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EVALUATION OF WELL-TYPE Ge(Li) DETECTORS FOR LOW-LEVEL RADIOCHEMICAL ANALYSIS*

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SUMMARY

Well-type Ge(Li) detectors have been evaluated for low-level gamma-ray spectrometry and radiochemical analyses. The detectors were found to have good resolution, high peak-to-Compton ratios and low backgrounds. The use of an anticoincidence shield further improves the detector performance. The detector efficiencies and backgrounds are compared with those obtained with other Ge(Li) detector systems. The well detectors were found to have better detection efficiencies and as low backgrounds as either large coaxial detectors or opposed detector systems. Sum-coincidence effects are more pronounced in the well detector and use of this feature is discussed. Applications which utilize the low-energy response of the detector are described. Minimum detectable activity levels were determined for several nuclides.

INTRODUCTION

Well-type NaI(Tl) detectors have found wide applications in low-level gamma-ray spectrometry and radiochemical analyses. This results from the high efficiency and well-defined geometry of the well-type detector. Many

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applications require higher gamma-ray energy resolution than is available with NaI(Tl) detectors. Semiconductor Si(Li), Ge(Li) and Ge detectors without wells are now routinely used when high resolution is required. Several authors have reported on the construction and performance of well-type Ge(Li) detectors.[1-6] The small well sizes of Ge(Li) detectors limits their applications to small volume samples. However, the high efficiency and geometry control characteristics of well detectors make Ge(Li) well detectors attractive for small samples. This paper reports on an evaluation of well-type Ge(Li) detectors for gamma-ray spectrometric and radiochemical analysis applications.

DETECTOR PERFORMANCE

Resolution and Peak/Compton

The performance of two Ge(Li) well-type detectors has been compared with the performance of other high-resolution detector systems. Coaxial Ge(Li) detectors, planar Ge and Si(Li) detectors and opposed Ge(Li) and Ge detector pairs were used for this comparison. Characteristics of the detectors are listed in Table I. The resolution (FWHM) and peak/Compton measurements in Table I were made with point sources at the end cap of the detector.

The resolution and peak/Compton ratio of the well-type detectors were also measured at several energies with point sources in the well. Table II compares the results of the measurements in the well and at the end cap for the two well-type Ge(Li) detectors. Only a slight resolution loss was introduced in measurement with the source in the well. A significant improvement in the peak/Compton ratio was observed for measurements

TABLE I

DETECTOR CHARACTERISTICS

PNL No.	20	42	30	31	19	26	25	47	17
Type	Ge(Li) Well	Ge(Li) Well	Ge(Li) Coaxial	Two Ge(Li) Coaxial	Ge Coaxial	Ge Planar	Ge Planar	Two Ge Planar	Si(Li) Planar
Well Diameter (cm)	1	1							
Well Length (cm)	4	4							
Detector Diameter (cm)	4.8	5.3			5.4	25	16	25	20
Detector Length (cm)	5.3	4.4			5.7	1.0	0.8	1.0	0.5
Efficiency *(%)	11	13.2	10	20	26.3				
1.33 MeV FWHM (keV)	2.6	1.9	2.0	2.2	2.1				
661 keV FWHM (keV)	2.0	1.4	1.6	1.7	1.6				
122 keV FWHM (keV)	1.5	1.0	1.2	1.4	1.2	.53	.53	.57	.59
59.5 keV FWHM (keV)	1.5	0.9	1.2	1.4	1.1	.44	.42	.46	.44
Active Volume (cm^3)	55	70			120.				
Peak/Compton at 1.33 MeV	24.	36.	35.	29.	44.				
Peak/Compton at 661 keV	39.	62.	50.	45.	69.		15.	15.	

*Percent relative to 7.6x7.6 cm NaI(Tl) at 25 cm for 1.33 MeV.

TABLE II

COMPARISON OF RESOLUTION AND PEAK/COMPTON FOR POINT SOURCES AND WELL-TYPE Ge(Li) DETECTOR AT-CAPS AND IN-WELL

PNL Detector No.	Gamma Source	Gamma Energy	Resolution (FWHM)		Resolution (FWTM)		Peak/Compton	
			At Cap	In Well	At Cap	In Well	At Cap	In Well
	(Nuclide)	(keV)	(keV)	(keV)	(keV)	(keV)		
20	^{241}Am	59.5	1.49	1.48	2.84	2.65		
	^{57}Co	122.	1.53	1.68	2.84			
	^{137}Cs	662.	1.99	2.14	3.91	4.27	39.0	67.0
	^{60}Co	1173.	2.50	2.79	5.39	6.41		
	^{60}Co	1332.	2.62	2.90	5.59	6.72	23.6	29.2
42	^{241}Am	59.5	0.92	0.91	1.72	1.68		
	^{57}Co	122.	0.99	0.99	1.82	1.78		
	^{137}Cs	662.	1.42	1.43	2.59	2.64	61.9	102.4
	^{60}Co	1173.	1.80	1.83	3.33	3.43		
	^{60}Co	1332.	1.91	1.96	3.54	3.62	36.1	45.6

in the well of sources without coincident gamma-rays. Sum-coincidences effects the peak/Compton ratio of sources such as ^{60}Co which emit coincident gamma-rays.

Measurements were also made with one of the well-type detectors (No. 20) in a NaI(Tl) annulus. The NaI(Tl) annulus was 24 cm in diameter by 23 cm long with an 8 cm diameter hole along its axis. The Ge(Li) detector spectra in anticoincidence and coincidence with the annulus were accumulated in separate sections of the multichannel analyzer memory. Figure 1 shows ^{137}Cs spectra obtained with a source in the well of the Ge(Li) detector. The Compton region for normal and anticoincidence operation is shown. A factor of four reduction in the ^{137}Cs Compton region was obtained with the Ge(Li) detector in anticoincidence with the NaI(Tl) annulus.

Figure 2 shows ^{60}Co spectra obtained without the annulus, in anticoincidence with the annulus and in coincidence with the annulus. The sum peak at 2506 keV is not seen in the spectrum in coincidence with the annulus. The relative ^{60}Co peak areas from the spectra shown in Figure 2 are listed in Table III. The sum-coincident effect, which is much more important in well-type detectors results in more complex spectra but also supplies added information.[4,7] The high energy sum peaks are often unique and can be used for specific isotopic measurements in a mixture of radionuclides.

Efficiency

Well-type Ge(Li) detector efficiencies (counts/gamma-ray) were measured with the source in the well and were compared with those measured for the other detectors listed in Table I with the source at the end cap. A mixed-radionuclide gamma-ray solution standard obtained from the National Bureau of Standards (SRM-4254) was used to prepare the standard sources for the efficiency measurements.[8,9]

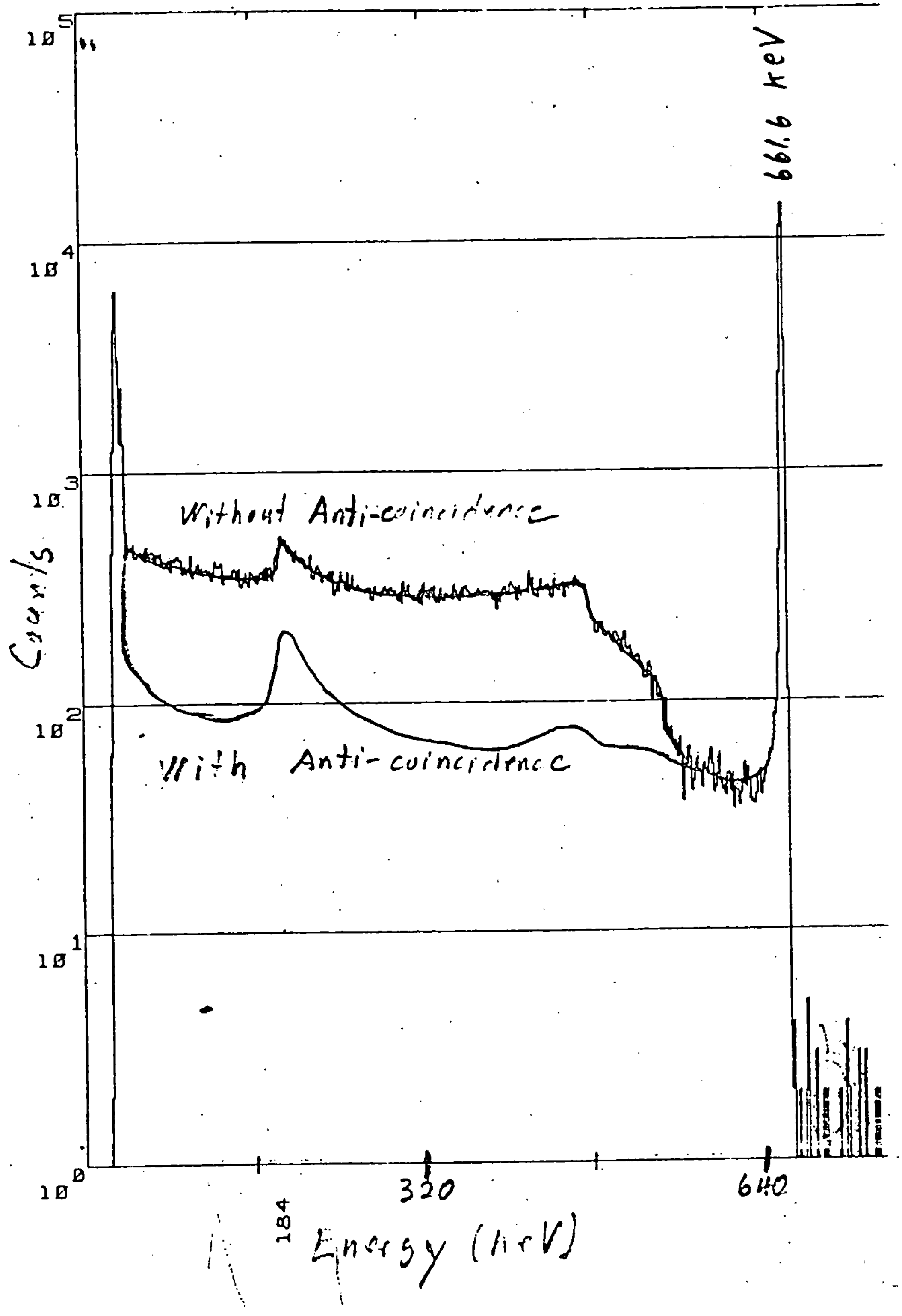

FIGURE 1. ^{137}Cs Spectra for a Well-Type Ge(Li) Detector Showing the Effect of an Anticoincidence Annulus.

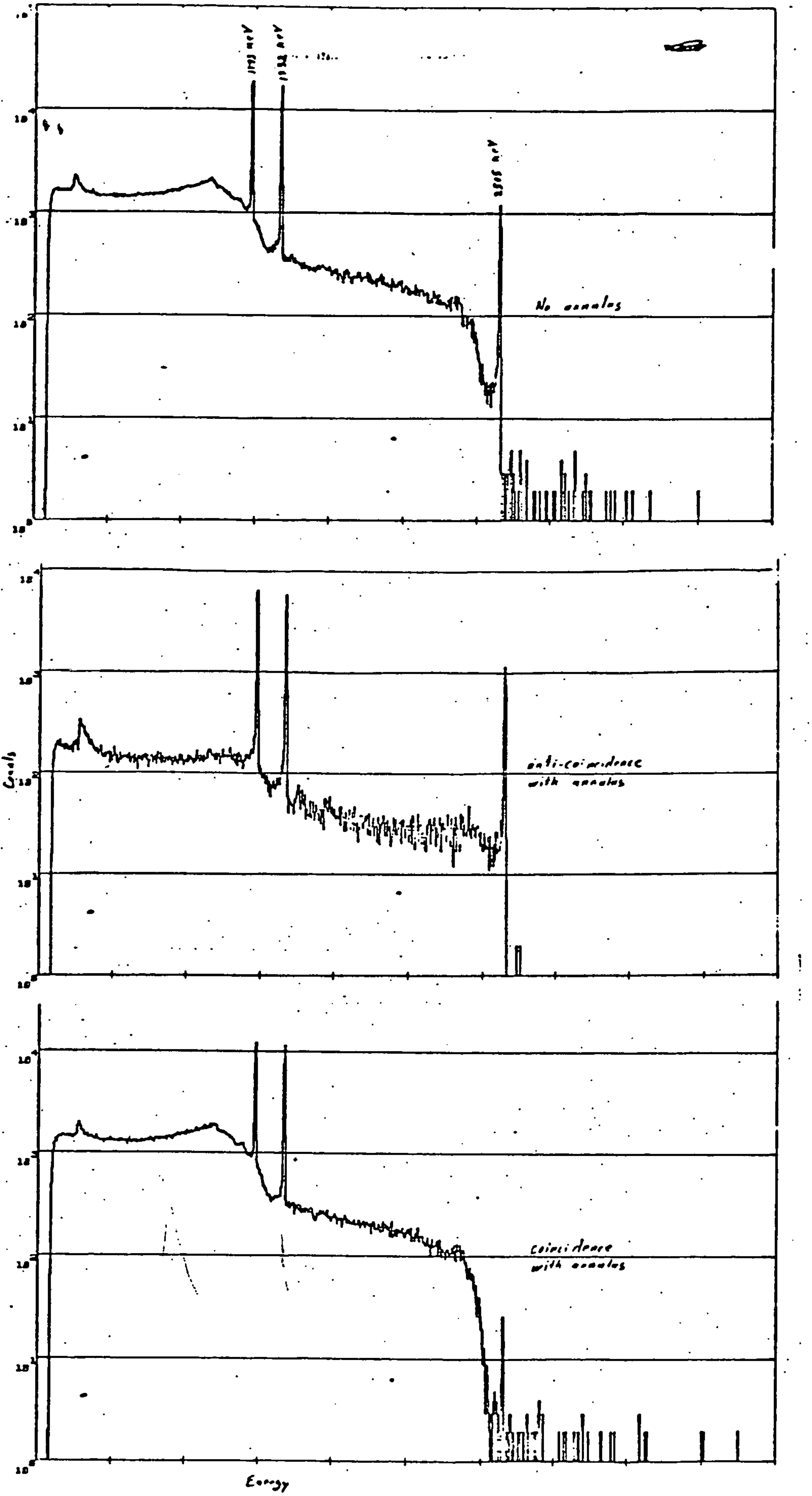

FIGURE 2. ^{60}Co Spectra for a Well-Type Ge(Li) Detector Showing the Effect of a NaI(Tl) Annulus.

TABLE III

RELATIVE PEAK AREAS FOR ^{60}Co SPECTRA IN A WELL-TYPE Ge(Li) DETECTOR

	Relative Peak Areas		
Annulus	1173 keV	1332 keV	2506 keV
None	100	84	6.4
Anticoincidence	37	30	6.6
Coincidence	63	54	

The spectrum of a NBS mixed standard source in a well Ge(Li) detector (No. 20) is shown on Figure 3. The NBS standard mixture contains several nuclides with coincident gamma-rays and x-rays which produce sum peaks. The sum-coincidence effect must be considered when the standard is used for well-type detector calibrations.[7] ^{241}Am was used for low-energy efficiency measurements. A well-type Ge(Li) detector (No. 42) spectrum of ^{241}Am is shown in Figure 4. Table IV summarizes the efficiency measurements for all the detectors used in this investigation.

The efficiency curves are shown on Figure 5 for a well-type Ge(Li) detector (No. 20), a pair of opposed, coaxial Ge(Li) detectors (No. 31), a single coaxial Ge(Li) detector (No. 30), and a planar Ge detector (No. 26). The well-type detectors were found to have significantly greater efficiencies at all energies for sources of about 1 cm^3 volume or less.

Background

Background measurements were made with the detectors in 10 cm thick lead shields. Measurements were also made with one of the well-type detectors (No. 20) in the NaI(Tl) annulus. The major background photopeak areas in counts per day are listed in Table V for each of the detectors. The well-type detector background photopeaks were found to have about the same area as similar size non-well Ge(Li) detectors. The NaI(Tl) annulus did not appreciably reduce the background photopeaks except for the lead x-ray peaks (which could be reduced by shield design) and the 511 keV

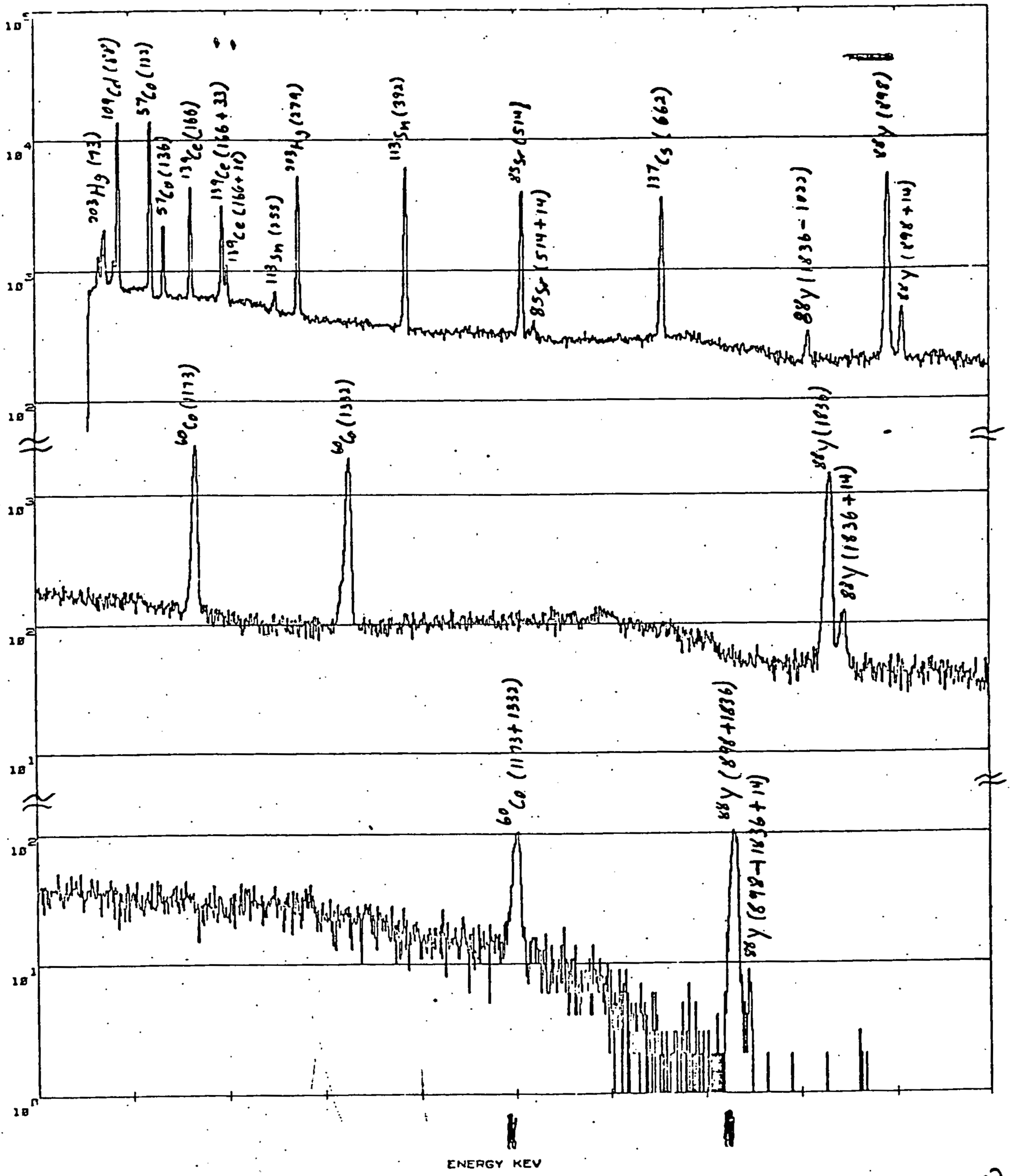

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FIGURE 3. Spectrum for a Mixed-Standard Source in a Well-Type Ge(Li) Detector

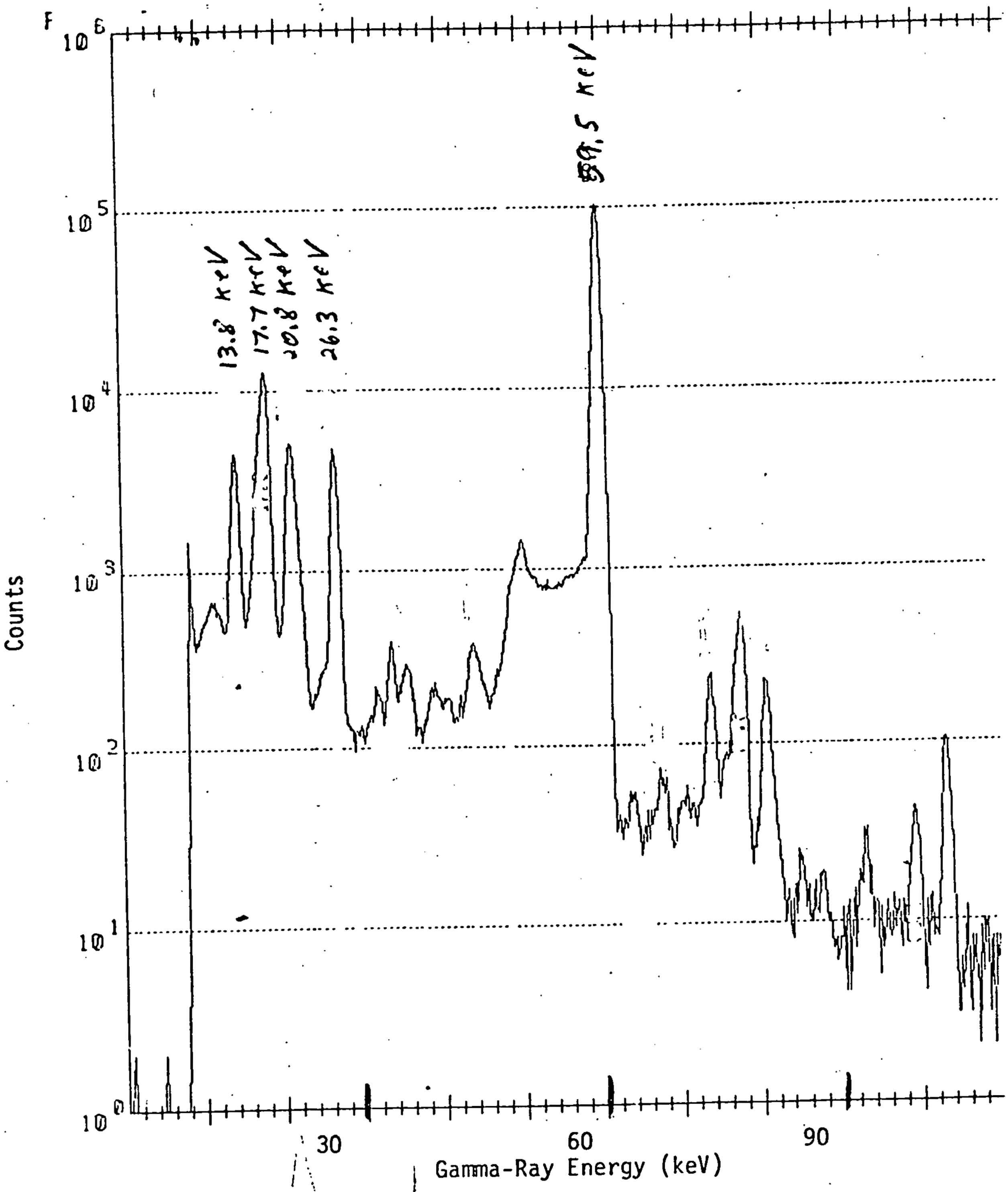

FIGURE 4. Spectrum for a ^{241}Am Source in a Well-Type Ge(Li) Detector

TABLE IV

EFFICIENCY MEASUREMENT RESULTS FOR VARIOUS DETECTORS

		Photopeak Efficiency* (Counts/gamma-ray)								
		Detector No.								
Radionuclide	X-ray or Gamma-Ray Energy (keV)	20	42	30	31	19	26	25	47	17
		Well	Well	Coaxial	2 Coaxial	Coaxial	Planar	Planar	2 Planar	Si(Li)
^{241}Am	13.8	.093	.069				.091	.034	.15	.025
^{241}Am	17.7	.23	.21				.081	.023	.13	.022
^{241}Am	26.3	.48	.44				.13	.050	.24	.027
^{241}Am	59.5	.77	.72				.15	.057	.27	
^{109}Cd	88.0	.82	.81	.13	.26	.32	.13	.061		
^{57}Co	122.1	.75	.75	.12	.28	.23	.094	.039		
^{139}Ce	165.8	.28	.29	.086	.20	.18	.050	.022		
^{203}Hg	279.2	.32	.36	.066	.11	.12				
^{113}Sn	391.7	.19	.20	.039	.086	.086	.010	.0021		
^{85}Sr	513.9	.13	.13	.024	.053	.064				
^{137}Cs	661.6	.10	.10	.021	.047	.049				
^{60}Co	1173.2	.034	.036	.011	.021	.030				
^{60}Co	1332.5	.027	.032	.0095	.019	.028				
^{88}Y	898.0	.044	.048	.013	.028	.037				
^{88}Y	1836.1	.016	.021	.0068	.013	.026				

*Not corrected for sum-coincidences.

Figure 5

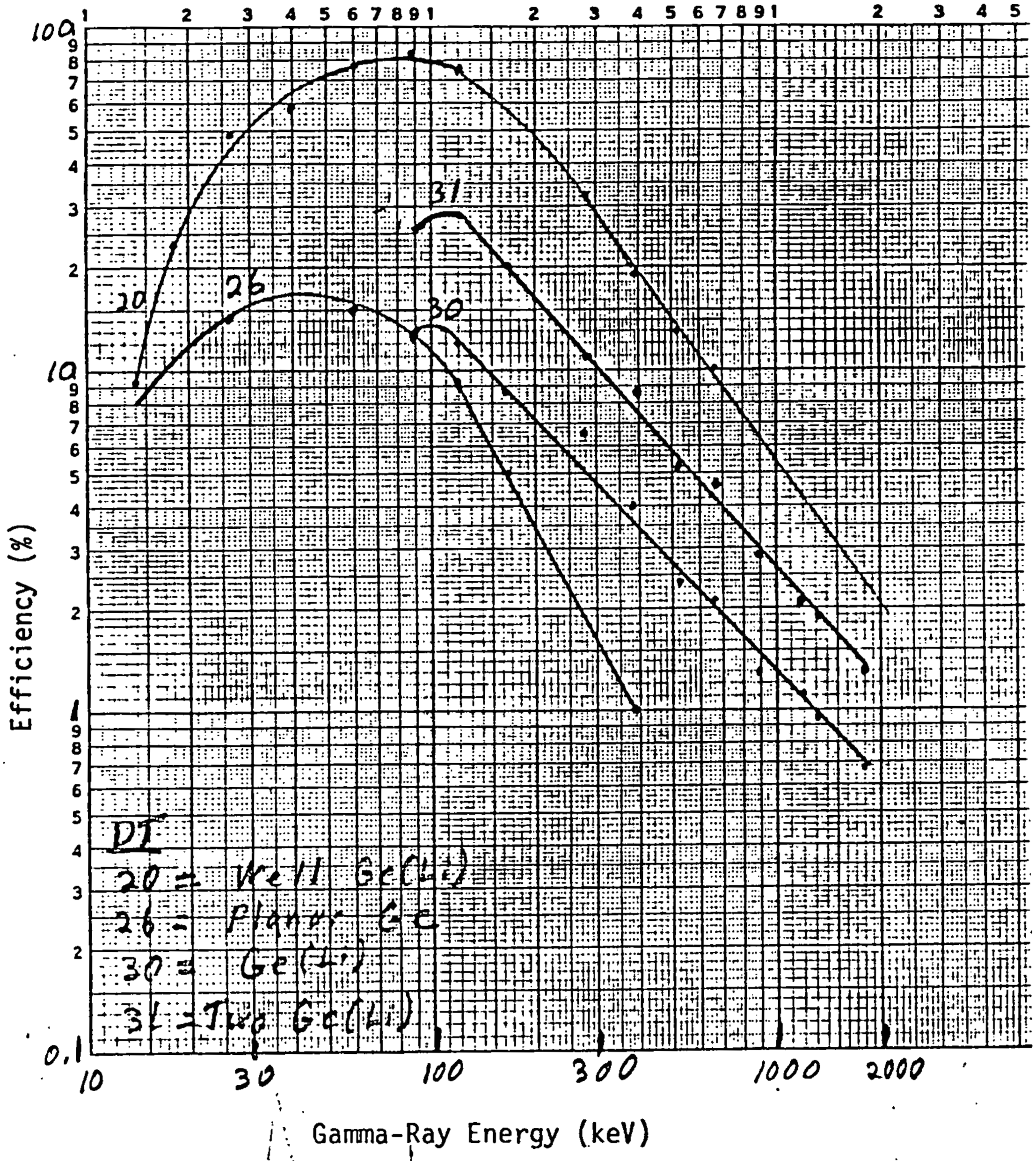

FIGURE 5. Efficiency Curves (Counts per Gamma) for Several Detector Types

TABLE V

MAJOR BACKGROUND PHOTOPEAKS IN 10 cm THICK LEAD SHIELD

Photopeak Energy (keV)	Radionuclide Parent	Radionuclide Daughter	Background Photopeak Areas (Counts/day) Detector No. 20	20*	42	30	31	19	26	25	47
63	^{238}U	^{234}Th	87	120	160	93	180		50		160
75		Pb x-ray	1600	230	740	780	1400	600	280	130	370
86		Pb x-ray	690	60	630	450	820	360	130	98	120
93	^{238}U	^{234}Th	280	370	710	170	510	75	130		370
185	^{226}Ra, ^{235}U		240	220	500	120	380	55			65
239	^{228}Th	^{212}Pb	380	340	260	390	1300	700			
295	^{226}Ra	^{214}Pb	84	45	130	64	290	71			
338	^{228}Ra	^{228}Ac	33	48		39	130	190			
352	^{226}Ra	^{214}Pb	160	100	130	110	440	86			
511			900	130	850	470	1300	1200			
583	^{228}Th	^{208}Tl	100	66	120	110	380	180			
609	^{226}Ra	^{214}Bi	140	93	140	90	350	75			
911	^{228}Ra	^{228}Ac	54	87		36	120	180			
969	^{228}Ra	^{228}Ac	16			27	66	82			
1120	^{226}Ra	^{214}Bi	27			19	81	12			
1461	^{40}K		170	370	84	79	210	70			
1764	^{226}Ra	^{214}Bi	29	27		18	58	21			
2615	^{228}Th	^{208}Tl	61	59	66						

*Well-type detector in anticoincidence with NaI(Tl) annulus.

peak. Increases in the ^{40}K and ^{234}Th peaks were observed when the NaI(Tl) annulus was used.

The background continuum between the photopeaks was significantly reduced by the anticoincidence NaI(Tl) annulus. This effect is seen in the background spectra shown in Figure 6.

The NaI(Tl) anticoincidence shield results in lower backgrounds for most radionuclide measurements. The background levels at several important energies for different detectors are listed in Table VI.

APPLICATIONS

The high-efficiency of the well-type Ge(Li) detectors obtained with no increase in background or decrease in resolution make them ideal for low-level gamma-ray spectrometry of small volume samples. The minimum detectable activity obtainable with a Ge(Li) well-type detector (No. 20) has been calculated for several radionuclides by the method described in reference 10. A counting time of one day and a 0.33 value for the peak area standard deviation divided by the peak area were used in the computations. The sample was assumed to be free of activities which increase the background under the photopeaks. The minimum detectable activities are listed in Table VII. Since the background of the well-type Ge(Li) detectors are not significantly different than backgrounds of similar size non-well Ge(Li) detectors, minimum detections limits for the detectors are directly related to the differences in efficiencies for detectors with similar resolution. The resolution and background data can be used to calculate detection limits as required for evaluation of specific measurement problems.

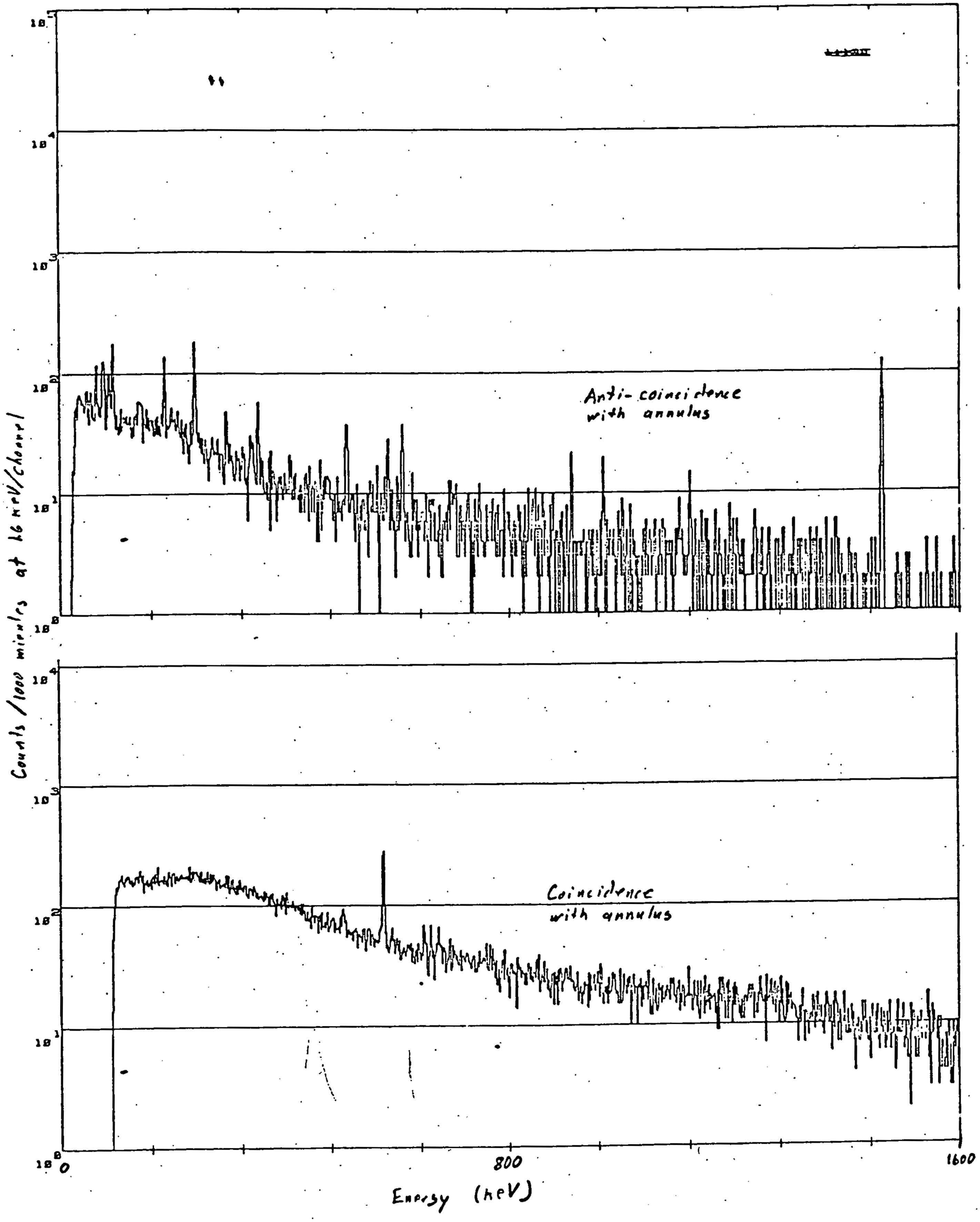

FIGURE 6. Background Spectra for a Well-Type Ge(Li) Detector with a NaI(Tl) Annulus.

TABLE VI

BACKGROUND LEVELS AT SEVERAL ENERGIES

Detector No.	Shield	Background Counts/Day for 1 keV at Energy (keV)						
		17	39	60	122	364	662	1332
20	Pb	500	220	290	220	76	30	11
20	Anticoincidence	160	50	45	40	16	4	3
20	Coincidence	280	150	150	140	85	36	14
42	Pb	580	300	350	190	78	30	11
30	Pb		100	160	86	45	17	7
31	Pb			420	340	110	50	20
19	Pb			390	250	98	50	43
26	Pb	100	66	70	39	6		
25	Pb	100	80	40	22	7		
47	Pb	130	110	170	95			
17	Pb	144						

TABLE VII

MINIMUM DETECTABLE ACTIVITY FOR SEVERAL RADIONULCIDES IN A WELL-TYPE Ge(Li) DETECTOR FOR 1440 MINUTE COUNTS

Radionuclide	Gamma-Ray Energy (keV)	Minimum Detectable Activity (pCi) No AC*	AC*
^{129}I	39.8	0.8	0.5
^{241}Am	59.5	0.14	0.08
^{131}I	364.5	0.16	0.08
^{137}Cs	661.6	0.23	0.10

*AC = Anticoincidence annulus for background reduction

The high efficiency of the well-type Ge(Li) detector at energies below 100 keV has application in the measurement of radionuclides such as ^{239}Pu, ^{241}Am, ^{125}I, ^{129}I and other low energy gamma-ray or x-ray emitters. Low detection limits were also obtained for higher energy gamgamma-ray emitters such as ^{131}I and ^{137}Cs. Several low-level measurement methods previously reported can be improved by use of well-type Ge(Li) detectors.[11,12,13] However, the limitation of small sample volume must be noted.

The presence of coincident gamma-rays or x-rays produces complex spectra as the result of sum-coincidence effects. The sum-coincidence effects are useful for nuclide identification but require special consideration in the calculation of activity levels.

The effect of source position in the well was investigated in order to determine how necessary it is to control the sample volume. A point source of mixed radionuclides was counted at the bottom of the well, at 1 cm from the bottom and at 2 cm from the bottom. The results of these measurements are listed in Table VIII. The data indicates that uncertainties in sample position of one cm from the bottom of the well can introduce about a 5% measurement error. However, the error becomes much greater as the source is moved to two cm. Sample volume control is required for accurate results.

TABLE VIII

RELATIVE ACTIVITIY OF A POINT SOURCE AS A FUNCTION OF POSITION IN THE WELL OF Ge(Li) DETECTOR

Energy (keV)	Relative Activiity		
	At Bottom	1 cm Up	2 cm UP
88.	1	.94	.58
122.	1	.94	.65
662.	1	.92	.59
898.	1	.96	.65
1173.	1	.97	.72
1332.	1	.87	.67

REFERENCES

1. J. Kemmer, "Ein Ge(Li) Gamma Spectrometer Für Geringe Gamma-Aktivitäten," Nucl. Instr. and Meth. 64, 268, 1968.

2. H. Büker, "Ein Hochauflosendes Gammaspektometer Mit Einem Bohrloch-Germanium-Detektor," Nucl. Instr. and Meth. 69, 293, 1969.

3. S. Santhanam, P. P. Webb and S. Monaro, "A Well-Type Ge(Li) Detector," IEEE Trans. Nucl. Sci., NS-16 p. 75 (1969).

4. S. Santhanam, and S. Monaro, "A Well-Type Ge(Li) Detector for Sum-Coincidence Studies," Nucl. Instr. and Meth., Vol. 76, p. 322, 1969.

5. P. Glasow, "Ge(Li)-Bohrloch-detektoren Zur Messung Geringer γ-Aktivitäten," Nucl. Instr. and Meth. 80, 141, 1970.

6. I. L. Fowler and R. J. Toone, "Large Hollow-Core Coaxial Ge(Li) Detectors," Nucl. Instr. and Meth, Vol. 98, p. 193, 1972.

7. J. C. Verplanke, "Full-Energy Peak Efficiency Calibration of a Well-Type Ge(Li) Gamma-Ray Detector," Nucl. Instr. and Meth., Vol. 96, p. 557, 1971.

8. B. M. Coursey, "Use of NBS Mixed-Radionuclide Gamma-Ray Standards for Calibration of Ge(Li) Detectors Used in Assay of Environmental Radioactivity," Proc. of a Symposium on Measurement for the Safe Use of Radiation, Gaithersburg, Maryland, 1976.

9. A. T. Hirshfeld, D. D. Hoppes and F. J. Schima, "Germanium Detector Efficiency Calibration with NBS Standard," Proc. of ERDA Symposium on X-and Gamma-Ray Sources and Applications, Ann Arbor, Michigan, p. 90, 1976.

10. G. V. Walford, J. A. Cooper and R. M. Keyser, "Evaluation of Standardized Ge(Li) Gamma-Ray Detectors for Low-Level Environmental Measurement," IEEE Trans on Nucl. Sci., Vol. NS-23, p. 734, 1976.

11. F. P. Brauer and J. H. Kaye, "Detection Systems for the Low-Level Radiochemical Analysis of Iodine-131, Iodine-129 and Natural Iodine in Environmental Samples," IEEE Trans on Nucl. Sci., Vol. NS-21, p. 496, 1974.

12. F. P. Brauer, J. M. Kelley, R. W. Goles and J. E. Fager, "Measurement of Environmental ^{241}Am and the Pu/^{241}Am Ratio by Photon Spectrometry," IEEE Trans. on Nucl. Sci., Vol. NS-24, p. 591, 1977.

13. F. P. Brauer, J. E. Fager and W. A. Mitzlaff, "Evaluation of a Large Ge(Li) Detector for Low-Level Radionuclide Analysis," IEEE Trans. on Nucl. Sci., Vol. NS-20, p. 57, 1973.