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IMPROVED ALPHA SPECTROSCOPY USING A MODIFIED FRISCH GRID IONIZATION CHAMBER AND A 256-CHANNEL ANALYZER

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IMPROVED ALPHA SPECTROSCOPY USING A MODIFIED FRISCH GRID IONIZATION CHAMBER AND A 256-CHANNEL ANALYZER

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ABSTRACT

Adaptation of a Frisch Grid alpha ionization chamber and its preamplifier to an Argonne-type 256-channel pulse height analyzer for alpha spectroscopy is described. Overall resolution of better than 1% is achieved. Circuit modifications to the commercial preamplifier are given, and a procedure for electrodeposition of thin samples is described.

I. INTRODUCTION

This laboratory has been engaged in precision alpha spectroscopy in the 5-Mev range, using a 256-channel pulse height analyzer which has a resolution limit of about 0.4%. Overall resolutions of isotope analysis in the range of 1 to 5%, which were being achieved, prompted examination of the causes of loss of resolution. Good resolution is very important in areas such a burnup studies in which quantitative measurements are made. Wide "skirts" of a predominating isotope, due to poor resolution, tend to mask out other isotopes of energies close to the predominating one, making differential concentration computations difficult.

II. RESOLUTION ASPECTS IN ALPHA SPECTROSCOPY

Resolution in spectroscopy is defined herein as follows: given a mono-energetic alpha emitter, the resolution of energy measurement is the ratio of the width of the measured energy distribution curve to the energy at the peak of the curve. The width is taken at the points of the curve which are one-half the peak value of the curve.

The following are the major causes contributing to imperfect resolution in alpha spectroscopy using ionization and pulse height analysis techniques:

a) Statistical variation in the energy of the alpha particles escaping the sample,

b) Statistical variation in the ion pair production in the ionization chamber,

c) Imperfect resolution in the pulse height analyzer.

The variations due to the statistical effects are directly additive. The third effect is not strictly statistical, so that its effect is not directly additive.

The first effect is due to self-absorption in the sample, and is minimized by preparing very thin samples. Statistical variation in the ion pair production is minimized by use of gas with the lowest ionization potential. The resolution of the pulse height analyzer is limited by the number of channels in the analyzer; the larger the number of channels, the better the resolution. The resolution is also adversely affected by drift of channels and imperfect pulse handling in the analyzer.

III. ION CHAMBER RESOLUTION

The ionization chamber delivers pulses which have a Poisson distribution. The resolution of this distribution is given by Equation 6 (Appendix):

$$\text{Res.} = \frac{(8 \ln 2)^{1/2}}{m}$$

where:

$$m = \frac{E}{I}$$

E is the energy of the alpha particle in electron-volts and I is the energy expended per ion pair created, in the same units. The energy per ion pair for argon gas is 27 ev.

This distribution is a statistical property of the chamber and is determined by the ionization potential of the chamber gas. For a 5.3-Mev alpha particle and argon gas, the resolution is 0.53%.

IV. PULSE ANALYSIS CIRCUITRY

A block diagram of the ionization chamber and the pulse handling circuit is shown in Figure 1. This system was examined in detail and modified to achieve the best resolution obtainable from the pulse height analyzer.

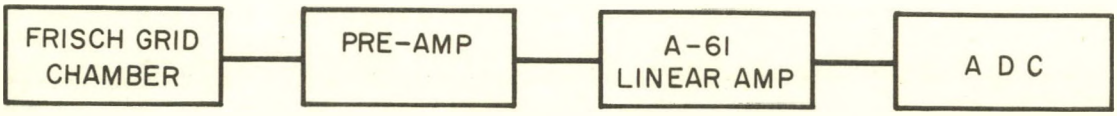


Figure 1. Pulse Circuit

The ionization chamber used is a Tracerlab RLD-1 Frisch Grid chamber[1] with an electron collection time of about 1 μsec. To preserve the linearity of the chamber, the time constant of the effective chamber capacity and the load resistor must be long, compared with the collection time. The proper load resistance value was selected by increasing the resistance until further increase did not increase the amplitude of the output pulse. For this chamber, the value was 9.5×10^{11} ohms.

The chamber is followed by a preamplifier, designed by Tracerlab for use with the RLD-1 chamber. This unit is composed of two feedback-stabilized linear amplifiers. The output of the first amplifier is matched tc a shorted delay line pulse shaper which clips away the trailing portion of the input pulse.[2] The pulse output of the preamplifier is further amplified by an RCL A-61 Linear

Amplifier and delivered to the analog-to-digital converter (ADC) in the analyzer. RCL states that rise times shorter than 0.6 μsec cause amplitude distortion. The electron collection time of 1 μsec in the ionization chamber avoids this difficulty.

The ADC digitizes the input pulse by:[3]

a) Charging a storage capacitor through a diode in a pulse stretcher,

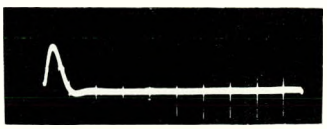
b) Starting a linear ramp generator and a 2-Mc oscillator when the storage capacitor is fully charged by the input pulse,

c) Stopping the oscillator when the ramp signal equals the voltage on the storage capacitor,

d) Storing a count in the channel which corresponds to the number of cycles generated by the 2-Mc oscillator during the ramp interval. The greater the input pulse amplitude, the greater will be the time required for the ramp signal to reach the voltage on the storage capacitor. Hence, the greater will be the number of cycles of the 2-Mc signal.

a. 20-μsec Pulse Delivered by Standard Preamplifier

b. 4-μsec Pulse Delivered by Preamplifier Using 2-μsec Delay Line



c. 2-μsec Pulse Delivered by Preamplifier Using 1-μsec Delay Line

Figure 2. Preamplifier Output Pulses with Various Amounts of Delay Line Clipping

The 2-Mc oscillator and the ramp generator are triggered when the charging pulse's voltage drops slightly below the voltage on the storage capacitor. If the charging pulse decays slowly, it degrades the accuracy and resolution of the ADC by allowing discharge of the storage capacitor through the only slightly reverse-biased charging diode.

An abruptly decaying charging pulse was provided the ADC by reducing the time delay of the delay line shaper in the preamplifier. A single section shorted line with L = 1 mh and C = 0.001 μfd gave a total time delay of about 1 μsec and provided a sharply falling pulse, 2-μsec wide, without peak amplitude clipping. Figure 2 shows waveforms with several amounts of time clipping. Figure 3 shows the revision to the preamplifier. It should be noted that excessive pulse clipping will result in severe amplitude distortion.

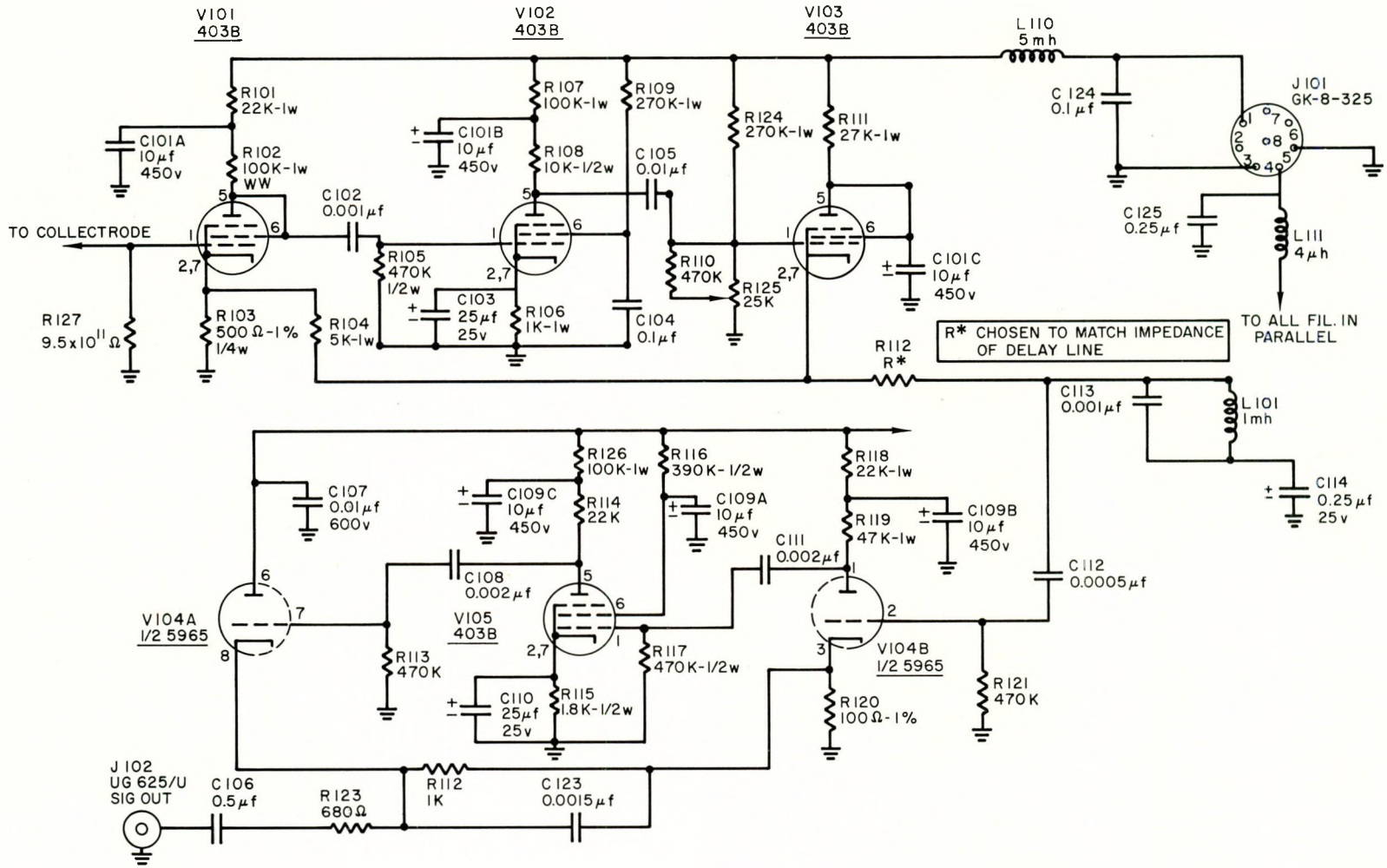


Figure 3. Tracerlab Preamplifier, as Modified

V. SAMPLE PREPARATION

The disc used in the electrodeposition was polished platinum having a mirror finish. The electroplating cell used is shown in Figure 4, and is the cell suggested by Mitchell.[4]

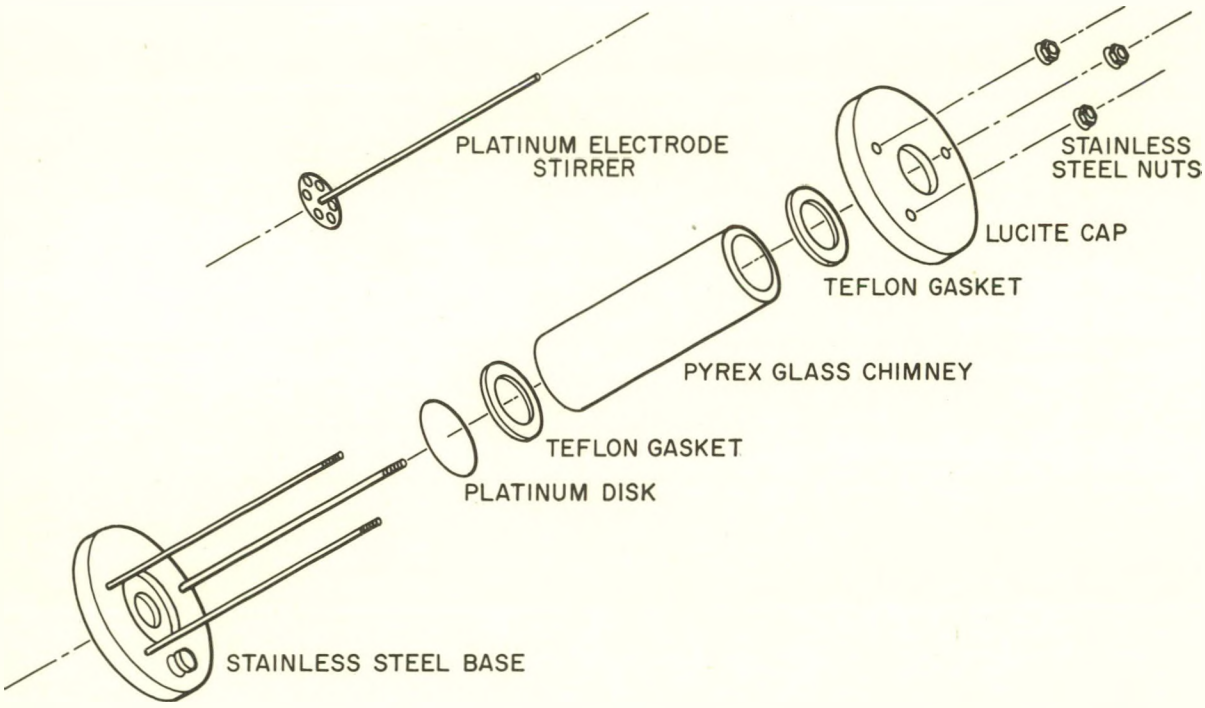


Figure 4. Electroplating Cell Assembly

The method used in preparing samples for the alpha spectrometer, to meet the requirements for thin, uniform sources, was to electrodeposit the alpha-emitting elements from an electrolyte of ammonium chloride and hydrochloric acid, as used by Mitchell.[4] In this method, the sample is evaporated to dryness several times with HCl and the residue finally taken up to 1 ml of concentrated HCl. This solution is transferred to the plating cell with two, 1-ml washes of concentrated HCl, and 1 drop of methyl red indicator is added. Ammonium hydroxide is added until the end point is reached, and the solution is brought back to the acid side with 2N HCl. The plating volume and chloride ion concentration at this point is that required for electrodeposition. The sample is

now electroplated for 30 min (aliquots less than 1 μgm) with a current density of 0.6 to 1.0 amp/cm^2 and with a stirrer speed of 60 rpm. Before stopping the deposition, the electrolyte is made ammoniacal with concentrated NH_4OH, and the current is stopped. The electrolyte is decanted and the disc is flamed to a dull red.

VI. EXPERIMENTAL RESULTS

Before modifying the preamplifier, an electroplated standard source of Th^{230}, Pu^{239}, and Am^{241} yielded a spectrum having a resolution of 1.2%.

After the electronic modification of the preamplifier, the same source yielded the spectra shown in Figures 5 and 6. In Figure 5, the equipment counted for 1000 sec and produced a spectrum with a resolution of 0.77%. In Figure 6, the equipment counted for 50,600 sec (overnight). Drift in the equipment probably caused the slightly increased resolution (0.81%).

An electroplated standard source of Po^{210}, when counted for 200 sec, yielded the spectrum shown in Figure 7, which has a resolution of 0.72%. This source had the highest specific activity of any sample measured.

Evaporated samples were then analyzed, but the results were very poor. The resolution obtained with such samples was about 5%, due to the existence of thick spots in otherwise thin sources. It was then decided to use electroplated samples, using the procedure described above.

A thick source was prepared, containing 100 μgm of uranium and 140 dpm of Pu^{239}. The Pu^{239} was added as a tracer to verify that all the radioactive material was plated out. The alpha spectrum of this source is shown in Figure 8. It has a resolution of 1.05%. The effect of source thickness is shown in the low energy tails on each of the alpha peaks. Even with this source thickness, the U^{235} peak shows clearly above the background.

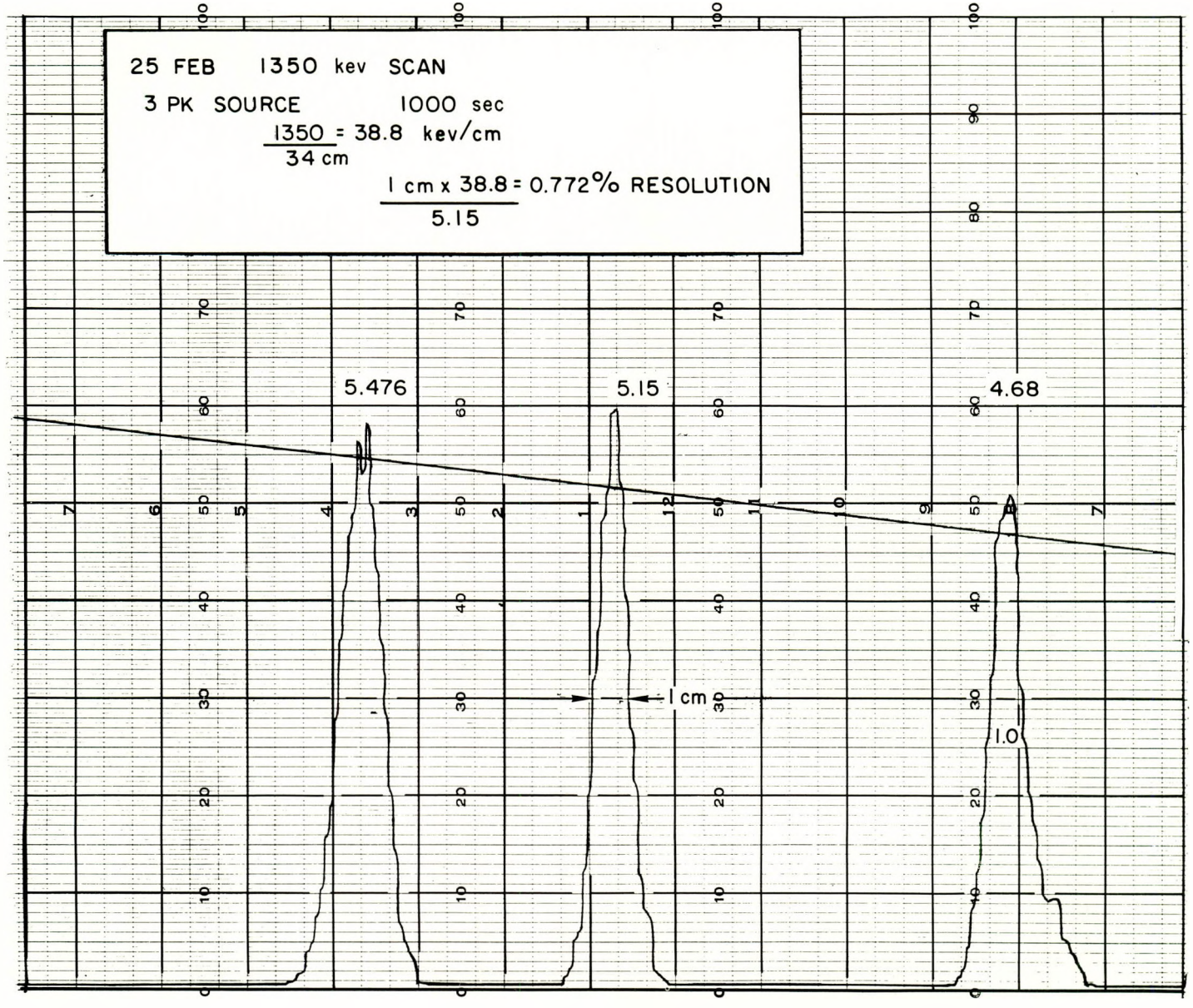


Figure 5. Plot of 1000-sec Count of Th^{230}, Pu^{239}, and Am^{241}, Showing Resolution of 0.77%

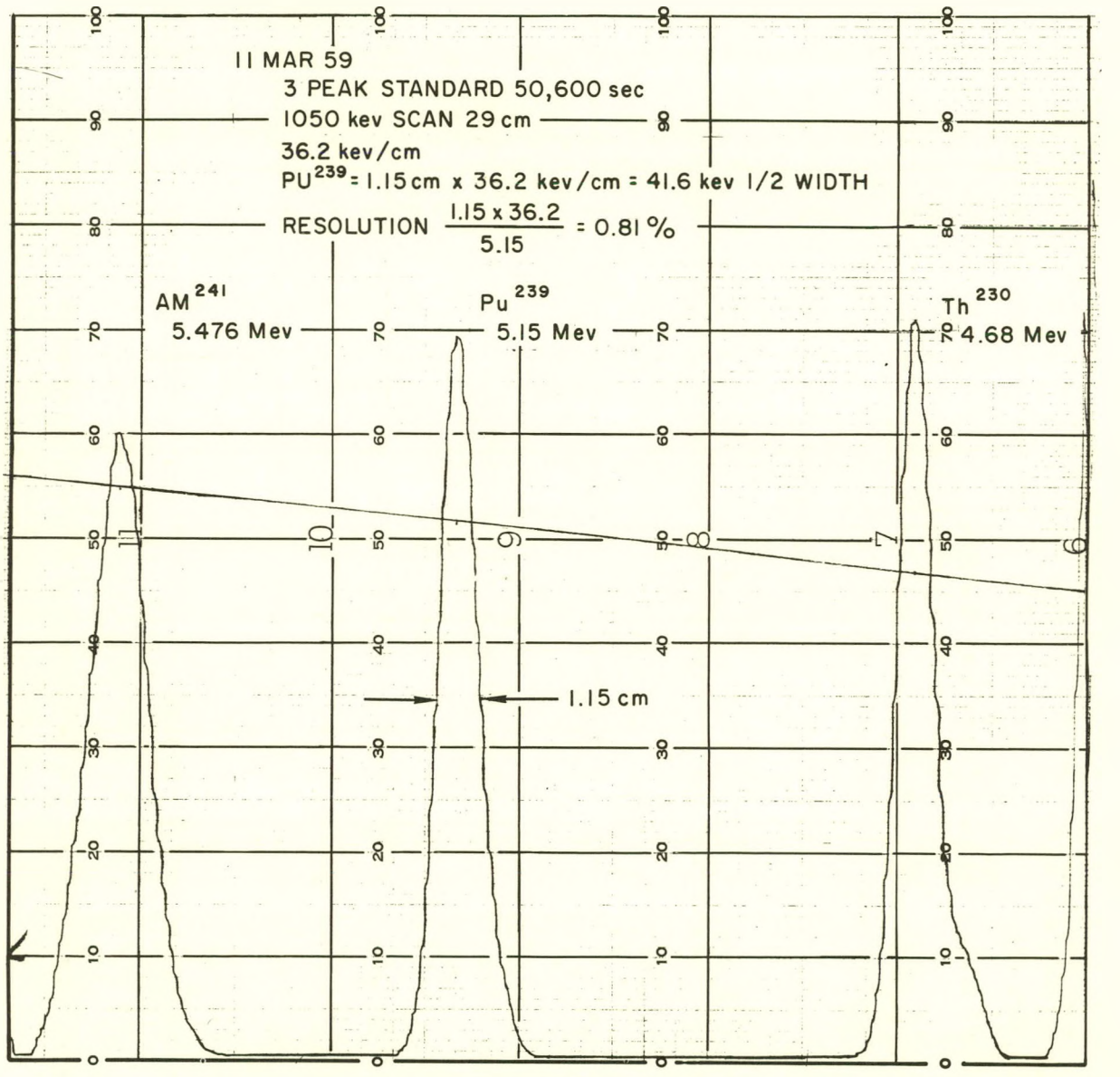


Figure 6. Plot of 50,600-sec Count of Th^{230}, Pu^{239}, and Am^{241}, Showing Resolution of 0.81%

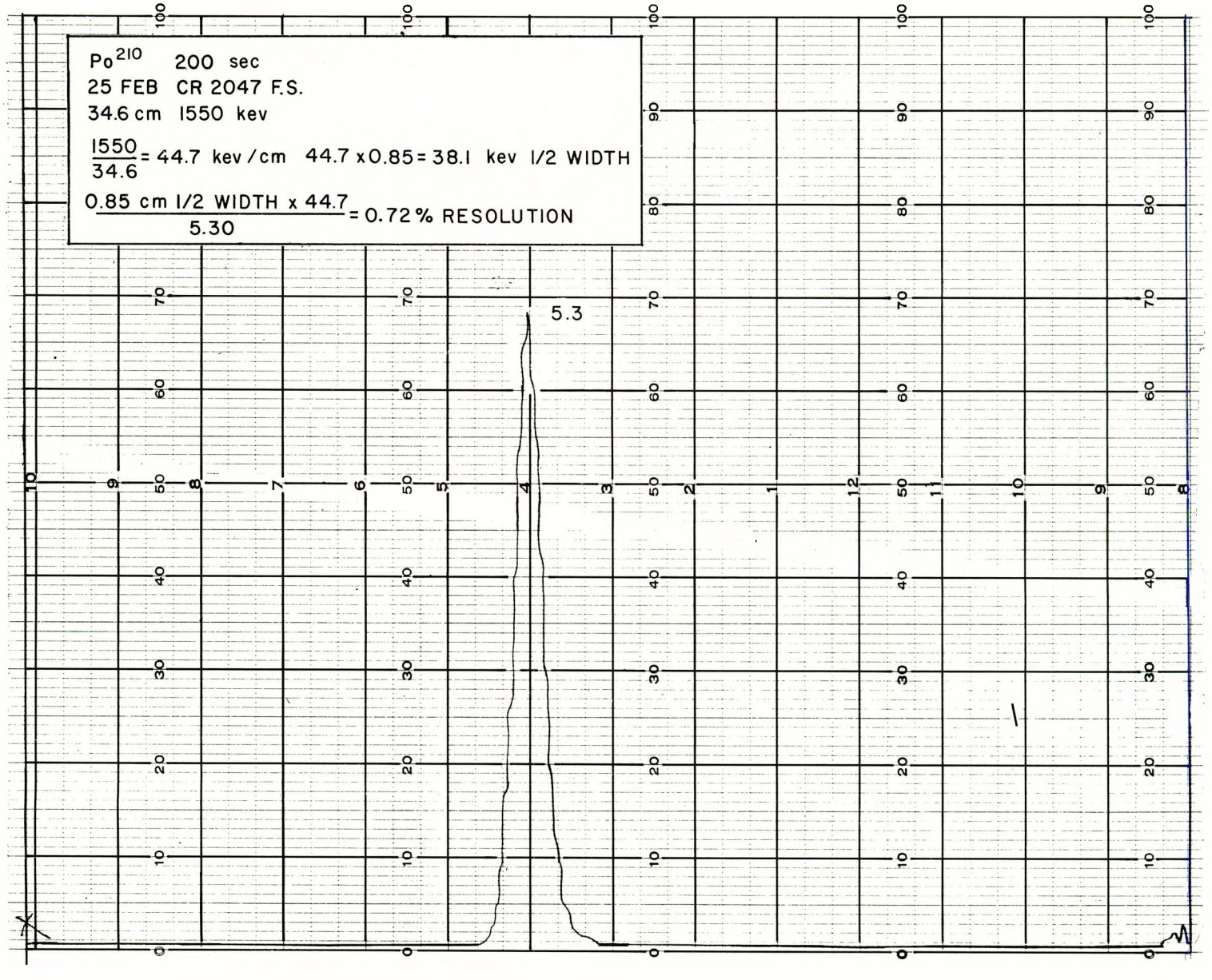


Figure 7. Plot of 200-sec Count of Po^{210}, Showing Resolution of 0.72%

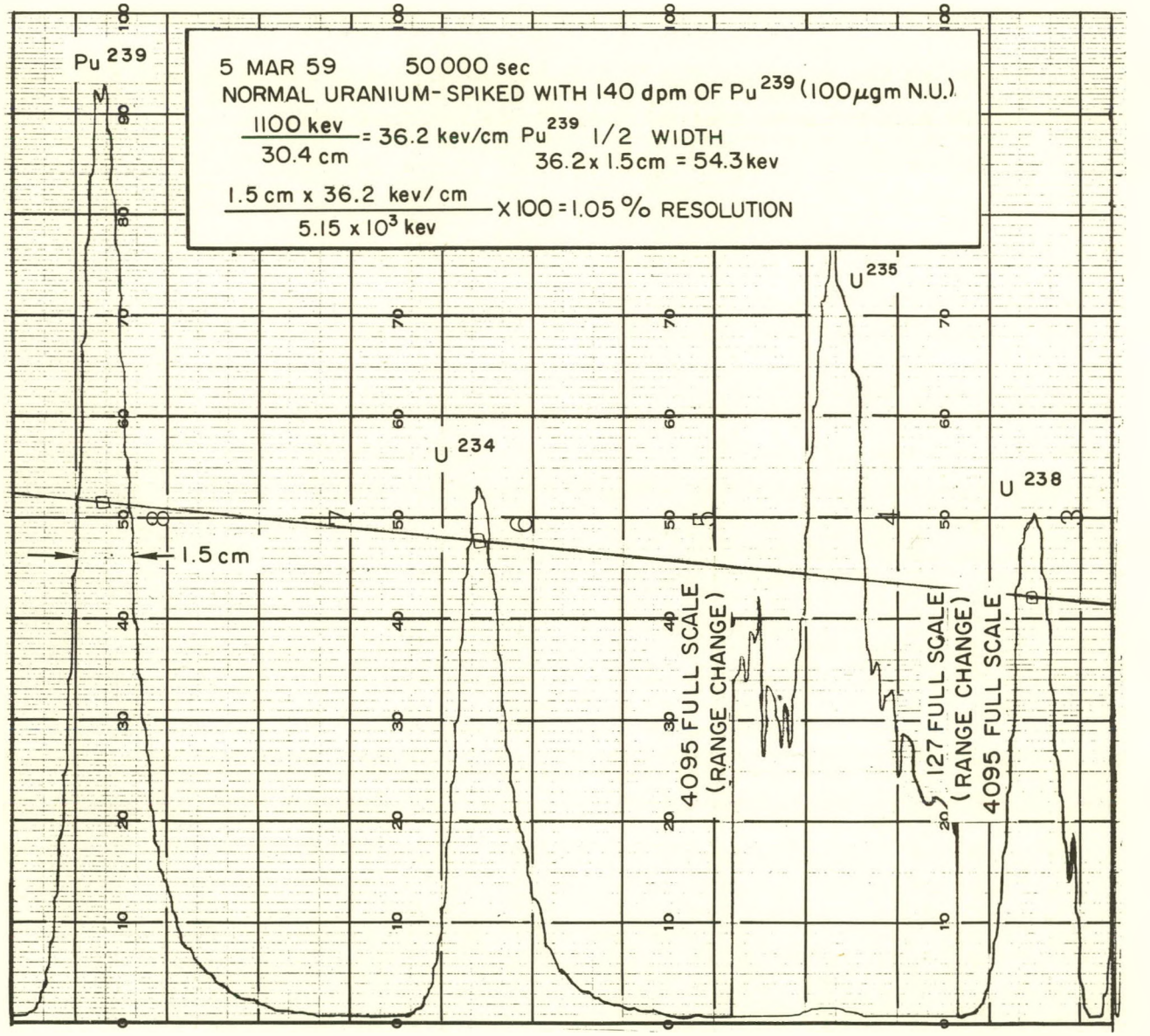
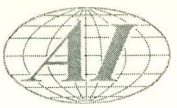


Figure 8. Alpha Spectrum of 100 μgm of Uranium and 140 dpm of Pu^{239}, Showing Resolution of 1.05%



VII. DISCUSSION OF OVERALL RESOLUTION

The best resolution obtained in the analyses described was 0.72%, which was for a 200-sec count of Po^{210}. The ion chamber distribution curve alone has a resolution of 0.537%. Perfect resolution in the pulse height analyzer would therefore improve the overall system resolution by less than a factor of 2.

To minimize the variance due to channel drift, the counting should run only long enough to achieve good statistics from the ion chamber.

APPENDIX

ION CHAMBER RESOLUTION

The ionization process in the ionization chamber produces pulses which have a Poisson distribution which is given by

$$P_{(x)} = \frac{m^x e^{-m}}{x!} \quad . \qquad \ldots(1)$$

The resolution of this expression is not easily determined directly. However, it can be shown (by comparing first and second moments) that, for large numbers of events, the Poisson distribution approaches a special case of the normal distribution. The normal distribution in general is

$$P_{(x)} = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(m-x)^2}{2\sigma^2}\right] \quad . \qquad \ldots(2)$$

The special case equivalent to the Poisson distribution occurs when $\sigma = (m)^{1/2}$. Making this substitution in Equation 2, we have, as a valid expression for the ionization chamber's pulse height distribution, the following:

$$P_{(x)} = \frac{1}{\sqrt{2\pi m}} \exp\left[-\frac{(m-x)^2}{2m}\right] \quad . \qquad \ldots(3)$$

Since we are looking for the width of the curve at the probability $P_{(x)} = 1/2\ P_{(m)}$, we may write

$$\frac{P_{(x)}}{P_{(m)}} = \frac{1}{2} = \exp\left[-\frac{(m-x)^2}{2m}\right] \qquad \ldots(4)$$

or

$$(2m \cdot \ln 2)^{1/2} = \left| x - m \right| \quad .$$

The width of the curve at $P_{(x)} = \frac{1}{2} P_{(m)}$ is

$$W = 2 \left| x - m \right| = 2(2m \cdot \ln 2)^{1/2}, \qquad \ldots (5)$$

so that for resolution we may write

$$\text{Res} = \frac{W}{m} = \frac{2(2m \cdot \ln 2)^{1/2}}{m} = \frac{(8 \ln 2)^{1/2}}{m} \qquad \ldots (6)$$

The mean pulse height delivered by an alpha ionization chamber is proportional to the mean number of ionizations caused by an alpha particle. Given monoenergetic alpha particles of 5.3 Mev and argon gas of 27-ev mean ionization energy, the mean number of ionizations per alpha particle is

$$m = \frac{5.3 \times 10^6}{27} = 1.96 \times 10^5 \quad . \qquad \ldots (7)$$

Inserting this number in Equation 6, we have

$$\text{Res} = 0.53\% \quad . \qquad \ldots (8)$$

REFERENCES

1. B. B. Rossi and H. H. Staub, Ionization Chambers and Counters, National Nuclear Energy Series, Manhattan Project Technical Section, Div. 5, Vol. 2 (New York, McGraw-Hill Book Co., 1949) p 39, 123

2. W. C. Elmore and M. Sands, Electronics, National Nuclear Energy Series, Manhattan Project Technical Section, Div. 5, Vol. 2 (New York, McGraw-Hill Book Co., 1949) p 43

3. Radiation Counter Laboratories, Manual of Instruction, 256 Channel Analyzer (Skokie, Illinois)

4. R. F. Mitchell, "Electrodeposition of Actinide Elements at Tracer Concentration," (private communication to T. B. Crockett, 1958)

5. R. D. Evans, The Atomic Nucleus (New York, McGraw-Hill Book Co., 1955) p 750