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Gamma-Ray Emission Probabilities of the Daughters of ^{238}U

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In the past several years a wealth of decay data has been obtained and reported, much of it in "Decay Data of the Transactinium Nuclides", IAEA Technical Reports Series No. 261 (1986). The decay data for the daughters of ^{238}U have been notable by their absence in such compilations; and since there is a need for such data, a set of experiments has been performed to measure the gamma-ray emission probabilities. Uranium samples of known mass and isotopic concentration in aqueous solution are analyzed with a high-purity germanium gamma-ray spectrometer. Various samples have also in solution multi-line calibration sources with well-known relative intensities. The well-known emission probabilities of the ^{235}U gamma rays are used to provide an absolute intensity reference. Since self-absorption of the sample is included in the effective detector efficiency, there is no need for a separate calculation of this absorption. Gamma-ray emission probabilities for the energy range 63 to 1938 keV are reported. Sources of error, including those in the efficiency curve, are discussed.

1. Introduction

Gamma-ray emission probabilities for the actinides have been studied extensively in the past few years [1]. The results of these studies are useful in the identification of nuclides in nuclear fuel, waste, etc. Furthermore, well-known gamma-ray emission probabilities permit quantitative assessment of the nuclides. One application of quantitative assessment is the determination of ^{235}U in uranium, i.e. enrichment. The two principal isotopes for such a determination are ^{235}U and ^{238}U. While the gamma-ray emission probabilities for ^{235}U are well known [1], those for ^{238}U and its daughters are not. Compilations [2,3] give gamma-ray emission probabilities for the 1001 keV line of ^{234m}Pa which disagree by 10%, while the results of Moss [4] and Gunnink and Tinney [5] are in good agreement but disagree with the earlier results by 30-40%.

Since Moss only measured the emission probabilities of some of the more intense gamma rays, and we have been unable to find other reported

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measurements, the authors proceeded to design and carry out experiments to measure the emission probabilities for a wide range of gamma-ray energy and intensity in the decay of ^{238}U and its daughters. This paper describes the source preparation, detector, detector shield, data analysis methods and results. Briefly, the approach is to use a liquid source with uniformly distributed internal calibration sources which allows an accurate determination of the detection efficiency without requiring attenuation corrections. An additional important point is that the data analysis methods include estimates of the error in our results due to realistic uncertainty in the detection efficiency. The results include emission probabilities for 92 gamma-ray transitions, ranging in energy from 63 to 1937 keV. Errors (1σ) in the emission probabilities range from 0.46% to 11.2%.

2. Experimental Apparatus

2.1 Source Preparation

Uranium samples of known mass and isotopic concentration in aqueous solution were used in this measurement. These samples were produced for this measurement by Isotope Products Laboratories of Burbank, CA. The solution was prepared by dissolution of uranium metal in nitric acid. The uranium metal was certified for purity and isotopic content (0.2008 ± 0.0002 Wt.% ^{235}U) by the New Brunswick Laboratory of the U. S. Department of Energy. After drying, the resulting nitrate was dissolved in pure water producing a solution containing approximately 4 grams of uranium with a volume of nearly 15 ml and flame-sealed in standard laboratory 20 ml glass ampoules. The weights and volumes of the various samples are shown in table 1. Six samples were prepared in this manner. To certain of these samples were added small amounts of calibration sources. The added sources and their nominal activities are also shown in table 1.

2.2 Detector, Detector Shield and Source Holder

The detector used in these measurements was a high-purity germanium (HPGe) detector manufactured by EG&G Ortec, Inc with a relative efficiency of 35.7% and energy resolution of 1.74 keV at

1332 keV. Materials used in the detector assembly and cryostat were selected for low radioactivity content. The shield was made of low-activity lead and was lined with cadmium and copper. The central cavity was 30x30x40 cm. A background spectrum collected for the same time as that for the uranium samples is shown in fig. 1. A source holder fabricated of acrylic plastic was used to position the 2.3x7.7 cm ampoules approximately 6.5 cm above the detector. Figure 2 shows the source ampoule, source holder and detector.

2.3 Electronics

Standard spectroscopy-grade nuclear electronics were employed for the measurements; this included using pulse pile-up rejection and dead-time correction due to losses in the linear amplifier. The amplifier was a Canberra, Inc. Model 2020. The Nuclear Data, Inc. ND9900 pulse-height analyzer recorded 8192 channels of data using a ND 583 analog-to-digital converter. Due to the low specific activity of uranium and the small (<100 nCi) quantities of the calibration sources, the dead time was typically less than 1%. The system was quite stable since the energy resolution remained the same for counts of a few thousand seconds duration and for counts of up to 200,000 seconds.

3. Data Collection

Soon after we obtained these uranium solution sources, measurements were performed to determine if there was any evidence of settling-out of the uranium material. No such evidence was found.

Since the count rate from these samples was very low, the counting times necessary for statistical uncertainties less than 1% on even the most intense lines were quite long. The data collection for this work consisted of a series of 200,000 second counts for each of the six sources spanning approximately a month; followed by a two month waiting period and another month-long set of measurements. Since no changes, other than those consistent with counting statistics, were observed in the peak intensities, the results of all measurements were included in a weighted average.

4. Data Analysis

Gamma-ray spectra from the uranium and calibration sources were analyzed for peak position (energy) and intensity by the program HYPERMET [6,7] using a Digital Equipment Company MicroVax II computer.

The driving force behind this work has been to measure accurately and with high precision the gamma-ray emission probabilities of ^{238}U and its daughters. This required the detection efficiency of the spectrometer to be known to high precision as well. Since there may be considerable uncertainty in determining the self-absorption correction that must be made when using metallic foils of uranium, the authors opted for eliminating this correction entirely by homogeneously mixing the source material used to determine the detection efficiency throughout the attenuating (uranium) matrix.

The detection efficiency as a function of energy was determined by making use of the well-known absolute gamma-ray emission probabilities of ^{235}U [1] and the well-known relative gamma-ray emission probabilities of ^{152}Eu [8], ^{88}Y [3], ^{125}Sb [9], ^{60}Co [3], ^{133}Ba [10] and ^{228}Th [1,3]. Since the mass of uranium, isotopic abundance of ^{235}U and the gamma-ray emission probabilities of ^{235}U are all known accurately, the detection efficiency can be determined accurately in the energy range 143-205 keV. Using the multi-line internal calibration sources, segments of the efficiency curve can be determined on a relative basis. All of the efficiency calibration data were included in a model where the logarithm of the efficiency was described by a fifth order polynomial in the logarithm of the gamma-ray energy. With this model, not only the coefficients of the polynomial terms were determined, but also the normalization constant for each of the relative multi-line internal calibration sources was obtained. Since the masses, volumes and geometries of the various sources were highly consistent (well within the errors discussed below), the calibration lines in all sources were used in order to produce one overall efficiency curve applicable to all sources.

The model which was used to describe the functional form between detection efficiency and gamma-ray energy is given by

$$y = \ln(\varepsilon) = \sum_{j=0}^{5} a_j \ln\left(\frac{E_{max} + E_{min}}{2E}\right)^j , \quad (1)$$

where ε denotes the detection efficiency, E the gamma-ray energy, and the six coefficients a_j are determined from a minimization of χ^2, given by

$$\chi^2(a_j,b_k) = \sum_{i=1}^{N} \left(\frac{y_i - Y_i}{\sigma_i}\right)^2 . \quad (2)$$

The standard deviation in the measured values Y_i is σ_i, and Y_i for the N measured efficiencies is expressed as

$$Y_i = \ln(\varepsilon_i) + \ln(b_k) . \quad (3)$$

The parameters b_k are also determined by this minimization technique and are the normalization factors for the activity of the calibration sources. Table 2 shows a summary of the results of the χ^2 minimization for the detection efficiency as well as the normalization factors, b_k. For each multi-line source there is only one factor, b_k, which applies to all of the nuclide's gamma rays. The normalization factor for ^{235}U is fixed at 1.

The errors associated with the measured detection efficiency values include contributions from statistical errors in the peak intensities, uncertainties in the gamma-ray emission probabilities, decay correction errors due to half-life uncertainties, uncertainties in the coincidence summing correction and uncertainties in the normalization factors for the calibration sources as determined from the χ^2 minimization. The ^{235}U gamma rays, while not having an uncertainty due to the normalization factor, do have errors due to uncertainties in the mass of ^{235}U in the samples. No effort was made to correct for random summing, since for

the most active source the full spectrum count rate was less than 200 sec^{-1} and electronic pulse-pileup and live-time corrections were employed. These errors for each measurement have been added in quadrature. Weighted averages of the independent measurements were computed with the stated uncertainty being the larger of the external or internal errors [8]. The uncertainties in the coincidence summing corrections were arbitrarily taken to be 50% of the correction. These errors were typically less than 0.7%. An additional error in the peak intensity of the 898.1-keV gamma ray of ^{88}Y is due to the interference with a weak 898.5-keV gamma ray from the decay of ^{238}U. The largest single source of errors in the efficiency determinations was the uncertainties in the source normalization factors. These ranged from 1.2% for ^{228}Th to 1.6% for ^{125}Sb.

Following the minimization of eq. (2) with respect to the parameters a_j, the full covariance matrix has been used [11] to estimate the uncertainties in efficiency values predicted by the model. The fractional uncertainty associated with the predicted values of efficiency is shown in fig. 3 and the efficiency curve for gamma-ray energy greater than 122 keV is shown in fig. 4.

The detection efficiency for the energy region less than 100 keV has been established from the measured efficiencies for the 59.54-keV gamma ray from ^{241}Am and the 80.99-keV gamma ray of ^{133}Ba.

5. Results

Figure 5 illustrates a 200,000-second spectrum of one of the uranium samples without an added internal calibration source.

Since two spectra were recorded for each sample, there were twelve spectra which were analyzed. In a few cases the internal calibration sources interfered with weak uranium peaks. For those peaks, only six or eight peak areas were determined; for the remainder, all twelve values were obtained. Weighted averages were used to obtain the gamma-ray emission probabilities. A half-life of $(4.468 \pm 0.0005) \times 10^9$ years was used in the calculation. Errors from the efficiency curve, the half-life, and the mass of ^{238}U in the sample were combined in quadrature with the

maximum of the internal or external error of the weighted averages. Table 3 shows the results obtained in this work for the gamma-ray emission probabilities of ^{238}U and its daughters in equilibrium. The errors (1σ) are also given. No attempt was made to perform precise energy measurements, but the authors believe that the stated energy values shown in table 3 have errors of approximately ± 0.1 keV. The energies of strong gamma rays agree to within approximately 0.1 keV with those of Gunnink and Tinney [5].

The emission probabilities of the most prominent gamma rays are compared with those of Moss [4] and Gunnink and Tinney [5] in table 4. It should be noted that the errors of [5] include only statistical effects. The gamma-ray emission probabilities of [5] and the present work are in good agreement at all except the 946-keV gamma ray. Quoted errors are considerably better in the present work for the two lowest-energy gamma rays, are slightly better up through the 1001-keV gamma ray and are comparable at higher energies. Moss's [4] emission probabilities are generally in agreement with the present work; but except for the 1001-keV gamma ray, his quoted errors are much larger. Moss's emission probability (and error) for the 1001-keV gamma ray is in excellent agreement with the present work.

References

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[11] Louis Lyons, "Statistics for Nuclear and Particle Physicists" (Cambridge University Press, Cambridge, 1986).

Figure Captions

Fig. 1 Background spectrum accumulated for 200,000 seconds with the uranium source ampoule replaced by an empty glass ampoule.

Fig. 2 Spatial relationship between the germanium detector, source holder and the uranium source ampoule.

Fig. 3 Fractional error in the efficiency values predicted by model.

Fig. 4 Efficiency curve for the energy region from 122 keV to 2614 keV.

Fig. 5 Gamma-ray spectrum of uranium sample accumulated for 200,000 seconds.

Table 1
Samples used in the experiment

Sample	Uranium Mass (g)	Source Volume (ml)	Calibration Source	
			Nuclide	Nominal Activity (nCi)
1	3.9953	14.97	^{241}Am	60
			^{60}Co	14
			^{88}Y	12
2	3.9947	14.97	^{152}Eu	11
			^{125}Sb	4
3	3.9941	14.96	^{133}Ba	15
			^{228}Th	3
4	3.9937	14.96		
5	3.9943	14.96		
6	4.0032	14.94	^{241}Am	60
			^{60}Co	14
			^{88}Y	12

Table 4
Comparison of results with other works

Energy (keV)	Gamma-Ray Emission Probability (%)		
	Moss [4]	Gunnink and Tinney [5]	Present Work
238		0.073 ± 0.002	0.0730 ± 0.0003
743	0.097 ± 0.006	0.095 ± 0.002	0.0946 ± 0.0007
766	0.333 ± 0.007	0.313 ± 0.003	0.322 ± 0.002
786	0.055 ± 0.004	0.055 ± 0.001	0.0554 ± 0.0005
946	0.034 ± 0.004	0.0355 ± 0.0007	0.0335 ± 0.0003
1001	0.834 ± 0.007	0.828 ± 0.008	0.839 ± 0.005
1510		0.0130 ± 0.0003	0.0129 ± 0.0002
1738		0.0212 ± 0.0002	0.0212 ± 0.0002
1831		0.0175 ± 0.0002	0.0172 ± 0.0002
1937		0.00287 ± 0.00006	0.00290 ± 0.00007

Table 2 Summary of detection efficiency determination for HPGe spectrometer with aqueous uranium samples

Nuclide	Ref.	Normalization Factor [a]	Energy (keV)	Normalized [b] Efficiency ($\times 10^3$)	Exper. Error (%)	Model Efficiency ($\times 10^3$)	Difference (%)
^{235}U	[1]		143.76	4.25	0.32	4.27	-0.20
			163.33	4.96	0.47	5.00	-0.67
			185.72	5.52	0.17	5.50	0.22
			205.31	5.69	0.45	5.72	-0.66
^{241}Am	[3]		59.54 [c]	0.56	0.77		
^{60}Co	[3]	1.00	1173.24	2.44	1.70	2.44	-0.01
			1332.50	2.23	1.71	2.23	0.00
^{88}Y	[3]	1.01	898.07	2.92	1.67	2.91	0.46
			1836.08	1.73	1.66	1.73	-0.51
^{133}Ba	[10]	0.99	80.99 [c]	3.17	4.47		
			276.39	5.61	1.43	5.65	-0.69
			302.85	5.45	1.36	5.48	-0.52
			356.00	5.10	1.35	5.09	0.16
			383.84	4.92	1.40	4.89	0.42
^{152}Eu	[8]	1.03	121.78	3.24	2.26	3.07	5.57
			244.69	5.82	1.54	5.79	0.44
			344.29	5.17	1.34	5.18	0.01
			778.92	3.22	1.56	3.18	1.03
			964.11	2.76	1.36	2.78	-0.76
			1085.89	2.58	1.39	2.57	0.10
			1112.08	2.53	1.38	2.53	-0.23
			1408.06	2.15	1.35	2.14	0.44
^{125}Sb	[9]	0.97	427.88	4.63	1.82	4.60	0.50
			463.37	4.36	2.44	4.39	-0.75
			600.50	3.70	1.90	3.75	-1.20
			635.89	3.67	2.39	3.61	1.55
^{228}Th	[1,3]	0.92	238.63	5.79	1.52	5.80	-0.22
			583.14	3.81	1.67	3.81	-0.05
			2614.60	1.24	2.21	1.24	0.20

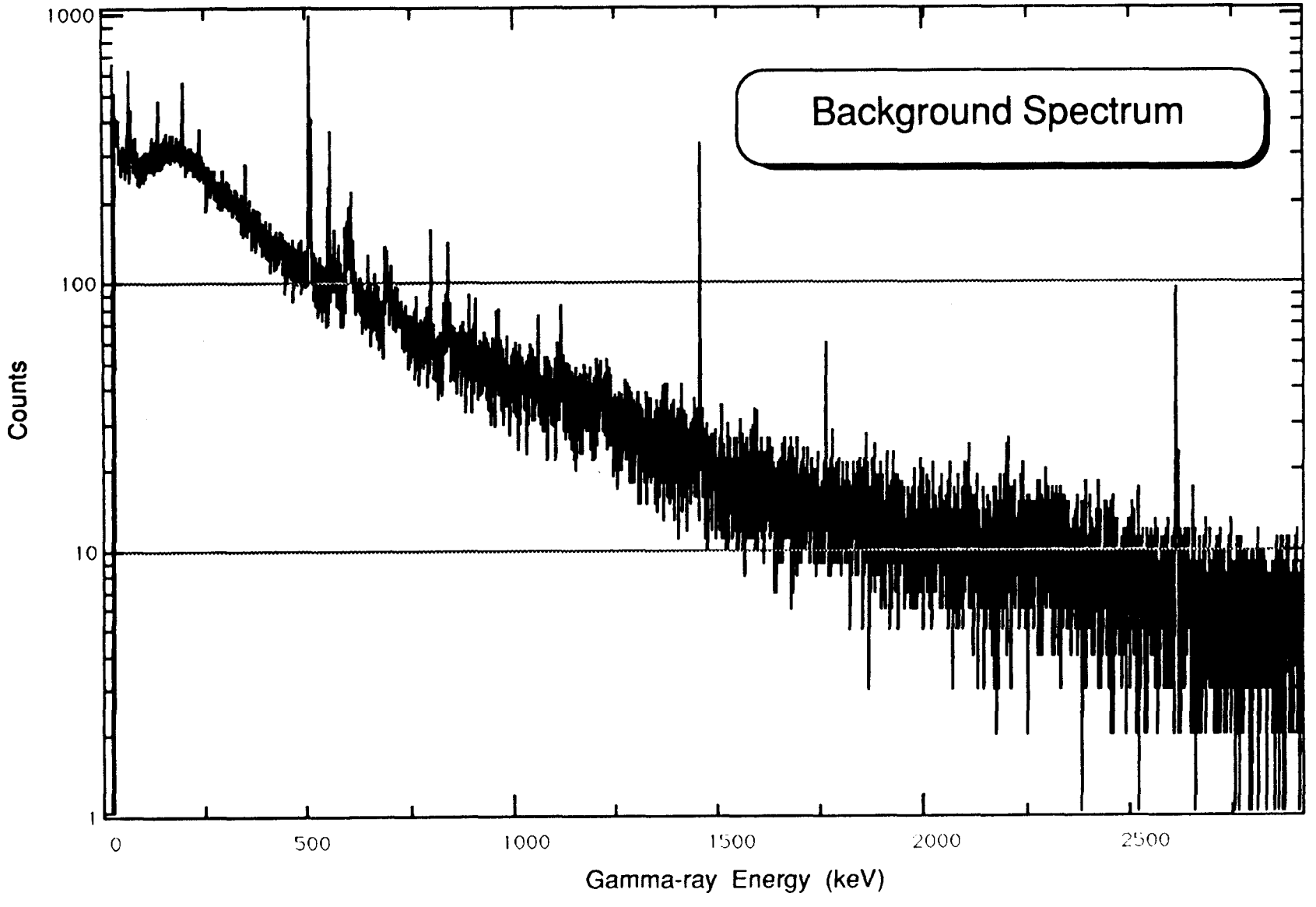
a) Determined from χ^2 minimization. Calibration source activity normalization factor.

b) Measured efficiency multiplied by normalization factor.

c) Not included in χ^2 minimization.

Table 3 Emission probabilities for gamma rays following the decay of ^{238}U

E_γ (keV)	P_γ (x10^2)	Error (%)	E_γ (keV)	P_γ (x10^2)	Error (%)
63.24	3.6	3.0	921.70	0.0127	1.1
131.31	0.0286	1.4	924.98	0.0142	1.2
152.76	0.0083	3.7	926.61	0.0192	1.1
203.12	0.0027	8.0	941.94	0.0025	4.2
226.95	0.0167	1.3	945.90	0.0335	0.86
249.21	0.0035	4.7	947.43	0.0031	4.4
258.26	0.0730	0.46	980.42	0.0045	3.0
272.20	0.0018	9.1	984.09	0.0030	4.2
293.74	0.0049	3.1	994.93	0.0057	2.1
369.52	0.0044	3.5	1000.99	0.839	0.56
372.02	0.0023	6.9	1041.70	0.0012	8.0
450.96	0.0030	5.2	1061.86	0.0023	5.2
453.58	0.0019	8.4	1084.25	0.0012	7.5
458.63	0.0020	8.0	1124.93	0.0042	3.1
468.44	0.0023	6.8	1193.69	0.0135	0.96
475.75	0.0023	6.5	1220.37	0.0009	10.2
506.70	0.0035	5.5	1237.24	0.0053	1.8
543.98	0.0036	4.7	1292.66	0.0009	11.2
569.30	0.0203	1.3	1352.80	0.0019	4.1
654.37	0.0022	7.6	1393.57	0.0039	2.5
666.42	0.0015	9.8	1413.88	0.0023	4.2
669.64	0.0017	8.9	1434.13	0.0097	1.3
691.08	0.0090	2.1	1452.63	0.0012	7.3
699.02	0.0059	2.6	1510.20	0.0129	1.2
702.05	0.0071	2.4	1527.27	0.0024	3.7
705.90	0.0065	2.4	1548.12	0.0014	5.9
733.38	0.0115	1.5	1553.74	0.0081	1.6
737.88	0.0021	8.3	1570.67	0.0011	7.8
739.95	0.0118	2.1	1591.65	0.0019	5.2
742.77	0.0946	0.70	1593.88	0.0027	3.6
755.00	0.0021	8.1	1668.44	0.0012	6.2
766.37	0.322	0.65	1694.08	0.0013	5.9
781.73	0.0078	2.2	1737.73	0.0212	1.1
786.25	0.0554	0.93	1759.81	0.0014	4.4
796.42	0.0054	4.3	1765.44	0.0087	1.4
805.74	0.0088	1.8	1809.04	0.0037	2.1
808.20	0.0026	10.0	1819.69	0.0009	7.3
819.21	0.0037	3.9	1831.36	0.0172	1.3
824.94	0.0068	2.6	1863.09	0.0012	4.3
831.39	0.0078	1.9	1867.68	0.0092	1.4
851.57	0.0070	2.0	1874.85	0.0082	1.5
875.94	0.0042	3.0	1877.21	0.00165	3.4
880.45	0.0212	0.90	1893.50	0.00219	2.9
887.28	0.0071	0.18	1911.17	0.0063	1.6
898.52	0.0059	2.2	1925.42	0.0005	10.1
883.22	0.0211	0.90	1937.01	0.00290	2.3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0
500
1000
1500
2000
2500
Gamma-ray Energy (keV)

Figure 1

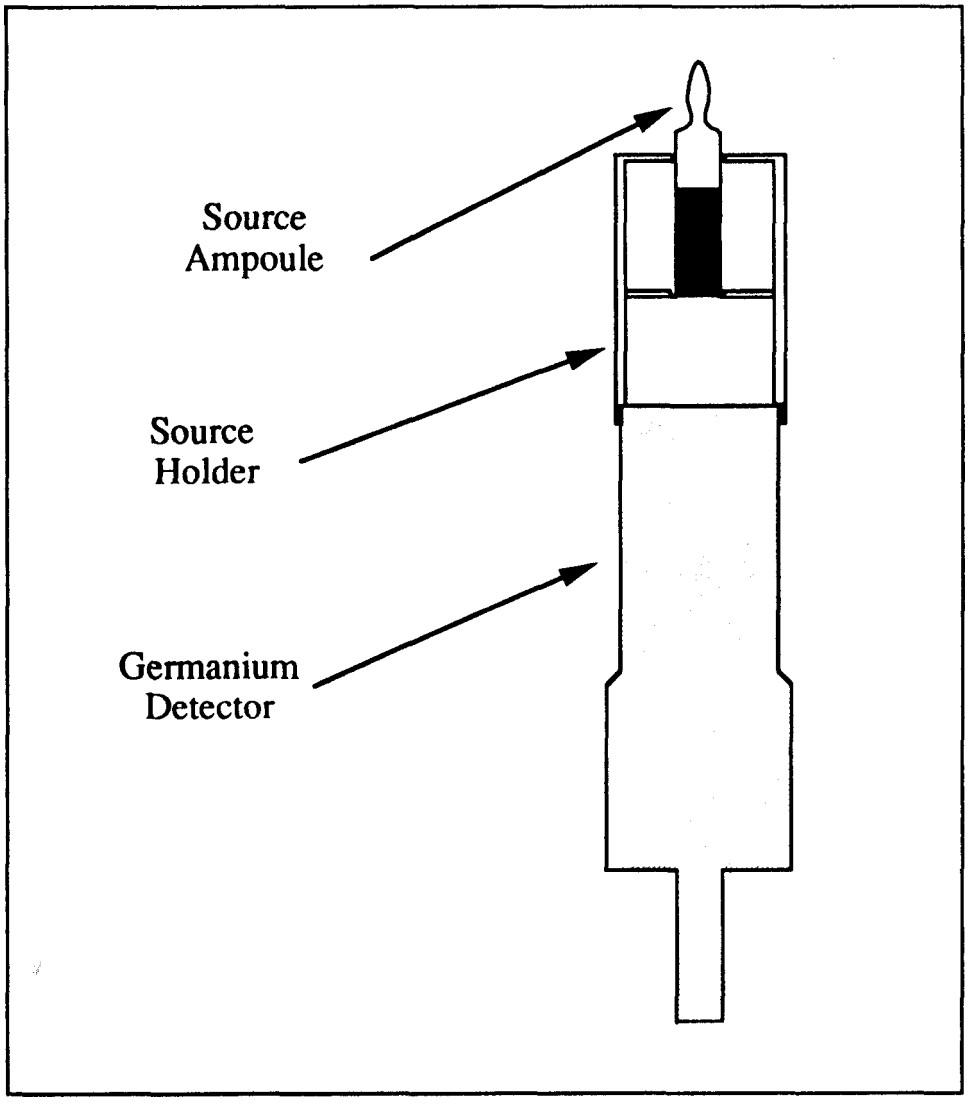


Figure 2

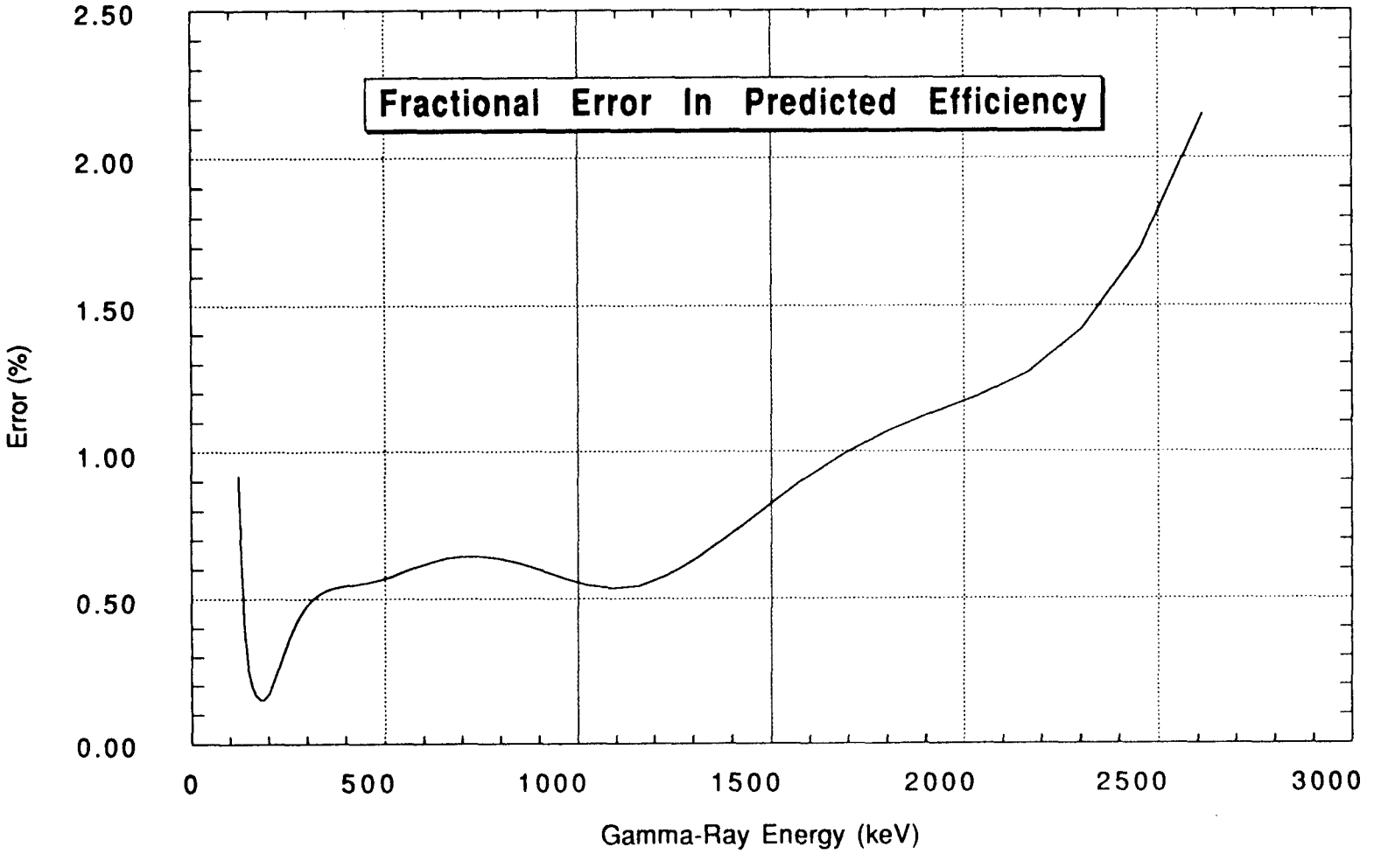
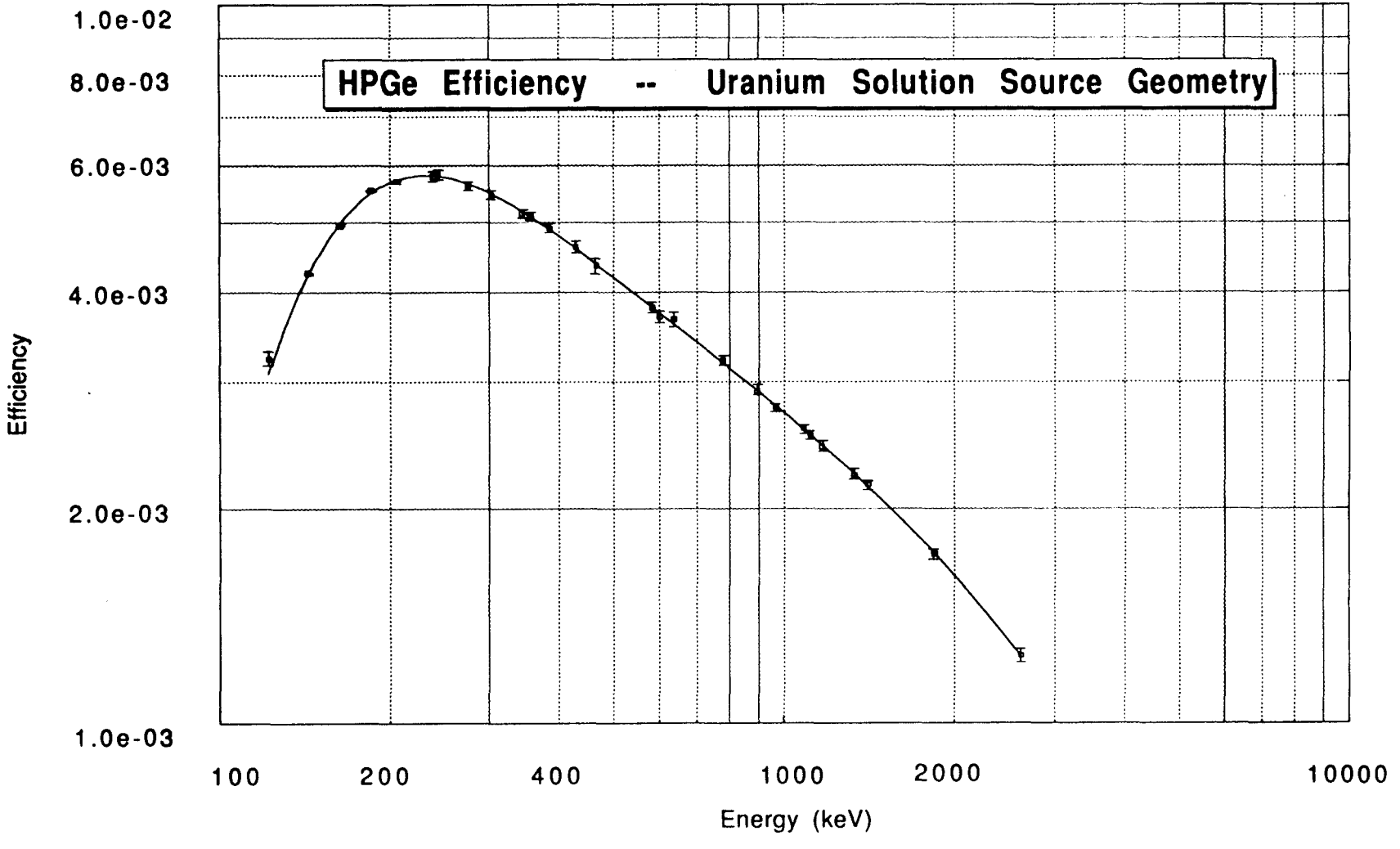

Fractional Error In Predicted Efficiency
Error (%)
2.50
2.00
1.50
1.00
0.50
0.00
0
500
1000
1500
2000
2500
3000
Gamma-Ray Energy (keV)

Figure 3


HPGe Efficiency -- Uranium Solution Source Geometry
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8.0e-03
6.0e-03
4.0e-03
2.0e-03
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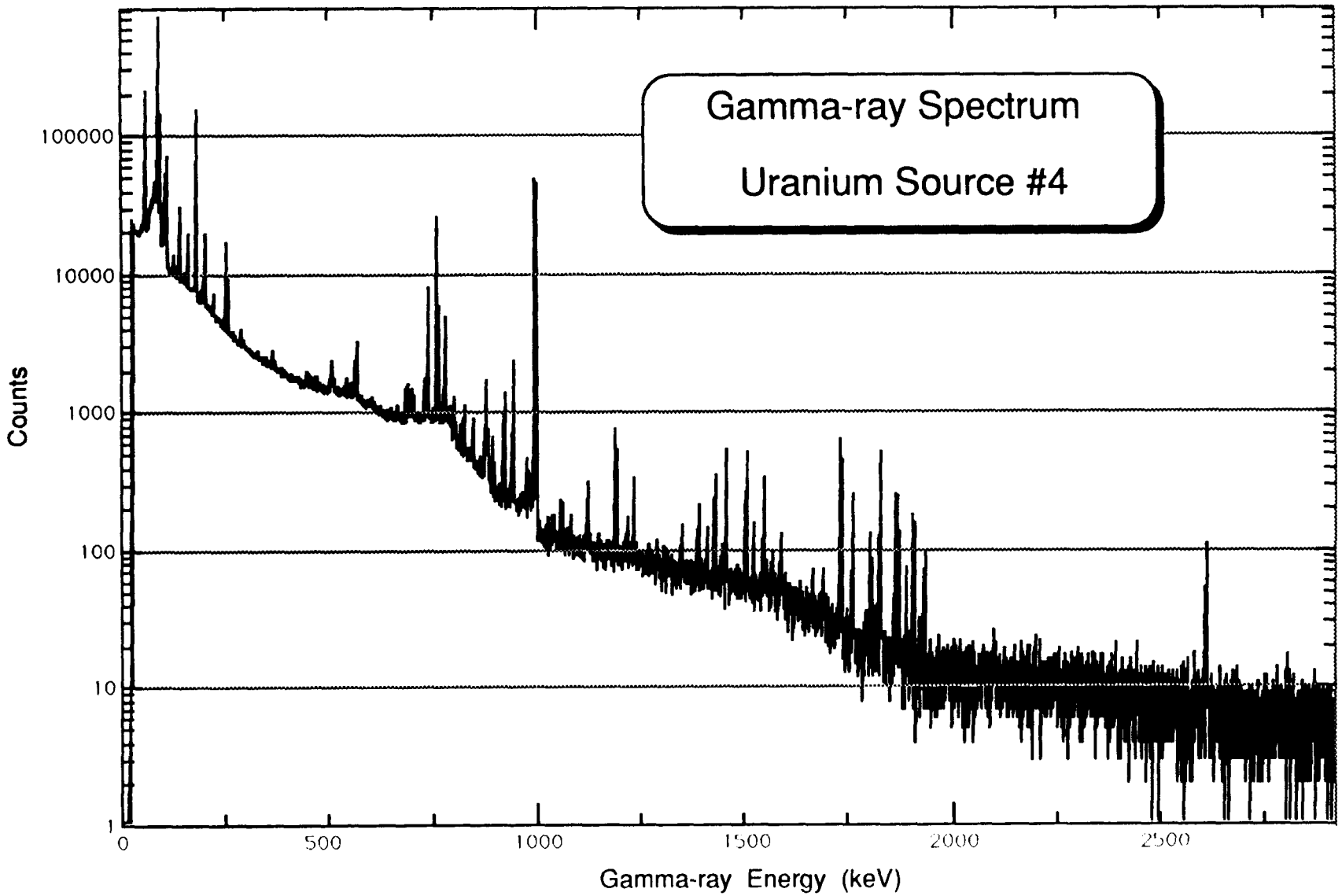
Efficiency
100
200
400
1000
2000
10000
Energy (keV)

Figure 4


Gamma-ray Spectrum
Uranium Source #4
Counts
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10000
1000
100
10
1
0
500
1000
1500
2000
2500
Gamma-ray Energy (keV)

Figure 5