and Ba^{137} by weight and mass spectrometry. Average of 3 measurements.	J. H. Gillette[52] 1965
30.72 ± 0.12	Determination of CS^{137} specific activity by weight, curie content, and mass spectrometry. Average of 20 measurements.	J. H. Gillette[52] 1965
30.76 ± 0.5	Weighted estimate from specific activity ($4\pi\beta$ counting and calibrated γ-ray spectrometry), ASTM-CS^{137} standard, and published values.	This work.

A half-life was calculated from the certified atoms/ml calibration of the ASTM-Cs^{137} standard and the disintegrations/min/ml value measured by absolute ($4\pi\beta$) counting.

The following equation was used:

$$N\lambda e^{-\lambda t} = (E/min/ml) \times \left[1 + \frac{P_1 \alpha}{1 + \alpha} \right]^{-1}$$

where: $N = 1.72 \times 10^{14}$ Cs^{137} atoms/ml on 8/8/62.

t = time in minutes from 8/8/62 to 2/13/65.

$E/min/ml = 7.542 \times 10^{6}$ electrons emitted/min/ml on 2/13/65.

$\alpha = 0.110$

$P_1 = 0.954$

The equation was solved by iteration using a desk calculator giving:

$\lambda = 4.2375 \times 10^{-8}$ min^{-1}, from which,

$T_{1/2} = \frac{\ln 2}{\lambda} = 1.6357 \times 10^{7}$ min = 31.12 yr was calculated.

Calculation of $T_{1/2}$ from the certified atoms/ml and γ-ray emission rate calibrations using the same decay scheme values yields a value for $T_{1/2}$ of 30.94 yr.

The spread in the Cs^{137} half-life values reported by different investigators is such that an uncertainty of about 5%, in the direction of a mean value, must be placed on several of these half-life figures. The half-life of 30.76 yr given in Figure 1 is an estimate based on measurements made here on the ASTM-Cs^{137} standard, certified calibration values of that standard, and published half-life values. The estimate is weighted to minimize the descrepancies among the bases.

The probable uncertainty in the atoms/ml value of the ASTM-Cs^{137} standard is about 1%, based on the 0.7% uncertainty reported by Rider[2] for the Cs^{137} determination and allowing for systematic error introduced

during subsequent dilutions and aliquoting. The best estimate of 1.71×10^{14} atoms/ml on 8/8/62 given in the summary was computed from the best estimates adopted for the other parameters.

Absolute Counting

Cs^{137} standards were calibrated for Hanford distribution by $4\pi\beta$ counting using published decay schemes to correct for internal conversion electrons. Standards purchased from off-site which were calibrated in terms of γ-ray emission were used as cross-check sources. Published decay schemes were used to convert γ-ray emission rates to disintegration rates. For the 8/1/61 standard from the Nuclear Chicago Corporation, the disintegration value based on $4\pi\beta$ counting was about 7.4% lower than the value computed from the certified γ-ray emission rate.

All of that difference can now be explained on the basis of the decay scheme values used and by taking into account the effect of Cs^{134} contamination in the Cs^{137} standards.

Cs^{134} causes a positive bias in γ-ion chamber measurements used by both Nuclear Chicago and the National Bureau of Standards to calibrate Cs^{137} γ-ray standards. The magnitude of the bias caused[38] is about 2.6 times the fraction of the Cs^{134} present. This stems from the fact that the decay of Cs^{134} is accompanied by several times more γ-ray energy than is the decay of Cs^{137}. Originally computed values and corresponding values after adjustment for Cs^{134} contamination and computed using the decay scheme shown in Figure 1 are given in Table IV.

Early measurements were made by simple $4\pi\beta$ counting. Aliquots of the solutions to be standardized were taken with calibrated micro pipettes and dried on gold coated VYNS films[39] which were supported on 0.004 in. thick aluminum rings. Colloidal silica treatment[40] was used to minimize self absorption losses. A correction for absorption losses in the film mount was calculated by the method of Seliger and Cavallo.[41, 42] Although shown to be invalid in some cases,[43] particularly for low energy beta emitters, the method gives reasonably good results for beta emitters of moderate maximum beta energy such as Cs^{137}.

TABLE IV

THE EFFECT OF Cs^{134} ON γ-ION CHAMBER AND ABSOLUTE BETA COUNTING STANDARDIZATIONS OF Cs^{137}

Standard[a]	Date[b] 4πβ	γ-Calib.[c] Disintegration/min/ml	4πβ[d] Disintegration/min/ml	Corrected for Cs^{134}[e] γ-Calib. Disintegration/min/ml	Corrected for Cs^{134}[e] 4πβ Disintegration/min/ml
NC(8/1/61)	6/11/62	6.42×10^7	6.01×10^7	6.08×10^7	5.94×10^7
	2/13/65	6.42×10^7	6.08×10^7	6.08×10^7	6.05×10^7

(a) Nuclear Chicago Corporation Cs^{137} γ-ray standard, reference date 8/1/61. All disintegrations/min/ml values are corrected for radioactive decay to the reference date.

(b) Date of 4πβ measurement. (Data are corrected to standard reference date.)

(c) Computed from certified γ-ray emission value using the decay scheme given in Figure 1.

(d) Computed from Battelle-Northwest 4πβ counting measurements using the decay scheme given in Figure 1.

(e) Values from (c) and (d) after correction for Cs^{134} contamination (2.07% of Cs^{137} on 8/1/61).

A recent measurement (2/13/65) of the Nuclear Chicago (7/2/64)Cs^{137} standard was made by the 4πβ-γ coincidence efficiency tracing method[40, 44-46], using Sc^{46} as the tracer. Accurately measured aliquots of the Cs^{137} and Sc^{46} were mounted together on the same film. The 4πβ, Sc^{46} γ-ray, and Sc^{46} β-γ coincidence counting rates for the mixed source were measured simultaneously under conditions for maximum beta counting efficiency. Both sides of the source were covered with aluminum leaf absorber (~180 $\mu g/cm^2$) and the source recounted. The process was repeated for several additional layers of aluminum and leaf and for several sources. The contribution from Sc^{46} to the mixed source 4πβ counting rate was determined from independent measurements, and subtracted after making the usual background and deadtime corrections. The net 4πβ counting rate due to the Cs^{137} was plotted versus the Sc^{46} 4πβ counting efficiency for each measurement. Extrapolation by least squares to 100% efficiency gives the total electron emission rate.

Comparisons of Battelle-Northwest disintegration values for Cs^{137} obtained by 4πβ counting of off-site standards versus corresponding values

computed from certified γ-ray emission and atoms/ml values are shown in Table V and VI, respectively. The decay scheme values in Figure 1 were used to convert the electron and γ-ray emission rates and the atoms/ml value to disintegration rates.

TABLE V

ABSOLUTE BETA COUNTING VALUES FOR OFF-SITE Cs^{137} STANDARDS VERSUS VALUES BASED ON CERTIFIED GAMMA-RAY EMISSION RATES

Standard[a]	Date 4πβ[b]	γ-Calibration[c] Disintegration/ min/ml x 10^{-6}	4πβ[d] Disintegration/ min/ml x 10^{-6}	Relative[e] Difference
NC(8/1/61)	6/11/62	60.8	59.4	-2.30%
	2/13/65[f]	60.8	60.5	-0.49%
NC(8/8/62)	1/31/63	7.33	7.36	+0.38%
	8/3/64[f]	7.33	7.30	-0.41%
	2/13/65[f]	7.33	7.30	-0.45%
NC(7/2/64)	2/13/65[g]	87.6	86.7	-0.94%
NBS(8/1/63)	2/13/65[f]	3.48	3.45	-1.07%
IAEA(12/12/63)	3/19/64	22.5[h]	22.9	+1.74%
	8/3/64	22.5[h]	22.7	+0.86%

(a) Standard and reference date of standard. NC = Nuclear Chicago Corp.; NBS = National Bureau of Standards; IAEA = International Atomic Energy Agency, Vienna.

(b) Date of 4πβ measurement. (Data are corrected to standard reference date.)

(c) Certified γ-ray emission rate converted to disintegrations/min using decay scheme in Figure 1. Corrections for Cs^{134} made when necessary.

(d) Values from Battelle-Northwest 4πβ counting using decay scheme in Figure 1.

(e) (d) minus (c) divided by (c).

(f) Indirect determination by γ-ray spectrometer comparison with Cs^{137} solution calibrated by 4πβ counting.

(g) 4πβ-γ coincidence efficiency tracing method using Sc^{46} as the tracer.

(h) IAEA value by 4πβ-γ coincidence efficiency tracing with Co^{60} tracer.

TABLE VI

ATOMS Cs^{137} VERSUS DISINTEGRATIONS Cs^{137} FOR ASTM STANDARD

Standard	Date[a] $4\pi\beta$	Certified Value Atoms/ml	Certified Value[b] Disintegration/ min/ml x 10^{-6}	$4\pi\beta$[c] Disintegration/ min/ml x 10^{-6}
NC(8/8/62) (ASTM STD.)	1/31/63	1.72×10^{14}	7.37	7.36
	8/3/64	1.72×10^{14}	7.37	7.30[d]
	2/13/65	1.72×10^{14}	7.37	7.30[d]

(a) Date of $4\pi\beta$ measurement. (Data are corrected to standard reference date.)

(b) Atoms per milliliter converted to disintegrations/min/ml using a 30.76 yr half-life.

(c) $4\pi\beta$ measurements converted to disintegrations. min/ml using decay scheme in Figure 1.

(d) Indirect $4\pi\beta$ measurements by γ-ray spectrometer comparison with a Cs^{137} solution calibrated by $4\pi\beta$ counting.

Total electron emission rate measurements of Cs^{137}-Ba^{137m} solutions by the tracer method and $4\pi\beta$ counting have an uncertainty of 1 to 2%, an estimate based upon the 2% uncertainty cited by the IAEA for their Cs^{137} standard, the work of Merritt and Taylor[3] and the measurements made in this laboratory.

Gamma-Ray Emission Measurements

The primary γ-ray emission rate calibrations of the Nuclear Chicago and National Bureau of Standards Cs^{137} standards are made with well-type γ-ionization chambers. These ion chambers were calibrated using radionuclide sources standardized by absolute counting methods.

Measurements on the γ-ray emission rates of the ASTM, NC (8/8/62), and the NBS 8/1/63) Cs^{137} standards were made here using a calibrated 3 by 3 in. NaI(Tl) crystal γ-ray spectrometer. The γ-ray emission rates

were determined using photopeak response curves obtained with radionuclide sources standardized by absolute counting methods. The maximum error in the response curves in the 660 keV γ-ray energy region used, is estimated to be less than ±2.5%. The measured γ-ray emission rates of both standards checked their respective certified values to within 0.5%.

The uncertainty in the measurement of the 662 keV γ-ray emission rate of Cs^{137}-Ba^{137m} was estimated to be about 1%. The excellent agreement between independent measurements at the NBS and at this laboratory, and good agreement between measurements made here and those by Merritt and Taylor[3] at the Chalk River Nuclear Laboratories, deduced from data presented in their paper, were used to justify this estimate.

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