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GAMMA-RAY ASSAY OF PLUTONIUM-238 IN WASTE CANS

PART I: SINGLE-CHANNEL ASSAY
PART II: MULTICHANNEL ASSAY

W. W. Strohm, A. B. Combs and J. Y. Jarvis

AEC Research and Development REPORT

MONSANTO RESEARCH CORPORATION

A SUBSIDIARY OF MONSANTO COMPANY

MOUND LABORATORY

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

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SUMMARY

Two techniques are described for assaying the amount of plutonium-238 in cans of contaminated waste by measuring the intensity of the 765-keV gamma-ray from the decay of plutonium-238 with a NaI(Tl) detector. In both techniques, the transmission T of the 765-keV gamma-ray through the waste material in the can is determined from the weight w of the can. The determination of $T(w) = 1.04e^{-0.000212w}$ and its very small dependence on the material in the can is described. The first technique is a single-channel analysis of the 765-keV photoelectric peak developed for an in-line assay. For these measurements, one standard deviation is +59% and -14%. The second technique utilizes a multichannel analyzer, and contributions of higher-energy gamma-rays to the 765-keV photoelectric peak are subtracted. The accuracy of this technique is ±15% at the 95% confidence level when the can contains 0.100 g or more of plutonium-238.

INTRODUCTION

In part of the plutonium-238 recovery process at Mound Laboratory, waste contaminated with plutonium-238 is processed in the plutonium-238 recovery line and packaged in 0.5-gal (13.8 cm in diameter and 14.9 cm high) cans which are removed from the recovery line and then sealed in #12 cans (15.7 cm in diameter and 22.2 cm high). The #12 cans are then assayed by the 99.8-keV gamma scan facility.[1] Those cans of non-burnable waste containing less than 0.336 g of plutonium-238 are marked for disposal. However, a significant number of the cans contain more than 0.336 g and these must be sent back through the recovery line for further processing of the waste. In addition, the #12 cans containing this waste are cut up, processed and packaged, thus generating more cans of waste.

It was desired to significantly reduce the number of cans of non-burnable waste sent back through the recovery line by performing an in-line assay of the 0.5-gal cans before they are sealed in the #12 cans. Those cans containing more than 0.336 g of plutonium-238 can be reprocessed immediately without opening and cutting up #12 cans for processing and packaging.

Part I of the report describes a technique for this in-line assay consisting of a single-channel analysis of the intensity of the 765-keV gamma-ray from the decay of plutonium-238 in the can, detected by a NaI(Tl) detector. A correlation between the weight (w) of the can of waste and the transmission (T) of the 765-keV gamma-ray in the can was established. It is shown that T(w) has a very small dependence on the contents of the can, assuming the can is uniformly packed with waste. This technique is based in part on the assay technique for drums developed at Mound Laboratory[2] and later applied to cans.[3]

Part II describes a technique for accurately assaying the plutonium-238 content of the cans of waste utilizing a multichannel analyzer to analyze the 765-keV photoelectric peak using the gamma-ray stripping procedure used in the drum assay technique[2,3] to subtract contributions of higher-energy gamma-rays. The transmission T(w) described in Part I was utilized for these measurements.

PART I: SINGLE-CHANNEL ASSAY OF PLUTONIUM-238 IN WASTE CANS

Apparatus

The apparatus for these measurements was set up in a "cold" laboratory and is shown in Figure 1. The detector is a 3-in. x 3-in. (7.6-cm x 7.6-cm) NaI(Tl) Harshaw Integral Line assembly with a resolution of 7.6% for the 661-keV peak of cesium-137. The detector is shielded by a 3-in. (7.6-cm) thick, cylindrical lead shield with an inside diameter of 6 in. (15.2 cm). A 1/8-in. (0.32-cm) lead plate covers the front of the shield to reduce the intensity, at the detector, of the very intense 99.8-keV gamma-radiation from the plutonium-238 in the can.

The turntable, on which the 0.5-gal can of waste is placed, is positioned so the center of the can is 25.0 cm from the face of the detector and the axis of the detector passes through the center of the can. The turntable has a variable speed motor and is operated between 10 and 15 rpm. An aluminum pedestal was designed to hold a standard calorimetry can containing approximately 5 g of plutonium-238 for use in the transmission measurements discussed later. The pedestal was positioned so the axis of the detector passed through the center of the calorimetry can. For the in-line assay, the turntable sits inside a glove box and the detector and shield are outside the glove box. A 1/8-in. (0.32-cm) stainless steel plate was inserted between the turntable and the detector to simulate the glove box wall.

Figure 2 is a close-up view of the turntable and the detector in the shield with the lead and the stainless steel plates removed to show more clearly the counting geometry used.

The signal from a Hamner Model 356 preamplifier was analyzed by a Hamner Model 302 amplifier and single-channel analyzer. High voltage to the photomultiplier tube was supplied by a Hamner Model N401 power supply.

The readout from the single-channel analyzer was recorded by a Hamner Model N240 decade scaler controlled by a Hamner Model N850R mechanical timer.

The cans were weighed on the triple beam balance seen in Figure 1.

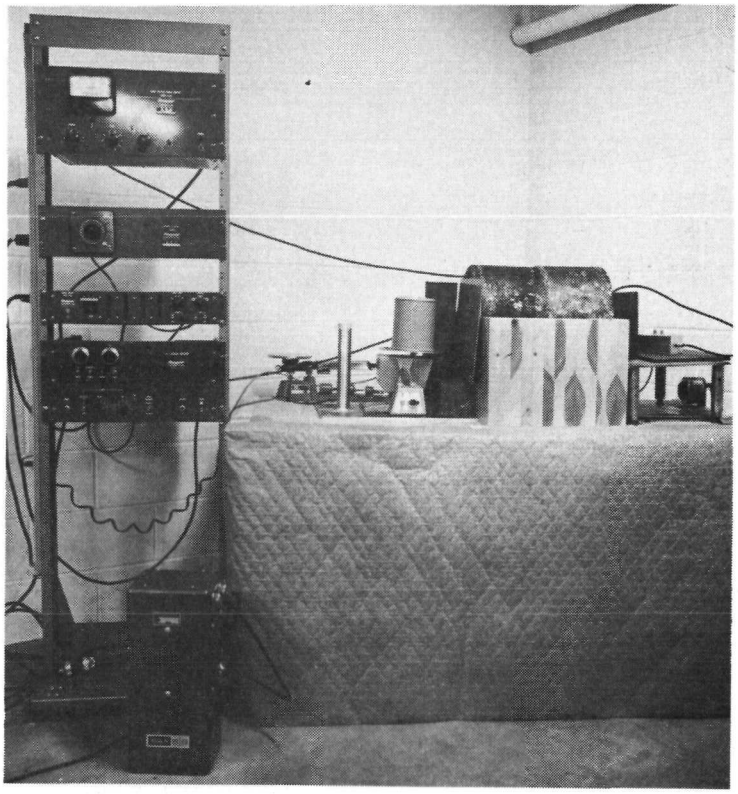

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FIGURE 1 - Apparatus for the single-channel assay measurements.

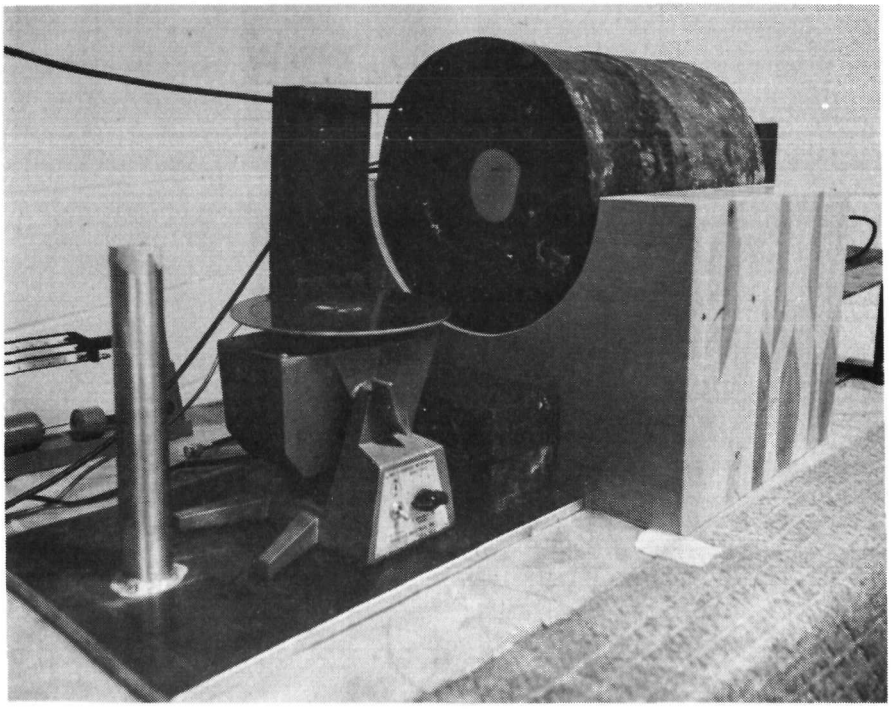

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FIGURE 2 - Close-up view showing counting geometry used in the single-channel measurements.

Transmission T(w)

In a quantitative measurement of the amount of an isotope in a thick source such as a can of waste by measurement of the intensity of a gamma-ray from the decay of the isotope, correction must be made for the attenuation of the gamma-ray intensity by the material in the thick source. Table 1[4] shows the mass attenuation coefficients, μ/ρ, (in cm^2/g) of several common materials. These coefficients are from the equation:

$$T = \frac{I}{I_o} = e^{-(\mu/\rho)m} \qquad (1)$$

where μ is the linear gamma-ray attenuation coefficient, ρ is the density of the absorber, and $\underline{m}$ is the thickness (mass per unit area) of the absorber.[5] It is seen that the coefficients for a 0.8-MeV photon (the nearest value given to 765 keV) can be written 0.0752 ± 0.0084 (or ±11%) cm^2/g.

Table 1

MASS ATTENUATION COEFFICIENTS[a]

Photon Energy (MeV)	Water	Aluminum	Iron	Lead
0.1	0.167	0.160	0.342	5.29
0.15	0.149	0.133	0.182	1.84
0.2	0.136	0.120	0.138	0.895
0.3	0.118	0.103	0.106	0.335
0.4	0.106	0.0922	0.0918	0.208
0.5	0.0967	0.0840	0.0828	0.145
0.6	0.0894	0.0777	0.0761	0.114
0.8	0.0786	0.0682	0.0668	0.0837
1.0	0.0706	0.0614	0.0595	0.0683
1.5	0.0576	0.0500	0.0484	0.0514
2.0	0.0493	0.0431	0.0422	0.0451
3.0	0.0396	0.0353	0.0359	0.0410
4.0	0.0339	0.0310	0.0330	0.0416
5.0	0.0302	0.0284	0.0314	0.0430
6.0	0.0277	0.0266	0.0305	0.0455
8.0	0.0242	0.0243	0.0298	0.0471
10.0	0.0221	0.0232	0.0300	0.0503

[a] μ/ρ, in cm^2/g

The fact that these coefficients are similar and near their minimum value for 765-keV gamma-rays is the principal reason for choosing the low-intensity 765-keV gamma-ray instead of the higher-intensity 99.8 and 153-keV gamma-rays for quantitative analysis of plutonium-238 in waste containers.[2]

The economics of the recovery operation dictate that the 0.5-gal cans can be packed uniformly full. This and the similarity of the mass absorption coefficients have allowed a correlation to be established between the weight of the can and the transmission of the 765-keV gamma-rays through the material in the can. This measurement, described below, consisted of measuring the transmission of the 765-keV gamma-rays from an external plutonium-238 source through several cans fully packed with various uncontaminated materials. The square root of these values is taken to be the transmission T of the 765-keV gamma-rays from the plutonium-238 in similar cans of contaminated waste. This assumes the gamma-rays from the plutonium-238 in the contaminated cans encounter one-half the amount of absorber as the gamma-rays from an external source passing through the can. It is at this point that the significance of rotating the can can be seen, since the varying thicknesses of absorber are effectively averaged to an equivalent thickness of a uniform absorber, giving meaning to the mass per unit area $\underline{m}$.

For the transmission measurements, an external 5-g plutonium-238 source was placed on the pedestal shown in Figures 1 and 2. The window of the single-channel analyzer was centered on the 765-keV photoelectric peak. Eighteen uniformly packed 0.5-gal cans with a variety of materials and having a variety of weights up to 3,500 g were weighed on the triple beam balance and then placed on the turntable and rotated. A 1-min count, N, was taken. The can was removed and another 1-min count, N_1, was taken. The room background was subtracted from each count. The transmission T(w') was determined by

$$T(w') = \sqrt{\frac{N - BG}{N_1 - BG}} \qquad (2)$$

where w' is the weight of the particular can whose transmission is being measured. No gamma-ray stripping of the contribution of higher-energy gamma-rays was used since the same external source was used in all measurements. The contribution of higher-energy gamma-rays was small and the fractional change of this contribution due to different amounts of absorber was ignored.

Table 2 shows the results of these measurements. A least-squares fit of the function Ae^{-kw} to the data points yields

$$T(w) = 1.04e^{-0.000212w} \qquad (3)$$

Table 2

TRANSMISSION MEASUREMENTS

Mock Can No.	Can Contents	Can Weight (g)	Experimental Transmission (T)	Calculated[a] Transmission	Deviation from Least Squares Fit (%)
1	Can Minus Top & Bottom	124.0	0.996	1.013	-1.7
2	Empty Can	262.7	0.980	0.984	-0.4
3	Loose Steel Turnings	521.0	0.940	0.931	+1.0
4	Copper Turnings	580.4	0.935	0.919	+1.7
5	Aluminum Turnings	670.0	0.855	0.902	-6.2
6	Cloth	595.0	0.917	0.917	0
7	Al+Cu Turnings	744.4	0.895	0.888	+0.8
8	Tightly Packed Steel Turnings	826.5	0.890	0.873	+1.9
9	Non-Burnable Plastic	841.4	0.917	0.917	0
10	Lucite Blocks	1270.8	0.765	0.794	-3.7
11	Lead-Lined Rubber Gloves	1712.9	0.730	0.723	+1.0
12	Broken Glass	1957.3	0.687	0.687	0
13	Water	2058.3	0.660	0.672	-1.8
14	Scrap Metal	2208.1	0.690	0.651	+6
15	Scrap Metal	2603.1	0.590	0.599	-1.5
16	Al+Cu+Water	2933.6	0.530	0.558	-5.0
17	Scrap Metal	3362.6	0.500	0.510	-2.0
18	Glass & Water	3472.9	0.520	0.498	+4.1

[a]The values in this column were calculated from the formula $T(w) = 1.04e^{-0.000212w}$ which is a least square fit of the function Ae^{-kw} to the experimental values of T.

with a standard deviation of ±3.3%. This curve and the data points are shown in Figure 3. The curve was plotted to 5,000 g, but it is not expected that many cans will exceed 3,500 g.

It is seen that for w = 0, T = 1.04. This occurs because the heavy rims on the top and bottom of the 0.5-gal cans add to the weight but do not enter into the transmission measurements. The top and bottom together weigh approximately 130 g. From Figure 3, T = 1.01 for 130 g, which is very close to 1.00, as it should be.

Technique

Figure 4 shows a typical gamma-ray pulse height spectrum from a container of waste material with the room background subtracted. In the procedure developed for assaying the plutonium-238 content of waste drums,[2,3] an analysis is made of the nine-channel region centered on the photoelectric peak. The area (A+B) is the contribution to this region from the 765-keV gamma-ray from the decay of plutonium-238, and the area C is the contribution from higher-energy gamma-rays. The area (A+B) is measured, corrected for the transmission T of the 765-keV gamma-ray, and compared to a calibration curve of (A+B)/T as a function of weight of plutonium-238. This is written as

$$W = \frac{(A + B)}{c \cdot t \cdot T} \qquad (4)$$

where: W = weight of plutonium-238 in grams

c = 313 counts/min/g of plutonium-238 for a source-to-detector distance of 2.00 m and is the slope of the calibration curve used for the drum assay[2]

t = counting time in minutes

The procedure reported here for the in-line assay of plutonium-238 in waste cans is a simplification of the above procedure. Referring again to Figure 4, a single-channel analysis of the 765-keV photoelectric peak was made of the region bounded by the vertical lines E and E+ΔE. This was accomplished by adjusting the gain so the 765-keV peak fell at 765 on the E dial of the single-channel analyzer and 900 on the ΔE dial corresponded to 90 keV. The 90-keV window defined by E and E+ΔE was centered on the 765-keV photoelectric peak. Thus, the region of the 765-keV photoelectric peak analyzed by the single-channel analyzer was the same as the nine-channel region analyzed in the drum scan procedure. For this single-channel analysis:

$$W = \frac{N - BG}{D \cdot c \cdot t \cdot T(w) \cdot T_{Pb} \cdot T_{SS}} \qquad (5)$$

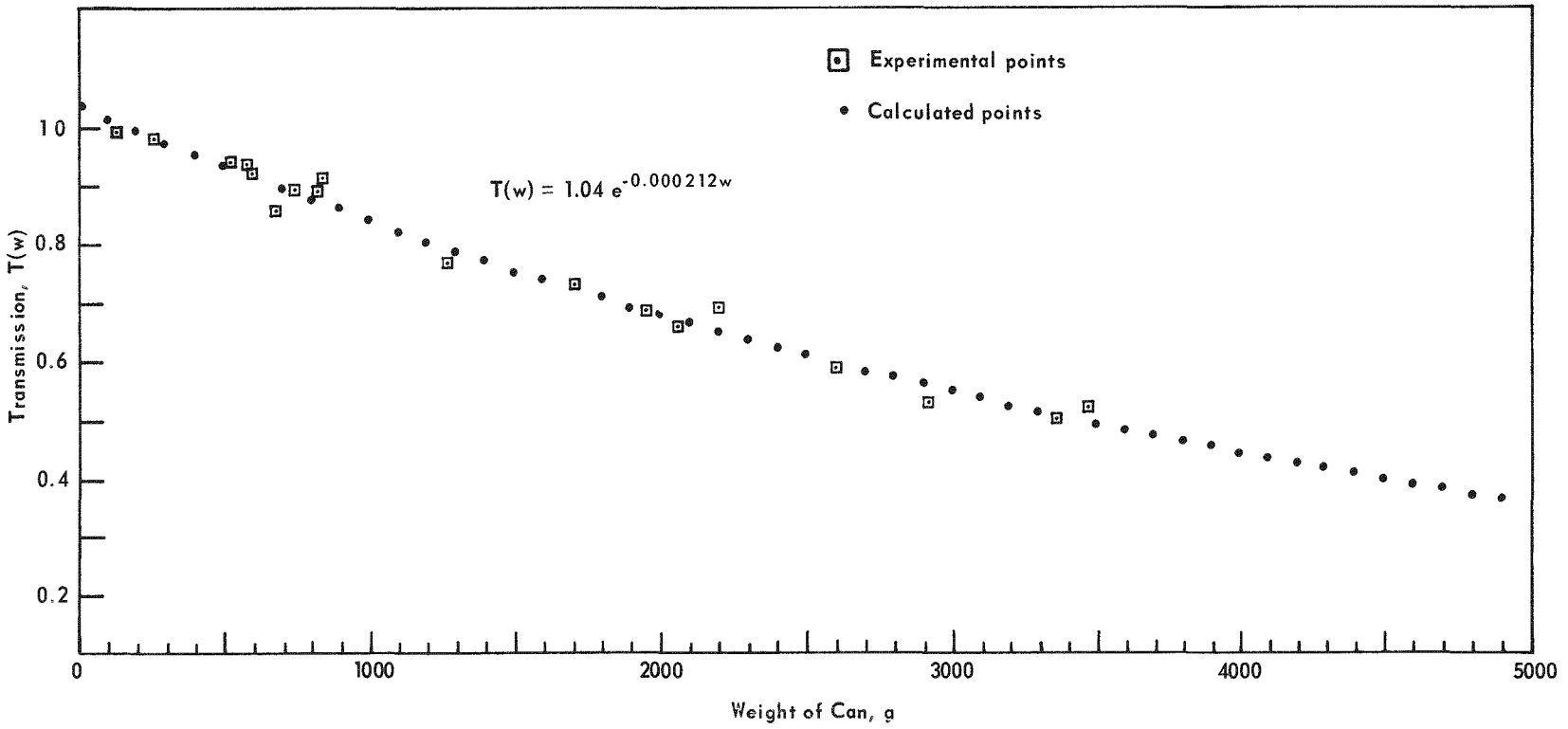

FIGURE 3 - Transmission as a function of the weight of the can plus contents (the curve is a least-squares fit of the function Ae^{-kw} to the experimental points).

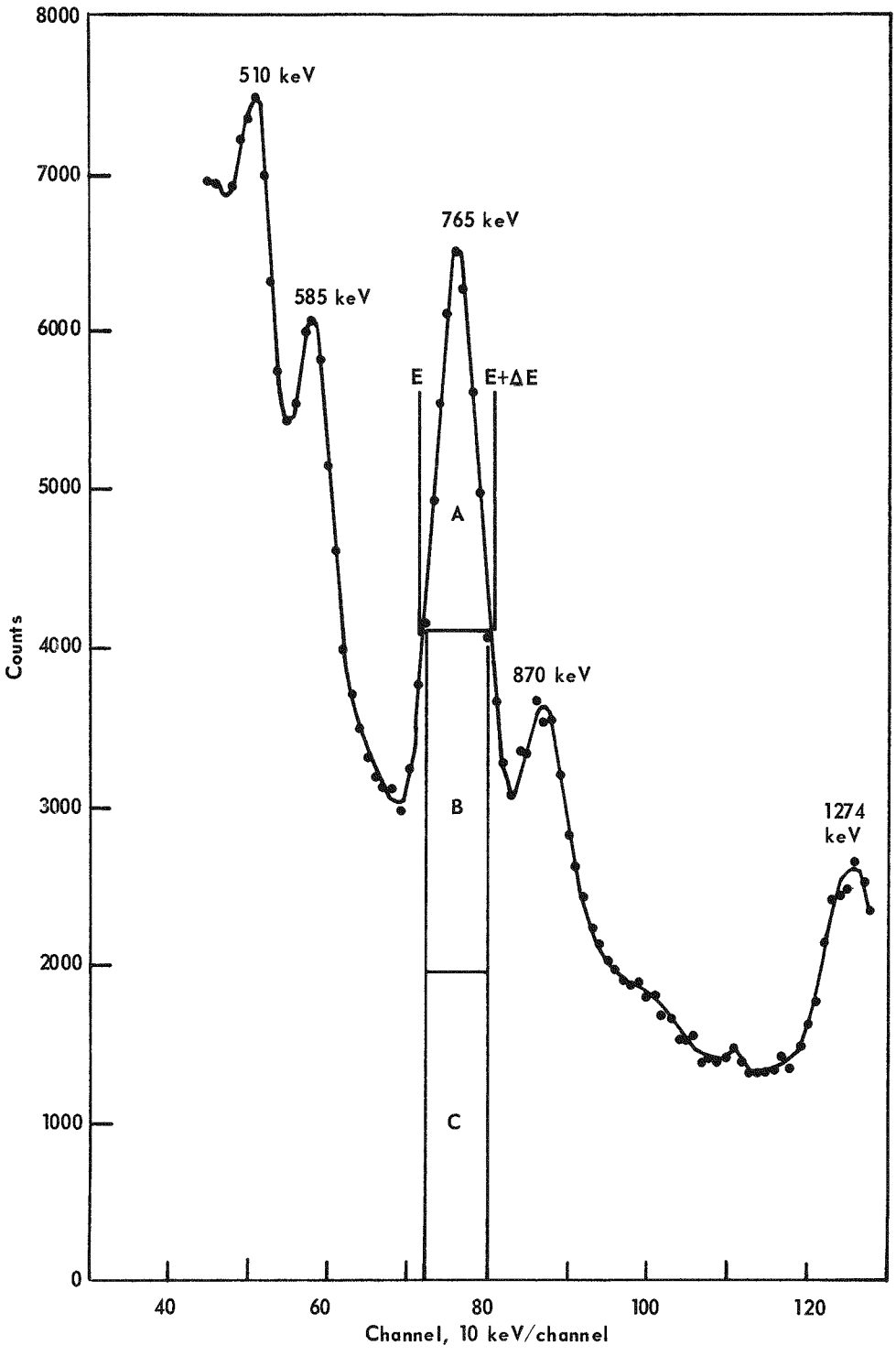

FIGURE 4 - Gamma-ray spectrum from a can containing waste contaminated with plutonium-238 showing a moderate contribution at 870 keV and at 1274 keV.

where: N = number of counts from the can

BG = background

D = 64 and is the inverse square law correction of the slope c from 2.00 m to 25.0 cm

t = the counting time used to obtain both N and BG

T(w) = transmission taken from Figure 3 after determining the can weight w

$T_{Pb} \cdot T_{SS}$ = 0.71 and is the transmission of the 765-keV gamma-ray through both the 1/8-in. (0.32-cm) lead plate and the 1/8-in. (0.32-cm) stainless steel plate

Related to Equation 4, this is:

$$W = \frac{A + B + C}{D \cdot c \cdot t \cdot T(w) \cdot T_{Pb} \cdot T_{SS}} \tag{6}$$

and it is seen the area C is not subtracted. This procedure tends to give a high value for the assay weight W, but this is favorable to the purpose of this in-line assay. An analysis of numerous gamma-ray spectra from containers of contaminated waste showed that few of the spectra had a contribution C less than 10% of the total area (A+B+C), so a factor of 1.10 was introduced into the denominator of Equation 5. Thus, Equation 5 becomes

$$W = \frac{N - BG}{D \cdot c \cdot t \cdot T(w) \cdot T_{Pb} \cdot T_{SS} \cdot 1.10} \tag{7}$$

or

$$W = \frac{N - BG}{15{,}645 \cdot t \cdot T(w)} \tag{8}$$

Measurements

Several 0.5-gal cans of contaminated waste were prepared with known amounts of plutonium-238. The material in the cans consisted of a variety of burnable and non-burnable waste. Each of these cans was then sealed in a #12 can. These cans were used for all measurements reported.

A counting time t = 1.0 min was used for these measurements. Thus Equation 8 becomes

$$W = \frac{N - BG}{15{,}645 \cdot T(w)} \tag{9}$$

It is seen that three measurements are required and may be made in any order. They are:

1) A 1.0-min count of the can is obtained.
2) A 1.0-min count of the background is obtained.
3) The can is weighed and the weight of the #12 can (362 g) is subtracted to determine the weight w. The transmission T(w) is determined from Figure 3.

Results and Error

Measurements were made on 17 standard cans in which a weighed amount of plutonium-238 was added or the calorimetry value was known. The results of these measurements are shown in Table 3. Two runs were made on separate days to demonstrate reproducibility.* Since these measurements tend to give a high value, the positive and negative errors are treated separately. From the results in Table 3, one standard deviation is +59% and -14%.

Table 3

SINGLE-CHANNEL ASSAY OF PLUTONIUM-238 IN CANS OF WASTE[a]

				Run #1		Run #2	
Can No.	Weight of can (g)	T(w) [Fraction]	Actual ^{238}Pu Content (g)	Measured ^{238}Pu Content (g)	Error (%)	Measured ^{238}Pu Content (g)	Error (%)
1	1083	0.826	0.1196	0.0993	-17	0.0970	-19
2	1076	0.829	0.2804	0.2470	-12	0.2433	-13
3	4104	0.437	0.1980	0.2839	+43	0.2910	+46
4	3487	0.498	0.3912	0.5013	+28	0.4872	+22
5	1445	0.767	0.5007	0.4581	-9	0.4355	-13
6	2207	0.652	0.5490	0.6252	+14	0.5863	+7
7	2167	0.657	0.5600	0.5060	-10	0.4878	-13
9	1194	0.806	0.4463	0.3788	-15	0.3573	-20
15	608	0.920	1.775	1.3799	-22	1.4279	-20
16	705	0.894	0.503	0.4688	-7	0.4803	-5
17	1050	0.833	1.800	---	---	4.032	+224
21	689	0.899	0.681	0.6060	-11	0.6493	-5
22	2350	0.631	0.422	0.5144	+17	0.5154	+17
23	1148	0.813	0.060	0.0775	+29	0.0800	+33
26	629	0.914	0.102	0.1078	+5	0.1041	+2
27	1580	0.743	0.201	0.2170	+5	0.2182	+9
31	1118	0.823	0.080	0.0914	+14	0.1012	+18

[a]Distance from can to detector was 25.0 cm and the counting time was 1.0 min.

*Can #17 was measured only once.

Pulse height spectra of all the standard cans are shown in Figures 5 through 21. It is instructive to note that the single-channel assay values of W run high for those spectra showing large contributions at 870 and 1274 keV.

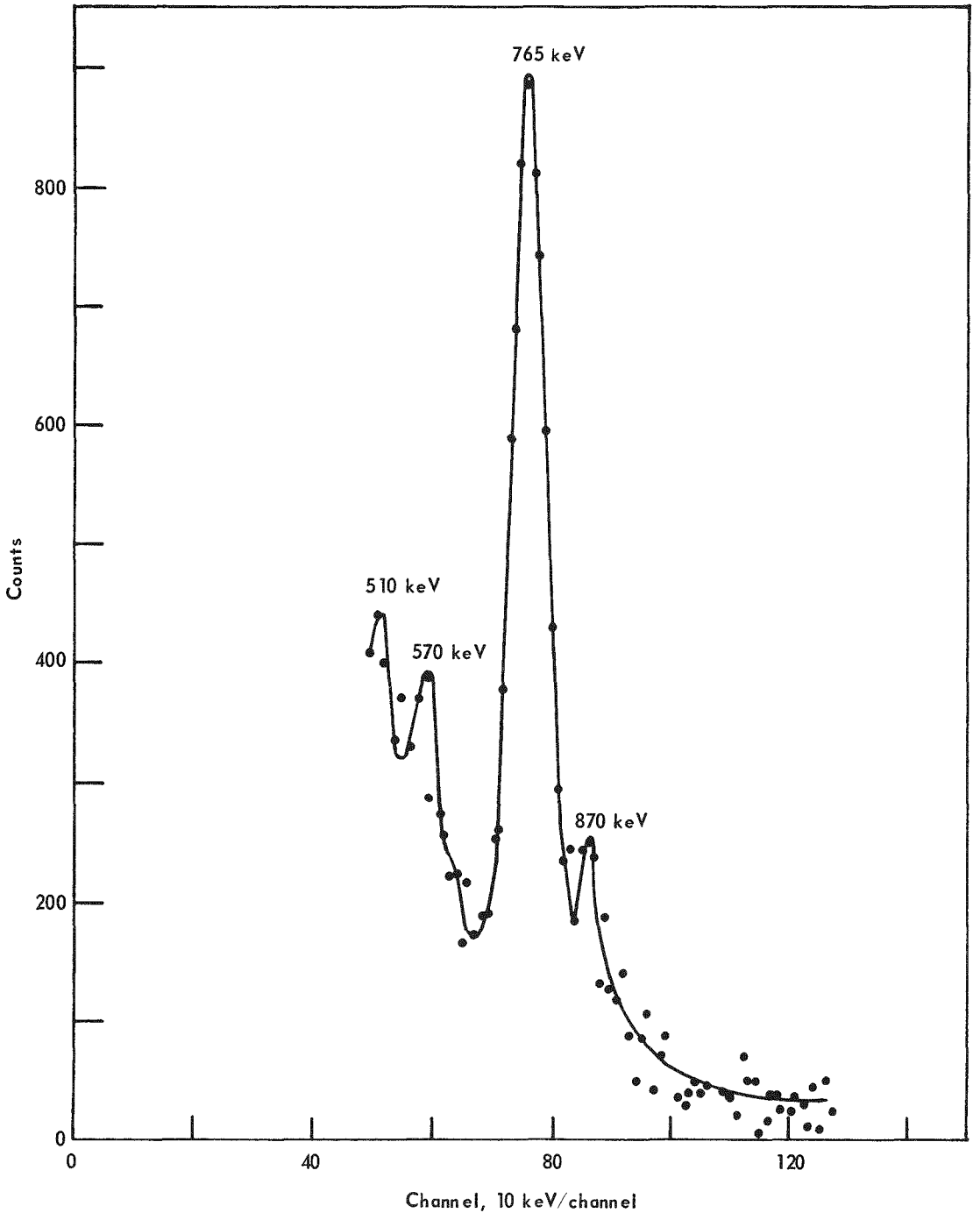

FIGURE 5 - Gamma-ray spectrum from can No. 1.

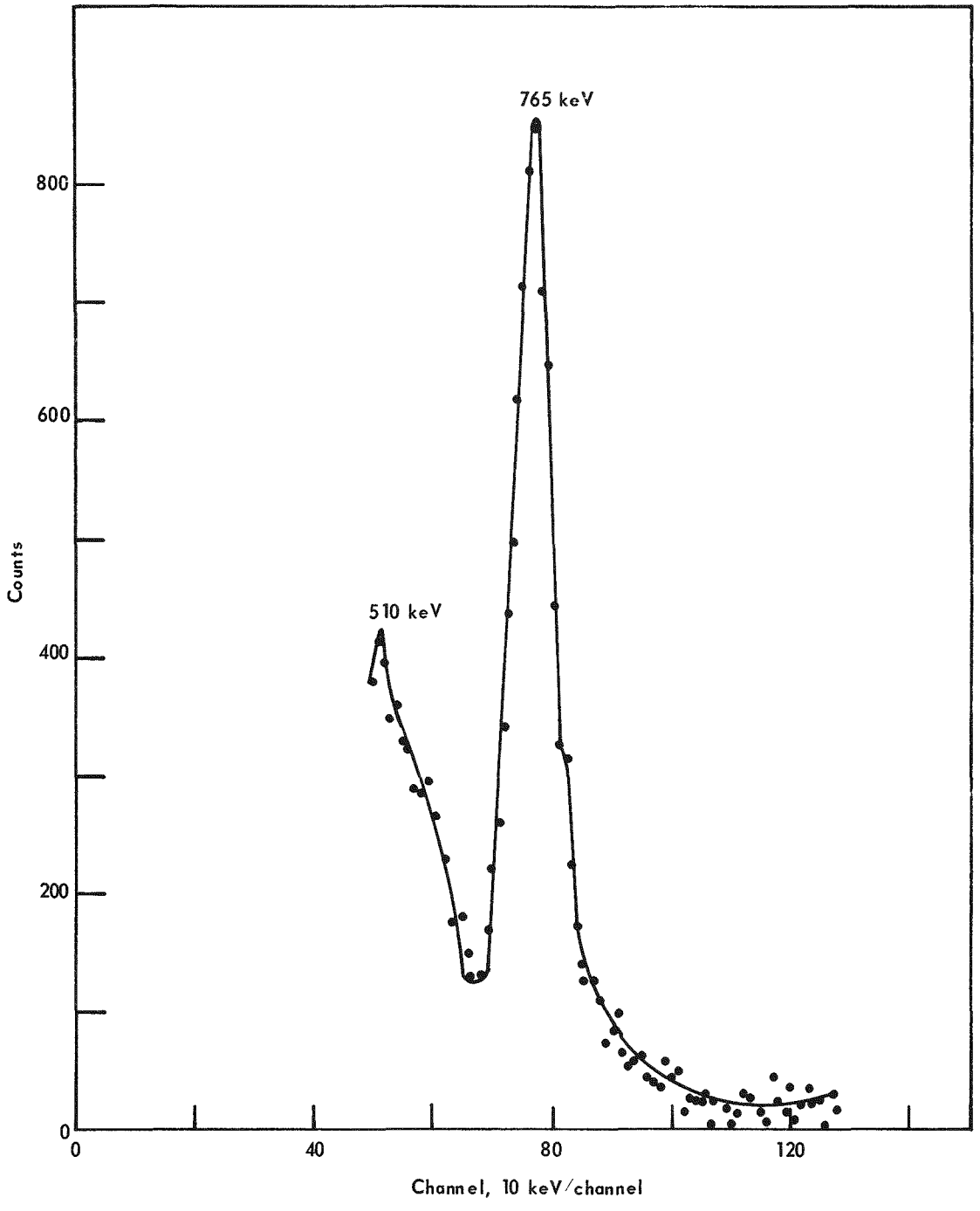

FIGURE 6 - Gamma-ray spectrum from can No. 2.

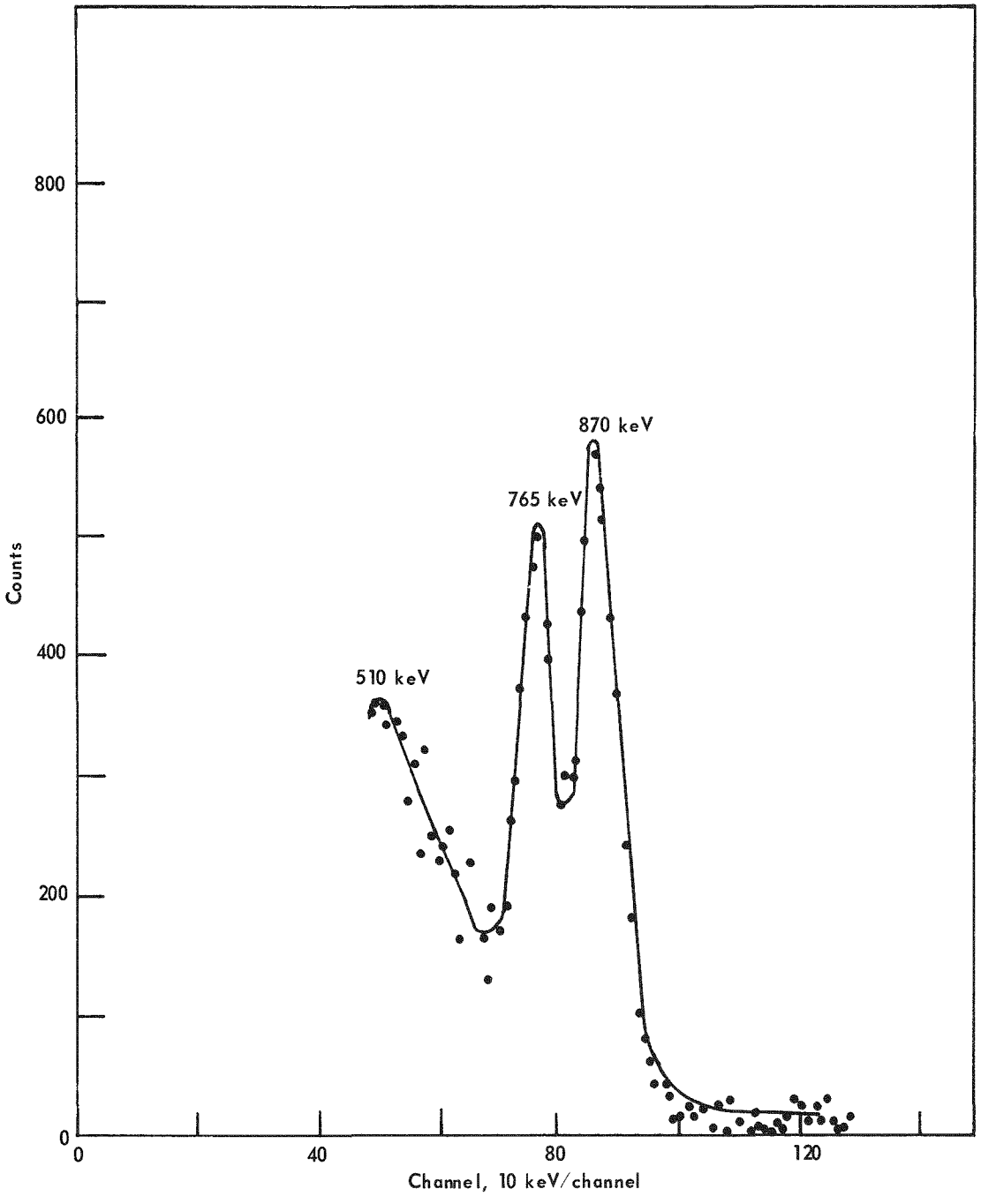

FIGURE 7 - Gamma-ray spectrum from can No. 3.

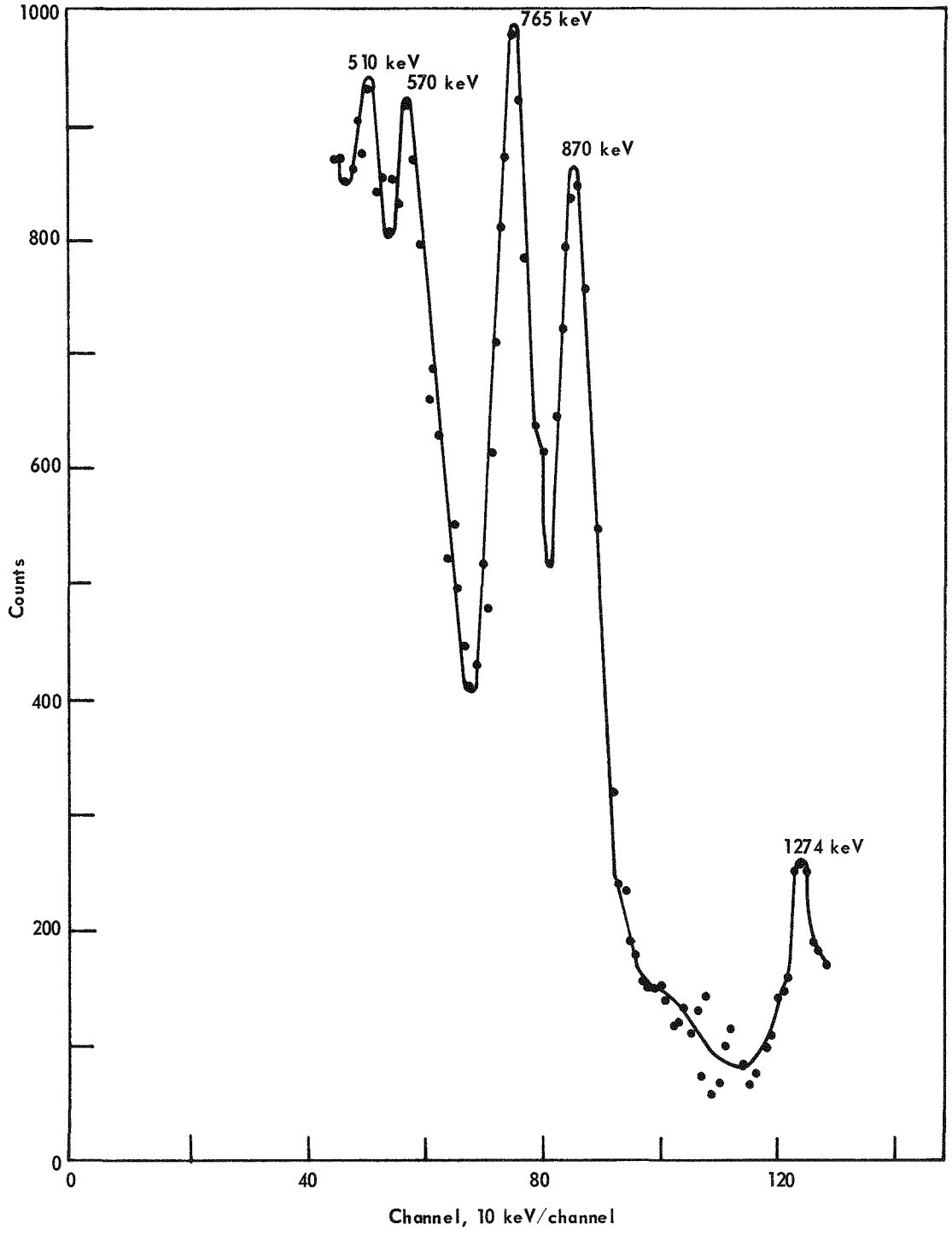

FIGURE 8 - Gamma-ray spectrum from can No. 4.

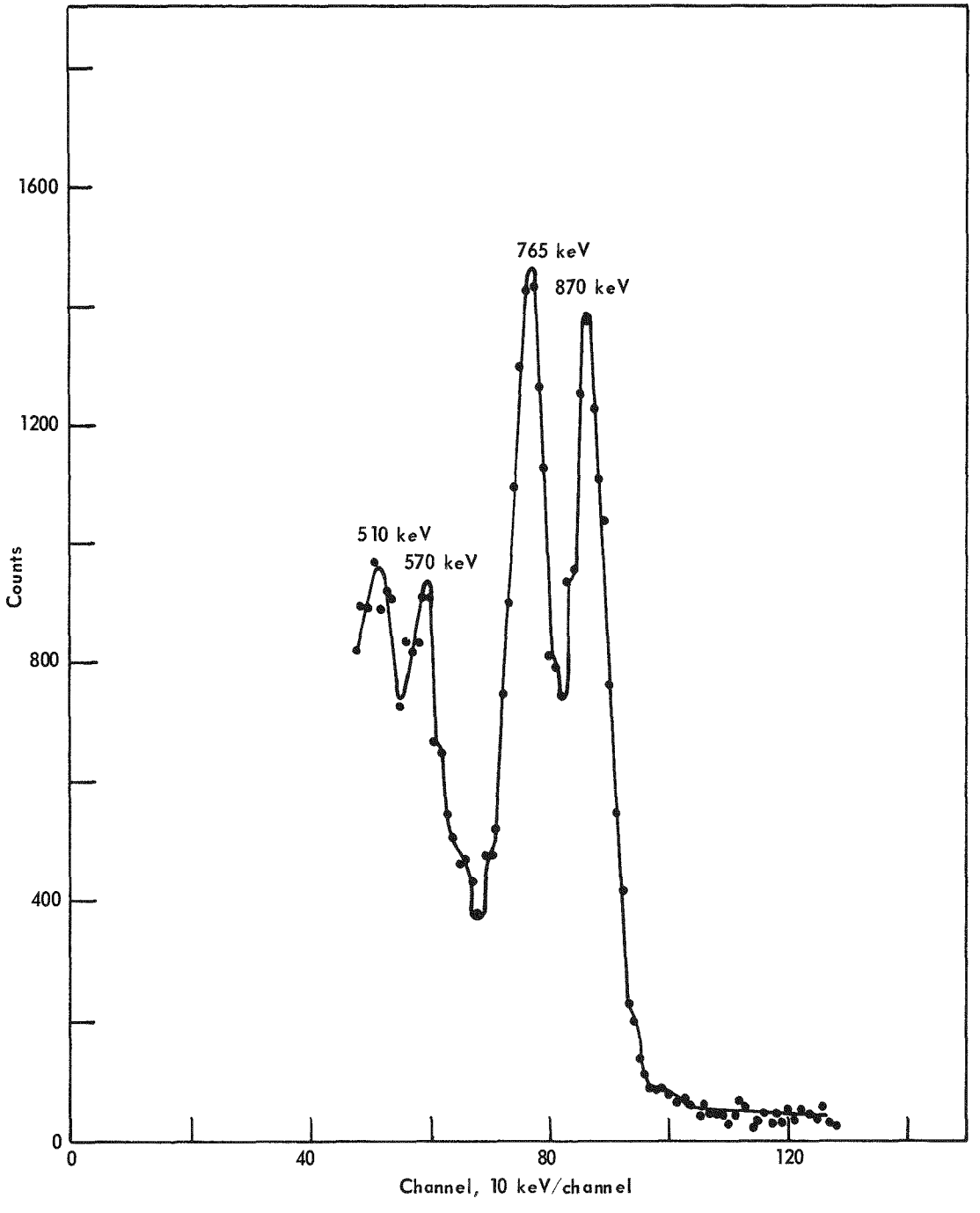

FIGURE 9 - Gamma-ray spectrum from can No. 5.

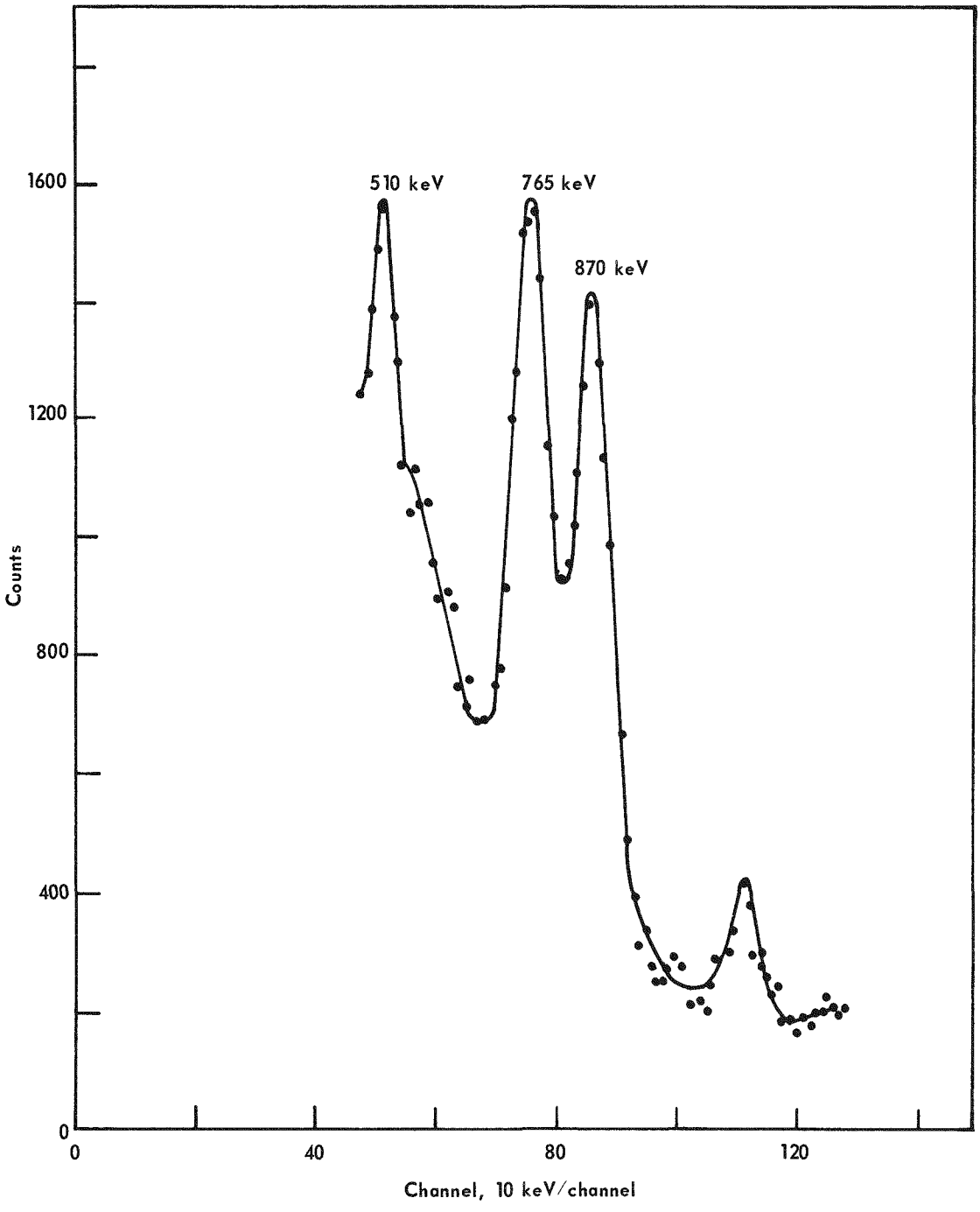

FIGURE 10 - Gamma-ray spectrum from can No. 6.

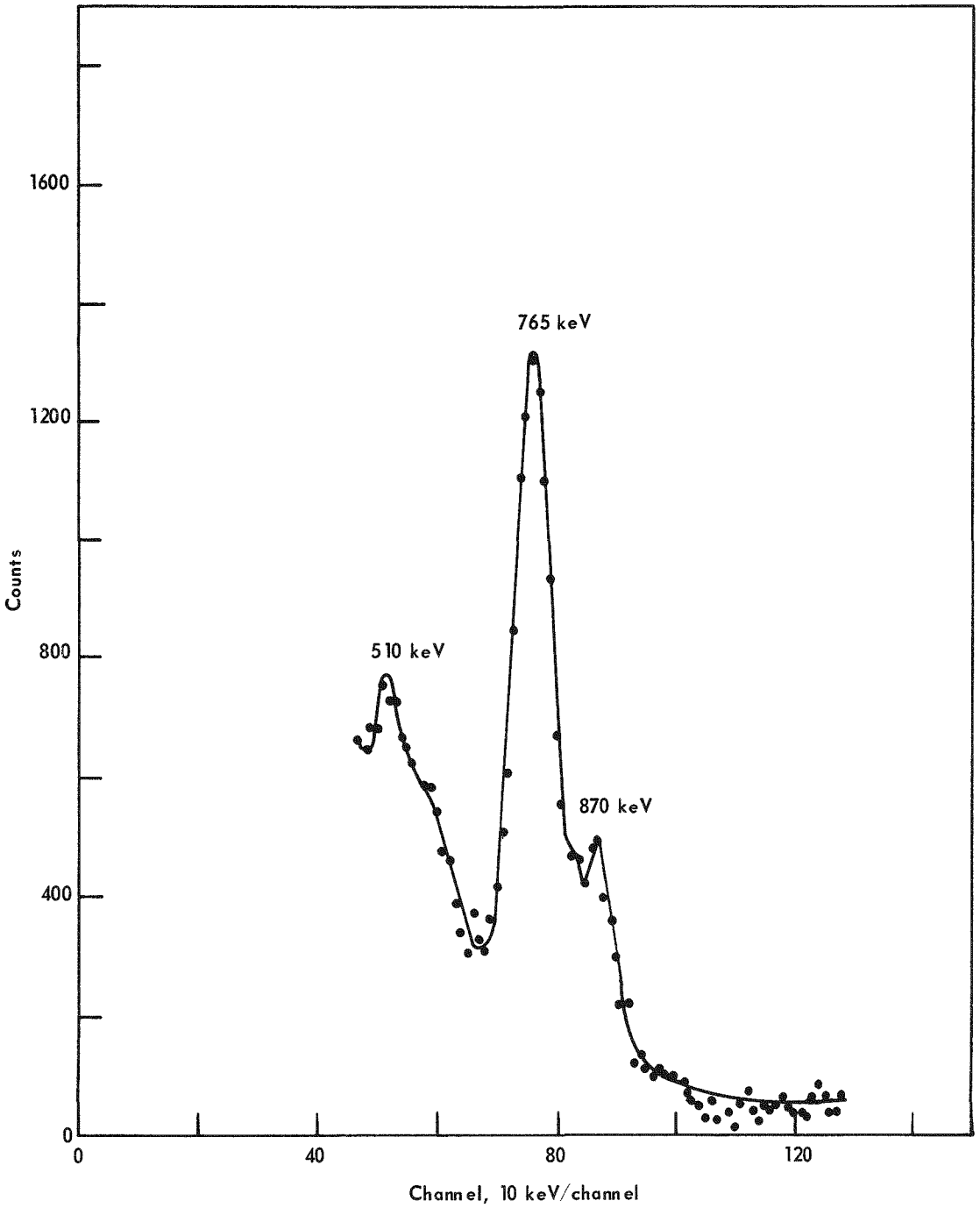

FIGURE 11 - Gamma-ray spectrum from can No. 7.

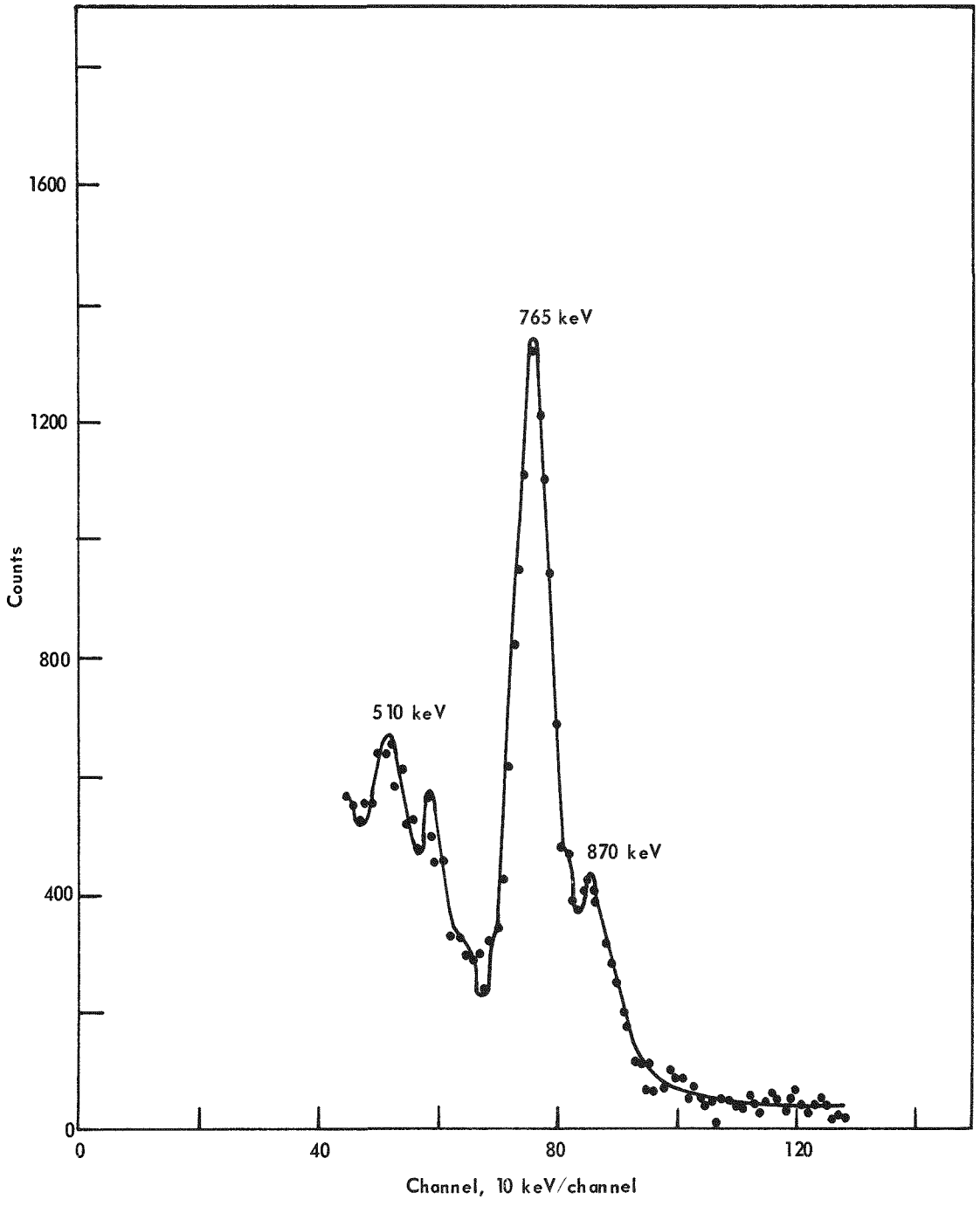

FIGURE 12 - Gamma-ray spectrum from can No. 9.

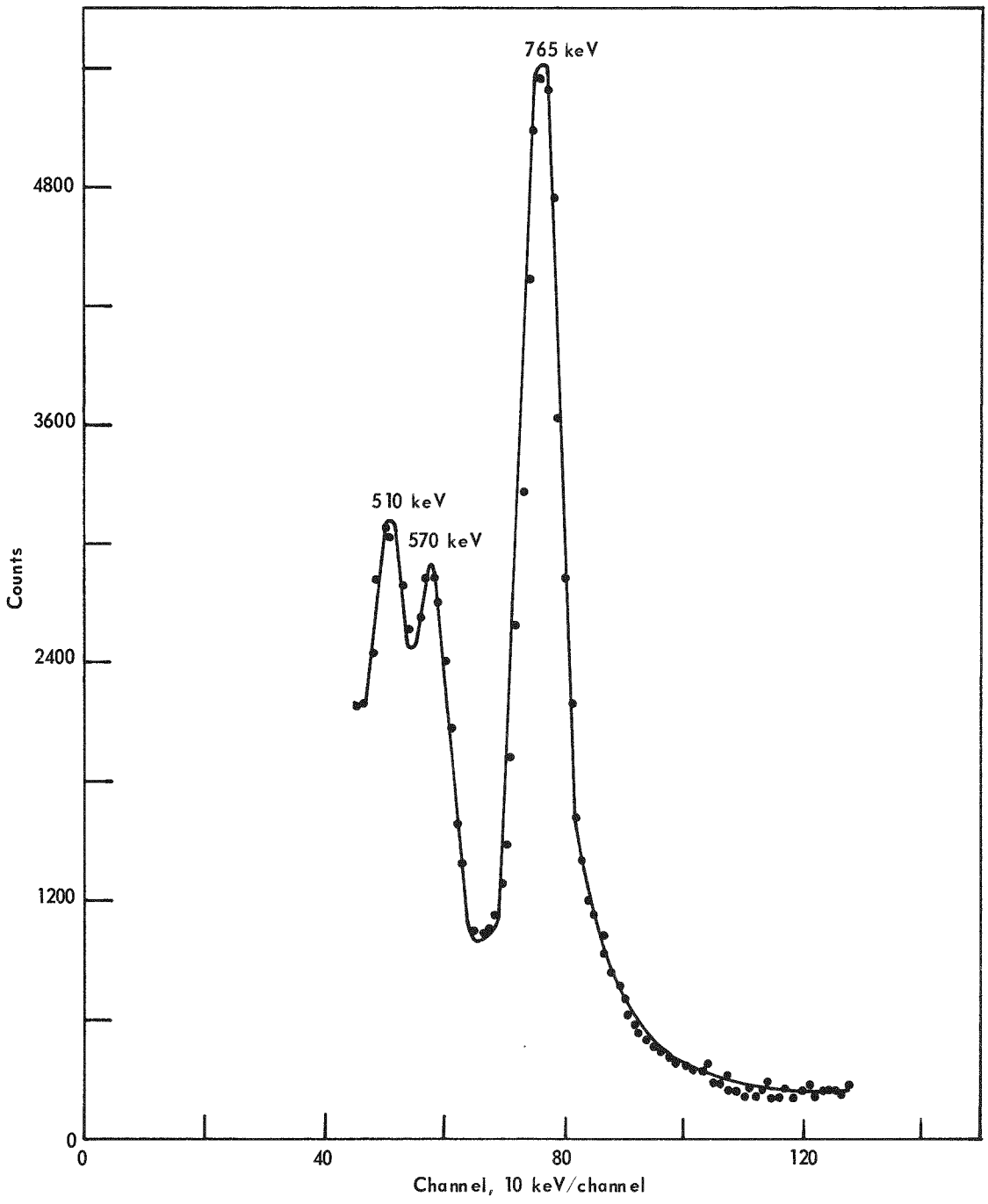

FIGURE 13 - Gamma-ray spectrum from can No. 15.

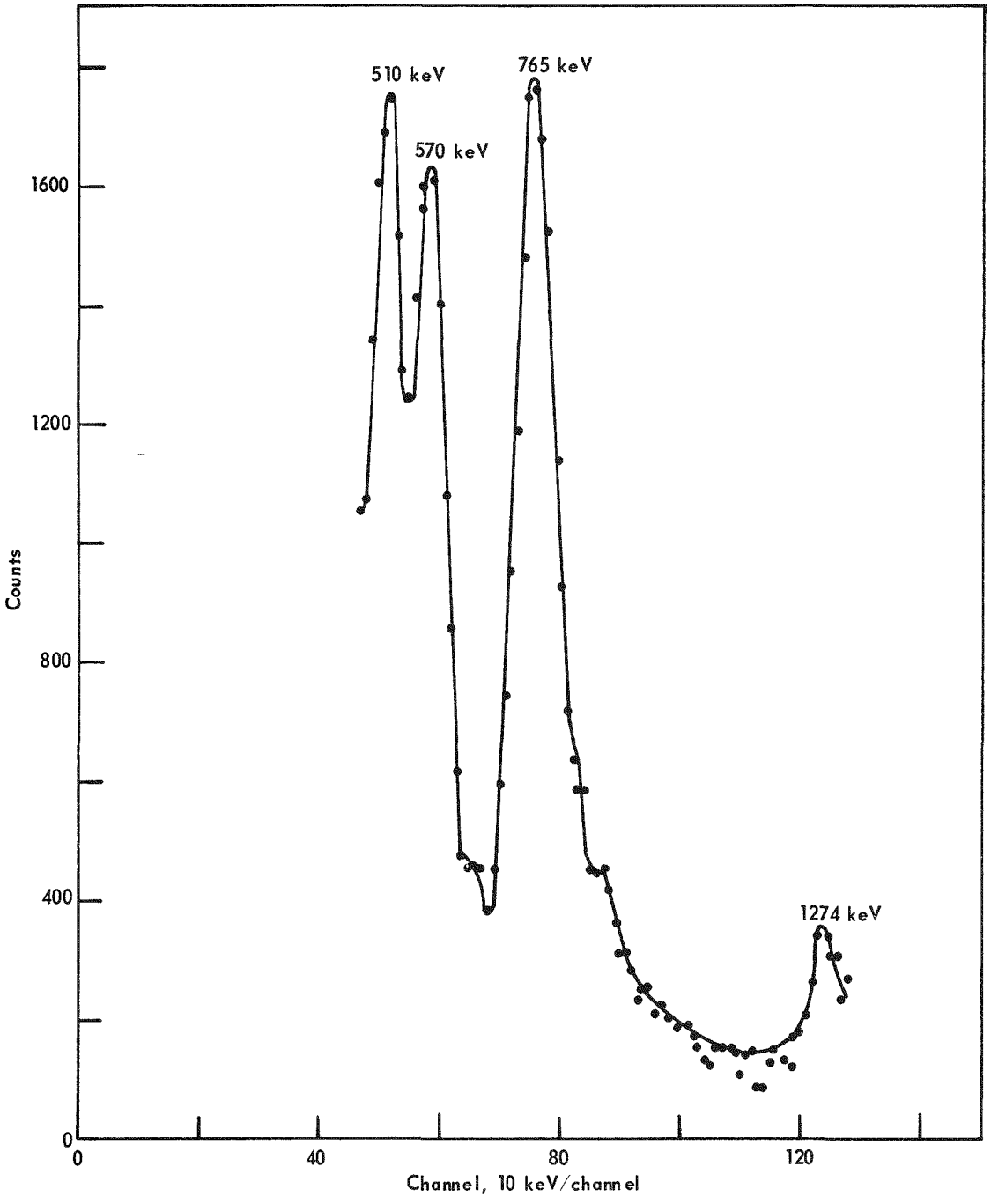

FIGURE 14 - Gamma-ray spectrum from can No. 16.

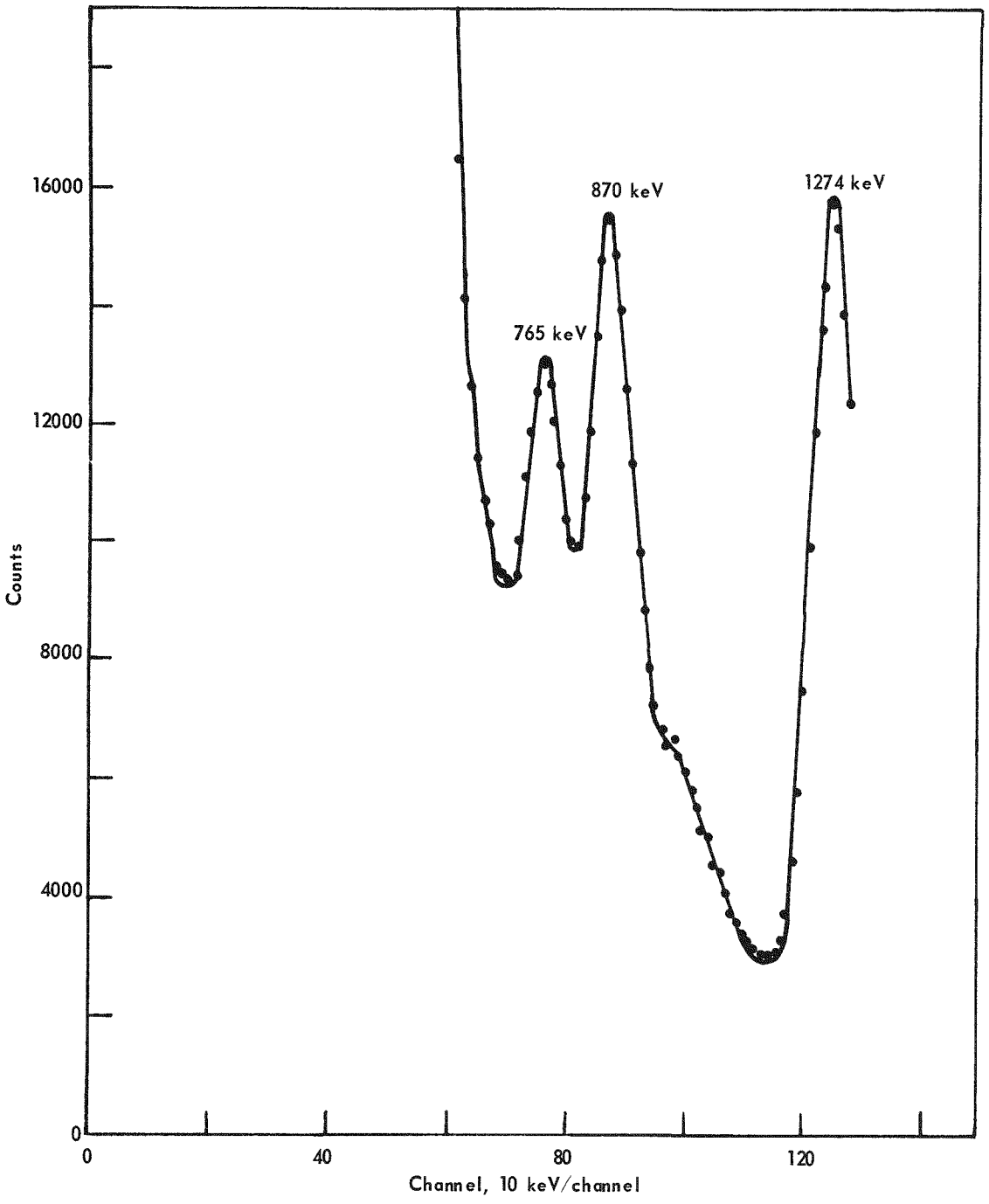

FIGURE 15 - Gamma-ray spectrum from can No. 17.

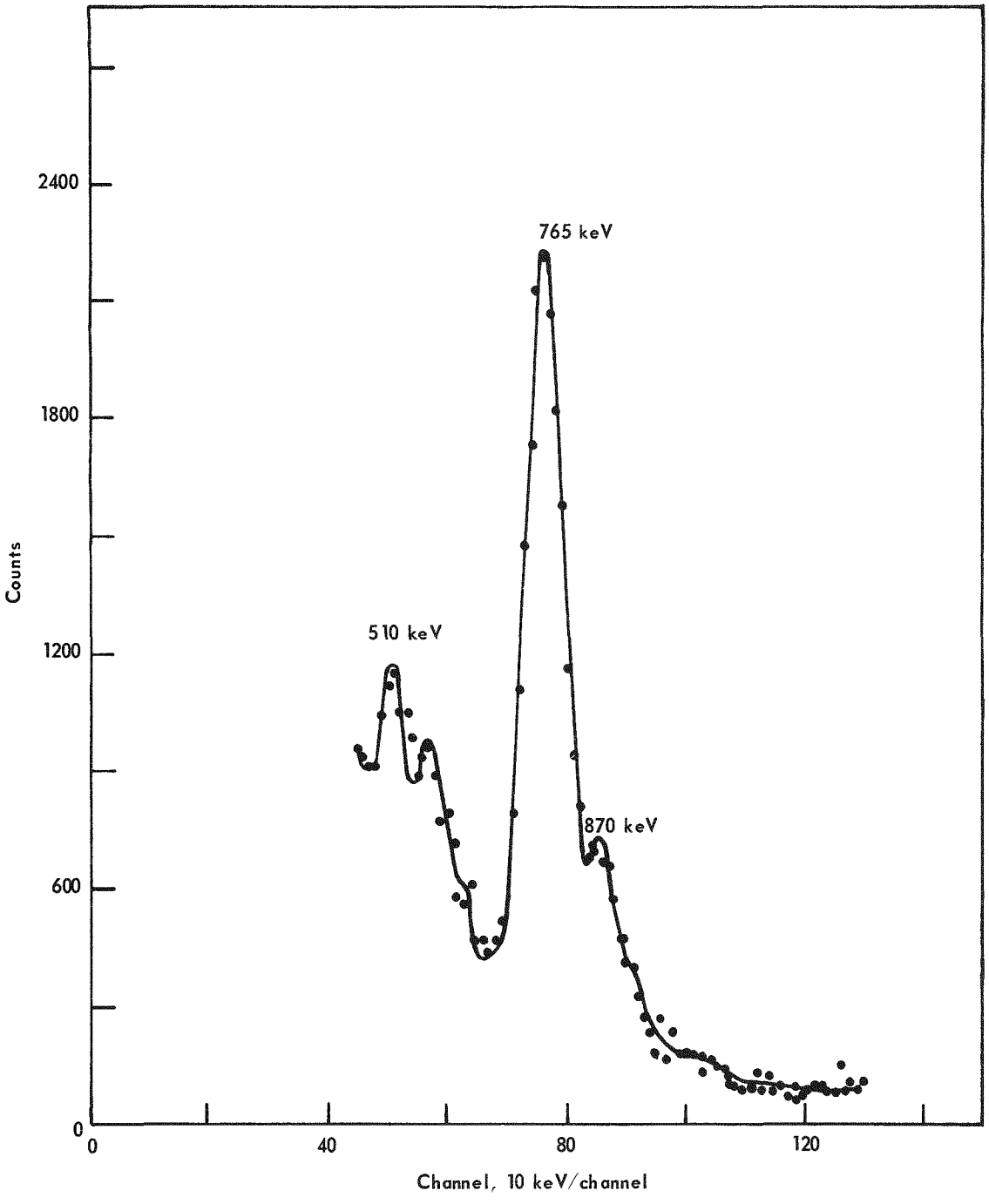

FIGURE 16 - Gamma-ray spectrum from can No. 21.

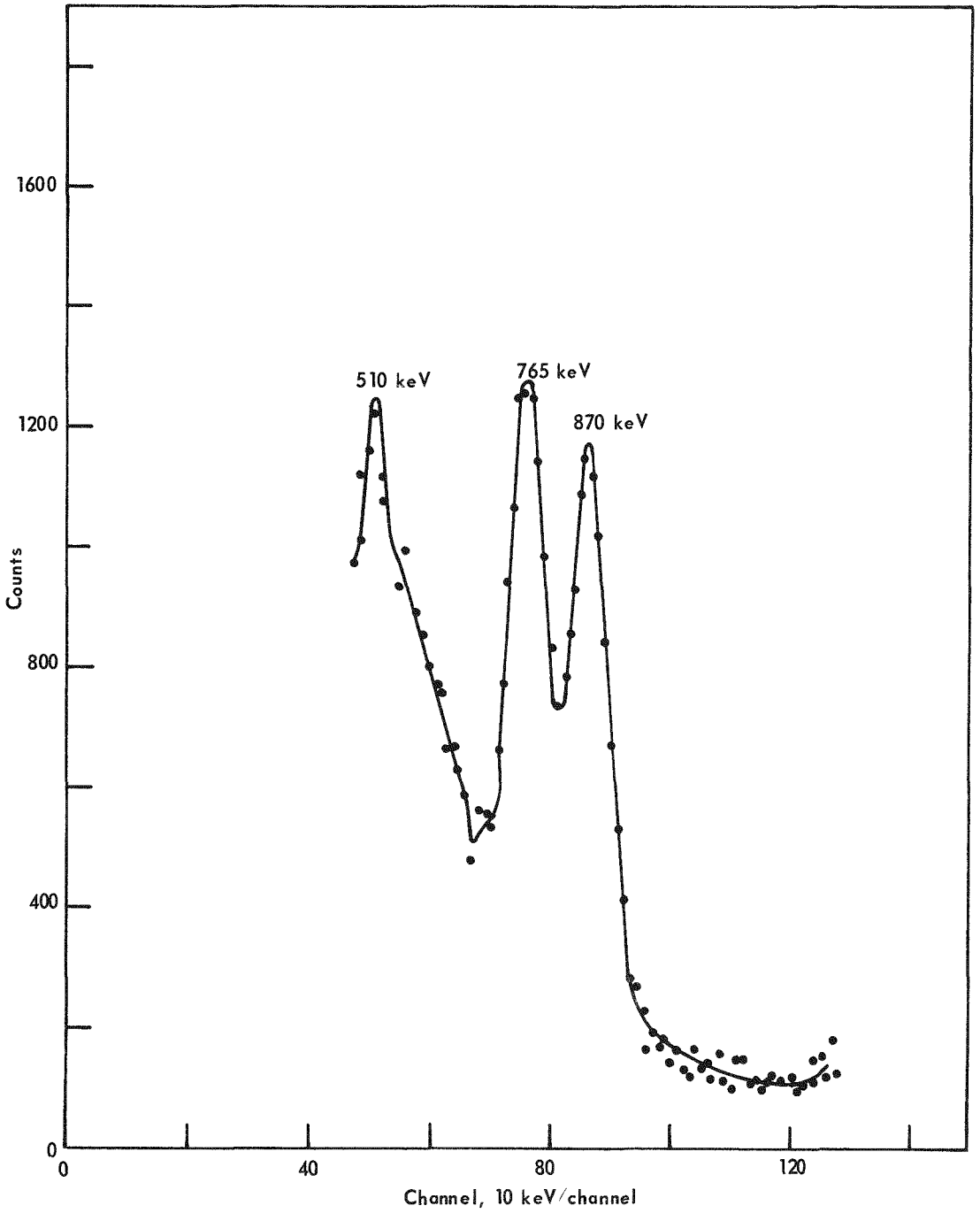

FIGURE 17 - Gamma-ray spectrum from can No. 22.

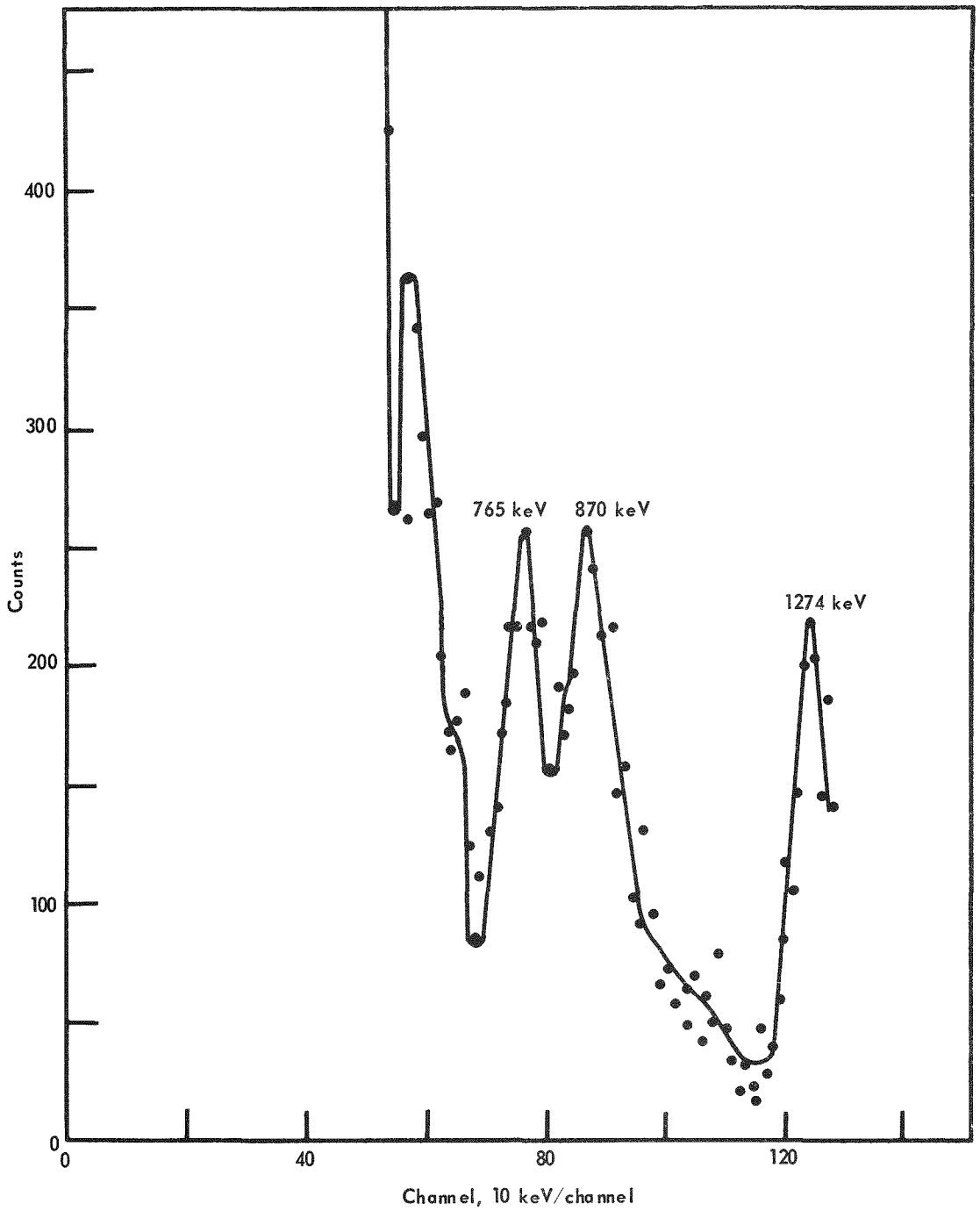

FIGURE 18 - Gamma-ray spectrum from can No. 23.

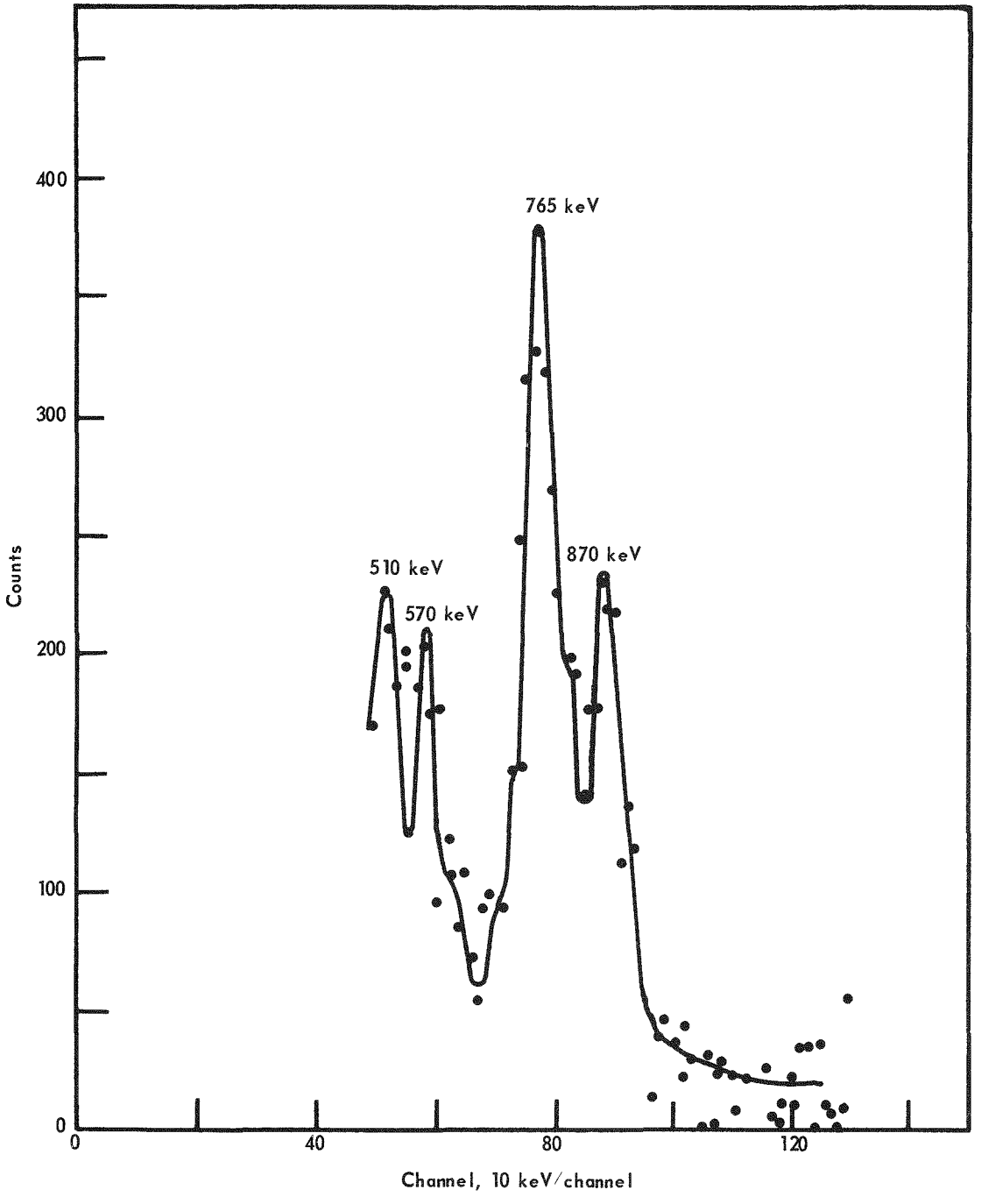

FIGURE 19 - Gamma-ray spectrum from can No. 26.

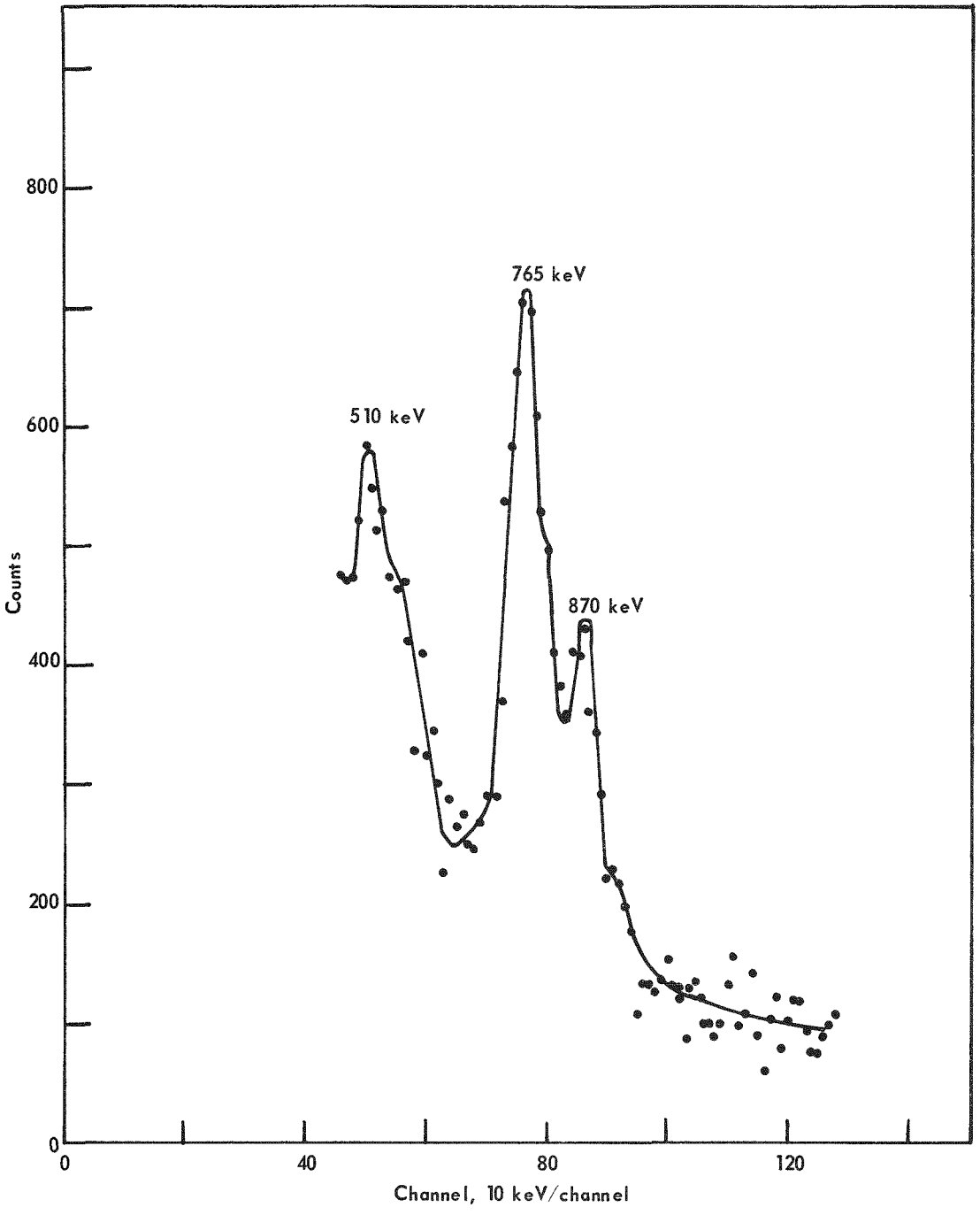

FIGURE 20 - Gamma-ray spectrum from can No. 27.

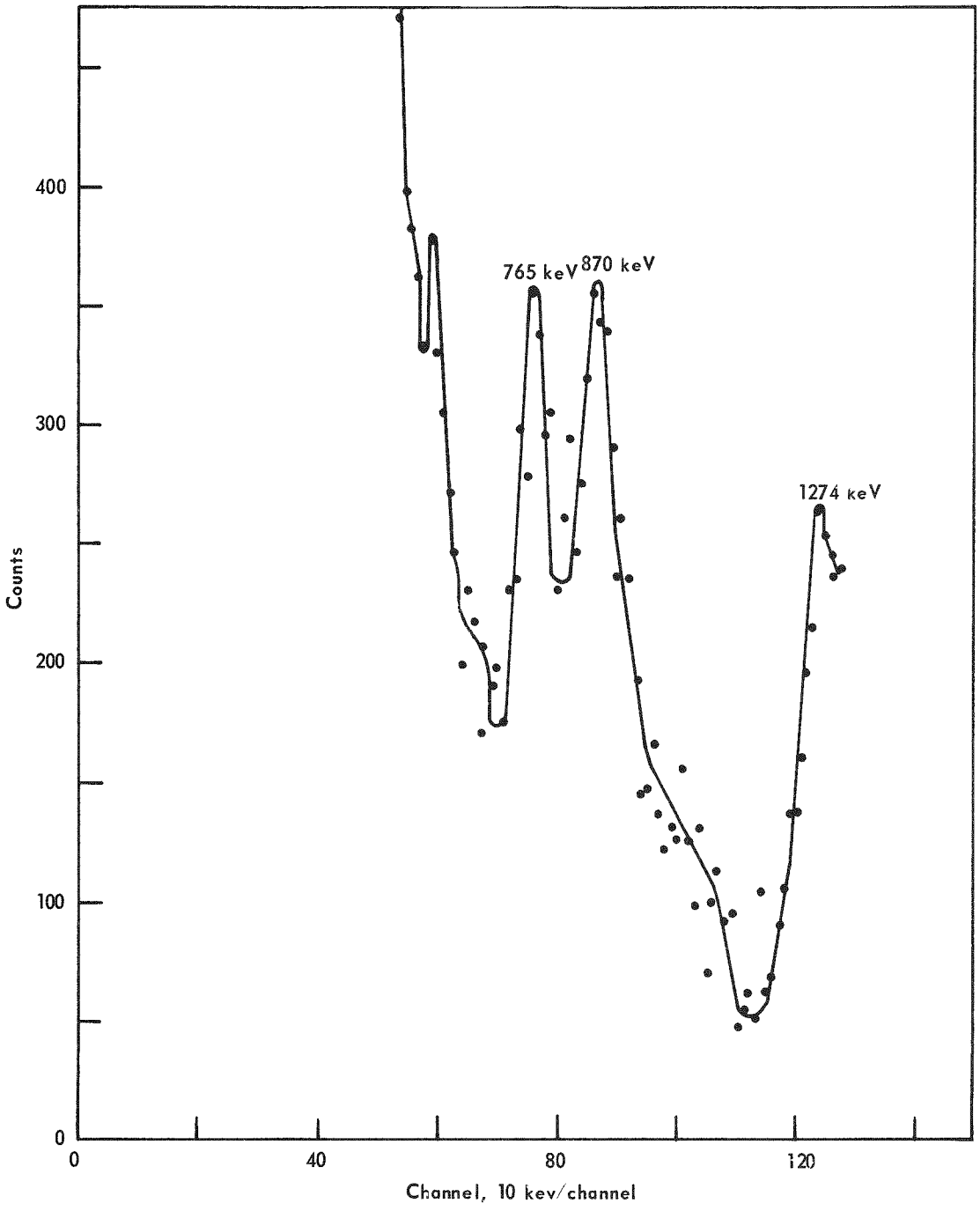

FIGURE 21 - Gamma-ray spectrum from can No. 31.

PART II: MULTICHANNEL ASSAY OF PLUTONIUM-238 IN WASTE CANS

Apparatus

The apparatus for the multichannel assay is shown in Figure 22. A T.M.C. 256-channel pulse height analyzer with a printer readout analyzed the signal from the Hamner amplifier. The gain control was set for 10 keV per channel. Measurements were taken with the cans centered 25.0 and 40.0 cm from the face of the detector. Otherwise, the apparatus was the same as described in Part I.

Technique

Referring again to Figure 4, the area C is the contribution to the nine-channel region centered on the 765-keV photoelectric peak, from the 870 and 1274-keV and higher energy gamma-rays. It is seen in the gamma-ray spectra of the standard cans used in these measurements (Figures 5 through 21) that the 870 and 1274-keV gamma-rays vary considerably in intensity relative to the intensity of the 765-keV gamma-ray from the alpha decay of plutonium-238. The 870-keV gamma-ray arises from the $^{14}N(\alpha,p)^{17}O$ reaction,[6,7] and the 1274-keV gamma-ray is assumed to be from the $^{19}F(\alpha,n)^{22}Na$ reaction. The varying intensities of these two gamma-rays means the area C varies as a fraction of the total area (A+B+C). Hence, an accurate plutonium-238 assay requires the area C be subtracted from the total area (A+B+C) of the nine-channel region of the 765-keV photoelectric peak selected for analysis. This is confirmed by the single-channel analysis of Part I since the assay weight was high for those cans whose gamma-ray spectra had a large contribution at 870 and 1274-keV and was low when these contributions were very small.

The gamma-ray stripping procedure used in the drum assay technique[2] was applied to the multichannel spectra obtained from the same standard cans used for the measurements in Part I. In that technique, the weight W (in grams) of plutonium-238 in the drum is given by[2]

$$W = \frac{\left[\sum_{i=p-4}^{p+4} N_i - 9\left(\frac{N_{p+4} + N_{p-4}}{2}\right)\right]\frac{(A+B)}{A}}{c \cdot t \cdot T} \qquad (10)$$

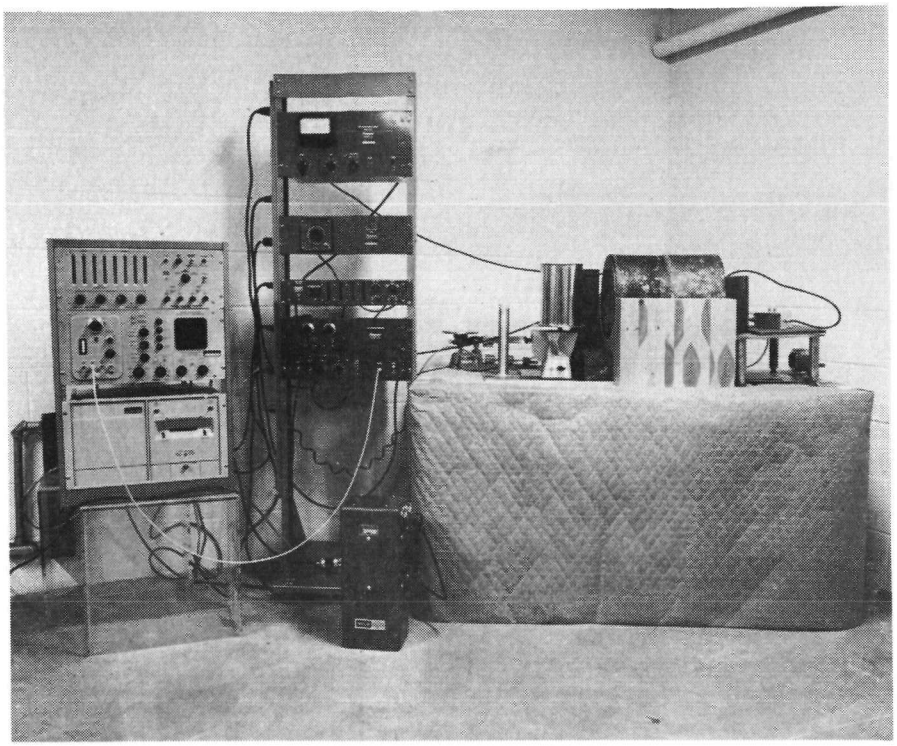

FIGURE 22 - Apparatus for the multichannel assay measurements.

where: N_p = number of counts in the center channel, p, of the 765-keV photoelectric peak

c = 313 counts/min/g for a source-to-detector distance of 2.00 m and is the slope of the calibration curve used for the drum assay

t = counting time (live time on the pulse height analyzer) in minutes

T = transmission of the 765-keV gamma-radiation in the can expressed as a fraction

$\frac{(A+B)}{A}$ = 2.56 ± 0.14 for the detector used in these measurements

In writing Equation 10 and in the following discussion it is assumed that the room background has been subtracted in the multichannel analyzer before printout of the data. In the drum assay, the room background is reasonably constant and can be stored in one part of the multichannel analyzer for use in all measurements taken for one or two days.

The numerator of Equation 10 is the area (A+B) shown in a representative spectrum in Figure 4. For the measurements reported here, the transmission T is the function T(w) shown in Figure 3. Thus, as in Part I, the transmission T was determined by weighing the can and subtracting the weight of the #12 can. Measurements of the cans were made at 25.0 and 40.0 cm, and the inverse square law correction was applied to the slope c of the calibration curve used for drum measurements at 2.00 m. Thus, for the cans, the weight (in grams) of the plutonium-238 contents is given by

$$W = \frac{\left[\sum_{i=p-4}^{p+4} N_i - 9\left(\frac{N_{p+4} + N_{p-4}}{2}\right)\right] 2.56}{D \cdot 313 \cdot t \cdot T(w) \cdot T_{pb} \cdot T_{ss}} \qquad (11)$$

where: D = 64 for the measurements at 25.0 cm

D = 25 for the measurements at 40.0 cm

$T_{pb} \cdot T_{ss}$ = 0.71 as in Part I

The counting time for all measurements was 4.00 min live time on the multichannel analyzer. Thus, Equation 11 becomes

$$W = \frac{\left[\sum_{i=p-4}^{p+4} N_i - 9\left(\frac{N_{p+4} + N_{p-4}}{2}\right)\right] 2.56}{D \cdot 888.9 \cdot T(w)} \qquad (12)$$

Measurements

Three measurements are required:

1) The can is rotated and counted for 4.00 min live time on the pulse height analyzer.
2) A 4.00-min background is subtracted from the data in the analyzer before printout. As discussed above, if the background is relatively constant it can be stored in one part of the multichannel analyzer, and this step requires only a momentary push of a switch on the multichannel analyzer.
3) The can is weighed and the transmission T(w) is taken from Figure 3.

Results and Error

Measurements were made on the same 17 standard cans used in Part I. The results of the measurements at 25.0 cm are presented in Table 4 and the results of the measurements at 40.0 cm are presented in Table 5. Two runs were made at each distance to demonstrate reproducibility.

Table 4

QUANTITATIVE ANALYSIS OF PLUTONIUM-238 IN CANS OF WASTE[a]

Can No.	Actual ^{238}Pu Content (g)	Run #1 Measured ^{238}Pu Content (g)	Run #1 Error (%)	Run #2 Measured ^{238}Pu Content (g)	Run #2 Error (%)
1	0.1196	0.1363	+14	0.1243	+4
2	0.2804	0.2944	+5	0.2832	+1
3	0.1980	0.2317	+17	0.2416	+22
4	0.3912	0.4381	+12	0.4264	+9
5	0.5007	0.3956	-21	0.4306	-14
6	0.5490	0.5061	-8	0.4831	-12
7	0.5600	0.5320	-5	0.5264	-6
9	0.4463	0.4121	-12	0.4151	-7
15	1.775	1.758	-1	1.491	-16
16	0.5030	0.4518	-9	0.4879	-3
17	1.800	1.584	-12	1.799	0
21	0.6810	0.7559	+11	0.7422	+9
22	0.4220	0.4302	+2	0.4262	+1
23	0.0600	0.0420	-30	0.0410	-32
26	0.1020	0.1193	+17	0.1255	+23
27	0.2010	0.2270	+13	0.2030	+10
31	0.080	0.066	-30	0.068	-28

[a]Distance from can to detector was 25.0 cm and the counting time was 4.0 min. The weights of the cans and the transmission T are given in Table 3.

The errors are expressed for amounts of plutonium-238 greater than 0.100 g. Two of the standard cans contain less than this amount and, while the results are presented in Tables 4 and 5, they were not used for the error determination.

For the measurements at 25.0 cm one standard deviation is ±12%, yielding an accuracy at the 95% confidence level of ±25%.

For the measurements at 40.0 cm one standard deviation is ±7.4%, yielding an accuracy at the 95% confidence level of ±15%.

Table 5

QUANTITATIVE ANALYSIS OF PLUTONIUM-238 IN CANS OF WASTE[a]

Can No.	Actual ^{238}Pu Content (g)	Run #1 Measured ^{238}Pu Content (g)	Run #1 Error (%)	Run #2 Measured ^{238}Pu Content (g)	Run #2 Error (%)
1	0.1196	0.1247	+3	0.1157	-3
2	0.2804	0.2779	-1	0.3090	+10
3	0.1980	0.2098	+6	0.2424	+22
4	0.3912	0.3328	-15	0.3805	-3
5	0.5007	0.4628	-8	0.5203	+4
6	0.5490	0.5101	-7	0.4659	-15
7	0.5600	0.5666	+1	0.5611	0
9	0.4463	0.4017	-10	0.4169	-7
15	1.775	1.1184	-8	1.7194	-3
16	0.5030	0.5096	+1	0.5309	+6
17	1.8000	1.8186	+1	1.7460	-3
21	0.6810	0.6507	-4	0.6431	-6
22	0.4220	0.4153	-2	0.4371	+4
23	0.0600	0.0531	-11	0.0594	-10
26	0.1020	0.1092	+1	0.1160	+8
27	0.2010	0.1999	-3	0.2117	+2
31	0.080	0.0664	-17	0.0546	-32

[a]Distance from can to detector was 40.0 cm and the counting time was 4.0 min. The weights of the cans and the transmission T are given in Table 3.

The measurements at 40.0 cm are considerably more accurate than those at 25.0 cm. It would seem reasonable that the inverse square law correction to those finite sources would be responsible. However, this would only mean the factor D would be in error and would not result in a larger scatter in both the plus and minus directions. Apparently the detector is much more sensitive to the actual distribution of waste in the can at 25.0 cm than at 40.0 cm.

It is seen that the smallest amount of plutonium-238 assayed was 0.080 g in can No. 31 and the error was large. The measurement could be improved by increasing the counting time t, perhaps to 6.0 min.

CONCLUSIONS

The single-channel analysis seems suitable for the purpose of the in-line assay. In practice it may be desirable not to include the factor 1.10 in the denominator of Equation 9 to account for a nominal contribution of higher-energy gamma-rays. This would raise the assay weights 10%. One significant improvement in the electronics would be to use a pulse height analyzer equipped with a single-channel analyzer instead of the amplifier and single-channel analyzer used for the measurements reported here. It would be easier to adjust the gain, and a 1-min live time count could be taken. The single-channel analyzer used in these measurements operates on clock time, and with a high count rate some counts would be lost because of dead time in the single-channel analyzer.

The multichannel measurements should be made at 40.0 cm. These measurements show that this technique is competitive in time and accuracy with the 99.8-keV gamma-scan technique[1] and has the advantage of not requiring the cans to be categorized by the type of waste or the amount of plutonium-238 present. Also, as reported in Reference 3, a high-energy gamma-ray assay is significantly more accurate for heavy cans than is the 99.8-keV gamma-ray assay.

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