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THE HALF-LIFE
AND
GAMMA-RAY ABUNDANCE
OF Cs-137

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<p>SUMMARY</p> <p>The half-life of cesium-137 has been determined to be 29.2 ± 0.3 years by an all mass spectrometric method.</p> <p>The abundance of the 662 Kev gamma-ray in the decay scheme of Cs-137 was found to be 0.82 ± 0.03 per beta disintegration.</p> <p>In the course of the work, the isotopic distribution of natural barium was determined with improved accuracy to be $(72.11 \pm 0.06)\%$ 138, $(11.18 \pm 0.05)\%$ 137, $(7.77 \pm 0.02)\%$ 136, $(6.42 \pm 0.03)\%$ 135, $(2.33 \pm 0.02)\%$ 134, $(0.091 \pm 0.003)\%$ 132, and $(0.098 \pm 0.002)\%$ 130.</p>		

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ABSTRACT

The half-life of cesium-137 has been determined to be 29.2 ± 0.3 years by an all mass spectrometric method.

The abundance of the 662 Kev gamma-ray in the decay scheme of Cs-137 was found to be 0.82 ± 0.03 per beta disintegration.

In the course of the work, the isotopic distribution of natural barium was determined with improved accuracy to be $(72.11 \pm 0.06)\%$ 138, $(11.18 \pm 0.05)\%$ 137, $(7.77 \pm 0.02)\%$ 136, $(6.42 \pm 0.03)\%$ 135, $(2.33 \pm 0.02)\%$ 134, $(0.091 \pm 0.003)\%$ 132, and $(0.098 \pm 0.002)\%$ 130.

SECTION I

INTRODUCTION

The nuclide Cs-137 is a fission product commonly used for measurement of uranium burnup in irradiated uranium fuel by the fission product to uranium ratio method. In the application of this method, the largest single error introduced in the measurement of burnup is the uncertainty in the half-life of Cs-137⁽¹⁾. The Cs-137 half-life has been reported from specific activity measurements to be 28.4 ± 1.4 ⁽¹⁾, 26.6 ± 0.4 ⁽²⁾, 30.0 ± 0.4 ⁽³⁾, $28.6 \pm \frac{2}{1}$ ⁽⁴⁾, 32.6 ± 1.6 ⁽⁵⁾, 27 ⁽⁶⁾ and 29 ± 1 years⁽⁷⁾, and from mass spectrometric measurements to be 30.4 ± 0.4 years⁽⁸⁾.

Because of the uncertainty in this value and its importance in nuclear fuel burnup analysis, a reinvestigation was undertaken to obtain a more accurate value using the mass spectrometric method.

SECTION II

SUMMARY

The half-life of Cs-137 has been determined from the rate of formation of daughter Ba-137 in a measured amount of Cs-137 by isotopic dilution mass spectrometry. Only one such measurement, 30.4 ± 0.4 years⁽⁸⁾, has been previously reported.

The barium daughters initially present in a 200 mc. source of fission product cesium were removed by Dowex-50 cation exchange techniques. After sampling for mass analysis, the cesium eluate was added to a polyethylene flask containing accurately weighed aliquots of standardized Cs-133 and 98.6% enriched Ba-138 spike solutions.

The Cs-133 solution was a primary standard prepared by weight from spectrographically pure natural CsCl. The Ba-138 was standardized by isotopic dilution with a primary standard prepared by weight from analyzed C.P. natural $\text{BaCl}_2 \cdot 2 \text{H}_2\text{O}$. By isotopic dilution, the initial number of Cs-137 and Ba-138 atoms in the flask were known to a standard deviation of 0.68% and 0.42% respectively.

To establish the rate of growth of Ba-137, small samples of the master solution were analyzed for barium isotopic abundances over a period of 100 days. The barium was freed of cesium by cation exchange prior to analysis. Accurately known sample volumes or chemical yields were not necessary because the initial solution contained the enriched Ba-138 spike.

Two solid emission mass spectrometers, a two-stage magnetic analyzer, using an electron multiplier detector and a single-stage magnetic analyzer with a Faraday cage detector and vibrating reed electrometer, were used to determine the mass ratios.

In barium samples, the mass 133 position was used to monitor for Cs-137 contamination, which was found to be negligibly small. The mass-136 position was used to correct for natural barium contamination. Small corrections were applied for Ba-137 content of the Ba-138 spike and the effect of Cs-137 decay.

The slope of the Ba-137 growth curve (Figure 1) was determined to a standard deviation of 0.50% from a weighted least squares analysis of the data. From these data, the half-life was determined to be 29.2 ± 0.3 years at one standard deviation. This value is consistent with the average of the values referenced and has a smaller uncertainty than any of the previous measurements.

The gamma-ray abundance of Cs-137 was measured by gamma counting ampoules with a known number of atoms of Cs-137 on a sodium iodide gamma spectrometer calibrated with Nuclear Chicago Standard Cs-137 sources. The abundance of the 662 Kev gamma ray in the decay scheme of Cs-137 was found to be 0.82 ± 0.03 per beta disintegration.

Figure 1. RATE OF GROWTH OF Ba^{137}

SECTION III

EXPERIMENTAL AND RESULTS

A description of the experimental work performed to measure the Cs-137 half-life can be logically arranged into the following sections: (a) development of an ion exchange separations scheme for isolating barium daughters from a Cs-137 sample in preparation for mass spectrometry; (b) measurement of the number of parent Cs-137 atoms and the number of Ba-138 tracer atoms present in the stock solution by isotopic dilution mass spectrometry; (c) description of the mass spectrometers used in this work; (d) measurement of the growth of Ba-137 atoms with respect to the Ba-138 spike as a function of time; (e) calculation of Cs-137 half-life; and finally, (f) measurement of the gamma ray abundance of Cs-137. Each section will be described in some detail.

A. Ion Exchange Studies

The ion exchange behavior of barium and cesium on cation resin in HCl systems was investigated with small columns of 2.5 mm I.D. containing 10 mm length of 200 - 400 mesh Dowex-50 x 8. The volume of resin per column was about 50 μ liters. The Cs-137 activity was loaded on the column and eluted with 1M HCl at a flow rate of 1 cc/min. The specific activity of each four-drop fraction (18 drops per ml) is plotted in Figure 2. After 52 drops, no cesium activity above background could be detected.

A similar study was performed with Ba-139 tracer using 1M HCl as the eluant (Figure 2). Very little barium was eluted in the first 40 drops (18 drops per ml). By drop 60, about 2% of the barium activity was lost from the column, and by drop 80, barium losses totaled 8%. After drop 80, the eluant was changed to 6M HCl to elute the barium. Over 80% of the barium was eluted in the first 10 drops of 6M HCl.

Figure 2. CESIUM AND BARIUM ELUTION
2.5mm x 10mm Column of Dowex - 50 x 8 (200 - 400 mesh)

Based on these observations, the following procedure was adopted for the purification of barium daughters from Cs-137. After loading up to 10 drops of Cs-137 in 1M HCl, the column was washed with 80 drops of 1M HCl to remove all Cs-137. The barium daughters were then eluted with 10 drops of 6M HCl, evaporated to dryness, and mounted with 0.05M HCl on a rhenium filament for mass analysis.

Every effort was made to minimize natural barium contamination. Constant boiling (6M) HCl was distilled and stored in leached polyethylene bottles. The 1M HCl was prepared by dilution of this acid with deionized distilled water. The glass columns, pipets, and test tubes used to collect the eluate fractions were boiled in 6M HCl and rinsed in deionized distilled water. Any natural barium contained in the resin was removed by elution with 40 column volumes of 6 M HCl for each column; this was followed by conditioning with 1M HCl prior to sample loading.

B. Isotopic Dilution Techniques

Having established an ion exchange separation procedure for separation of cesium and barium, a 200 millicurie batch of ORNL Cs-137 was passed through a large Dowex-50 column and eluted with twenty column volumes of 1M HCl in order to strip out all barium daughters present. The resulting Cs-137 solution was mass analyzed for a total of 47 mass spectrum scans on 3 filaments. The atom ratios of 133/137 and 133/135 were observed to be 1.246 ± 0.006 and 2.505 ± 0.009 , respectively. For consistency, all errors quoted are at one standard deviation. This solution was added to a leached polyethylene bottle containing a teflon covered stirring bar plus weighed aliquots of standardized Cs-133 and enriched Ba-138 solutions and was thoroughly stirred to achieve isotopic exchange. These two isotopic diluents were selected because they were available in high purity.

The Cs-133 solution was a primary standard prepared by weight from chemically pure CsCl. The most common impurity in this chemical is usually rubidium. The rubidium content of the salt was therefore

analyzed flame photometrically and found to be 0.02% by weight. The salt was then analyzed by emission spectroscopy to be 99.95% pure. The impurities found are listed in Table I. The isotopic analysis of the cesium standard was verified to be 100% Cs-133 with other mass positions from 129 to 138 less than 0.01% of the 133 peak. The analysis of the master Cs-137 stock solution isotopically diluted with Cs-133 standard was found from 183 scans on ten filaments. The observed atom ratios of 133/137 and 133/135 were 50.83 ± 0.32 and 104.9 ± 0.72 respectively. Appendix II shows the calculation of the number of Cs-137 atoms present initially in the master solution to be $1.044_4 \times 10^{19} \pm 0.68\%$.

The Ba-138 standard solution was standardized by isotopic dilution with a primary standard prepared by weight from C.P. $\text{BaCl}_2 \cdot 2 \text{H}_2\text{O}$ analyzed by the BaSO_4 gravimetric procedure to be 99.88% pure. The principal impurities present are listed in Table I. The mass analysis of natural barium was obtained from 86 scans on 8 filaments and the mass analysis of the enriched Ba-138 was obtained from 63 scans on 5 filaments. Both analyses are summarized in Table II. Four weighed aliquots of the enriched Ba-138 standard solution were isotopically diluted with weighed portions of the natural barium standard solution. The calculations of the Ba-138 tracer atoms in the master solution as calculated from 129 scans on 7 filaments are shown in Appendix III. A weighted average of all determinations show $1.175_2 \times 10^{17} \pm 0.43\%$ atoms Ba-138 tracer atoms present in the master solution.

C. Mass Spectrometry

All isotopic analyses of cesium samples, and natural enriched barium samples, before and after isotopic dilution, as well as most of the Ba-137 daughter growth samples, were performed on single-stage solid emission mass spectrometer employing a Faraday cage detector and vibrating reed electrometer. This instrument makes use of a secondary emission suppressor and measures only primary beam current. For this reason, and because ions are accelerated at constant potential while mass positions are scanned magnetically, mass discrimination should not

TABLE I

Impurity Analyses of Primary Cesium and Barium Standards

CsCl		BaCl ₂ · 2 H ₂ O	
Impurity	ppm	Impurity	ppm
Rb	250 ± 50	Sr + Ca	1000
Na	100	Insolubles	50
Fe	100	Heavy Metals	50
Al	50	Fe	20
Ca	10		
Si	10		
Zr	5		
Sn	5		
Mo	5		
Cu	5		
Co	5		
Cr	5		
Tc	4		
B	1		
Mg	1		
Mn	1		
Ag	< 1		
Cd	< 1		
K	< 1		
Li	< 1		

TABLE II

Isotopic Analysis of Natural Barium and
Enriched Ba-138 Used in This Study

Isotope	Natural Ba, Atom %	Enriched Ba-138, Atom %
Ba-138	72.11 $\pm$ 0.06	98.56 $\pm$ 0.01
Ba-137	11.18 $\pm$ 0.05	1.24 $\pm$ 0.01
Ba-136	7.77 $\pm$ 0.02	0.136 $\pm$ 0.004
Ba-135	6.42 $\pm$ 0.03	0.052 $\pm$ 0.001
Ba-134	2.33 $\pm$ 0.02	0.0134 $\pm$ 0.0004
Ba-132	0.091 $\pm$ 0.003	0.00035 $\pm$ 0.00004
Ba-130	0.098 $\pm$ 0.002	0.00054 $\pm$ 0.00005

occur nor was it observed. Ba-137 growth samples were chemically processed almost daily for the first two weeks and approximately every other day thereafter.

Initially, Ba-137 growth samples were analyzed on a more sensitive two-stage magnetic analyzer using an electron multiplier detector because of the greater sensitivity of this instrument. This instrument uses a fixed magnetic field while the mass positions are scanned electrostatically. Mass discrimination occurs at the first dynode electron multiplier due to variation in secondary emission with ion velocity. This mass discrimination was measured and a correction made.

D. Measurement of Ba-137 Daughter Growth

The growth of Ba-137 daughter was observed by noting the increase in the mass 137/138 ratio with time. In order to avoid interference in the mass 137 position from (a) Cs-137, (b) Ba-137 from natural barium contamination, and (c) Ba-137 traces in the Ba-138 spike and in order to avoid interference from natural Ba-138 contamination in the mass 138 position, an equation was developed (Appendix IV) to allow for correction of each of these interferences by making use of the 133 mass position as a cesium indicator and the 136 mass position as an indicator for natural barium contamination. The final equation is expressed:

$$\frac{\text{Atoms Ba-137 daughter}}{\text{Atoms Ba-138 spike}} = \frac{1 + \frac{\left[\frac{M_{8/6}}{S_{8/6}} - 1 \right]}{1 - \frac{M_{8/6}}{N_{8/6}}}}{M_{8/7} + \frac{\left[\frac{M_{7/3}}{H_{7/3}} - 1 \right]}{M_{8/7}}} - \frac{1}{S_{8/7}} \frac{\left[\frac{M_{8/6}}{S_{8/6}} - 1 \right]}{N_{8/7}}$$

where the symbols are defined in Appendix I.

In practice, only four of the fifty-four data points showed much Cs-133 present and the correction in these cases amounted to less than 1%. Natural barium contamination was larger, however, and was in the order of 13% correction at 100 days of growth and was progressively larger near the origin. Care was taken to propagate into the final 137/138 ratios errors of each value substituted in the equations.

A plot of the Ba-137 daughter atoms per atom of Ba-138 spike as a function of time in days is found in Figure 1. The deviation of the points about the line evident between 20 and 40 days was of concern so that attempts were made to verify analysis of Ba-137 daughter growth samples on the less sensitive single stage instrument. This instrument showed sensitivity for barium much greater than anticipated and samples at 47 and 49 days ran well on both instruments. As a result, all samples from 60 to 75 days were analyzed on both instruments. Values obtained on the single-stage instrument agreed with those obtained on the two-stage instrument, exhibited a smaller scan to scan and point to point variation and eliminated the need for mass discrimination corrections.

After 75 days, all further measurements were made on the single-stage instrument because of its excellent performance. Five samples which had been set aside from the period of 6 to 12 days growth were also analyzed on this instrument. In addition, the final sample at 100 days was analyzed in triplicate because of the importance of last point in determination of the slope and to obtain a further check on the filament to filament variation.

The data in Figure 1 represent the growth curve for a stable daughter in a radioactive parent and do not form a straight line. Daughter growth is proportional to $(1 - e^{-\lambda t})$. However, each point has been multiplied by the factor $\left(\frac{\lambda t}{1 - e^{-\lambda t}} \right)$ to correct for Cs-137 decay and obtain the 137/138 ratio which would be present if no decay of parent had occurred. The plot thus corrected is a linear function with time.

The equation for this line was found by least squares treatment (Appendix VI). The general equation is of the form $y = ax + b$ where y is the atoms Ba-137 daughter per atom Ba-138 spike, x is the time of growth in days, a is the slope, and b is the y intercept. By assuming no error in x (the time scale) since all time measurements were easily measured to 1/1000 of a day, and weighting values of y as the reciprocal of a function of the error in each value, the equation becomes

$$y = (0.00578_7 \pm 0.00002_9) x - (0.000_5 \pm 0.001_6).$$

This weighted treatment of the data was adopted to obtain minimum error in the slope after it was demonstrated that weighted treatment when compared with unweighted treatment of data from each individual mass spectrometer did not differ statistically from spectrometer to spectrometer or from weighted to unweighted treatment.

From the measurement made, it was possible to determine the half-life of Cs-137 and by some additional gamma measurements, the abundance of the 662 Kev gamma-ray per beta disintegration.

E. Calculation of Half-Life

The half-life, $T_{\frac{1}{2}}$, is related to the decay constant, λ , by the expression

$$T_{\frac{1}{2}} = \frac{0.69315}{\lambda}$$

Disintegration rate or activity is related to λ by the expression

$$\frac{-dN_{\text{Cs-137}}}{dt} = \lambda N_{\text{Cs-137}}$$

Solving the equation for λ , one obtains the expression

$$\lambda = \frac{-dN_{\text{Cs-137}}/dt}{N_{\text{Cs-137}}}$$

However, disintegration rate is equal to the atoms of Ba-137 formed per unit time. This quantity is the slope, a , of the equation for the growth line (Appendix VI) multiplied by the atoms of Ba-138 spike, $N_{\text{Ba-138}}$. Therefore

$$\lambda = a \left[\frac{N_{\text{Ba-138}}}{N_{\text{Cs-137}}} \right]$$

Since $N_{\text{Ba-138}}$ spike is $1.175_2 \times 10^{17} \pm 0.43\%$ (Appendix III) and $N_{\text{Cs-137}}$ is $1.044_4 \times 10^{19} \pm 0.68\%$ (Appendix II) and " a " is calculated to be $0.00578_7 \pm 0.50\%$ (Appendix VI), the value of λ becomes $6.51 \times 10^{-5} \pm 0.96\%$ days⁻¹ corresponding to a half-life of 29.2 ± 0.3 years.

F. Determination of Gamma Ray Abundance of Cs-137

Within two hours of the preparation of the master Cs-137 stock solution, seven weighed aliquots were sealed into glass ampoules similar to Nuclear Chicago Cs-137 standards with respect to liquid volume, glass dimensions, and thickness. Since the number of Cs-137 atoms in the master stock solution were known to an accuracy of 0.68% and each of these ampoules represent carefully measured aliquots, each of these ampoules serve as an accurately known number of Cs-137 atoms.

Appendix VI shows 69 comparisons that were made between these ampoules and two Nuclear Chicago Cs-137 standards. Both samples and standards were corrected for their Cs-134 contents. The Cs-137 atom concentration in the Nuclear Chicago standards were determined to 1%. These values are given in line 6 of Table III. It should be noted that the Cs-137 atoms/ml are more accurately known than the certified gamma activities of the standards.

If the atoms/ml of Cs-137 in line 6 are multiplied by λ in minute⁻¹ corresponding to 29.2 year half-life, one obtains the d/m/ml and finally d/s/ml on line 10. By dividing line 5 by line 10, one obtains a gamma abundance of 0.82 gamma photons at 662 Kev per disintegration on line 11.

TABLE III

Nuclear Chicago Cs-137 Standards
Used to Calculate 662 Kev Gamma Ray Abundance in Cs-137

Item	Batch #1	Batch #2
1. Date	2-27-59	8-1-61
2. γ /s/ml total	$6.33 \times 10^5 \pm 3\%$	$9.20 \times 10^5 \pm 3\%$
3. γ /s/ml 600 Kev Cs-134	$- 0.04 \times 10^5 \pm 10\%$	$- 0.18 \times 10^5 \pm 10\%$
4. γ /s/ml 800 Kev Cs-134	$- 0.04 \times 10^5 \pm 10\%$	$- 0.18 \times 10^5 \pm 10\%$
5. γ /s/ml 662 Kev Cs-137	$6.25 \times 10^5 \pm 3\%$	$8.84 \times 10^5 \pm 3\%$
6. Atoms/ml Cs-137 on date in line 1 above	$1.01 \times 10^{15} \pm 1\%$	$1.43 \times 10^{15} \pm 1\%$
7. $\times \lambda \text{ min}^{-1}$	$4.52 \times 10^{-8} \pm 1\%$	$4.52 \times 10^{-8} \pm 1\%$
8. = d/m/ml	$4.57 \times 10^7 \pm 1.4\%$	$6.47 \times 10^7 \pm 1.4\%$
9. $\div 60$	60	60
10. d/s/ml Cs-137	$7.62 \times 10^5 \pm 1.4\%$	$1.08 \times 10^6 \pm 1.4\%$
11. $\gamma/d = \text{line 5} \div \text{line 10}$	$0.82 \pm 3.3\%$	$0.82 \pm 3.3\%$

A large error exists in the 0.82 gamma ray abundance from the 3% error in γ/s certified in these standards. However, where the γ/s with a 3% error is divided by 0.82, the resulting d/s is accurate to $\pm 1.4\%$ due to cancellation of errors. Furthermore, when d/s is divided by λ corresponding to 29.2 years, the resulting atoms will be accurate to 1% because of further cancellation of errors. This accuracy, of course, applies only to this set of standards which was used to derive the 0.82 empirical factor.

Where Cs-137 analysis is performed to determine Cs-137 atom to uranium atom ratio as an indicator of uranium atom percent fission, it appears worthwhile to use a Cs-137 gamma counting standard calibrated directly in Cs-137 atom content known from isotope dilution mass spectrometry rather than one calibrated in gamma photon rate which requires the use of an accurate half-life and gamma abundance to convert to atom content.

SECTION IV

CONCLUSIONS

The half-life of Cs-137 has been determined to be 29.2 ± 0.3 years by an all mass spectrometric method. This value is in agreement with the average of the values referenced⁽¹⁻⁸⁾ and has the smallest uncertainty. This reduces significantly the uncertainty caused by half-life in the cesium to uranium ratio method for burnup determinations. The difference from the previous half-life value determined mass spectrometrically⁽⁸⁾ is attributed to the greater accuracy achieved in the present work by a greater number of observations over a longer period of time and by elimination of the need for chemical loss corrections. The abundance of the 662 KeV gamma ray in the decay scheme of Cs-137 was found to be 0.82 ± 0.03 per beta disintegration.

In the course of the work, the isotopic distribution of natural barium was determined with improved accuracy to be $(72.11 \pm 0.06)\%$ 138, $(11.18 \pm 0.05)\%$ 137, $(7.77 \pm 0.02)\%$ 136, $(6.42 \pm 0.03)\%$ 135, $(2.33 \pm 0.02)\%$ 134, $(0.091 \pm 0.003)\%$ 132, and $(0.098 \pm 0.002)\%$ 130.

SECTION V

ACKNOWLEDGEMENT

The authors are indebted to Dr. D. W. Gaylor for his statistical treatment of the data including an analysis of variance used in weighting the data. Gratitude is also expressed to W. E. Duffy, J. R. Asay and E. L. Kramer who made the mass spectrometric measurements reported here.

APPENDIX IDEFINITION OF SYMBOLS USED

The symbols used throughout the paper are defined as follows:

$N_{a/b}$ = a to b atom ratio in natural barium

$M_{a/b}$ = a to b atom ratio in a mixture of any spike and sample including the 54 Ba-138-spiked Ba-137 samples separated from the master solution.

$S_{a/b}$ = a to b atom ratio in the spike (enriched Ba-138 sol)

$R_{a/b}$ = a to b atom ratio in the ORNL fission cesium prior to isotopic dilution

$H_{a/b}$ = a to b atom ratio in master Cs-137 stock solution after isotopic dilution

k_a = atoms from spike of mass "a"

C_a = atoms from natural isotope of mass "a"

θ_a = atoms from the active Cs-137 solution of mass "a"

a, b, etc. the last digit of the mass number, i.e. 137 the number would be 7.

u = barium atoms of mass 138 in Ba samples separated from master solution

v = barium atoms of mass 137 in Ba samples separated from master solution

x = barium atoms of mass 136 in Ba samples separated from master solution

y = barium atoms of mass 135 in Ba samples separated from master solution

z = barium atoms of mass 134 in Ba samples separated from master solution

W = weight of material added

APPENDIX IICALCULATION OF THE TOTAL NUMBER OF Cs-137 ATOMSIN THE MASTER SOLUTION

Mass analysis was carried out on the fission cesium solution after removal of barium daughters. This solution was then added to a known weight of the natural cesium (mass-133). By using the total atoms of natural Cs-133, C_3 , and ratios $R_{a/b}$ before and ratios $M_{a/b}$ after spiking, the total number of Cs-137 atoms was calculated.

Atom Ratio	Cesium Ratio in Cs-137 Solution		Cesium atoms added (mass-133), C_3
	Unspiked, $R_{a/b}$	Spiked, $M_{a/b}$	
3/7	$1.246 \pm 0.43\%$	$50.83 \pm 0.63\%$	$5.182 \times 10^{20} \pm 0.54\%$
3/5	$2.505 \pm 0.36\%$	$104.9 \pm 0.69\%$	
7/5	---	$2.061 \pm 0.73\%$	

$$\text{Atoms of Cs-137 present} = C_3 \left[\frac{1}{M_{3/7} - R_{3/7}} \right]$$

$$= \frac{5.182 \times 10^{20}}{50.83 - 1.246}$$

$$= 1.045 \times 10^{19} \pm 0.84\%$$

$$\text{Atoms of Cs-137 present} = C_3 \left[\frac{1}{M_{3/5} - R_{3/5}} \right] \left[M_{7/5} \right]$$

$$= 5.182 \times 10^{20} \left[\frac{1}{104.9 - 2.505} \right] 2.061$$

$$= 1.043 \times 10^{19} \pm 1.16\%$$

By weighting these two values inversely proportional to the square of the percent error, the weighted average is obtained as follows:

$$1.045 \times 10^{19} \times \frac{1}{(0.84)^2} = 1.481 \times 10^{19}$$

$$1.043 \times 10^{19} \times \frac{1}{(1.16)^2} = 0.775 \times 10^{19}$$

$$\text{Total Wts.} \approx 2.160 \quad 2.256 \times 10^{19} = \text{Sum}$$

$$\text{Weighted average } N_{\text{Cs-137}} \quad 1.044_4 \times 10^{19} \pm 0.68\%$$

APPENDIX IIICALCULATION OF Ba-138 TRACER ADDED TO THE MASTER SOLUTION

The enriched Ba-138 standard solution, designated Solution A, was standardized by isotopic dilution with standard natural barium chloride dihydrate solutions. The aliquots taken of both enriched and natural barium solutions were weighed in order to minimize the aliquoting error (i.e. 1.0000 gram can be weighed to ± 0.0001). A weighed aliquot of the enriched barium solution was added to stock of Cs-137. The number of Ba-138 spike atoms added, which is the value k_8 in equation 25 of Appendix IV, is obtained from the expression

$$k_8 = (\text{weight of Soln. A added})(\text{atoms of Ba-138/gm of Soln. A}) \quad (1)$$

The value of atoms of Ba-138/gm of Soln. A can be calculated from

$$\frac{\text{Atoms Ba-138}}{\text{gm of Soln A}} = C_8 \left[\frac{\frac{M_{a/b}}{N_{a/b}} - 1}{1 - \frac{M_{a/b}}{S_{a/b}}} \right] \quad (2)$$

where C_8 is the number of natural Ba-138 added per gm of Soln. A. The values for $N_{8/7}$, $N_{8/6}$, $N_{8/5}$, $N_{8/4}$, $S_{8/7}$, $S_{8/6}$, $S_{8/5}$, and $S_{8/4}$ are constants and found in Appendix V. The ratio $M_{a/b}$ is the only ratio which changes (a/b stands for 8/7, 8/6, 8/5, or 8/4 in succession).

The weight of Soln. A added was calculated (Equation 3b) from the weight of the 1:100 dilution added to the Cs-137 stock solution.

$$\text{Wt of Soln. A added} = \frac{(\text{wt. of Soln. A})}{(\text{diluted wt.})} \times (\text{wt. of dilution added to Cs-137 stock}) \quad (3)$$

$$= \frac{0.4766}{50.7012} \times 6.0158 \quad (3a)$$

$$= 5.655_5 \times 10^{-2} \pm 0.021\% \text{ gms of Soln. A} \quad (3b)$$

added to 'Cs-137

Solving equation (1) for k_8 , the total number of Ba-138 atoms which were added to the Cs-137 stock is calculated, where aliquot weight of Soln. A is $5.655_5 \times 10^{-2}$ gm from equation (3b) and atoms of Ba-138 per gm of Soln. A equals $2.078_1 \times 10^{18}$ from Table IV on page 24 of this Appendix.

$$k_8 = (5.655_5 \times 10^{-2})(2.078_1 \times 10^{18})$$

$$k_8 = 1.175_2 \times 10^{17} \pm 0.43\% \text{ atoms Ba-138 in master Cs-137 stock.}$$

TABLE IV

Data Calculated for Ba-138 Spike Concentration

Filament	Atom Ratio of Spiked Barium Solution				Atoms Natural Ba-138 added per gm of Soln. A C_8	Atoms Enriched Ba-138 per gm. Soln A., (kg/gm. Soln. A)			
	$M_{8/7}$	$M_{8/6}$	$M_{8/5}$	$M_{8/4}$		Calc. by $M_{8/7}$ ($\times 10^{-18}$)	Calc. by $M_{8/6}$ ($\times 10^{-18}$)	Calc. by $M_{8/5}$ ($\times 10^{-18}$)	Calc. by $M_{8/4}$ ($\times 10^{-18}$)
J 583	-	131.8	178.0	526.9	1.2541×10^{17}	-	$2.025 \pm 1.6\%$	$2.054 \pm 1.6\%$	$2.161 \pm 2.2\%$
J 605	32.3	65.3	82.4	-	3.0891×10^{17}	$2.082 \pm 2.5\%$	$2.150 \pm 2.2\%$	$2.045 \pm 2.0\%$	-
J 604	32.3	67.2	84.9	-	3.0891×10^{17}	$2.082 \pm 2.5\%$	$2.125 \pm 3.3\%$	$2.120 \pm 3.6\%$	-
J 606	36.3	81.3	105.0	-	2.3839×10^{17}	$2.027 \pm 2.6\%$	$2.085 \pm 1.9\%$	$2.106 \pm 1.6\%$	-
J 607	36.4	80.6	103.0	-	2.3839×10^{17}	$2.039 \pm 3.2\%$	$2.062 \pm 3.0\%$	$2.058 \pm 4.0\%$	-
J 596	38.8	91.7	117.5	-	2.0706×10^{17}	$2.024 \pm 3.2\%$	$2.106 \pm 1.82\%$	$2.090 \pm 2.2\%$	-
J 595	39.0	92.7	120.6	-	2.0706×10^{17}	$2.047 \pm 3.6\%$	$2.135 \pm 2.6\%$	-	-
Weighted average* = $2.078_1 \pm 0.008_9 \times 10^{18}$ atoms Ba-138 spike per gm. of Soln. A.									

*Weighted as the reciprocal of the square of the error.

APPENDIX IV
DERIVATION OF EQUATIONS USED TO CALCULATE
ATOMS PRESENT IN VARIOUS SAMPLES

Symbols used in this section are defined in Appendix I.

(a) Number of Ba-138 Atoms, k_8 , in Spike

The ratios measured by the mass spectrometer are:

$S_{8/7}$, $S_{8/6}$, $S_{8/5}$, $S_{8/4}$, $N_{8/7}$, $N_{8/6}$, $N_{8/5}$, and $N_{8/4}$.

The weight of natural barium, W , added to the unknown amount of atoms of spike is known.

$$S_{8/7} = \frac{k_8}{k_7} \quad (1)$$

$$N_{8/7} = \frac{C_8}{C_7} \quad (2)$$

$$M_{8/7} = \frac{k_8 + C_8}{k_7 + C_7} \quad (3)$$

Solving equation (3) for k_8

$$k_8 = M_{8/7}k_7 + M_{8/7}C_7 - C_8 \quad (3a)$$

Solving equation (2) and equation (1) for C_8 and k_7 , respectively, and substituting in equation (3a)

$$k_8 = M_{8/7}k_7 + M_{8/7}C_7 - N_{8/7}C_7 \quad (3b)$$

$$k_8 = \frac{M_{8/7}k_8}{S_{8/7}} + (M_{8/7} - N_{8/7})C_7 \quad (3c)$$

$$k_8 - \frac{M_{8/7} k_8}{S_{8/7}} = C_7 (M_{8/7} - N_{8/7}) \quad (3d)$$

$$k_8 = \frac{C_7 (M_{8/7} - N_{8/7})}{\left(1 - \frac{M_{8/7}}{S_{8/7}}\right)} \quad (4)$$

Where:

$$C_7 = \frac{W \times \text{Avogadro Number}}{\text{Molecular Wt. of Natural Barium}} \left(\frac{\text{Atom \% of 137 in}}{\text{Natural Barium}} \right) \quad (5)$$

(b) Number of Cs-137 Atoms, k_7 , Present in Spike

The ratios measured by the mass spectrometer are:

$$S_{7/3}, S_{7/5}, \text{ and } S_{7/4}.$$

A check of the natural cesium showed only mass 133 to be present.

The sample was spiked with weight, W , of natural cesium.

$$S_{7/3} = \frac{k_7}{k_3} \quad (6)$$

$$M_{7/3} = \frac{k_7}{k_3 + C_3} \quad (6a)$$

Solving equation (6a) for k_7

$$k_7 = M_{7/3} (k_3 + C_3) \quad (6b)$$

Solving equation (6) for k_3 and substituting the resulting solution in equation (6a) and combining k_7 terms:

$$k_7 - k_7 \frac{M_{7/3}}{S_{7/3}} = M_{7/3} C_3 \quad (6c)$$

$$k_7 = C_3 \frac{M_{7/3}}{1 - \frac{M_{7/3}}{S_{7/3}}} \quad (7)$$

Where:

$$C_3 = \frac{W \times \text{Avogadro Number}}{\text{Molecular Wt. of Natural Cesium}} \quad (8)$$

(c) Number of Ba-137 daughter atoms grown into master solution

The mass spectrometer ratios measured are:

$N_{8/7}$, $N_{8/6}$, $N_{8/5}$, $N_{8/4}$, $M_{8/7}$, $M_{8/3}$, $M_{8/6}$, $M_{8/5}$, and $M_{8/4}$.

Ratios $H_{7/3}$, $S_{8/7}$, $S_{8/6}$, $S_{8/5}$, $S_{8/4}$ have been previously determined.

The general case where Cs-137 and natural barium contamination are present gives the following equation:

$$M_{8/7} = \frac{k_8 + C_8}{k_7 + C_7 + \theta_7 + v} \quad (9)$$

$$M_{7/3} = \frac{k_7 + C_7 + \theta_7 + v}{\theta_3} \quad (10)$$

$$S_{8/7} = \frac{k_8}{k_7} \quad (11)$$

$$S_{8/6} = \frac{k_8}{k_6} \quad (12)$$

$$H_{7/3} = \frac{\theta_7}{\theta_3} \quad (13)$$

$$N_{8/6} = \frac{C_8}{C_6} \quad (14)$$

$$M_{8/6} = \frac{k_8 + C_8}{k_6 + C_6} \quad (15)$$

$$N_{8/7} = \frac{C_8}{C_7} \quad (16)$$

Since the number of atoms of Ba-137, (v), is wanted in the terms of the added Ba-138, (k_8), and the ratios measured, equation (9) is solved for v as follows:

$$v = \frac{k_8 + C_8 - M_{8/7}k_7 - M_{8/7}C_7 - M_{8/7}\theta_7}{M_{8/7}} \quad (17)$$

Equations (12) and (14) are solved for k_6 and C_6 and resulting solutions substituted into equation (15).

$$M_{8/6} = \frac{k_8 + C_8}{\frac{k_8}{S_{8/6}} + \frac{C_8}{N_{8/6}}} \quad (17a)$$

Solving for C_8

$$C_8 = \frac{M_{8/6}}{S_{8/6}} k_8 + \frac{M_{8/6}}{N_{8/6}} C_8 - k_8 \quad (17b)$$

$$C_8 = \frac{\frac{M_{8/6}}{S_{8/6}} k_8 - k_8}{\left(1 - \frac{M_{8/6}}{N_{8/6}}\right)} \quad (18)$$

Solving equation (16) for C_7

$$C_7 = \frac{C_8}{N_{8/7}} \quad (19)$$

Substituting equation (18) and (19) solutions for C_7 and C_8 into equation (17)

$$v = \frac{k_8 + \left[\frac{\frac{M_{8/6}}{S_{8/6}} k_8 - k_8}{\left(1 - \frac{M_{8/6}}{N_{8/6}}\right)} \right] - M_{8/7} k_7 - C_8 \frac{M_{8/7}}{N_{8/7}} - M_{8/7} \theta_7}{M_{8/7}}$$

$$v = \frac{k_8 + \left[\frac{\frac{M_{8/6}}{S_{8/6}} k_8 - k_8}{\left(1 - \frac{M_{8/6}}{N_{8/6}}\right)} \right]}{M_{8/7}} - k_7 - \frac{C_8}{N_{8/7}} - \theta_7$$

Substituting equation (18) in the 3rd term and solution for k_7 from equation (11) for 2nd term.

$$v = \frac{k_8 \left[1 + \frac{\frac{M_{8/6}}{S_{8/6}} - 1}{\left(1 - \frac{M_{8/6}}{N_{8/6}}\right)} \right]}{M_{8/7}} - \frac{k_8}{S_{8/7}} - \frac{k_8}{N_{8/7}} \left[\frac{\frac{M_{8/6}}{S_{8/6}} - 1}{1 - \frac{M_{8/6}}{N_{8/6}}} \right] - \theta_7 \quad (20)$$

In order to simplify the equations, the term

$$\left[\frac{\frac{M_{8/6}}{S_{8/6}} - 1}{1 - \frac{M_{8/6}}{N_{8/6}}} \right]$$

will be designated by A. Equation (20) now becomes:

$$v = \frac{k_8 (1 + A)}{M_{8/7}} - \frac{k_8}{S_{8/7}} - \frac{k_8 A}{N_{8/7}} - \theta_7 \quad (21)$$

Cross multiplying equation (10)

$$M_{7/3} \theta_3 = k_7 + C_7 + v + \theta_7 \quad (21a)$$

Solving for θ_3 in equation (13) and substituting into equation (21a) and solving for θ_7 :

$$\frac{M_{7/3} \theta_7}{H_{7/3}} - \theta_7 = k_7 + C_7 + v \quad (21b)$$

$$\theta_7 \left[\frac{M_{7/3}}{H_{7/3}} - 1 \right] = k_7 + C_7 + v \quad (21c)$$

Substituting solution for k_7 from equation (11) and $\frac{C_8}{N_{8/7}}$ for C_7 from equation (19) where $C_8 = k_8 A$ from equation (18)

$$\theta_7 \left[\frac{M_{7/3}}{H_{7/3}} - 1 \right] = \frac{k_8}{S_{8/7}} + \frac{k_8 A}{N_{8/7}} + v \quad (22)$$

In order to simplify this equation, the term $\left[\frac{M_{7/3}}{H_{7/3}} - 1 \right]$ will be designated by B.

Equation (22) solved for θ_7 becomes:

$$\theta_7 = \frac{k_8}{S_{8/7} B} + \frac{k_8 A}{N_{8/7} B} + \frac{v}{B} \quad (23)$$

Substituting equation (23) in equation (21) and solving for v

$$\begin{aligned}
 v + \frac{v}{B} &= \frac{k_8 (1 + A)}{M_{8/7}} - \frac{k_8}{S_{8/7}} - \frac{k_8}{S_{8/7} B} - \frac{k_8 A}{N_{8/7}} - \frac{k_8 A}{N_{8/7} B} \\
 v &= \frac{k_8 (1 + A)}{M_{8/7} (1 + \frac{1}{B})} - \frac{\frac{k_8}{S_{8/7}} (1 + \frac{1}{B})}{(1 + \frac{1}{B})} - \frac{\frac{k_8 A (1 + \frac{1}{B})}{N_{8/7}}}{(1 + \frac{1}{B})} \\
 v &= k_8 \left[\frac{1 + A}{M_{8/7} + \frac{M_{8/7}}{B}} - \frac{1}{S_{8/7}} - \frac{A}{N_{8/7}} \right] \quad (24)
 \end{aligned}$$

In the case where no Cs-137 correction is needed because B approaches ∞ , the equation is simplified to:

$$v = k_8 \left[\frac{1 + A}{M_{8/7}} - \frac{1}{S_{8/7}} - \frac{A}{N_{8/7}} \right] \quad (25)$$

The quantity $\frac{v}{k_8}$ is selected as the quantity to plot on the ordinate against time in days on the abscissa in Figure 1. If the value of $\frac{v}{k_8}$ is multiplied by $\left(\frac{\lambda t}{1 - e^{-\lambda t}} \right)$ at each time, t, the exponential growth curve in Figure 1 becomes a straight-line function suitable for least square solution for straight-line equation in Appendix VI.

APPENDIX VTHE CONSTANT RATIOS USED IN CALCULATIONS IN APPENDIX IV

<u>Ratio</u>	<u>Value Used in Equations (24) and (25)</u>
$S_{8/7}$	76.69 ± 1.51
$S_{8/6}$	722.1 ± 19.9
$N_{8/7}$	6.448 ± 0.026
$N_{8/6}$	9.281 ± 0.036
$H_{7/3}$	0.01967 ± 0.00012
$\frac{1}{S_{8/7}}$	0.01255 ± 0.00024
$\frac{1}{N_{8/7}}$	0.1551 ± 0.0006

All values are arbitrarily reported to four significant figures to minimize rounding errors.

APPENDIX VITHE Ba-137 GROWTH LINE
AND WEIGHTED LEAST SQUARES SOLUTION OF ITS SLOPE

The ratio $\frac{\text{atoms of Ba-137 daughter}}{\text{atoms of Ba-138 spike}} = v/k_8$ is plotted against time (in days) in Figure 1. The slope of the line was solved after the values $\frac{v}{k_8}$ were multiplied by $\frac{\lambda t}{1 - e^{-\lambda t}}$ to correct for decrease of the atoms of Cs-137 by decay. In the least squares solution, each v/k_8 point was weighted inversely as a function of the percent error. These weights are listed in the following tables. The elapsed times were assumed to be without error because they were known to ± 2 minutes or ± 0.0014 days.

Let S_e^2 denote the variance of the average among the scans for any particular point. The standard deviations, S_e , are listed in the following tables for each point.

For the single stage analyses, the variance of the points from the fitted least squares line was less than the average value of S_e^2 . Due to the large differences in S_e^2 for the single stage results, weighting the points inversely proportional to S_e^2 would have resulted in extremely unequal weighting, whereas the precision among analyses generally do not vary markedly. Hence, the single stage points were weighted inversely proportional to S_e such that the sum of the weights equals 25, the number of single stage points, ignoring the 1.5 multiplying factor for the moment.

For the double stage instrument, the variance of the points from the fitted least squares line was greater than the average value of S_e^2 . Hence, there was evidence of a point to point variation, S_p^2 , in addition to the scanning error of a point, S_e^2 . The estimate of the point to point variation was 0.00009090. The values for S_p are listed in the

table. The points for the double stage instrument were weighted inversely proportional to $(S_p^2 + S_e^2)$ such that the sum of the weights equals 29, the number of double stage points, apart from the 0.569 multiplying factor.

For either instrument, the weighted analysis did not significantly alter the slopes from their respective unweighted analyses. Hence, the choice of two different weighting procedures dictated by the different error mechanisms for the two types of spectrometers actually had no significant influence on the slope of the final line. The purpose of weighting was to give those points with smaller errors more weight than those points with larger errors in order to reduce the standard deviations of the estimates of the slopes for each mass spectrometer.

The slopes of the lines for the two mass spectrometers did not differ significantly, hence, it was in order to pool the data from the two instruments and obtain one line. The variance of the points about the weighted least squares line for the single stage spectrometer was 0.00003134 and 0.00008248 for the double-stage instrument. Weighting the points from the two spectrometers inversely proportional to their variances gives relative weights of 2.64 to 1.00 for single-stage and double-stage points, respectively, when pooling the data. Multiplying the weights for the individual analyses on each spectrometer by 1.500 for the single-stage instrument and by 0.569 for the double-stage instrument (see the tables) provides weighting of the combined data in the ratio of $2.64 = 1.500/0.569$ such that the sum of the weights equals 54, the total number of points from the two spectrometers. The weighted least squares solution for the combined data was

$$y = (0.00578_7 \pm 0.00002_9)x - (0.000_5 \pm 0.001_6)$$

where y is the number of Ba-137 daughter atoms per atom of Ba-138 spike and x is time in days. The $\pm$ limits are the standard deviations on the slope and intercept. The percent error of the slope is $\pm 0.5\%$.

RESULTS FROM SINGLE STAGE INSTRUMENT

Sample No.	Δt	v/k_g	Decay Correction*	Corrected v/k_g	$\pm S_e$	Weight Assigned
6	6.029	0.03566	0.999812	0.03566	0.0039	1.68 x 1.5
8	8.033	0.04762	0.999750	0.04763	0.0164	0.40 x 1.5
9	9.755	0.05881	0.999696	0.05883	0.0023	2.83 x 1.5
11	11.013	0.06121	0.999656	0.06123	0.0040	1.63 x 1.5
12	12.085	0.06948	0.999623	0.06951	0.0026	2.51 x 1.5
23	47.071	0.26667	0.998531	0.26706	0.0082	0.80 x 1.5
24	49.042	0.27803	0.998470	0.27846	0.0043	1.52 x 1.5
27	60.139	0.34784	0.998124	0.34849	0.0043	1.52 x 1.5
28	61.045	0.34381	0.998096	0.34446	0.0064	1.02 x 1.5
29	63.019	0.36754	0.998034	0.36826	0.0060	1.09 x 1.5
30	66.031	0.37155	0.997940	0.37232	0.0051	1.28 x 1.5
31	68.051	0.40611	0.997877	0.40697	0.0407	0.16 x 1.5
32	69.982	0.39670	0.997817	0.39757	0.0065	1.01 x 1.5
33	73.032	0.41563	0.997722	0.41657	0.0500	0.13 x 1.5
34	75.057	0.42973	0.997659	0.43074	0.0056	1.17 x 1.5
35	80.043	0.46992	0.997503	0.47110	0.0081	0.81 x 1.5
36	84.009	0.47944	0.997379	0.48070	0.0160	0.41 x 1.5
37	86.988	0.50690	0.997286	0.50828	0.0104	0.63 x 1.5
38	89.040	0.51256	0.997222	0.51399	0.0081	0.81 x 1.5
39	91.078	0.51556	0.997159	0.51703	0.0098	0.67 x 1.5
40	97.009	0.56317	0.996974	0.56487	0.0226	0.29 x 1.5
41	98.023	0.57493	0.996942	0.57669	0.0282	0.23 x 1.5
42	99.994	0.58567	0.996880	0.58750	0.0081	0.81 x 1.5
43	99.994	0.58506	0.996880	0.58689	0.0081	0.81 x 1.5
44	99.994	0.58171	0.996880	0.58353	0.0084	0.78 x 1.5
*Values indicated are the reciprocal of $\lambda t/(1-e^{-\lambda t})$ described in text and are divided into v/k_g to correct for decay.					Total	25.00 x 1.5

RESULTS FROM DOUBLE STAGE INSTRUMENT

Sample No.	Δt	v/k_8	Decay Correction*	Corrected v/k_8	$\pm S_e$	Weight Assigned
1	0.070	-0.000286	0.999999	-0.000286	0.0023	1.26 x 0.569
2	3.067	0.026897	0.999905	0.026897	0.0105	0.60 x 0.569
3	4.074	0.026810	0.999873	0.026810	0.0041	1.13 x 0.569
4	4.829	0.029967	0.999850	0.029967	0.0042	1.12 x 0.569
5	5.076	0.029499	0.999842	0.029499	0.0012	1.33 x 0.569
7	7.070	0.044027	0.999780	0.044037	0.0013	1.32 x 0.569
10	10.090	0.068970	0.999685	0.068990	0.0070	0.87 x 0.569
13	14.067	0.084212	0.999562	0.084249	0.0016	1.30 x 0.569
14	17.048	0.092006	0.999468	0.092055	0.0039	1.15 x 0.569
15	21.050	0.12270	0.999343	0.12278	0.0021	1.28 x 0.569
16	24.071	0.12924	0.999249	0.12934	0.0021	1.28 x 0.569
17	28.076	0.14198	0.999124	0.14210	0.0105	0.60 x 0.569
18	31.073	0.15820	0.999031	0.15835	0.0680	0.03 x 0.569
19	33.067	0.19801	0.99897	0.19821	0.0032	1.20 x 0.569
20	35.084	0.21637	0.99891	0.21661	0.0084	0.75 x 0.569
21	38.065	0.20848	0.99882	0.20872	0.0048	1.07 x 0.569
22	40.053	0.21633	0.99875	0.21660	0.0032	1.20 x 0.569
23	47.071	0.28242	0.99854	0.28283	0.0092	0.69 x 0.569
24	49.042	0.29786	0.99847	0.29830	0.0044	1.10 x 0.569
25	53.056	0.30955	0.99835	0.31006	0.0045	1.09 x 0.569
26	56.907	0.33014	0.99823	0.33072	0.0046	1.09 x 0.569
27	60.139	0.35164	0.99812	0.35250	0.0056	0.99 x 0.569
28	61.045	0.35175	0.99809	0.35240	0.0058	0.98 x 0.569
29	63.019	0.35093	0.99803	0.35160	0.0055	1.00 x 0.569
30	66.031	0.39341	0.99794	0.39422	0.0066	0.90 x 0.569
31	68.051	0.38049	0.99788	0.38130	0.0056	0.99 x 0.569
32	69.982	0.39770	0.99782	0.39856	0.0064	0.92 x 0.569
33	73.032	0.43054	0.99720	0.43175	0.0069	0.88 x 0.569
34	75.057	0.42169	0.99766	0.42268	0.0069	0.88 x 0.569

*Values indicated are the reciprocal of $\lambda t/(1-e^{-\lambda t})$ described in text and are divided into v/k_8 to correct for decay.

Total 29.00 x 0.569

The weighted least squares solution used the following equations.

$$y = ax + b$$

where

$$y = v/k_8$$

$$x = t$$

$$a = \text{slope}$$

$$b = \text{y intercept}$$

Assuming no error in x and all errors to be in y, the least square solution becomes:

$$a = \frac{\sum(wt)\sum(wt)(xy) - \sum(wt)(x)\sum(wt)(y)}{\sum(wt)\sum(wt)(x^2) - [\sum(wt)(x)]^2}$$

$$b = \frac{\sum(wt)(x^2)\sum(wt)(y) - \sum(wt)(x)\sum(wt)(xy)}{\sum(wt)\sum(wt)(x^2) - [\sum(wt)(x)]^2}$$

APPENDIX VIICALCULATION OF GAMMA-RAY ABUNDANCE IN THE DECAY OF Cs-137

Aliquots of the master Cs-137 stock solution were weighed and sealed in vials of the same dimensions as the Nuclear Chicago Cs-137 vials. The sample vials were then counted on low geometry shelves 5, 6, 7, and 9 corresponding to a sample to sodium iodide crystal distance of 49.3, 69.7, 100.2 and 145.7 cm., respectively, and on two different analyzers in comparison with two different Nuclear Chicago Cs-137 standards. The following data were obtained for the γ/m at 662 Kev corrected to the total weight of the master Cs-137 stock solution.

SUMMARY OF COUNTING DATA

Ampoule	Shelf	RCL 256-Channel $\gamma/m \times 10^{-11}$	RIDL 200-Channel $\gamma/m \times 10^{-11}$
A	9	---	3.950
		---	3.952
		---	3.886
		---	3.890
		---	3.887
	7	---	3.868
B	9	---	3.877
		---	4.180
		---	4.275
		---	3.749
		---	3.900
		---	3.909
	7	---	3.895
		---	3.910
	6	4.010 3.901 3.846	
C	9	---	4.041
		---	3.962
		---	3.872
	7	---	3.885
		---	3.869
	6	3.854 3.856	---
		---	---
	5	3.573	---
D	9	---	3.914
		---	3.901
		---	3.846
	7	---	3.899
		---	3.894
	6	3.814 3.842	---
		---	---
	5	3.814 3.814	---
E	9	---	3.952
		---	3.915
		---	3.900
	7	---	3.903
		---	3.913
	6	3.828	---
		3.864	---
		3.825	---
		3.841	---
	5	3.883	---

SUMMARY OF COUNTING DATA (continued)

Ampoule	Shelf	RCL 256-Channel $\gamma/m \times 10^{-11}$	RIDL 200-Channel $\gamma/m \times 10^{-11}$
F	9	--	3.896
		--	3.878
		--	3.886
	7	3.711	3.894
		3.740	3.897
		3.769	--
	6	3.786	--
		3.789	--
		3.813	--
		3.764	--
	5	3.788	--
		3.772	--
		3.801	--
G	9	--	3.977
		--	3.948
		--	3.866
	7	3.721	3.902
		3.906	3.883
	6	3.817	--
		3.809	--
	5	3.821	--

<u>Sample</u>	<u>Average Value of γ/m Cs-137</u>
A	3.901×10^{11}
B	3.958×10^{11}
C	3.864×10^{11}
D	3.860×10^{11}
E	3.882×10^{11}
F	3.812×10^{11}
G	3.865×10^{11}

Grand Average $3.877 \times 10^{11} \pm 3.2\%$

(of which 3% is uncertainty in N-C Std.)

The 662 Kev gamma abundance per beta disintegration of Cs-137 is expressed as:

$$\text{gamma abundance} = \frac{\gamma/m \text{ Cs-137}}{(\text{atoms Cs-137})(\lambda \text{ min}^{-1})}$$

Since atoms of Cs-137 present = 1.0444×10^{19} (See Appendix II),
 $\lambda = 4.521 \times 10^{-8} \text{ min.}^{-1}$, and $\gamma/\text{m Cs-137} = 3.877 \times 10^{11}$,

$$\text{gamma abundance} = \frac{3.877 \times 10^{11} \pm 3.2\%}{1.0444 \times 10^{19} \pm 0.68\% \times 4.521 \times 10^{-8} \pm 1\%} = 0.82 \pm 3.3\%.$$

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