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ANALYSES OF URANIUM AND ACTINIUM GAMMA SPECTRA:

An Application to Measurements of
Environmental Contamination

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ABSTRACT

A system for the reduction of the complex gamma spectra of nuclides in the uranium, actinium, and thorium series, tailored to calculation of line intensities, analyses of errors, and identification of nuclides is described. This system provides an efficient technique for characterizing contamination in the environs of uranium mines and mills. Identification of the nuclides and calculation of their concentrations require accurate knowledge of gamma energies and absolute quantum intensities. For some spectral lines, there are no reported measurements of absolute quantum intensities and in some cases where reports are available the measured intensities are not in agreement. In order to improve this data base, the spectra of gamma rays (of nuclides in the uranium and actinium series) with energies between 40 and 1400 keV were measured using high-resolution germanium detectors. A brief description of the spectroscopy system, computational algorithms for deconvolution, and methods of calibration for energy and efficiency, are described. The measured energies and absolute quantum intensities are compared with those reported in the literature.

1. Introduction

Application of the gamma spectroscopy system, which utilizes the Ge(Li) detector, to measurement of environmental contamination requires deconvolution of spectra, search and identification of each gamma line, and calculation of activity for each detected radionuclide. Gamma line identification and activity calculation require calibration of the pulse height and the efficiency of the detector as a function of energy. Moreover, the absolute quantum yield for the gamma transitions must be known. In principle, the accuracy of the predicted activity depends on the accuracy of the deconvolution procedure, efficiency

calibration, and absolute quantum yield. In this article, I describe the calibration procedure and the measurement of intensities for the uranium and actinium series. The quantum yield for the radionuclides in the thorium series, detailed procedure for peak identification, and estimation of radionuclide activities will be reported elsewhere [1].

2. Spectral Measurements

The design of the spectrometer is shown in fig. 1. The detector is an upward true coaxial lithium-drifted germanium detector (ORTEC) with a resolution of 1.8 keV at the 1.33 MeV Co-60 photopeak, with a peak-to-Compton ratio of (44:1). The detector and the cooled FET (ORTEC) preamplifier are both housed in a 12-cm thick steel shield, which stands at a height of about 100 cm. The following systems were utilized: analog-to-digital converter (Ino-Tech model 2105, 50 MHZ), spectroscopy amplifier (Ortec Model 472A), livetime and pileup corrector (Canberra), random dual pulse generator (designed at Argonne National Laboratory), Ino-Tech ultima II multichannel analyzer (model 3408), 32K NOVA computer, and Tektronix systems 4014-1, 4054, and 4907.

The zero channel and the gain were software controlled. Two random, energy-stable nuclear pulses were generated, one at the lowest energy channel (first 10 channel) and the other at the highest. These reference pulses (1000 cps) were stored in a buffer memory and were scanned once every 100 seconds. Subsequently, the most likely centroid channels for these reference pulses were calculated and used to compute the zero channel and the gain. Comparison of the centroids calculated over a period of time for gamma lines of known intensity, within an energy range of 40 keV to 2 MeV, showed no evidence of drift in gain or degradation of peak resolution.

The detailed procedure for deconvolution will be reported elsewhere [1]. The peak shape function was a variable-width Gaussian function folded into an exponential and step function on the low energy side of the peak. The background continuum was represented by a polynomial. For deconvolution, the spectrum was divided into multiplets, and peak shape and background functions were fitted into the individual peaks within each of the multiplets. If a satisfactory chi-square (χ^2) was not achieved, the residuals were searched for hidden peaks. The fitting procedure is a non-linear, least-squares, variable-metric minimization method [2,3,4]. The peaks were identified by means of a square wave correlator convolution function with zero area [5].

3. Calibration

Primary gamma standards utilized for energy and efficiency calibration were: ^{226}Ra (uncertainty of 1.2%); ^{109}Cd (3.1%), ^{37}Co (2.7%), ^{139}Ce (1.3%), ^{203}Hg (2.3%), ^{113}Sn (3.1%), ^{85}Ar (2.5%), ^{137}Cs (2.3%), ^{88}Y (3.1%), ^{60}Co (1.6%), ^{88}T (2.5%), thorium ore ($1.01 \pm .01\%$ at a 95%-confidence level), and uranium pitchblend ore (53.55% U_3O_8) (0.04%). Additional gamma standards used were ^{241}Am , ^{210}Pb , and ^{75}Se from New Brunswick Laboratory.

The spectrum was subdivided into two adjoining energy regions, and a polynomial function was fitted to the data by a least-squares method. Figures 2 and 3 show the fit of the data to the calculated calibration curves. The efficiency curve was divided into three energy regions, and the data in each region was fitted to polynomial functions. Figure 4 shows the fit of the data to the calculated curves for the three energy regions.

4. Gamma Energies and Quantum Yields

Figures 5 and 6 show the spectra of nuclides, between 40 and 1400 keV, in a sample of natural uranium ore. Table 1 gives the energy and quantum yield of the observed lines. The detailed procedures for calculation of quantum yield and nuclide identification will be reported elsewhere [1]. The observed peaks are often composites of unresolved lines from several nuclides; e.g., the broad peak at 50 keV is a composite of lines from ^{223}Ra [6], ^{227}Th and ^{223}Fr [7]. Therefore, the composite peak location depends on the line intensities, their respective energies, and the concentrations of the nuclides in the sample. Estimation of the individual line intensities demands estimation of the relative contribution of each of the interference lines to the resolved lines.

The measured energies (table 1) are in agreement, within less than 1 keV, with those reported in the literature. The quantum yields for the major lines, i.e., those with intensities larger than 1×10^{-2} , are, in general, within the range of the reported values. For example, the quantum yield for ^{210}Pb is 4.1×10^{-2} (table 1), in agreement with the value reported by Adams and Gasparini [8]. The line at 67.67 keV is due to ^{230}Th with a quantum yield 3.63×10^{-3} . Smith and Wollenberg report a quantum yield of 3.81×10^{-3} at 67.8 keV for this line, and Kurcewicz et al. [9] report a yield of 3.73×10^{-3} at 67.7 keV. The major lines from ^{235}U are at 163.35 keV and 188.72 keV with the quantum yields 4.77×10^{-2} and 5.61×10^{-1} , respectively. Reported energies for these lines are 163.37 keV and 185.72 keV, with quantum yields 4.85×10^{-2} and 5.61×10^{-1} , respectively [7].

The major lines from ^{214}Pb are 295.16 keV and 351.89 keV with the quantum yields, respectively, 1.15×10^{-1} and 3.89×10^{-1} . In comparison, these lines are reported at 295.22 keV and 351.99 keV with the quantum yields 1.94×10^{-1} and 3.63×10^{-1} , respectively [7]. Similarly, the major lines for ^{214}Bi are at 609.27 keV and 1120.21 keV with the quantum yields, respectively, at 4.33×10^{-1} and 1.46×10^{-1} . The reported values [7] are 609.37 keV (4.28×10^{-1}) and 1120.4 keV (1.50×10^{-1}). The major line for determination of ^{238}U is 1001.04 keV (9.20×10^{-3}) from ^{234}mPa . The previously reported [7] values are 1001.10, with the quantum yield 8.28×10^{-3} .

5. Discussion

In general, the magnitude of the uncertainty associated with the computed quantum yield depends on the magnitude of the full width of the energy peak and the area under the peak, relative to the background continuum and the magnitude of the interferences. The uncertainty of estimating the contribution from the interference peaks is related to the magnitude of the uncertainty in the quantum yields and the uncertainty in the deconvolution method for resolution of the multiplets. The uncertainty in the quantum yields of less than 1×10^{-3} is smaller than 5% for the lines free from interference and more than 10% for the others. The uncertainty arises from the composite errors of deconvolution, the uncertainty in the absolute activity of the primary standards, and the magnitude of the interference. This gamma spectroscopy system and the procedure for data reduction permit measurements of concentrations of the nuclides in uranium ore and tailings with total errors of less than 5%, and, for typical environmental concentrations, with total errors of less than 15%.

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Table 1. Energy and quantum yield of selected nuclides
in the uranium and actinium series

Energy (keV)	Quantum Yield	Energy (keV)	Quantum Yield
²³⁴ Th		²¹⁴ Bi	
63.34	4.05 E-2	273.53	3.87 E-3
92.75	1.63 E-2	281.09	9.14 E-4
		286.12	3.00 E-4
		303.06	8.00 E-4
²³⁴ Pa		333.97	6.00 E-4
131.49	3.98 E-5	338.29	4.00 E-4
733.99	1.04 E-4	386.83	5.17 E-3
945.99	4.14 E-4	388.92	4.49 E-3
		405.74	1.56 E-3
		454.75	2.89 E-3
^{234m} Pa		462.24	2.25 E-3
258.44	3.31 E-4	469.86	1.18 E-3
742.54	5.00 E-4	474.60	1.06 E-3
766.28	3.68 E-3	543.56	7.67 E-4
786.27	6.60 E-4	572.70	1.00 E-3
1001.04	9.20 E-3	609.27	4.33 E-1
		615.98	9.33 E-4
²³⁴ U		633.26	6.10 E-4
53.18	1.18 E-3	639.57	2.96 E-4
		649.34	5.90 E-4
		665.42	1.25 E-2
		683.13	6.74 E-4
²³⁰ Th		698.23	3.34 E-4
67.67	3.63 E-3	703.22	3.93 E-3
144.24	2.76 E-4	710.84	7.11 E-4
		719.83	3.64 E-3
		734.30	4.99 E-4
²²⁶ Ra		752.97	1.25 E-3
186.04	5.05 E-2	768.37	4.97 E-2
		785.97	8.60 E-3
		806.28	1.04 E-2
²¹⁴ Pb		815.05	4.34 E-4
52.14	1.06 E-2	821.28	1.30 E-3
241.92	8.39 E-2	826.13	4.59 E-4
259.09	5.48 E-3	832.06	1.85 E-4
274.75	3.87 E-3	839.10	7.96 E-3
295.16	1.15 E-1	934.09	2.95 E-2
314.34	8.53 E-4	964.10	3.23 E-3
351.89	3.89 E-1	1032.24	1.18 E-3
461.92	2.25 E-3	1051.92	2.82 E-3
480.55	3.82 E-3	1069.95	1.98 E-3
487.16	3.62 E-3	1103.92	1.39 E-3
533.78	2.53 E-3	1120.21	1.46 E-1
543.56	1.17 E-3	1133.76	2.46 E-3
580.32	3.32 E-3	1155.12	1.60 E-2
785.97	8.60 E-3	1173.08	7.21 E-4
904.27	1.14 E-3	1207.62	4.30 E-3

Table 1. Contd.

Energy (keV)	Quantum Yield	Energy (keV)	Quantum Yield
1238.07	5.98 E-2	286.12	6.90 E-3
1280.95	1.33 E-2	299.93	3.89 E-3
1303.77	1.04 E-3	304.55	9.35 E-3
1377.82	4.91 E-2	314.34	4.40 E-3
1385.49	7.76 E-3	329.80	3.45 E-2
		333.97	5.11 E-4
²¹⁴ Po		²²³ Fr	
799.48	4.00 E-4	49.8	4.63 E-2
²¹⁰ Pb		²²³ Ra	
46.50	4.1 E-2	122.75	1.00 E-2
		144.24	5.86 E-2
		154.25	7.22 E-2
²³⁵ U		158.39	6.05 E-3
109.15	1.55 E-2	269.41	1.50 E-1
115.20	1.18 E-3	323.84	3.69 E-2
140.76	1.74 E-3	333.84	4.90 E-3
144.24	1.05 E-1		
163.35	4.77 E-2	²²³ Ra	
188.72	5.61 E-1	338.29	1.29 E-2
195.85	6.73 E-3	371.80	4.40 E-3
202.04	8.64 E-3	444.92	9.9 E-2
205.27	4.87 E-2		
221.38	1.23 E-3	²¹⁹ Rn	
240.93	7.85 E-4	130.75	9.66 E-3
246.83	6.17 E-4	271.20	1.20 E-1
²³¹ Th		401.84	7.98 E-2
163.64	1.09 E-2		
²³¹ Pa		²¹¹ Pb	
302.5	3.62 E-2	404.89	7.72 E-2
		426.90	1.74 E-2
		832.06	3.27 E-2
²²⁷ Th		²¹¹ Bi	
49.95	7.67 E-2	351.89	1.33 E-1
113.08	6.60 E-3		
206.10	3.59 E-3		
210.72	1.10 E-2		
235.90	1.10 E-1		
250.07	3.30 E-3		
256.18	7.75 E-2		
281.09	1.32 E-3		

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Fig 3

Fig 4

FIG 5

FIG 6