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A SYSTEM OF HIGH-PURITY GERMANIUM DETECTORS FOR THE DETECTION AND MEASUREMENT OF INHALED RADIONUCLIDES

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ABSTRACT

A system of eight high-purity germanium detectors mounted, four each, as arrays on two downward-looking cryostats is described. This system is designed to measure photons emitted by inhaled radionuclides and is efficient for photon energies between 10 keV and 200 keV. In routine operation since October, 1976, the system is used mainly to detect and measure inhaled plutonium and americium.

Each of the eight detectors has a surface area of 10 cm^2 and a resolution of 650 eV full-width half-maximum or better at 60 keV. This excellent resolution means that the photopeak can be placed in a narrow portion (about 1 keV) of the background energy spectrum. Consequences of this feature are: 1) The subject background count rate for a given photopeak is low (between 0.025 and 0.035 counts/min. per cm^2 detector surface area for a 1-keV band in the range of 15 to 100 keV); 2) The subject background can be well estimated from counts in the energy spectrum a few keV above the photopeak of interest; 3) Baseline resolution can be obtained for photons separated by as little as 1.5 keV in energy; and 4) Identification of unknown or unexpected radionuclides can be accomplished with good accuracy.

DISCLAIMER

Applications and capabilities of the system are discussed. The current application is for the measurement of plutonium via the measurement of the 59.54-keV gamma from ^{241}Am. The measurement of L x-rays from the decay of plutonium is also discussed.

INTRODUCTION

Detection and measurement of inhaled radionuclides involves the sensing of the photons emitted by the inhaled material by means of sensitive detectors placed against the surface of the chest. The detector system provides data about the photon energy spectrum at the surface of the subject's chest. The information is then analyzed to determine whether the count is positive or background for a given radionuclide and, if positive, to determine the amount of that radionuclide in the chest cavity.

For this purpose the ideal detector system should provide data from which a reliable determination of the subject's exposure status can be made at the lowest possible amount of the inhaled radionuclide. The requirements for such a detector system are an efficient collection of the characteristic photons emitted by the inhaled radionuclide and a low and well-establishable background count rate in the regions of interest in the photon energy spectrum. These requirements translate into the need for a detector system with a large surface area, negligible attenuation in the entrance window to the detector, total energy deposition and collection in the active region of the detector, good resolution, low noise, and stable operating characteristics.

This paper describes a detector system, in routine operation since October, 1976, which approaches the characteristics of the ideal detector system for the detection and measurement of inhaled radionuclides. This system consists of eight high-purity germanium detectors mounted, four each, as arrays on two downward-looking cryostats. Each of the eight detectors has a resolution of about 650 eV full-width half-maximum (FWHM) at a photon energy of 60 keV, an active surface area of 10 cm^2, and is efficient for photon energies between approximately 10 keV and 200 keV.

The system is used mainly to detect and measure inhaled plutonium and americium, although it can be used for any radionuclide emitting photons in the energy range of 10 to 200 keV. Discussion of the operating characteristics will focus on the use for measuring the 59.54-keV gamma from ^{241}Am and the L x-rays from the decay of plutonium.

PHYSICAL DESCRIPTION

Each of the eight detectors is a wafer of high-purity germanium with an active surface area of 10 cm^2 and a thickness of 1.0 cm. Four detectors are mounted, as a square array, on 10-cm areas on the lower surface of a downward-looking cryostat, as shown in Figure 1. The detectors are located 5 mm inside the lower face of the cryostat and are covered by a beryllium window, 0.26 mm thick, and by an additional Lexan cover, 0.127 mm thick. Transmission through the window is approximately 97 percent at 10 keV.

The detectors are cooled by liquid nitrogen, contained in the cryostats, to minimize detector noise during operation. Warming to ambient temperatures does not automatically destroy this type of detector as it would a lithium-drifted germanium detector. In fact, one of the arrays has accidentally warmed twice without detectable deterioration in performance. The capacity of each cryostat is 15 liters, providing cooling for about 5 days.

Each array system is mounted on mechanical support arms, shown in Figure 2, with adjustments for tilting up to 15° in two planes, for raising and lowering, for extension, and for movement in a horizontal arc. These adjustments allow the detector arrays to be positioned against the plane of the subject's upper chest as the subject lies supine.

The detector system is housed in a counting chamber with steel walls, ceiling, floor, and door 15.2 cm thick and covered on the interior with a graded shield of lead, tin, and zinc. This shielded chamber provides a reduction in the background count rate at 60 keV by a factor of over 100.

Electronically, the output of each of the detectors is routed separately throughout to maintain optimum resolution. The electronic setup is relatively simple. The pulse from the detector is preamplified in a unit mounted as an integral part of the cryostat to minimize the capacity effects on the input signal to the preamplifier. From the preamplifier the signal is brought from the counting chamber to the operating console through a coaxial cable, and is amplified (Ortec Model 472 amplifier). The signal is then directed through a signal processor (Ortec Model 4671), which was included to minimize the effect of baseline distortion in the low-energy region from large overload pulses and to eliminate saturated input conditions to the analog-to-digital converter (ADC), through a mixer-router with eight inputs (Northern Model NS-459B), through the ADC (Northern Model NS-621 8192 ADC), and into the appropriate memory group (256 channels) of the pulse height analyzer (Northern NS-636). The final display consists of eight individual spectra, each in separate memory groups in the pulse height analyzer. A more detailed description of the electronic setup of the system is presented elsewhere.[1]

For the analysis of the data presented by the eight spectra, a minicomputer (Data General Model 1220) with a dual disk system, for data and software storage, is used. The data from the eight spectra can be analyzed either separately or as a composite spectrum, summed by the minicomputer. Custom software is provided in-house to do the desired analysis. The dual disk capability offers a large storage capacity so that the spectra data for over 300 subjects can be stored on one disk cartridge and a variety of operating software programs can be stored on the other. Computer control and data output is via a cathode ray terminal (Tektronix Model 4012) with optional output to a hard copy unit (Tektronix Model 4631).

Table 1: Values of the Background Count Rate for a Typical Unexposed Male Subject for a 1-keV Energy Band and per 1 cm^2 Detector Surface Area

Photon Energy (keV)	Subject Background Count Rate (count/min per cm^2)
15	0.035
20	0.030
40	0.027
60	0.032
80	0.029
100	0.025

OPERATING CHARACTERISTICS

Each of the eight detectors has a resolution of 650 eV or better FWHM at 60 keV. This excellent resolution means that the photopeak of a characteristic photon can be placed in a narrow portion of the background energy spectrum. Consequences of this feature are: 1) The subject background count rate for a given photopeak is low; 2) The subject background count rate can be well-estimated from the count rate in the portion of the energy spectrum a few keV above the photopeak of interest; 3) Baseline resolution can be obtained for photopeaks separated by as little as 1.5 keV of energy; and 4) Identification of unknown or unexpected radionuclides can be accomplished with good accuracy.

The background count rate for a typical male subject is shown in Table 1 for photon energies from 15 to 100 keV. The values are for a 1-keV energy band, which is the width of a typical photopeak, and are presented in units of counts/min per cm^2 of detector surface area. The room background (with no subject present) is approximately 50 percent of these values.

Because 1) the photopeak is so sharply defined, 2) the spectrum returns to background levels immediately above the photopeak of interest, and 3) the background spectrum does not change detectably over a range of a few keV, the subject background count rate can be well established from the count rate in the region of the subject's spectrum one or two keV higher than the photopeak of interest. This feature is illustrated in Figure 3, which shows a subject spectrum for an exposure to ^{241}Am superimposed on a spectrum for an unexposed subject of similar body stature. The spectra are for the composite sum from the eight detectors and a counting time of 2000 seconds. The comparison shows the abrupt return to background levels in the spectrum immediately about the 59.54-keV photopeak from ^{241}Am. For such a case

and using the region of the spectrum from 61 to 75 keV to establish the subject background count rate for the photopeak at 59.54 keV, that value can be determined with a relative standard deviation of 2 percent.

Because baseline resolution can be obtained for characteristic photons separated by as little as 1.5 keV of energy, simultaneous measurement of different inhaled radionuclides can be accomplished as long as the energies of the characteristic photons are different by more than 1.5 keV. For example, Figure 4 shows the composite spectrum of a person exposed to both ^{241}Am and ^{238}U (63.2, 63.8 keV doublet from the ^{234}Th daughter). Both of the characteristic photopeaks are easily distinguishable and measurable with this system.

Identification of unknown or unexpected radionuclides can be accomplished easily with this detector system since the characteristic photons can be localized in a band in the energy spectrum of about 1 keV in width. Because of the method of determining the subject background in the region of interest (described above), the presence of such radionuclides does not appreciably affect the measurement of the characteristic photon of interest even though the subject background count rate in the region of interest may be significantly altered. The exception, of course, occurs when the interfering photopeak is within 1.5 keV of the photon energy of interest.

This type of system represents a significant procurement cost. The set of detector arrays, without the supporting electronic amplifying and analyzing system, alone cost 91,000 U.S. dollars. This cost, however, is decreasing as technology improves. A second high-purity germanium system, with a 50-percent increase in surface area and with similar resolution capability, was purchased in September, 1977 at a cost of 74,000 U.S. dollars.

APPLICATIONS

The detector system is designed for the primary application of the measurement of inhaled plutonium via the measurement of the 59.54-keV gamma from ^{241}Am. Figure 5 shows the spectrum of a subject (of approximately average stature) exposed to ^{239}Pu and ^{240}Pu, with 2750 ppm by weight ^{241}Am, presented in terms of count per minute per cm^2 of detector surface area. For comparison, the spectrum of that subject, obtained using a NaI-CsI "phoswich" detector system, is also presented. (The "phoswich" system consists of two detectors, each with a NaI crystal 4 mm thick by 10.2 cm in diameter backed by a CsI crystal 7.6 cm thick and the same diameter). Salient features of this presentation are 1) The sharp photopeak at 59.54 keV for the germanium system, 2) The similarity of the two systems for the measurement of the Compton-scattered portion of the spectrum, and 3) The relatively low count rate in the L x-ray region for both systems. For the germanium system only the count in the photopeak is measured while the "phoswich" measures both the photopeak and the photons Compton-scattered in the subject's body, since these two components are indistinguishable as a result of the poorer resolution of the "phoswich" system.

The minimum detectable activity for the measurement of ^{241}Am in the lungs of an "average man" subject is 0.129 nCi based on the method of Altshuler and Pasternak[2] for the conditions of 1) an accurately known subject background count, 2) a 0.05 risk of making either a Type I or a Type II error, and 3) a counting time of 2000 seconds. For the measurement, via the americium, of a mixture of ^{239}Pu and ^{240}Pu, with a combined specific activity of 0.070 Ci/g, the corresponding minimum detectable activity is 2.77 nCi of the plutonium at 1000 ppm by weight ^{241}Am.

Another measure of the measurement capability is the relative uncertainty in the measured amount. A relative standard deviation of 60 percent is obtained for the measurement of 0.073 nCi ^{241}Am and of 1.55 nCi plutonium at 1000 ppm ^{241}Am, calculated by the method of Falk.[3] This method includes considerations of uncertainties related to the subject and source parameters as well as counting statistics. Subject and source parameters include such items as chest thickness, effective thickness of the lung, the distance between the effective center of the source and the detector, and the fraction of the ^{241}Am in the plutonium mixture (for the plutonium measurement). The propagated relative uncertainty from just uncertainties in the subject and source parameters is considered to be 0.22 .

The system can be used for the measurement of plutonium via L x-rays and has several advantages over detector systems with poorer resolution. Figure 6 shows the low-energy portion of the spectrum for a plutonium source, accompanied by the component from ^{241}Am normalized to 1000 ppm by weight. The peak separation, a result of the good resolution of 650 eV FWHM, allows one to focus on the L_{beta} and L_{gamma} peaks, separately or together, for the measurement. The baseline separation of about three channels (at a calibration of 0.33 keV/channel) allows use of these channels to indicate the subject background for the adjacent peak region. The slight offset in the position of the L x-ray peaks from the decay of ^{241}Am lessens the interference from this source when a judicious choice of the channels to be included in the summation for the measurement of the plutonium is made.

Problems associated with the measurement of L x-rays from inhaled radionuclides and which are common for all detector systems, such as determining values for the subject and source parameters, will not be discussed here. The severe attenuation of L x-rays in the subject's lungs and chest wall remains a major problem which is independent of the sophistication of the detector system.

Based on calibration values the system capability for the measurement of L x-rays was calculated. For an average subject with a chest thickness of 2.5 cm, the calibration factor for the summed L_{beta} and L_{gamma} x-rays is 1.5 counts/min per nCi of $^{239,240}Pu$ (combined specific activity = 0.07 Ci/g) with a corresponding subject background of 5.0 counts/min. For these values and using the method of Falk,[3] the relative uncertainty (at one standard deviation) for the measurement of 16 nCi of plutonium via the L x-rays is 0.67 . The propagated relative uncertainty from only the uncertainties in the subject and source parameters is 0.49 for this calculation. A value for the minimum detectable activity is not presented because such a value, calculated by the method of Altshuler and Pasternak,[2] includes only considerations of counting statistics and does not include the significant uncertainties in the subject and source parameters.

SUMMARY

A system of arrays of high-purity germanium detectors represents the state of the art for the measurement of inhaled radionuclides. An indication of capability of this system is the measurement of 0.073 nCi ^{241}Am (59.54-keV gamma), and a corresponding value of 1.55 nCi $^{239,240}Pu$ at 1000 ppm by weight ^{241}Am, with a relative standard deviation of 60 percent. The system has several advantages over systems with poorer resolution for the measurement of L x-rays, but the relative standard deviation of 67 percent for the measurement of 16 nCi $^{239,240}Pu$ (combined specific activity of 0.07 Ci/g) reflects significant problems in the measurements of L x-rays not associated with the type of detector system.

Significant advantages are derived from the excellent resolution of 650 keV or better FWHM for each of the eight detectors. Subject background is low and can be well estimated (with a relative standard deviation of 2 percent for the measurement of the 59.54-keV photopeak from ^{241}Am). Interference from photon emissions from other radionuclides either can be avoided or minimized.

The electronic setup is relatively simple and stable. The output of each of the eight detectors is maintained separately to provide optimum resolution. The eight resulting spectra can be analyzed either separately or as a composite spectrum. Data analysis by computer is a necessary part of the system for routine counting conditions.

The surface area of 10 cm^2 per detector, positioned at eight locations over the upper chest, provides adequate coverage of the lung regions. Advances in technology, which now allows a surface area of at least 15 cm^2 per detector while maintaining good resolution, can be expected to improve further the detection and measurement capability of this type of detector system.

REFERENCES

(1) Tyree, W.H.; Falk, R.B.; Wood, C.B.; Liskey, R.W.; High-Purity Germanium Detection System for the in vivo Measurement of Americium and Plutonium, Rocky Flats Plant RFP-2525 (1976).

(2) Altshuler, B; Pasternak, B.; Statistical Measures of the Lower Limit of Detection of a Radioactivity Counter, Health Physics 9,293 (1963).

(3) Falk, R.B.; Lagerquist, C.R.; Error Analysis and Propagation of Uncertainties for the Measurement of Inhaled Transuranium Elements (Proceedings of the Workshop on Measurements of Heavy Elements in vivo, Seattle, 1976), BNWL-2088(1976)63.

FIGURE CAPTIONS

Figure 1 — The lower face of the two arrays of high-purity germanium detectors.

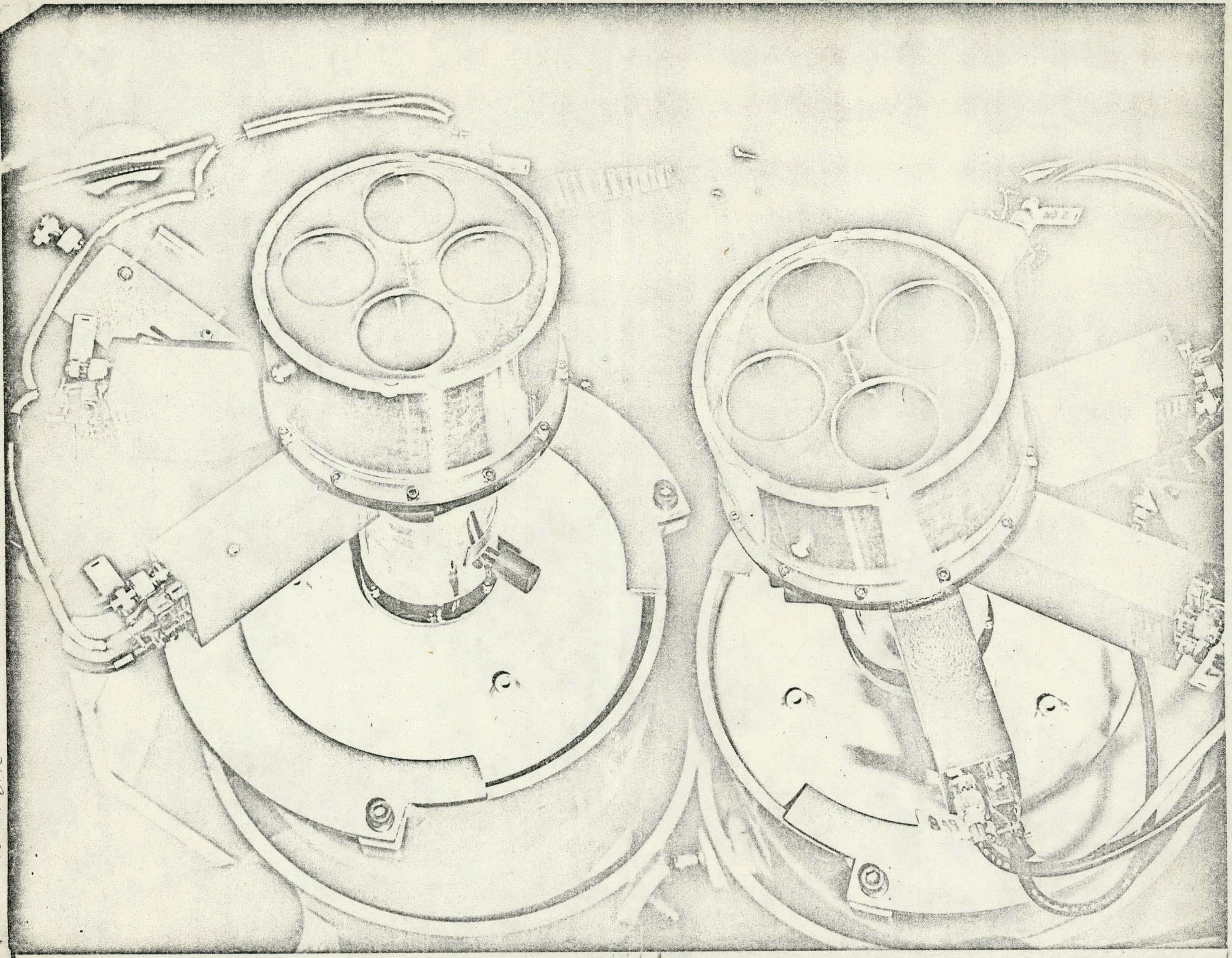
Figure 2 — One array mounted on the mechanical support arm inside the counting chamber.

Figure 3 — A spectrum of a subject exposed to a mixture of plutonium and ^{241}Am, superimposed on a subject background spectrum, illustrating the abrupt return to background levels immediately above the 59.54-keV photopeak.

Figure 4 — A spectrum of a subject exposed to both ^{241}Am and ^{238}U, showing the separation of the photopeaks at 59.54 keV and at 63.2, 63.8 keV (doublet).

Figure 5 — A spectrum of a subject exposed to a mixture of plutonium and ^{241}Am, for both the germanium system and a "phoswich" system, showing the comparison of these detector systems for the L x-ray, Compton-scattered, and photopeak regions of the spectrum.

Figure 6 — The L x-ray region of the spectrum for a plutonium source, accompanied by the component from ^{241}Am normalized to 1000 ppm by weight, illustrating the peak separation of the L_{alpha}, L_{beta}, and L_{gamma} x-rays obtained by one of the germanium detectors in the system.



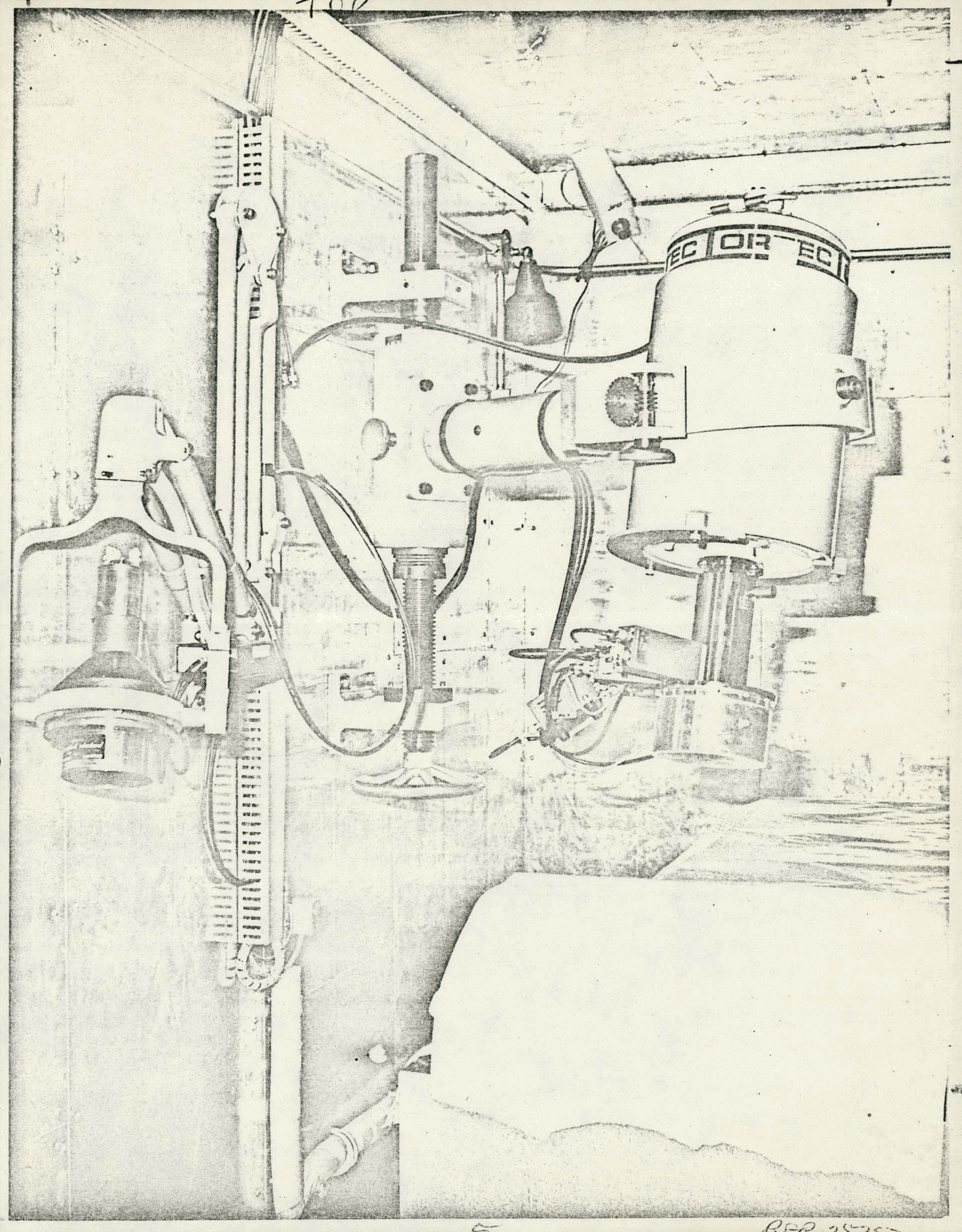

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Fig. 3

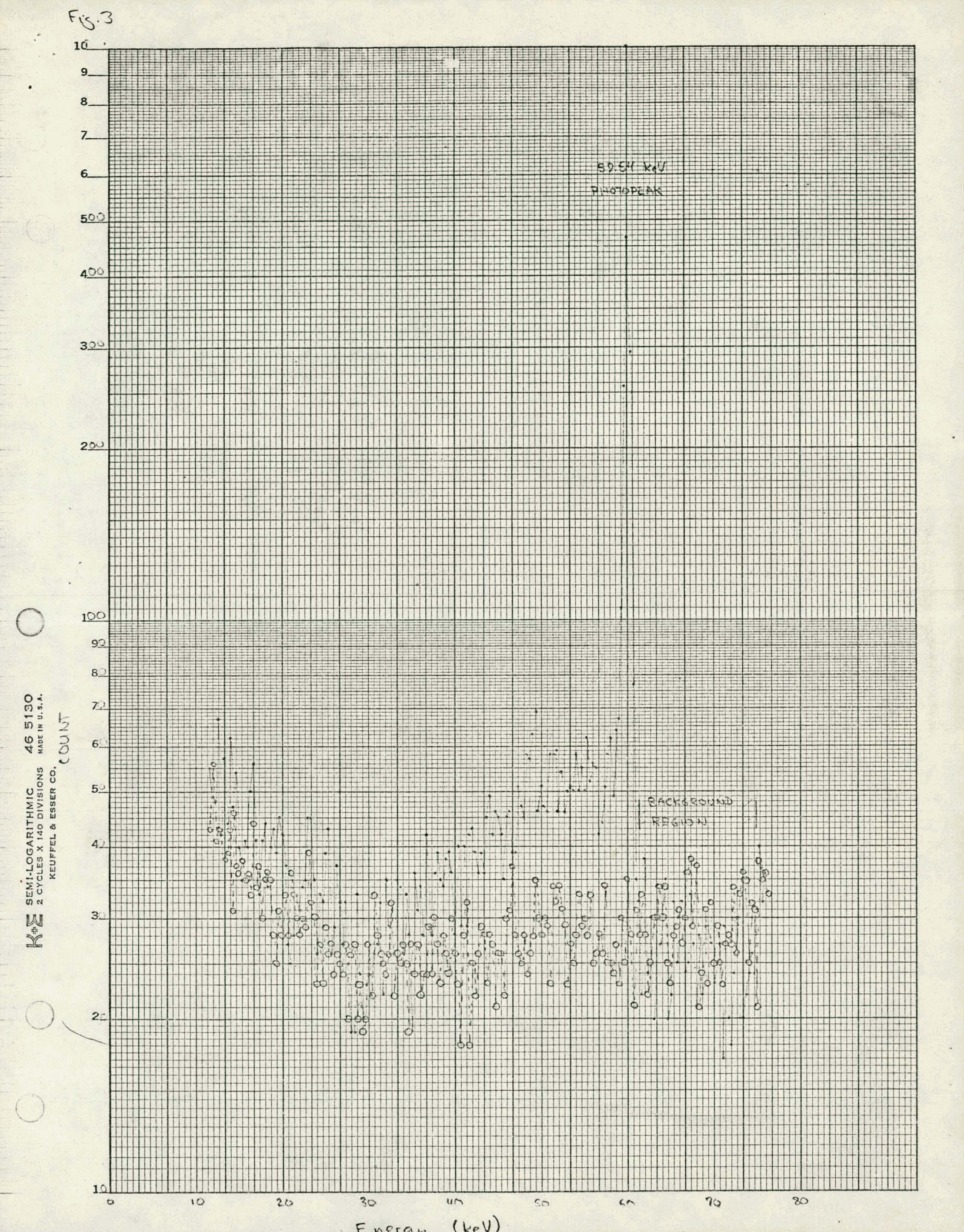
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Fig. 4

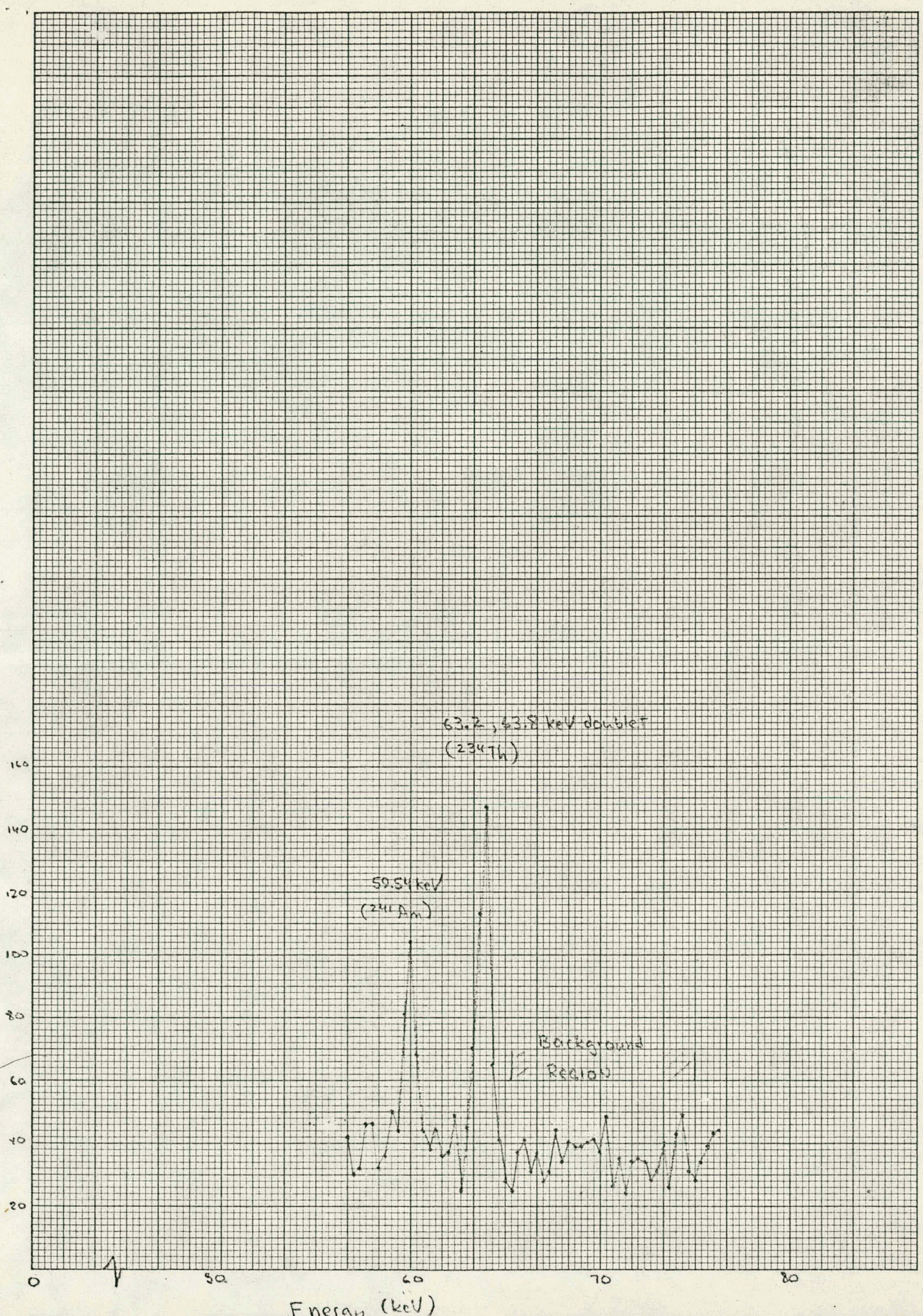
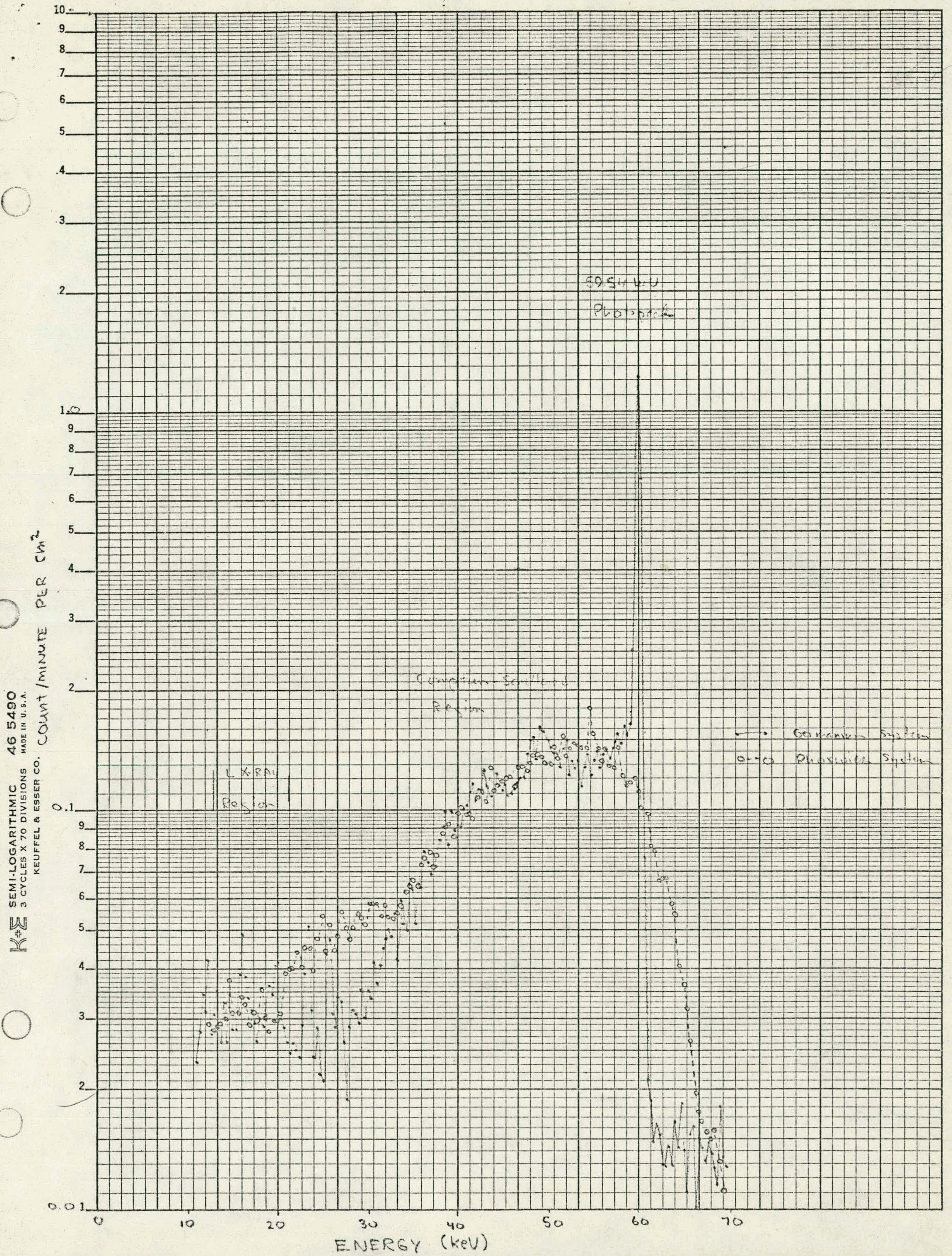


Fig. 5


K+E SEMI-LOGARITHMIC 46 5490
3 CYCLES X 70 DIVISIONS MADE IN U.S.A.
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ENERGY (keV)
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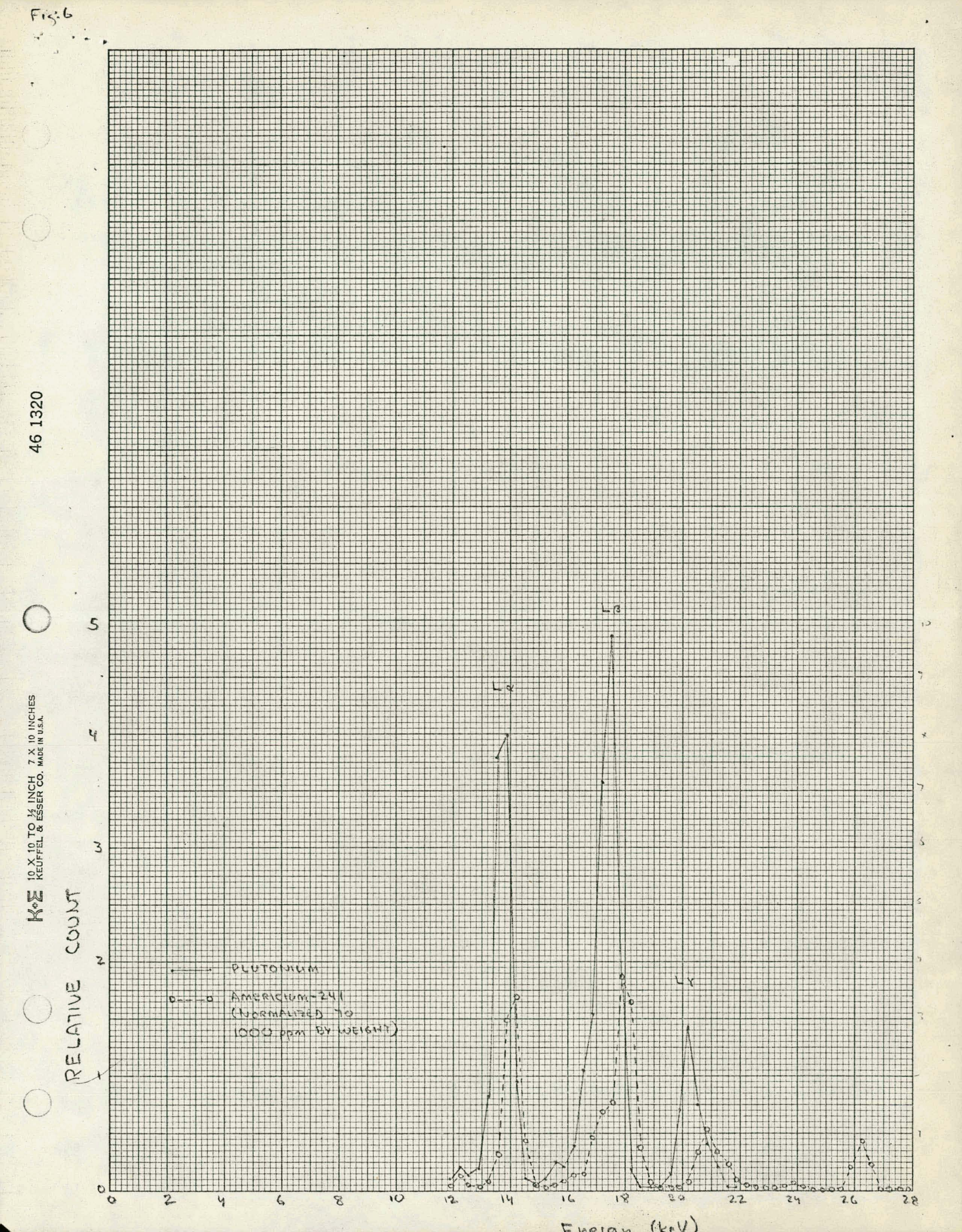
Fig. 6



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KEUFFEL & ESSER CO. MADE IN U.S.A.