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INSTRUMENT RESEARCH AND DEVELOPMENT

QUARTERLY PROGRESS REPORT

FOR PERIOD ENDING APRIL 20, 1952

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INSTRUMENT RESEARCH AND DEVELOPMENT REPORT

CHEMISTRY DIVISION

GAMMA-RAY ENERGY RESOLUTION WITH SODIUM IODIDE SCINTILLATION SPECTROMETERS

C. J. Borkowski

Various methods have been tried in the last year for obtaining a stable NaI-TlI scintillation spectrometer with good energy resolution, that is, less than 8% full width at half maximum for a 670-kev photoelectric peak. In early attempts lucite light pipers were used with aluminum foil as a reflector. Most of the assemblies showed a drift in pulse size over a period of weeks. The following method of crystal preparation and mounting has given long-time surface stability of the NaI-TlI and good energy resolution.

Crystal Preparation and Mounting. In most of the applications cylindrical crystals 1 1/2 in. in diameter and 1 in. in thickness were used. A radius was turned on one face of the crystal to conform to the curvature of the photocathode. This was done on the lathe while the crystal was kept wet with mineral oil. After the radius had been turned, all subsequent operations on the crystal were made in a dry box having a relative humidity of less than 10%. After surplus mineral oil was removed from the crystal with Kleenex, the crystal was ground dry with 180-grit carborundum or emery paper. No polishing was done, since diffuse reflection has given better energy resolution than specular reflection, especially for the larger crystals. All surfaces were rough ground. The crystal was optically coupled to the photocathode with Dow-Corning DC 200 fluid having a viscosity of 60,000 centipoises. However, if the crystal makes a very good fit to the photocathode, Dow-Corning silicone stopcock grease is preferred as the optical coupling medium.

Magnesium oxide is superior to aluminum foil as a reflector, especially for the larger crystals from which 30% more light was obtained. Prior to use, the magnesium oxide should be dried in an oven at 120°C. Figure 1 shows the method used for mounting the crystal on the type-5819 photomultiplier. The magnesium oxide should be firmly packed between the crystal and the thin aluminum tube. An aluminum housing was slipped over the thin

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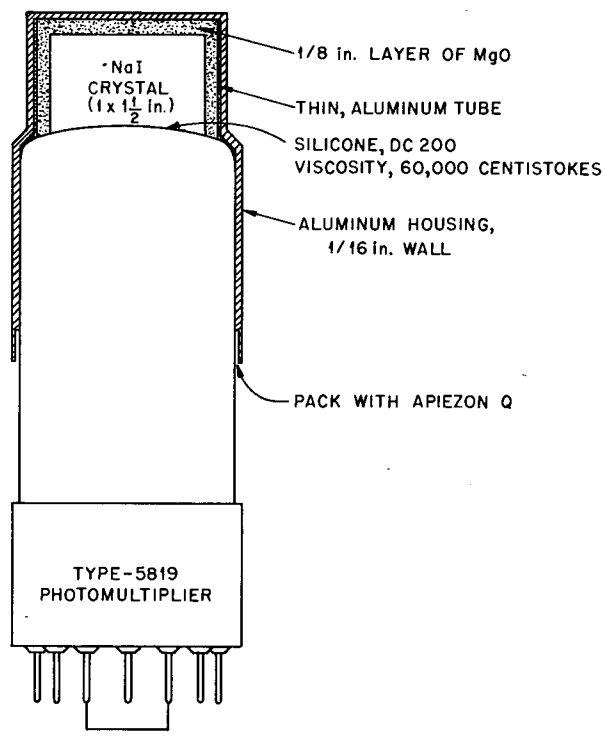

Fig. 1. Sodium Iodide Scintillation Spectrometer Crystal Mounting.

aluminum tube and crystal and sealed to the glass photomultiplier envelope with Apiezon Q. Black Scotch electrical tape was then wrapped around the Apiezon seal, the aluminum housing, and the remainder of the glass envelope to create a light-tight, hermetically sealed assembly. The top of the aluminum housing should be kept thin, about 0.015 inch.

Scintillation spectrometers with this type of seal have given over six months of service with negligible change in resolution or pulse size. Several portable scintillation counters made for uranium ore prospecting for U. S. Geological Survey were sealed in the same manner; they have given good service.

An alternate method of mounting that does not require machining of the sodium iodide crystal is the following: a flat, glass disk 1 1/2 in. in diameter and about 0.060 in. in thickness (made from photographic glass plate) is cemented to the photocathode with Gelva resin, which is highly transparent and has a melting point of 65°C. The photomultiplier, resin, and glass disk are heated to about 75°C in an oven to obtain optical coupling between the photocathode and glass disk. Heating to this temperature produces no change in the photomultiplier characteristics. The flat end of the 1 1/2- by 1-in., cylindrical, sodium iodide crystal is optically coupled to the flat, glass disk with silicone stopcock grease and mounted as shown in Fig. 1.

Lucite light pipers were not used because long-time surface stability of the sodium iodide could not be obtained with them. No improvement in resolution is obtained by using light pipers when magnesium oxide is used as a reflector.

Effect of Crystal Surface Treatment and Reflectors. The effect of surface treatment of the sodium iodide crystal and a comparison of magnesium oxide and aluminum-foil reflectors are shown in Table 1.

If the photomultiplier used gives a width around 9.0%, there is very little difference between solvent-polished and rough-polished crystals when the crystal is relatively small, that is, 1 1/2 by 1 in., and when magnesium oxide is used as the reflector. However, when a photomultiplier that gives excellent resolution, that is, 7.5% width, is used, better results are obtained with rough-polished crystals than with solvent-polished crystals. The preferred procedure is to rough grind all surfaces of the crystal and to use magnesium oxide as a reflector.

Energy Resolution. Figure 2 indicates the kind of resolution (7.2% half width) that is obtained for the 670-kev gamma ray from Cs^{137} with the use of a selected type-5819 photomultiplier and a 1 1/2- by 1-in. crystal. Figure 3 shows that a crystal 1 1/2 by 2 in. gives essentially the same resolution as a crystal half as thick. Using a 3- by 3-in., cylindrical, sodium iodide crystal with magnesium oxide as a diffuse reflector, a 9.0% width was obtained on Cs^{137}, as shown in Fig. 4. The 3- by 3-in. crystal was mounted in somewhat the same manner as than shown in Fig. 1. Since the crystal was much larger than the photocathode of the type-5819 photomultiplier, all crystal surfaces not in contact with the photocathode were surrounded with magnesium oxide.

Figure 5 shows the two Co^{60} gamma rays well resolved. The Compton distribution for the 1.33-Mev gamma ray can be seen on the tail of the 1.17-Mev Compton distribution.

Figures 6 and 7 show the difference in the energy distribution of recoil electrons, pairs, and photoelectrons from the gamma rays of Na^{24}. The data

TABLE 1

Effect of Surface Treatment of Sodium Iodide Crystals and a Comparison of Magnesium Oxide and Aluminum-foil Reflectors

CRYSTAL SIZE	SURFACE TREATMENT	TYPE OF REFLECTOR	RELATIVE PULSE SIZE	HALF WIDTH FOR Cs^{137} (%)	PEAK-TO-VALLEY RATIO	PHOTOMULTIPLIER TUBE NO.
1 1/2- by 1-in. cylinder	180-grit emery paper	MgO	100	9.5	28:1	650-265
	Acetone solvent polish	MgO	100	9.5	28:1	650-265
	Acetone solvent polish	Al	78	13.0	23:1	650-265
1 1/2- by 2-in. cylinder	180-grit emery paper	MgO	100	9.8	30:1	650-265
	Acetone solvent polish	MgO	79	10.9	31:1	650-265
	Acetone solvent polish	Al	63	13.6	21:1	650-265
1 1/2- by 1-in. truncated cone	180-grit emery paper	Al	90	9.7	38:1	650-265
	600-grit emery paper	MgO	100	9.0	42:1	650-265
	Acetone solvent polish	MgO	100	9.0	42:1	650-265
1 1/2- by 1-in. cylinder	180-grit emery paper	MgO	100	7.5	56:1	152-154
	180-grit emery paper	Al	73	8.6	45:1	152-154
	600-grit emery paper	MgO	100	8.3	48:1	152-154
1 1/2- by 1-in. cylinder with 3/4-in. Lucite light piper	600-grit emery paper	MgO	66	9.2	38:1	152-154
	Acetone solvent polish	MgO	100	8.8	37:1	152-154
	Acetone solvent polish	Al	76	10.2	29:1	152-154

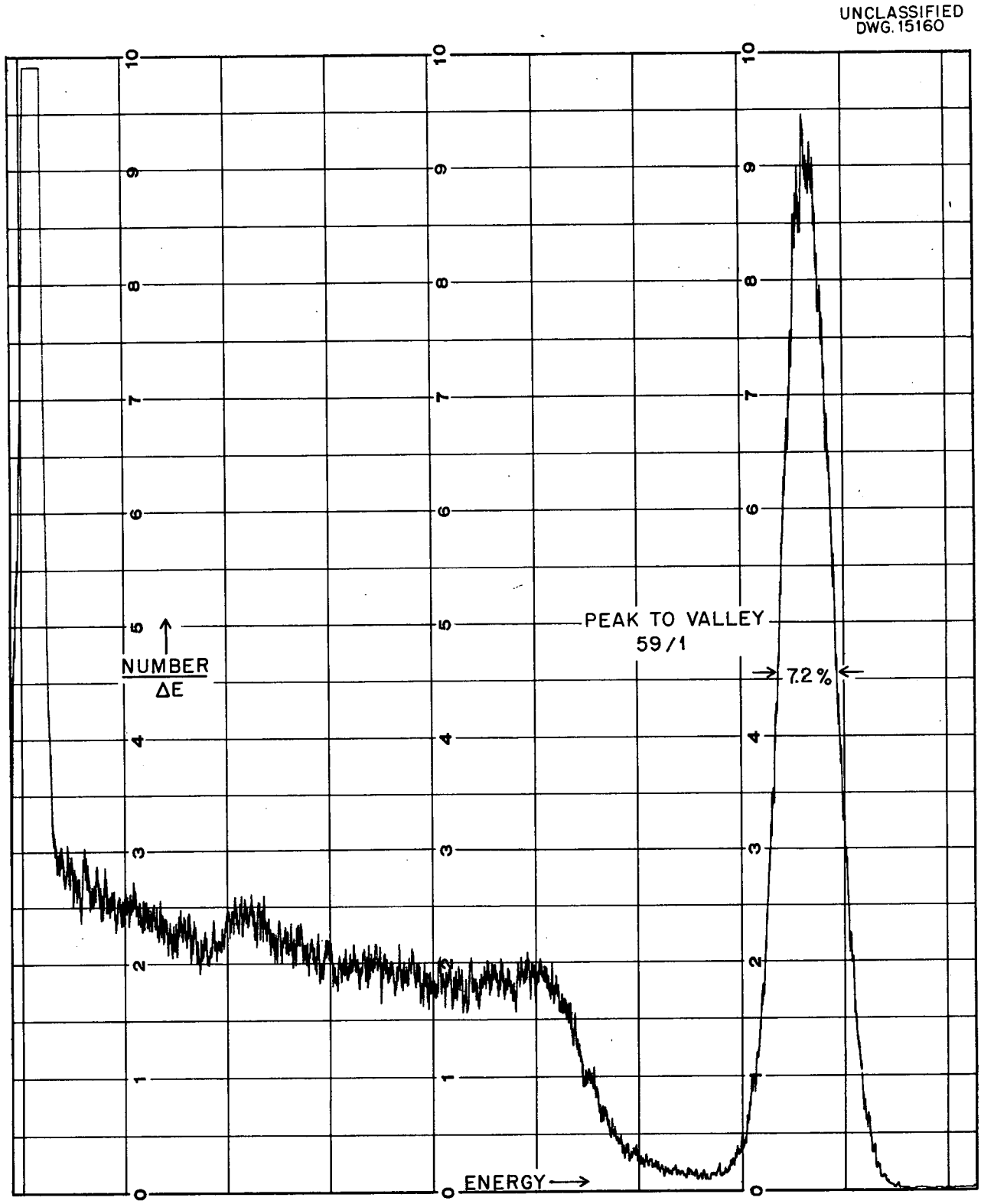

Fig. 2. Resolution of 670-kev Gamma Ray from Cs^{137} Obtained with a Type-5819 Photomultiplier and a 1 1/2- by 1-in. Sodium Iodide Crystal.

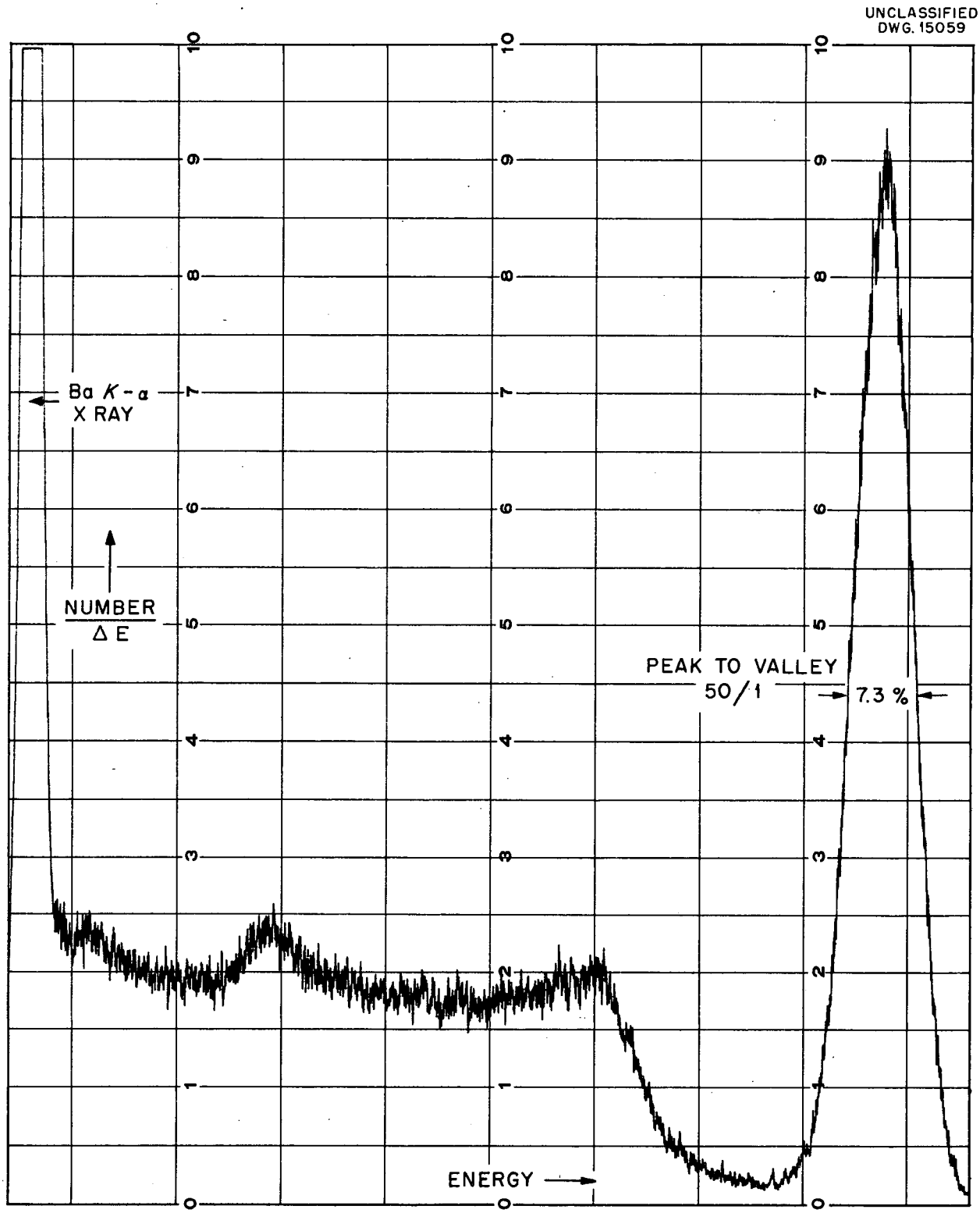

Fig. 3. Resolution of 670-kev Gamma Ray from Cs^{137} Obtained with a Type-5819 Photomultiplier and a 1 1/2- by 2-in. Sodium Iodide Crystal.

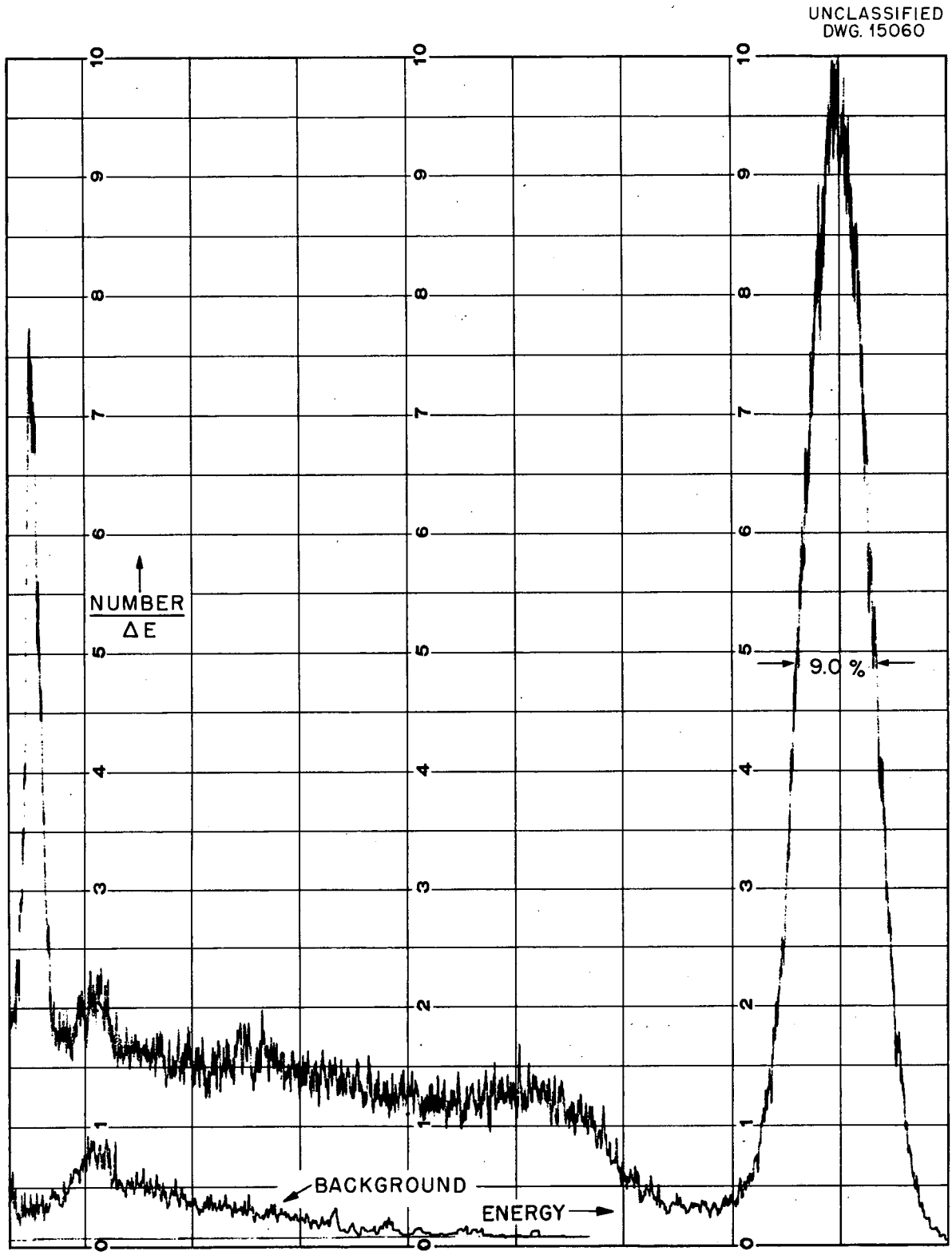

Fig. 4. Resolution of 670-kev Gamma Ray from Cs^{137} Obtained with a Type-5819 Photomultiplier and a 3- by 3-in., Cylindrical, Sodium Iodide Crystal with Magnesium Oxide as a Diffuse Reflector.

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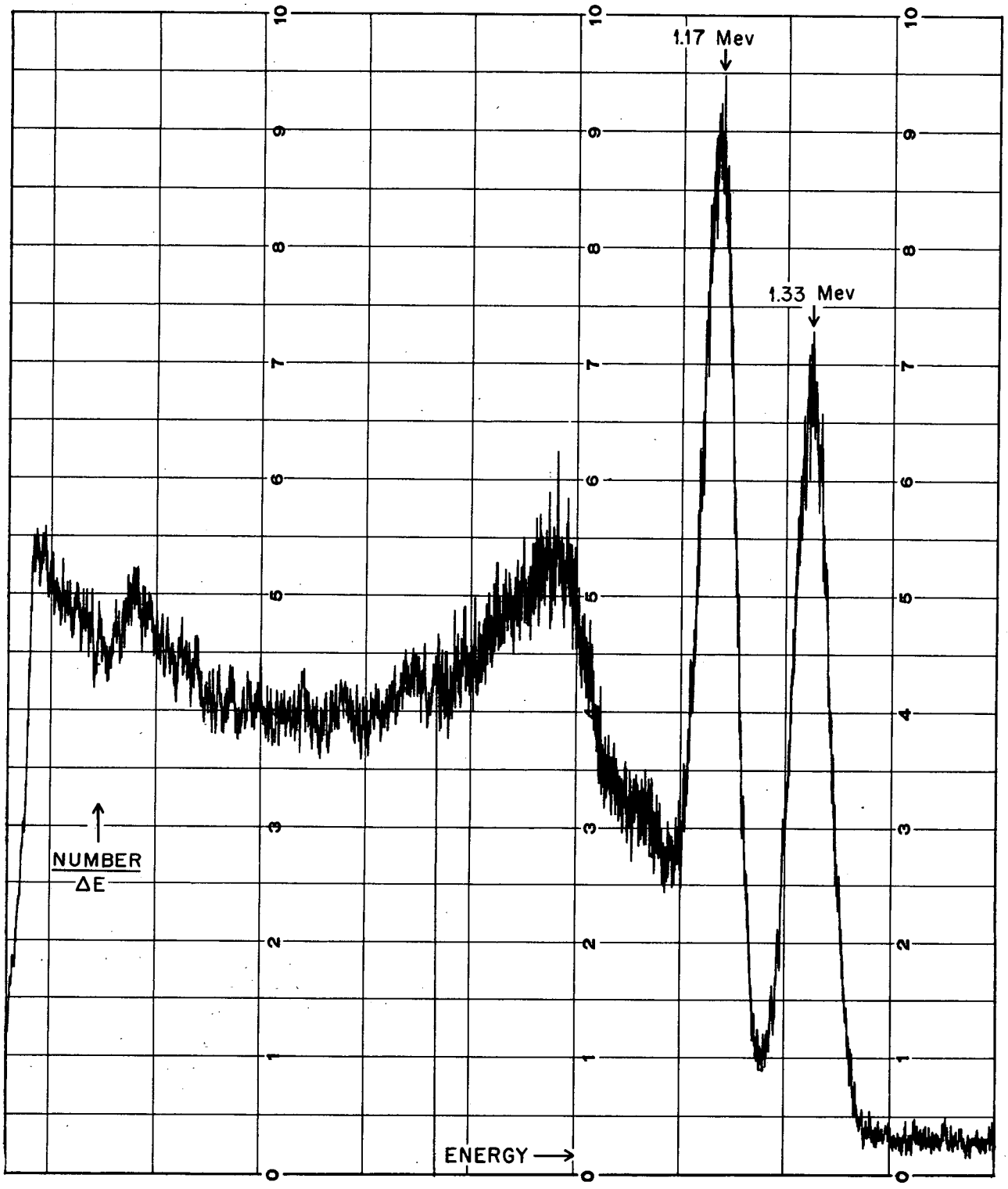

Fig. 5. Resolution of Two Co^{60} Gamma Rays Obtained with a Type-5819 Photomultiplier and a 1 1/2- by 1 1/2-in. Sodium Iodide Crystal.

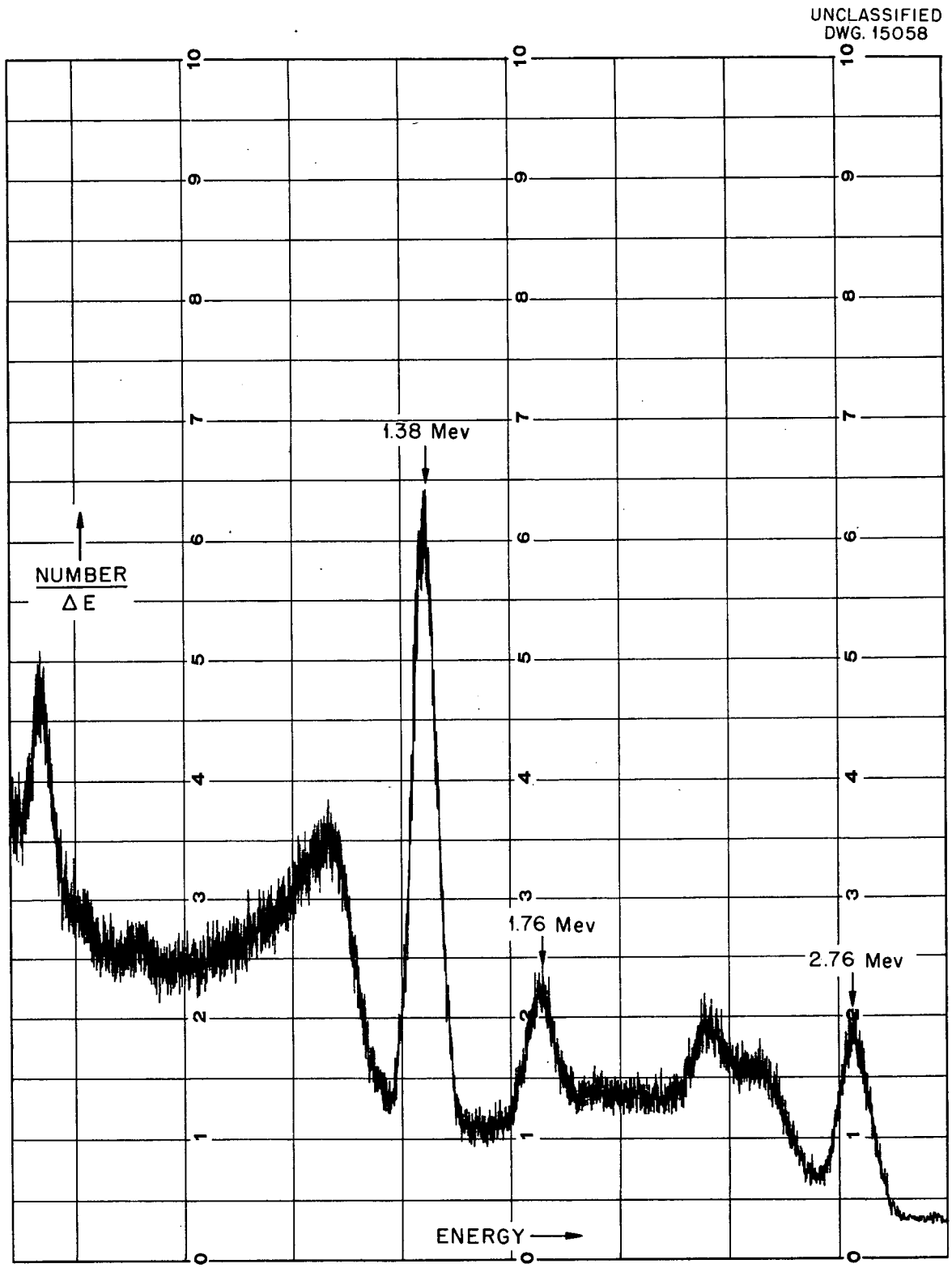

Fig. 6. Resolution of Na^{24} Gamma Rays Obtained with a Type-5819 Photomultiplier and a 1- by 1 1/2-in. Sodium Iodide Crystal.

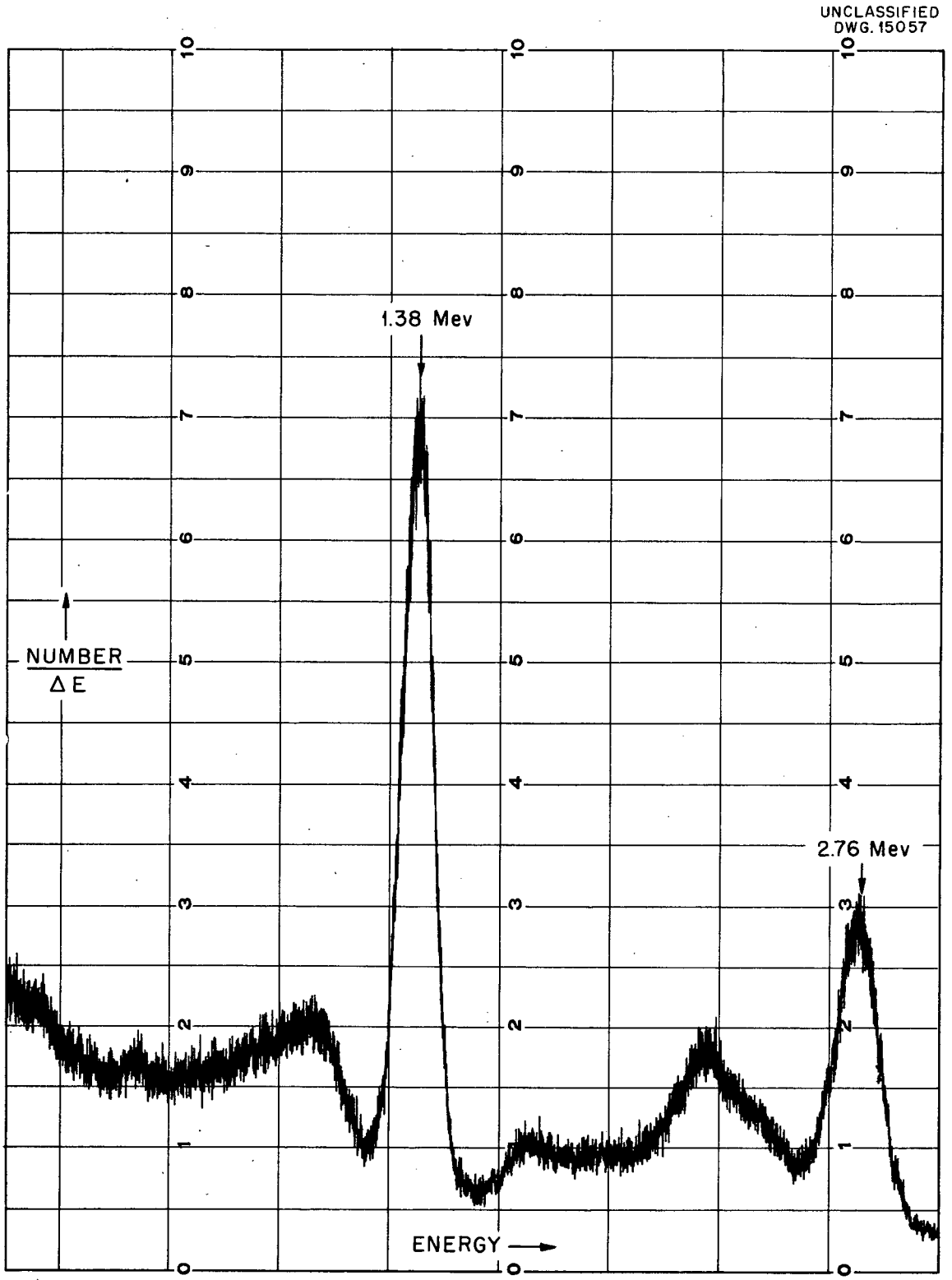

Fig. 7. Resolution of Na^{24} Gamma Rays Obtained with a Type-5819 Photomultiplier and a 3- by 3-in. Sodium Iodide Crystal.

were obtained by using a 1- by 1 1/2-in. and a 3- by 3-in. sodium iodide crystal. Note the absence of an "annihilation escape peak" for the larger crystal and the relatively small Compton recoil contribution. No collimation of the source was used in any of these experiments.

As shown by Morton,[1] the number of electrons actually entering the multiplier can be determined by extrapolating the integral pulse-height distribution curve to zero pulse height. Such a curve was obtained for a type-5819 photomultiplier tube that had a photocathode efficiency of 60 μa per lumen. To ensure that the pulses being counted originated at the photocathode and not at the first or second dynode, a small amount of light was directed on the photocathode. The dark-current pulse background was deducted from the illuminated photocathode pulse distribution to give the curve shown in Fig. 8. The pulse height in volts that correspond to a single photoelectron is found by taking the area under the distribution curve and dividing it by the number of photoelectrons entering the photomultiplier. For these particular conditions, a pulse height of 10.4 v corresponds to a single average electron. Using a 41.6-kev x-ray source and the sodium iodide crystal mounting shown in Fig. 1, an electron energy loss of 200 ev in the crystal releases a single photoelectron from the photocathode of this particular tube.

Figure 9 shows the dark-current pulse background as a function of electron energy loss in the crystal. It can be seen that even at 2 kv a background of only 34 cpm is obtained at room temperature for a good tube.

[1] G. A. Morton, *RCA Review*, p. 525, December 1949.

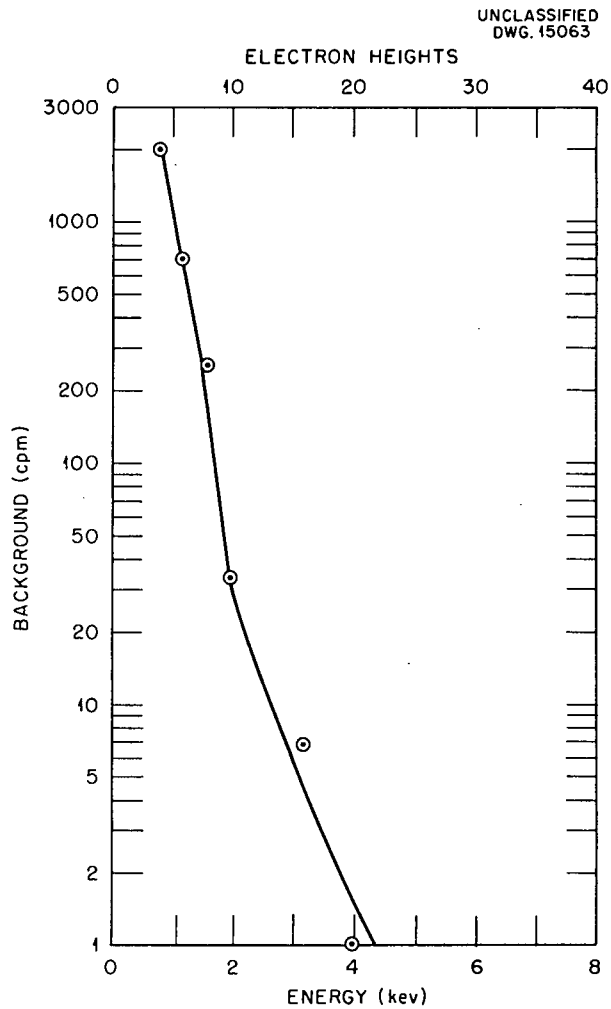

Fig. 8. Dark-Current Pulse Background for Type-5819 Photomultiplier at Room Temperature.

Variation in Energy Resolution of Photomultipliers. Table 2 shows the variation in energy resolution for 50 photomultiplier tubes (type 5819).

In general, tubes with the highest photocathode efficiency gave the best energy resolution. However, there were exceptions to this rule. In many of the exceptions the photocathode was visually nonuniform. In all the tests

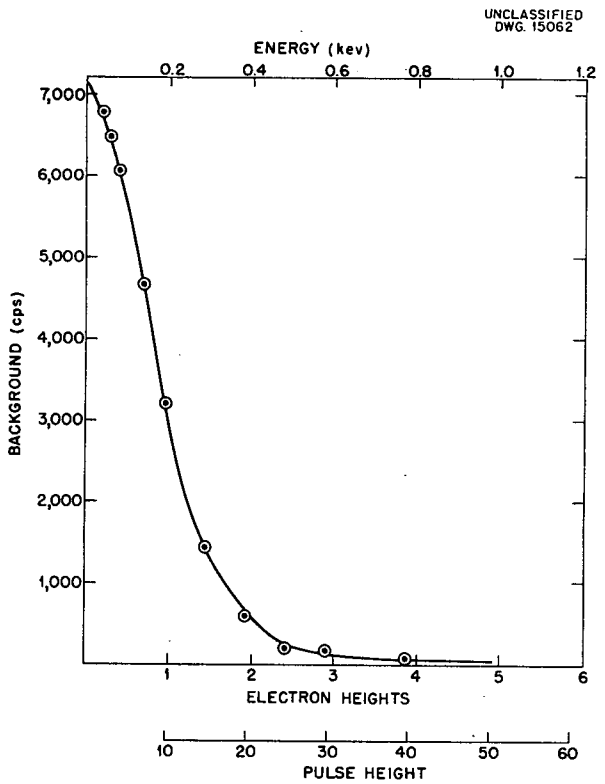

Fig. 9. Pulse-height Distribution of Photoelectrons from Illuminated Photocathode of the Type-5819 Photomultiplier.

a 5-1-1-etc., photocathode to first dynode to second dynode voltage ratio was used. The over-all voltage was 600 to 700 volts.

Variation in Harshaw Sodium Iodide Crystals. Table 3 indicates the variation in pulse size and energy resolution of 1 1/2- by 1-in., cylindrical, sodium iodide crystals.

TABLE 2

Variation in Energy Resolution for 50 Photomultiplier Tubes

FULL WIDTH AT HALF MAXIMUM FOR Cs^{137} PHOTO PEAK (%)	PERCENTAGE OF TUBES
7 to 8	11
8 to 9	23
9 to 10	18
10 to 12	26
over 12	22

Variations in energy resolution and pulse size for the various crystals is primarily due to variations in thallium iodide content and foreign inclusions in the crystal that absorb light. The crystals that showed no slight yellowish discoloration, which indicated a low thallium iodide content, invariably gave poor resolution and low light output.

A single-channel sweep type of differential discriminator designed by Fairstein and described in ORNL-1036[2] was used for all pulse height distributions shown.

The author wishes to express his thanks to R. L. Clark for the preparation and mounting of the crystals and his help in obtaining much of the counting data.

[2] *Chemistry Division Quarterly Progress Report for Period Ending December 31, 1950*, ORNL-1036.

TABLE 3

Variation in Pulse Size and Energy Resolution of 1 1/2- by 1-in., Cylindrical, Sodium Iodide Crystals

CRYSTAL NO.	FULL WIDTH AT HALF MAXIMUM FOR Cs^{137} PHOTO PEAK (%)	PEAK-TO-VALLEY RATIO	RELATIVE PULSE SIZE
1	7.6	52:1	82
2	7.8	46:1	95
3	8.0	35:1	80
4	8.0	45:1	85
5	8.3	42:1	89
6	8.4	37:1	90
7	9.0	37:1	78
8	9.1	40:1	78
9	9.5	28:1	53
10	9.7	38:1	72
11	9.7	29:1	83
12	11.7	32:1	84
13	13.2	16:1	61
14	14.0	13:1	64

INSTRUMENT DEPARTMENT

HIGH-VOLTAGE ACCELERATOR PROGRAM

R. F. King, R. W. Bennett, A. W. Prichard, H. Reese, J. P. Judish, H. E. Banta

Additional stabilization of the terminal voltage of the Cockroft-Walton generator has been achieved. A diode-limiter lens system for the No. 2 accelerator position has been installed and initial consideration given to an electron accelerator for the Health Physics Division.

A new accelerator tube for the 2-Mev Van de Graaff generator has been constructed. Automatic valves to shut off the system in the event of a vacuum failure have been installed on the No. 2 accelerator.

Control instrumentation for the 2-Mev machine was built up in final form and is ready for installation in the new High-Voltage Building. This work had to be carried on without disrupting the work of the machine, since it was functioning as a proton accelerator.

Zirconium deuteride targets have been prepared. This work exactly parallels the zirconium tritide target preparation, except that separate take-up systems must be used.

SERVICE WASTE-WATER MONITORING SYSTEM

I. D. Groves, B. C. Behr

The service waste-water monitoring system was designed to continuously

monitor a waste-water disposal system to provide permanent records of the flow-rate and water contamination. The system will detect beta activity in water in which the concentration of certain radioactive substances is less than the permissible tolerance. The monitor records activity level and simultaneously sounds an alarm when the activity rate reaches a predetermined level. The water flow is measured and recorded, and a sample is removed for chemical analysis. The monitor gives a total integration of the activity of the waste water plus a record of the activity at any given time.

To compensate for background radiation, the system contains two monitoring cells that operate concomitantly: one counts radiation from waste water and the other counts background radiation while being purged with tap water. The pulses from both counting tubes are fed into differential electronic counting-rate meter circuits; this automatically cancels the background count and leaves only the activity rate from the waste water.

Owing to the intrinsic geometry of the water cell, high counting efficiency is obtained without contamination of the counter tubes. Water is continuously injected into the cell tangentially to its inner surface. Thus, the stream is deflected into a circular path around the inner wall and out the bottom of the cell. When equilibrium is reached inside the cell, a cylindrical wall of water is formed that has an inside diameter just large enough to permit the insertion of a Geiger-Mueller tube, which remains dry and free from contamination.

The recording of information obtained from the service waste-water monitoring system is achieved by the use of weir stations for measuring and recording the water flow, a Brown Electronik potentiometer for recording the activity level, and a watt-hour meter to indicate the total activity. A signal is picked up from the weir station that is proportional to waste-water flow. This signal is fed to counting-rate recorders and modified in accordance with the instantaneous counting rate, and as a result a signal proportional to the product of the instantaneous counting rate and waste-water flow is obtained. The time integral of this function is determined by using the watt-hour meter. The total activity that passes the weir station is indicated by the reading on the watt-hour meter.

CONCENTRATION-MEASUREMENT INSTRUMENT, Q-1231

B. M. Hildebrant

A concentration-measurement instrument, Q-1231, has been developed for recording the relative concentration of a liquid composed of two nonconducting constituents of widely varying dielectric constant. The design of the device is based on the principle of liquid flowing through a cell that serves as a frequency-determining capacitor in a stable oscillator. The oscillator output is fed to a limiter and discriminator. The output of the discriminator is linear with respect to relative concentration of the liquid over a limited region. Model Q-1282, which is a more simple version of the instrument, has also been designed and constructed for making similar measurements on laboratory sample quantities. It is believed that this device may be made into a null-balance instrument for other applications.

"WALSH" GENERAL-PURPOSE SOURCE UNIT

K. H. Kline

The "Walsh" general-purpose source unit was built specifically to supply power to produce a pulsed spark for use in spectrographic analysis. The pulsed spark is produced by charging a bank of condensers to approximately 250 v and then discharging them across an analytical gap. A high-voltage trigger circuit serves to ionize the atmosphere surrounding the gap and thereby provide a conducting medium. The sparks occur in uniformly spaced pulses. The energy of the spark is adjustable by changing the value of the storage condensers. The number of discharges per unit time is adjustable by increasing or decreasing the control gap of the instrument. Full-wave or half-wave operation may be obtained by switching either one or both rectifiers into the circuit, as the occasion demands.

HIGH-MEGOHM BRIDGE AND STANDARD RESISTOR, Q-1196-1

F. M. Glass

The high-megohm bridge and standard resistor, Q-1196-1, accurately measures resistance values from 10^7 to 10^{14} ohms. The device is accurate to 0.5% for measurements up to 10^{12} ohms.

The standard resistor assembly used as a calibration reference in measuring resistances is composed of 200 five-megohm, precision resistors wound with Evan-Ohm resistance wire. The standard resistor assembly is sealed in an aluminum cylindrical chamber containing an atmosphere of nitrogen. Fluorothene insulation is used for mounting the resistor assembly and terminals of the resistor. The accuracy of the resistor assembly is 0.1% or better. Resistance measurements are obtained by a null-balance indication employing a vacuum-tube electrometer circuit. The variable element in the bridge is a 15-turn helical potentiometer of 0.05% linearity. The bridge contains 16 combinations; provision is made to calibrate each combination individually.

DELAYED-PULSE GENERATOR FOR THE NEUTRON VELOCITY SPECTROMETER

W. G. James D. D. Walker

A delayed-pulse generator has been designed for testing the neutron velocity spectrometer. The generator will deliver a delayed pulse upon receiving a triggering pulse; the delay of the pulse is continuously variable from 4 to 100 microseconds. A Schmitt trigger circuit was designed into the input of the generator to accept a positive pulse. The output of the circuit triggers the Phantastron circuit by means of a pentagrid converter tube. The linearly decreasing output voltage of the Phantastron is then fed into a biased diode. This causes the diode to conduct at a given time, t, which is determined by the setting of a helipot connected in the plate circuit of the diode section. The pulse from the diode is reshaped in another Schmitt trigger circuit and transmitted to a blocking oscillator that turns out a fast, sharp pulse. The maximum delay time of the delayed pulse is 150 microseconds.

HRE-REACTOR SIMULATOR, Q-1063

D. M. Collier J. P. Palmer
L. A. Meeks

The kinetic equations that express reactor power and temperature as functions of time for various perturbations have been derived and coefficients appropriate to the HRE have been evaluated. The equations constitute a system of eight simultaneous nonlinear differential equations that are not readily solvable by any of the ordinary analytical methods. Because

of this problem, an electronic analog computer, the HRE-reactor simulator, Q-1063, has been developed to solve the equations for various conditions of operation. Since the computer is to be used for training operating personnel as well as for experimental purposes, it has been necessary to design the machine without expansion of the time scale. The computer thus follows the same time periods encountered in operating the reactor itself.

Essential components of the computer include a stable, high-gain, d-c amplifier and an electronic multiplier. For the sake of reliable, drift-free operation a differential system was chosen at a rather early stage in the development of the instrument. An amplifier has been developed to give a maximum output signal of 100 v, to have a differential drift of less than 1 mv referred to the input, and to have a gain of about 20,000. A reliable electronic multiplier was designed with one variable input to cover the range from 1 to 100 v, whereas the other has a usable range of 5 to 75 volts. The input variables can change at any rate up to 100 cps without causing more than 2% error in the product. The output is linear to within 5% for a range of 0.06 to 20 volts.

The instrument has been completed and is operating satisfactorily. The last few weeks have been spent obtaining solutions for numerous cases corresponding to extremes in operating conditions. The solutions have been quite accurate in every case in which checks are possible. Many cases that could not be attempted with a reactor have been explored. A report containing detailed information relative to the computer and the solutions obtained will appear in the near future.

TRIPLE-COINCIDENCE PROTON-RECOIL COUNTER

R. E. Zedler

A triple-coincidence proportional proton-recoil counter has been designed and assembled that is the culmination of an effort in which several experimental counters were built and tested. This counter is used to determine the neutron energy spectrum from 1.2 to 11 Mev. Recoil protons from an external hydrogenous radiator enter the counter assembly through a mica window approximately 1.5 mg/cm^2 in thickness. Each successive counter tube from the mica window is larger in diameter than the preceding one. The counter tubes are filled with krypton to a pressure of 10-cm Hg.

SCINTILLATION COUNTER SPECTROMETER

D. J. Knowles

Equipment to be used as a scintillation counter spectrometer has been constructed. The various components are: an A-1 linear amplifier with a differential pulse-height discriminator, a counting-rate meter, a high-voltage supply, a Brown recorder, and a detector assembly. In addition, a dry box has been built for use in mounting sodium iodide crystals. This equipment will be used to select crystals and photomultiplier tubes for good resolution and good peak-to-valley ratio. The equipment has been set up as a facility for those in the laboratory who need to select tubes and crystals for scintillation spectrometers.