# Determination of gamma ray energies and abundances of $^{229}$Th

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Gamma-ray energies and intensities in the alpha decay of $^{229}$Th were precisely determined using a high-resolution Ge detector. Twenty new gamma rays were observed whereas 16 gamma rays earlier reported could not be observed. A modified energy level diagram for $^{225}$Ra is proposed using the present results.

[RADIOACTIVITY $^{229}$Th; measured $E_\gamma$, $I_\gamma$. Ge detector. $^{225}$Ra deduced levels. Radiochemistry.]

## INTRODUCTION

The decay of $^{229}$Th has been studied by Tretyakov *et al.*[1] using a low-resolution Ge(Li) detector (FWHM 5−6 keV) and electron spectroscopy. However, their investigations yielded only crude estimates of the gamma-ray intensities. The thorium activity used in Ref. 1 contained comparable activities of both $^{228}$Th and $^{229}$Th. The presence of $^{228}$Th ($T_{1/2}=1.927$ yr) and of the short-lived daughter products (cf. Fig. 1) of $^{228}$Th and $^{229}$Th greatly complicates the gamma-ray spectrum. Most of the reported[1,2] gamma-ray abundances were calculated from internal-conversion electron intensities and suggested multipolarities, and for some of the gamma rays, no abundance was reported. The present work was undertaken to redetermine the $^{229}$Th gamma-ray energies and abundances.

During the preparation of this paper Dickens and McConnell[3] reported gamma ray intensities of $^{229}$Th and its daughter products. The source used in their work contained $^{228}$Th and $^{229}$Th along with their respective daughter products in equilibrium. The abundances for 18 gamma rays of $^{229}$Th were reported by them, taking the 440 keV gamma ray of $^{213}$Bi as standard with an abundance of 27.4%.

## EXPERIMENTAL

### A. Purification and activity estimation of $^{229}$Th

Thorium along with its daughter products was separated from an old (~6 yr) $^{233}$U sample by an ion exchange method[4] using Dowex 1×8 in the $Cl^-$ form in a 6*M* HCl medium. The thorium was purified from its daughter products by another ion exchange method[5] using Dowex 2×8 in the $NO_3^-$ form in a 7.5*M* $HNO_3$ medium. The purity of the thorium fraction was checked by gamma spectrometry. In addition to $^{229}$Th, the sample was known to contain isotopic impurities of $^{228}$Th (<0.1%) and $^{230}$Th (<0.4%). The gamma rays due to $^{230}$Th (<0.4%) were not observed due to its very long half-life ($7.7\times10^4$ yr).

The purified thorium activity was used to prepare

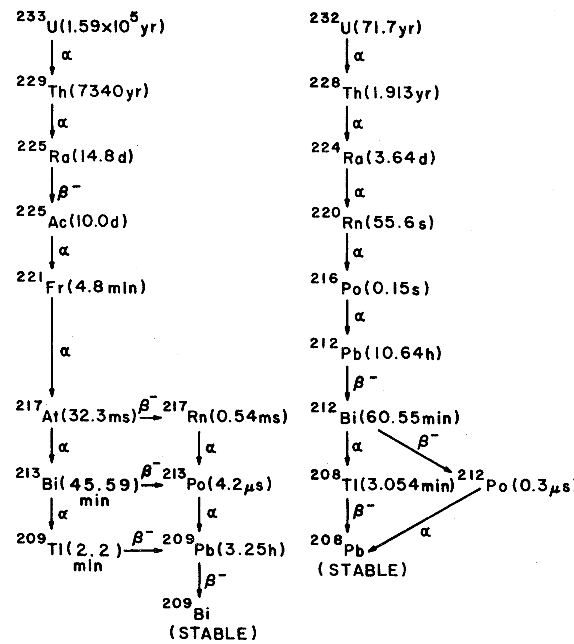


FIG. 1. Decay series of $^{232}$U and $^{233}$U.

 

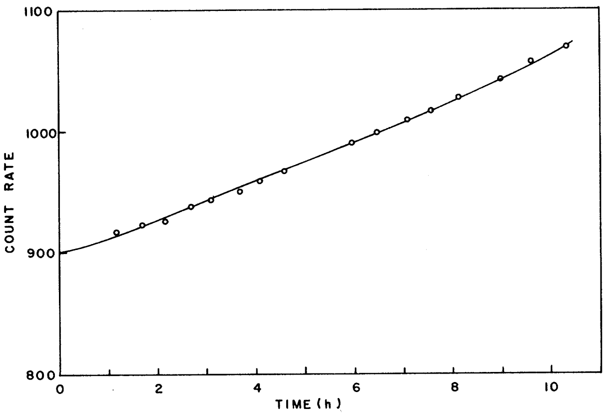


FIG. 2. Growth of total alpha activity in the estimation of thorium ($^{228}Th + ^{229}Th$) as a function of time.

samples for gamma counting and gross alpha counting. Three samples on polished stainless steel disks and three samples in liquid scintillation vials were prepared by a weight transfer method for gross alpha counting. The solid samples were followed on a gas proportional counter (efficiency = $50.0 \pm 0.5$ %) and the liquid samples on a liquid scintillation counter (efficiency $\geq 99.5\%$) over a period of 10–15 hours. Figure 2 gives a typical plot of growth in total alpha activity with time due to formation of alpha active daughter products. The alpha activity due to thorium ($^{228}Th + ^{229}Th$) was determined by extrapolating the growth curve to the time of purification by least squares fitting of the data to a quadratic equation.[6]

An electrodeposited source of thorium was

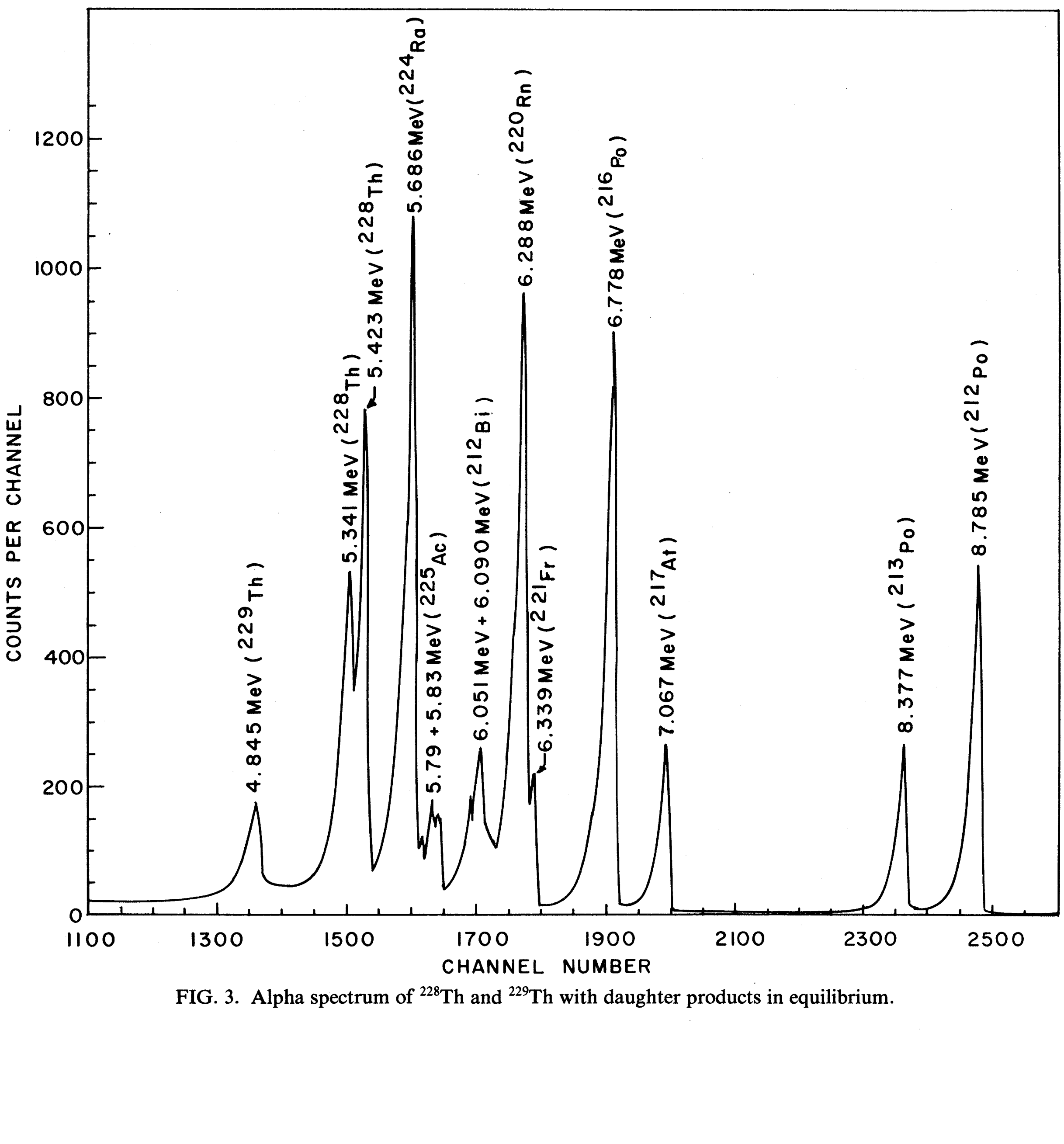


FIG. 3. Alpha spectrum of $^{228}Th$ and $^{229}Th$ with daughter products in equilibrium.

prepared on platinum backing using a thorium solution having the daughter products in equilibrium. The alpha spectrum of thorium (Fig. 3) was taken on a high resolution silicon surface barrier detector (resolution 20 keV at 5.486 MeV). The area under the well-separated alpha peaks of $^{213}Po$, $^{217}At$ (daughter products of $^{229}Th$), and $^{212}Po$ (the daughter product of $^{228}Th$) were used to determine the activity ratio of $^{229}Th$ and $^{228}Th$. The total alpha activity and the activity ratio of $^{229}Th$ and $^{228}Th$ were used[6] to determine the individual activities of $^{229}Th$ and $^{228}Th$.

### B. Gamma counting and spectrum analysis

Three independent experiments were carried out for measurements of gamma ray energies and their abundances. In each experiment a freshly purified fraction of thorium of the purified stock solution was deposited in a standard counting vial and followed for a suitable length of time with a 2 $cm^3$ Ge detector (resolution 600 eV at 122 keV) coupled to a 4096 channel analyzer. Figure 4 shows a typical gamma spectrum of the purified $^{228,229}Th$. After 12 hours another gamma count was taken under identical conditions in order to identify, through growth, the lines due to daughter products of $^{228}Th$ and $^{229}Th$. The total count rate was always less than 1000/sec. Hence the errors due to pileup effects were neglected.

Standard activities of $^{57}Co$, $^{133}Ba$, and $^{241}Am$ were used to calibrate the detector for gamma ray energy and efficiency in the required geometry. The gamma ray spectrum analysis was carried out using program SAMPO.[7] The gamma ray energies of $^{57}Co$, $^{133}Ba$, and $^{241}Am$ and their respective peak positions were least squares fitted to develop the energy calibration curve. The efficiency values for the energy region above 120 keV were found to lie in a straight line on a log-log scale.

## RESULT AND DISCUSSION

The relative emission rates of the $^{229}Th$ gamma rays were determined using the developed energy versus efficiency calibration curves. The absolute abundances of the gamma rays were then calculated using the estimated $^{229}Th$ activity. Table I gives the gamma ray energies and their absolute abundances obtained in this work, along with the reported data.[2]

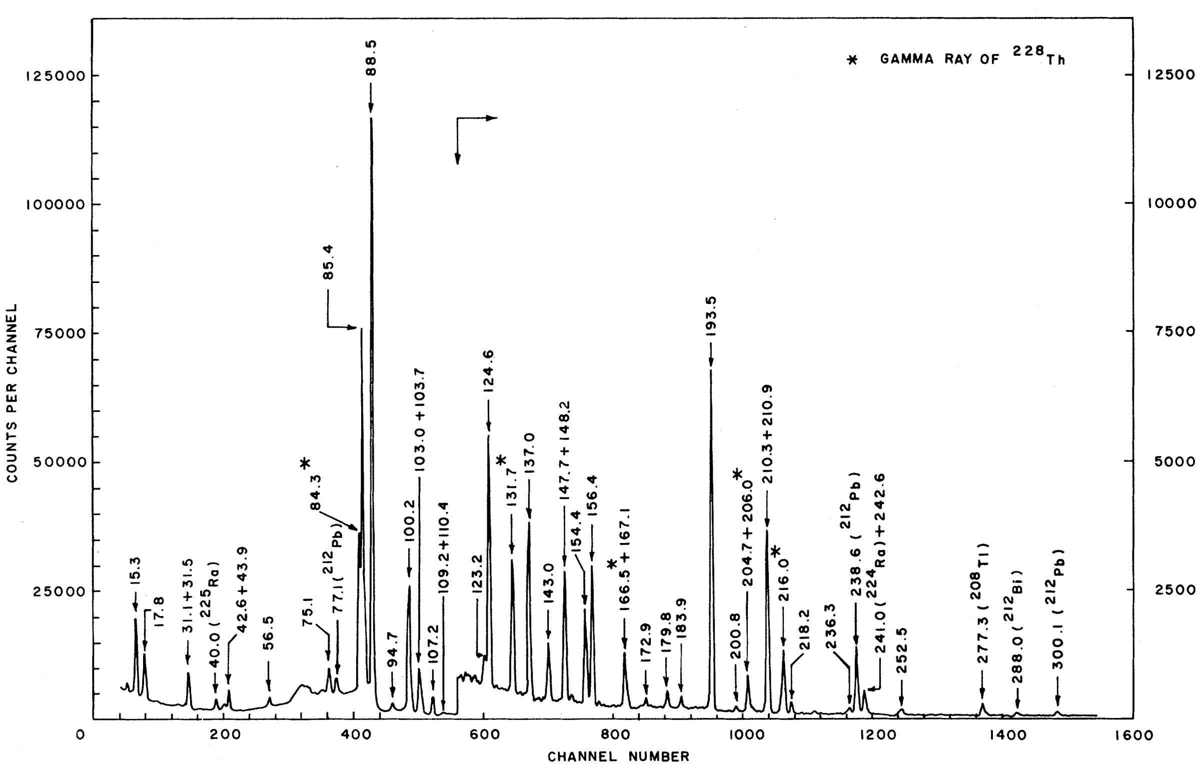


FIG. 4. Gamma spectrum of $^{228}Th$ and $^{229}Th$.

TABLE I. Gamma ray abundances of $^{229}$Th.

| Present | | Reported (Ref. 2) | |
|---|---|---|---|
| Energy (keV) | % abundance | Energy (keV) | % abundance |
| | | 11.1±0.1 | |
| 12.33±0.04[a] | 5.960±0.536 | | |
| 14.81±0.02[a] | 9.381±0.781 | | |
| 15.25±0.02[a] | 42.480±1.592 | | |
| | | 17.36±0.03 | 0.17 |
| 17.82±0.02[a] | 17.033±0.772 | | |
| 18.31±0.03[a] | 4.068±0.403 | | |
| | | 25.39±0.02 | 0.035 |
| 28.50±0.14 | 0.117±0.024 | | |
| | | 30.30±0.10 | |
| 31.13±0.03 | 0.896±0.080 | | |
| 31.53±0.04 | 1.692±0.085 | 31.30±0.20 | 4.0 |
| | | 37.80±0.10 | |
| 42.63±0.02 | 0.188±0.010 | 42.76±0.03 | 0.16 |
| 43.96±0.02 | 0.604±0.020 | | |
| 53.84±0.09 | 0.017±0.003 | 53.20±0.10 | |
| 56.50±0.03 | 0.246±0.006 | 56.60±0.03 | 0.32 |
| 68.05±0.08 | 0.052±0.014 | 68.18±0.07 | 0.10 |
| 68.80±0.07 | 0.060±0.013 | 68.90±0.04 | 0.11 |
| 75.10±0.05 | 0.420±0.043 | 75.20±0.07 | 0.51 |
| | | 75.30±0.10 | |
| 85.43±0.04[a] | 9.820±0.017 | | |
| | | 86.30±0.10 | 0.37 |
| 86.35±0.04 | 2.732±0.074 | 86.44±0.05 | 3.0 |
| 88.48±0.04[a] | 16.681±0.251 | | |
| 94.72±0.02 | 0.232±0.006 | | |
| 99.47±0.02[a] | 2.245±0.070 | | |
| 100.18±0.02[a] | 3.927±0.086 | | |
| 102.99±0.02[a] | 1.443±0.046 | | |
| 103.71±0.03 | 0.451±0.035 | | |
| 107.15±0.02 | 0.656±0.009 | 107.17±0.05 | 0.82 |
| 109.21±0.06 | 0.023±0.004 | | |
| 110.38±0.03 | 0.107±0.004 | | |
| 118.21±0.09 | 0.015±0.005 | | |
| 120.16±0.08 | 0.017±0.003 | | |
| 123.19±0.03 | 0.120±0.004 | | |
| 124.59±0.02 | 1.040±0.012 | 124.50±0.10 | 1.2 |
| | | 124.70±0.10 | 0.6 |
| 126.76±0.09 | 0.013±0.004 | | |
| | | 131.97±0.05 | 0.32 |
| | | 132.60±0.10 | |
| 134.33±0.08 | 0.015±0.003 | 134.80±0.10 | |
| | | 135.71±0.07 | |
| 136.99±0.03 | 0.904±0.018 | 137.03±0.06 | 1.6 |
| | | 140.30±0.20 | |
| 142.97±0.03 | 0.314±0.006 | 142.95±0.10 | 0.42 |
| 147.66±0.03 | 0.183±0.014 | 147.80±0.10 | |
| 148.17±0.03 | 0.708±0.017 | 148.30±0.20 | 1.36 |
| 149.91±0.04 | 0.042±0.003 | 150.20±0.30 | |
| | | 151.60±0.30 | |
| 154.37±0.02 | 0.612±0.012 | 154.40±0.70 | 0.65 |
| 156.41±0.02 | 0.972±0.018 | 156.48±0.04 | 1.1 |

TABLE I. (*Continued.*)

| Present | | Reported (Ref. 2) | |
|---|---|---|---|
| Energy (keV) | % abundance | Energy (keV) | % abundance |
| 158.42±0.04 | 0.034±0.003 | 158.50±0.07 | |
| 160.48±0.56 | 0.005±0.003 | 161.60±0.30 | |
| | | 165.70±0.30 | |
| 167.14±0.04 | 0.113±0.010 | | |
| 171.59±0.07 | 0.020±0.005 | | |
| 172.91±0.04 | 0.093±0.006 | 172.90±0.10 | 0.22 |
| 179.75±0.03 | 0.176±0.005 | 179.80±0.20 | 0.50 |
| 183.95±0.03 | 0.118±0.006 | 184.00±0.10 | 0.23 |
| | | 190.20±0.20 | |
| 193.53±0.02 | 3.769±0.075 | 193.63±0.06 | 4.5 |
| 200.81±0.03 | 0.066±0.005 | | |
| 204.70±0.02 | 0.495±0.012 | 204.90±0.30 | |
| 210.31±0.05 | 0.210±0.033 | | |
| 210.90±0.05 | 2.467±0.063 | 210.97±0.10 | 3.2 |
| 215.16±0.08 | 0.146±0.016 | | |
| 218.15±0.04 | 0.149±0.037 | 218.10±0.20 | 0.14 |
| 221.31±0.09 | 0.022±0.003 | | |
| 225.25±0.06 | 0.048±0.004 | | |
| 236.31±0.06 | 0.158±0.028 | 236.20±0.20 | 0.035 |
| 242.61±0.07 | 0.065±0.007 | 242.60±0.30 | |
| | | 243.50±0.30 | |
| 252.49±0.05 | 0.089±0.005 | | |
| 259.15±0.05 | 0.033±0.011 | | |
| | | 261.00±0.50 | |
| | | 290.00±0.50 | |

[a]x rays of radium.

Table II gives the absolute gamma ray abundances of $^{228}$Th determined during the present investigation, along with the reported data.[8] Our values for $^{228}$Th are in fair agreement with the literature. Table III gives a comparison of our data on $^{229}$Th with those of Dickens and McConnell[3] in which only 18 gamma rays were reported.

The earlier data[1,2] on the gamma ray energies of $^{229}$Th used for developing the energy level diagram[9] of $^{225}$Ra had considerable uncertainties. Based on the present data and the previous measurements,[1] a modified level diagram is proposed (Fig. 5). Table IV gives the gamma ray energies and their positions in the level scheme. It is clear from the table that most of our data fit well in the level scheme. In the present work the following new gamma rays were observed: 28.50, 31.13, 43.96, 94.72, 103.71, 109.21, 110.38, 118.21, 120.16, 123.19, 126.76, 167.14, 171.59, 200.81, 210.31, 215.16, 221.31, 225.25, 252.49, and 259.15 keV. Some of the earlier reported[1] transitions (25.39, 30.3, 37.8, 75.3, 124.7, 131.97, 132.6, 135.71, 140.3, 151.6, 165.7, 190.2, 243.5, 261.0, and 290.0 keV) were not observed. In some cases, this may be a result of the transition having a large internal conversion coefficient. The 43.96 keV gamma ray could not be placed in the present level diagram. The 53.84, 68.80, 118.21, 124.59, and 218.15 keV gamma rays have been placed twice in the level diagram. Since $\gamma$-$\gamma$ coincidence data are not available, the latter gamma rays could not be placed unambiguously. The gamma-ray transitions 75.10, 124.59, 154.37, 210.90, 218.15, 242.61, and 259.15 keV are postulat-

TABLE II. Gamma ray abundances of $^{228}$Th.

| Present | | Reported (Ref. 8) | |
|---|---|---|---|
| Energy (keV) | % abundances | Energy (keV) | % abundances |
| 84.29±0.05 | 1.351±0.064 | 84.5 | 1.6 |
| 131.74±0.03 | 0.214±0.004 | 132.0 | 0.19 |
| 166.48±0.03 | 0.136±0.003 | 167.0 | 0.12 |
| 205.97±0.10 | 0.022±0.004 | 205.0 | 0.03 |
| 216.00±0.06 | 0.285±0.002 | 216.0 | 0.29 |
| | | 234.0 | 0.00007 |

TABLE III. Comparison of gamma ray energies and abundances with those of Ref. 3.

| Present data | | Dickens and McConnell data (Ref. 3) | |
|---|---|---|---|
| Energy (keV) | % abundance | Energy (keV) | % abundance |
| 31.13±0.03 | 0.896±0.080 | 31.24 | 1.43 ±0.05 |
| 31.53±0.04 | 1.692±0.085 | | |
| 42.63±0.02 | 0.188±0.010 | 42.79 | 0.272±0.011 |
| 56.50±0.03 | 0.246±0.006 | 56.57 | 0.427±0.015 |
| 86.35±0.04 | 2.732±0.074 | 86.38 | 2.94 ±0.09 |
| 107.15±0.02 | 0.656±0.009 | 107.20 | 0.95 ±0.03 |
| 124.59±0.02 | 1.040±0.012 | 124.68 | 1.62 ±0.05 |
| Not observed | | 132.00 | 0.433±0.015 |
| 136.99±0.03 | 0.904±0.018 | 137.06 | 1.51 ±0.05 |
| 142.97±0.03 | 0.314±0.006 | 143.05 | 0.532±0.019 |
| 148.17±0.03 | 0.708±0.017 | 148.18 | 1.26 ±0.04 |
| 154.37±0.02 | 0.612±0.012 | 154.36 | 1.13 ±0.04 |
| 156.41±0.02 | 0.972±0.018 | 156.45 | 1.26 ±0.04 |
| 172.91±0.04 | 0.093±0.006 | 173.01 | 0.130±0.006 |
| 179.75±0.03 | 0.176±0.005 | 179.85 | 0.262±0.010 |
| 183.95±0.03 | 0.118±0.006 | 184.0 | 0.091±0.009 |
| 193.53±0.02 | 3.769±0.075 | 193.59 | 5.89 ±0.18 |
| 204.70±0.02 | 0.495±0.012 | 204.74 | 0.75 ±0.04 |
| 210.90±0.05 | 2.467±0.063 | 210.93 | 4.00 ±0.13 |

TABLE IV. Gamma ray transitions and their position in the level scheme.

| Energy of gamma ray (keV) | Levels associated with the transition | Energy of gamma ray (keV) | Levels associated with the transition |
|---|---|---|---|
| 28.50 | 272.05-243.48 | 147.66 | 248.13-100.47 |
| 31.13 | 236.26-205.13 | 148.17 | 179.72- 31.55 |
| 31.53 | 31.55- 0.0 | 149.91 | 149.91- 0.00 |
| 42.63 | 42.72- 0.00 | 154.37 | 179.72- 25.37 |
| | | 156.41 | 268.05-111.63 |
| 43.96 | | 158.42 | 394.79-236.26 |
| 53.84 | 203.62-149.91 | 160.48 | 272.05-111.63 |
| | 272.05-218.15 | | |
| 56.50 | 236.26-179.72 | 167.14 | 268.05-100.47 |
| 68.05 | 179.72-111.63 | 171.59 | 272.05-100.47 |
| 68.80 | 111.63- 42.72 | 172.91 | 284.48-111.63 |
| | 100.47- 31.55 | 179.75 | 179.72- 0.0 |
| 75.10 | 100.47- 25.37 | 183.95 | 284.48-100.47 |
| 86.35 | 236.26-149.91 | 193.53 | 236.26- 42.72 |
| 94.72 | 205.13-110.41 | 200.81 | 243.48- 42.72 |
| 103.71 | 321.66-218.15 | 204.70 | 236.26- 31.55 |
| 107.15 | 149.91- 42.72 | 210.31 | 321.86-111.63 |
| 109.21 | 326.79-218.15 | 210.90 | 236.26- 25.37 |
| 110.38 | 110.41- 0.00 | 215.16 | 326.79-111.63 |
| 118.21 | 321.86-203.62 | 218.15 | 243.48- 25.37 |
| | 268.05-149.91 | | 218.15- 0.0 |
| 120.16 | 392.21-272.05 | 221.31 | 321.86-100.47 |
| 123.19 | 326.79-203.62 | 225.25 | 268.05- 42.72 |
| 124.59 | 236.26-111.63 | 236.31 | 236.26- 0.0 |
| | 149.91- 25.37 | 242.61 | 268.05- 25.37 |
| 126.76 | 394.79-268.05 | 252.49 | 284.48- 31.55 |
| 134.33 | 284.48-149.91 | 259.15 | 284.48- 25.37 |
| 136.99 | 179.72- 42.72 | | |
| 142.97 | 243.48-100.47 | | |

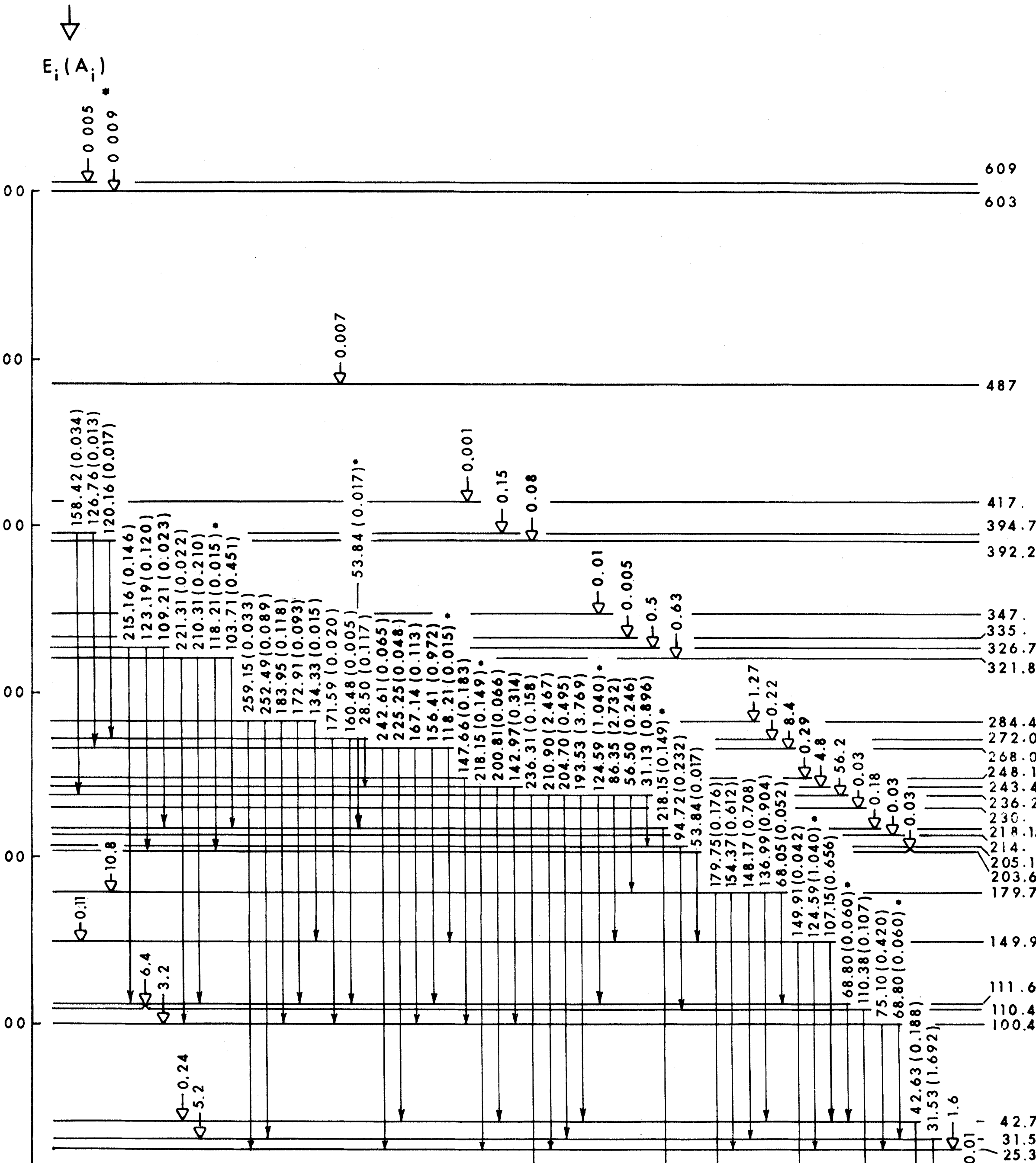


FIG. 5. Level scheme of $^{225}_{88}Ra_{137}$. % feeding by alpha decay of $^{229}Th$ (Refs. 2 and 9). $E_i$ is the energy of the gamma ray in keV and $A_i$ is the absolute gamma ray abundance. $\gamma$ placed twice in the level scheme.

ed to populate the 25.37 keV level. However, the 25.37-keV gamma ray could not be observed; we estimate its gamma-ray abundance to be less than 0.01%. Presumably this transition is of $M1$ multipolarity and is thus highly converted. From the levels 609, 603, 487, 417, 347, 335, 230, and 214 keV, which are quite weakly populated in the alpha decay of $^{229}Th$, no transition could be observed.

[1]E. F. Tretyakov, N. I. Tretyakova, V. F. Konyaev, Y. V. Khrudev, A. C. Beda, G. F. Kartashev, and I. N. Vishnevskii, Izv. Akad. Nauk SSSR, Ser. Fiz. 34, 856 (1970) [Bull. Acad. Sci. USSR, Phys. Ser. 34, 763 (1971)].

[2]K. S. Toth, Nucl. Data Sheets 24, 263 (1978).

[3]J. K. Dickens and J. W. McConnell, Radiochem. Radioanal. Lett. 47, 331 (1981).

[4]K. A. Kraus and F. Nelson, in *Proceedings of the International Conference on Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 7, p. 113.

[5]L. R. Bunney, N. E. Ballou, J. Pascual, and S. Foti, Anal. Chem. 31, 324 (1959).

[6]S. S. Rattan, A. V. R. Reddy, V. S. Mallapurkar, R. J. Singh, and Satya Prakash, J. Radioanal. Chem. 67, 95 (1981).

[7]J. T. Routi, University of California Radiation Laboratory Report No. UCRL-19452, 1969 (unpublished).

[8]G. Erdtmann and W. Soyka, J. Radioanal. Chem. 26, 375 (1975).

[9]C. Maples, Nucl. Data Sheets 10, 643 (1973).