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A 48-CHANNEL PULSE HEIGHT ANALYZER FOR ALPHA ENERGY MEASUREMENTS

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1. Introduction

In the program of investigation of new artificial heavy nuclides* carried on at this laboratory, it was found almost essential to be able to identify and trace specific alpha-emitters in the presence of other alpha-emitters. Because many of the nuclides at the upper end of the periodic table are alpha-emitters, the bombardment of a particular starting material (generally an alpha-emitter) with neutrons, deuterons or helium ions usually resulted in the formation of new nuclides which were also alpha-emitters. The products formed were often present in very low concentrations (with alpha activities much lower than that of the starting material), and the chemical properties of the element to be separated were often poorly understood. In either case, a good analytical tool was necessary to make possible the tracing of a particular nuclide through a long series of chemical separations. In addition there were a number of cases where the nuclides formed were not chemically separable, being isotopic (e.g., a mixture of Pu^{236}, Pu^{238} and Pu^{239}), so that chemical identification was impossible. The instrument described here was developed for use in such tracing, and has also proved

*Nuclide. "A species of atom characterized by the constitution of its nucleus, in particular by the numbers of protons and neutrons in its nucleus."(1)

useful in following the chemical separation and purification of the natural alpha-emitters. It has also been used for the determination of the alpha energies of many of the new isotopes. Although the accuracy of the present instrument is less than that of other instruments, the fact that it could be used at 50% geometry often made energy measurements feasible where little activity could be used.

The alpha-particle energy of an alpha-emitting nuclide is almost specific to that nuclide. In fact, if it were possible to make convenient alpha-energy measurements with very high resolution and accuracy, it would probably be possible to make unambiguous determinations of the identity and quantity of any known alpha-emitter present in sufficient concentration. There are some cases, however, where two (or more) nuclides emit alphas of nearly equal energies*, and the limited resolving power and accuracy of the instruments available makes identification very difficult, if not impossible. This ambiguity can often be removed by other methods, such as chemical separation or the use of a mass spectrograph, or in some cases, by fission measurements, either spontaneous or neutron-induced.

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* Examples of such pairs are U^{232} and Po^{210}, U^{234} and Np^{237}, and Pu^{238} and Am^{241}. (See Table IV)

2. Methods of Alpha Energy Measurement

In using the measurement of alpha-energy as an analytical tool for chemical experiments, a prime consideration is simplicity in the sample preparation, in the operation of the instrument and in the interpretation of the results. This factor made the use of the magnetic alpha-energy analyzer* undesirable, because of the necessity of using carefully prepared line sources, because of the bulky and elaborate equipment required, and because of the low geometry involved ($< 10^{-3}$). Good geometry, preferably 50%, was highly desirable because of the low total amounts of alpha activity available in some measurements, or because of low specific activities in the sample. It was also desirable that the resolving power and accuracy of energy measurement be as good as was consistent with the requirements of relative simplicity and high geometry.

The measurement of ranges was tried, using uniform foil (mica) absorbers with surface densities from 1 to 8 mg/cm^2, at 50% geometry (parallel plate ionization chamber).[2] The method is described elsewhere.[2] Although the mica absorber method has high geometry, it has relatively poor resolving power and it was decided to use other methods instead. Other methods of range measurement were considered undesirable, since the best high geometry method (3) has poor resolution, while the methods having good resolution have quite low geometry.[2,4,5]

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* Such as those used by Rosenblum, Rutherford, Briggs, Chang and others. See Ref. 2 for references.

It was decided that the best results could be gotten from the measurement of the total ionization due to an alpha particle in a suitable ionization chamber. This method has at least one advantage in that the ionization collected is very closely proportional to the alpha particle energy, at least* within the energy region covered by the natural alpha-emitters (4 to 10 Mev.).

The method involves: (1) an ionization chamber in which the alpha particle expends all of its energy and in which the charge is rapidly collected, generating across the chamber capacity a voltage pulse (of the order of a millivolt), whose magnitude is at least approximately proportional to the particle energy, (2) a linear pulse amplifier which amplifies this pulse up to a voltage suitable for detection, and (3) a device, electronic or otherwise, for determining the relative number of pulses of various sizes. Plotting relative number versus pulse size essentially gives a plot of relative number versus alpha energy (assuming a correct calibration of the pulse height-energy relationship).

This method has been extensively used, [6-28] for measuring the energies of alphas, protons and fission fragments.

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* There is fairly good evidence that this holds even below 4 Mev.[4]

3. Total Ionization Method for Alpha-Energy Measurement

3.1. Chambers.-- Two types of chambers have been in general use. Air has been used as a chamber gas, usually with parallel plate electrodes, or free electron gases* (such as H_2, N_2, rare gases) have been used either with parallel electrodes or with electrodes with cylindrical symmetry (thin rod as collectrode). More recently, chambers with grids have been used, with free electron gases.

The optimum amplifier design for a particular type of chamber involves a reconciliation between the requirements: (1) preservation of pulse height and (2) ability to use high counting rates. If, as is often the case, the charge collection time varies from one alpha-particle to another because of the angular distribution of the alpha-particle paths, the pulse size may also vary if the amplifier is incorrectly designed.

3.2. Amplifiers.-- In all practical circuits, the charge collected in the ion chamber is passed on to the sensitive element of the detector, usually the grid of the first amplifier tube. The capacity of the chamber and tube and the grid input resistance form a parallel RC system which determines the shape of the pulse passing into the amplifier. The electric charge collected in the chamber charges up the capacity relatively rapidly, and then discharges slowly through the resistance. The characteristic pulse shape (voltage vs time) passing out of most pulse ion chambers consists, therefore, of

*Sec. 4.2

a rapidly rising part followed by a slow exponential decay, with the pulse height (for sufficiently large input resistance) proportional to the charge collected. One method of preserving the relative pulse sizes due to various alpha-particles independently of collection time, is to carefully reproduce the original pulse shapes in the recording apparatus. In the first use of this method, very sensitive electrometers[29,30] with very high input resistance were utilized. For this purpose, however, such electrometers have been largely displaced by D.C. amplifiers using electrometer tubes.[6,7,8,12,13,15] Recently, there has been developed[31] a highly sensitive dynamic condenser electrometer*, with great stability which has made it a useful substitute for electrometer tube circuits, and it has been utilized for alpha energy measurements by Jesse.[32]

Because of the very slow decay caused by the large input RC value ($\geq$ 1 sec) of such electrometer circuits, the useful counting rates must be low ($<$1/sec). If the counting rate is too high, excessive superposition of pulses results.

For energy measurements, the relation of the frequency response to the collection time is subject to more stringent requirements than is the case where only counting of particles is to be done. It can be shown[33] that for circuits whose frequency response is determined only by R-C elements, the pulse

* A very high resistance vibrating condenser changes a small D.C. voltage signal to an A.C. voltage, which is amplified through a tuned, high-gain, feed-back-stabilized A.C. amplifier, and recorded on a pen writing recorder, such as the Brown recorder.

height is essentially preserved if the decay time is at least 100 times the collection time. Because of its very long input time constant, the electrometer circuit (either D.C. or vibrating condenser) may be used with chambers having relatively long (> 1 millisecond) collection times. It may thus be used with air-filled chambers (where charge mobility is relatively low) with electric fields no larger than are necessary to prevent recombination of positive and negative ions (i.e., the saturation field). Of course, free-electron gases may also be used, since faster collection times do not affect the preservation of pulse heights.

To use higher counting rates than are possible with electrometer circuits, it is necessary to reduce the decay time by raising the low frequency cut-off of the amplifier. To preserve the relative pulse heights, the collection times must then be decreased, which can be achieved by higher electric fields in air-filled chambers or by the use of free electron gases. Corresponding to the increase in the low-frequency cut-off, the high frequency cut-off is also increased, in order to preserve the rise-time of the pulse.

A number of applications of the total ionization method have used amplifiers in the audio-frequency range with chambers containing either air or free-electron gases, although with air the electric fields must be high (to decrease collection time) and the low frequency cut-off must be moderately low. In general, chamber fillings of free electron gases

are the most useful, even with slow amplifiers, since the field strengths required, either for saturation or for short collection times, are considerably smaller than for air.

Besides allowing higher counting rates, the faster decay time allows greater tolerance for the presence of betas and gammas. With an electrometer circuit, the superposition of the small pulses caused by betas may seriously interfere by increasing the "background noise", and therefore the "line-width" of the pulse distribution. With faster-decaying pulses, the superposition is less likely, so that a given amount of beta-activity affects the noise-level much less.

In recent years, full advantage has been taken of the fast collection possible in free electron gases by using amplifiers with quite high frequency response (approximately one megacycle bandwidth) and correspondingly higher low frequency cut-off. These systems can use much higher counting rates and can tolerate much greater backgrounds of betas and gammas.

3.3. Pulse height recording and Measurement.-- In the detection process the pulses are sorted on the basis of magnitude and recorded in some suitable fashion. The most widely used methods have involved one of two procedures: (1) recording (on film or paper) a pulse voltage-vs.-time trace whose vertical displacement is proportional to the pulse height, and subsequently measuring the trace size (the pulse height sorting is usually done manually) or (2) sorting the pulse sizes electronically and recording them with registers, with or without scalers.

Trace recording has generally been carried out by photographing the movement of a fast string galvanometer, the mirror of a loop oscillograph or the trace of a cathode-ray oscillograph, with photographic film running past the instrument. This method has been extensively used in the Austrian, German and Swiss laboratories, while the electronic method has been more widely used in England, Canada and the U.S.A. Recently, Jesse[32] has used the Brown strip recorder as a precise recording device working out of the "vibrating-reed" or dynamic condenser electrometer. In most cases, the trace sizes are measured with a rule and then sorted into groups according to size. In a recent publication from Switzerland,[34] an ingenious photographic recording method has been described where a fixed photographic plate is used throughout an experiment and the number of pulses of a given size measured by determining plate blackening as a function of height on the plate.

For electronic detection, sensitive "relay" circuits have been used, such that a pulse exceeding a critical voltage by a small amount (< 0.1 volt) causes the recording of that pulse, whereas one smaller than the critical voltage is not recorded. Among the most commonly used of such relay circuits are thyratrons and vacuum tube trigger pairs. Variation of the D.C. bias on the detector grid serves to vary the cut-off point, and since D.C. voltages are easily measured, the cut-off point can be precisely determined and reproduced.

With a single variable bias detector of this type, an "integral" curve may be measured, i.e., the number of pulses above a certain voltage may be measured as a function of that voltage. Thus for alphas of a single energy in a total ionization chamber, varying the bias voltage would give a number versus voltage curve with a flat plateau which dropped off suddenly at the pulse height corresponding to the alpha energy. The derivative of this integral curve is the pulse-height distribution curve. In determining the differential curve, the number of counts at one point is subtracted from the number at the neighboring point. Since each point is independently measured, the statistical fluctuations of both points enter into the error in the difference. This statistical fluctuation may be greatly reduced by using two detector circuits whose grid biases are kept a fixed voltage apart.(11) The difference between the counts recorded through the two detectors determines directly the number of pulses at the given pulse height and the counting error in the difference is due solely to the statistical fluctuation of the difference*. The pulse distribution (or differential curve) may be determined directly by sweeping the biases of the two detectors over the desired range.

*E.G., In an integral curve taken by a single biased detector, the number of counts at one voltage might be 1089 ± 33 (statistical fluctuation error), while the number of counts at the next voltage step might be 900 ± 30. The number of pulses within this voltage interval would then be 189 ± 45. If two biased detectors had been used the number of counts on one might be 1100, with 904 on the other. The number of pulses in this voltage interval would be 196 ± 14, or 1/3 the statistical error.

Instead of subtracting the total counts passing through one detector from the total of the other, the differences may be determined directly through the use of a cancellation circuit which allows the recording of only those pulses which exceed one bias but not the other.(35,36)

The photographic method has the serious disadvantage of requiring the tedious measurement of the pulse traces on the film. In the form in which it was often used (with a mechanical oscillograph) it had the further limitation of requiring low counting rates. This limitation is not inherent in the method, since a cathode ray oscillograph may be used. It is relatively easy to collect a moderately large number of pulses in a reasonable time, since pulses of all sizes are recorded as they come through. Nevertheless, the tedium of measurement makes the collection of a large number of pulses impractical. With only a small number of measured traces, the statistical fluctuations make accurate measurements rather difficult. In addition, the problems involved in making accurate measurements of the film traces also makes good accuracy difficult to attain. The precision of Jesse's method, using the vibrating reed and the Brown recorder, is better than has been achieved thus far by any photographic method.

The electronic method makes it possible to avoid making visual measurements of a trace by referring measurements to a precisely determined D.C. voltage (pulse height selector bias). It has the disadvantage that only one pulse height is measured

at a time. With the differential pulse selector, for example, the pulses measured at any time lie within the narrow band determined by the difference between the two pulse selectors. Thus, in order that each point be measured with adequate statistical accuracy, it is necessary to spend considerable time in the complete sweeping of the energy region to be measured. At any one time, a large fraction of the pulses formed in the ion chamber are not recorded*, resulting in a corresponding increase in the counting time. This carries with it the added disadvantage of possible difficulties if the chamber or amplifier characteristics drift with time.

3.4 Multichannel Systems.-- It would obviously be desirable to combine the advantages of both methods--recording all the alpha pulses formed in the ionization chamber, while using the precision and measuring ease of the pulse height selector method. To achieve this combination a multi-channel system was constructed, using a series of pulse-height selectors with biases differing by fixed voltage increments. In this instrument, all the pulses from the ion chamber are recorded, each pulse being applied to every channel, but recording only in the channel whose bias corresponds to its pulse-height. Of course, because of the finite widths of the channels, it is not

* This comment applies also to the case where all pulses above a certain size are recorded, since only those pulses just getting through the pulse height selector are significant in the numerical differentiation process carried out later to get a number versus pulse height curve.

possible to attain a continuous description of the pulse distribution. This, however, is rarely a limitation, since if the channel width is somewhat smaller than the natural width of the pulse distribution curve, no significant loss in resolution or accuracy occurs. (Sec. 9.10, Fig. 23).

After this instrument was built, it came to our attention that a number of such multichannel instruments had been built elsewhere, with quite different designs, although there was an overall similarity. Due to the compartmentalization of the various projects during the war-time, it was only after our analyzer had been in operation for a while that we found that the British had constructed[18,37] several such instruments, as had the Canadians.[38] Still another one had been built at Los Alamos.[39] A description of one of the Canadian analyzers has been recently published,[40] and in it, there is also a discussion of some of the problems involved in designing and using a multichannel selector.

While the instrument described here is far from having optimum characteristics, it has been used for analyzing thousands of samples, and it was felt, therefore, that a description of its construction and operation was worthwhile. Work is now going on to improve its stability and resolution.

4. Apparatus

4.1. General.-- In Fig. 1 is a block diagram. The pulse from the chamber is amplified in the preamplifier and then the first amplifier. It is then passed through a pre-discriminator stage, which subtracts the same voltage from each pulse.

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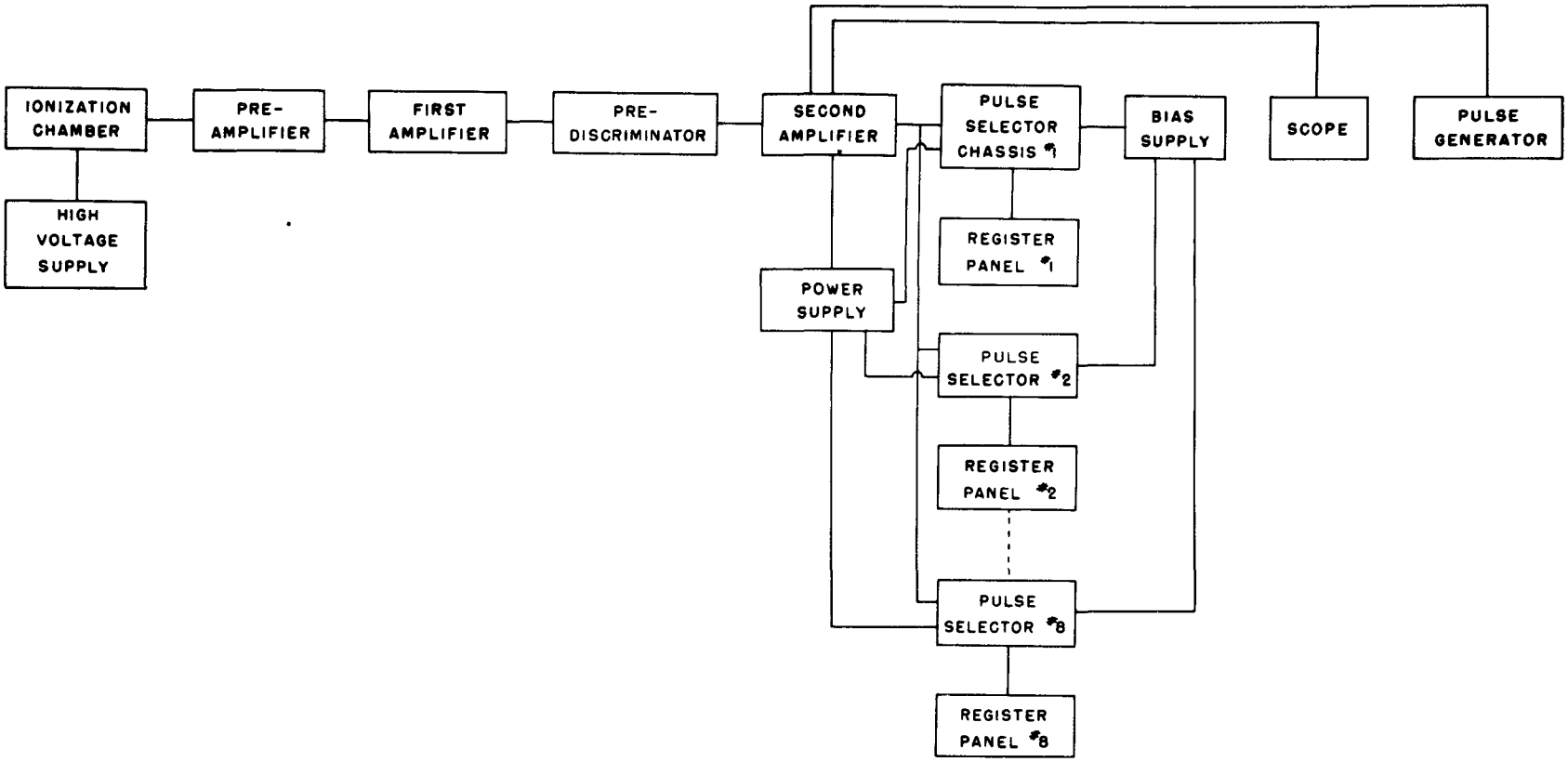
BLOCK DIAGRAM



48-CHANNEL PULSE ANALYZER FOR ALPHA ENERGY MEASUREMENTS.
FIGURE 1

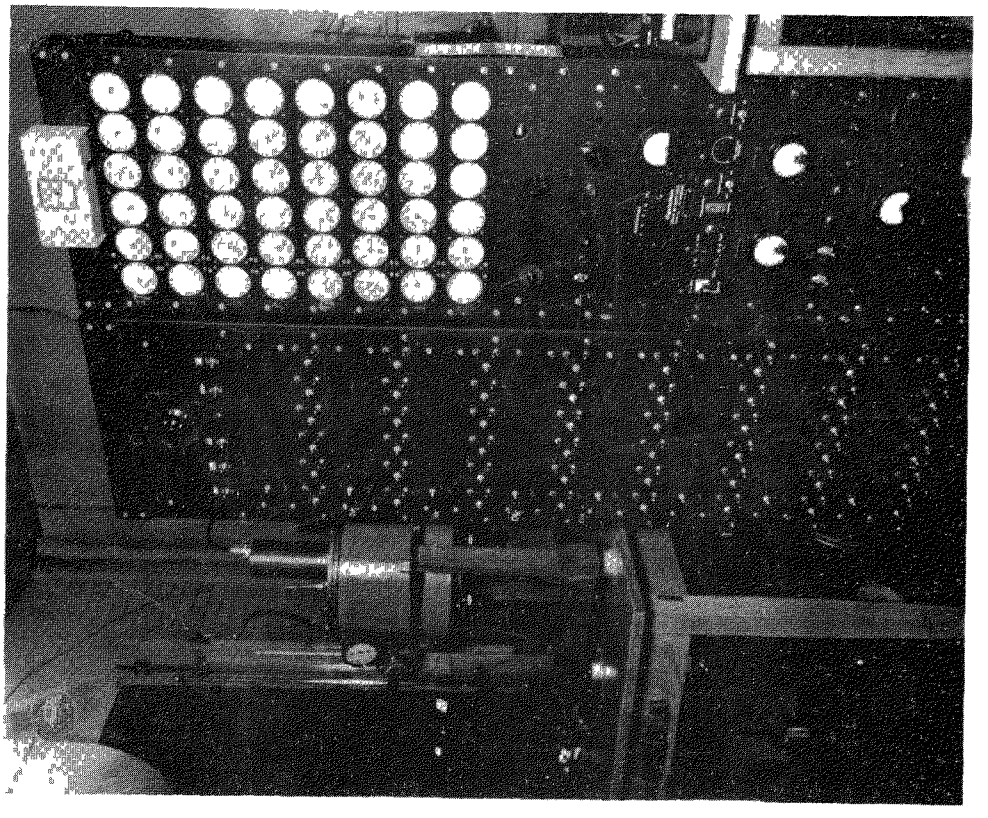


FIG 2

The remaining portion of each pulse is amplified in the second amplifier, the output of which is a cathode follower. (Fig.4). The output stage then feeds into the 48 channels through a subsidiary set of cathode followers. By varying the bias in the pre-discriminator and by varying the gain of each amplifier separately, the region of interest can be spread over all the 48 channels. The ability to concentrate all the detectors on a small energy region (thus accentuating energy differences) is important, since most of the alpha-emitters of interest have energies between 4 and 6 Mev. Every detector whose bias is less than the pulse size is fired by a pulse, but by means of the cancellation circuit described below the resulting signal is not transmitted to the recorder unless the pulse has not tripped the detector with next higher bias. The counts in each channel are registered by fast mechanical recorders. A photograph of the entire unit is shown in Fig. 2.

4.2. Chamber. The design of the chamber is greatly simplified by the use of "free electron" gases[41,42,43], instead of air, because of the low electric fields necessary in such gases.* Low fields suffice, because (1) the great mobility of electrons in free electron gases enables rapid collection of charge even in small electric fields and (2)

*Such gases as nitrogen, hydrogen or the noble gases, in which the capture of electrons to form negative ions has very low probability, and hence electrons carry the negative current in an electric field. Oxygen, on the other hand, is highly electronegative, having a high probability of capturing electrons.

the field necessary to prevent recombination of positive and negative charges (i.e., the saturation field) is much smaller than in air.

With comparable electric fields, the electron velocities are of the order of a thousand times larger than those of the positive ions, so that during the time in which the electrons are collected, the positive ions hardly move at all. Due to the electrostatic induction effect, the charges affect the collectrode* before they actually strike it.(44,46,47,48) At the instant of charge creation by the alpha-particle, the net induction effect is zero, since both charges of each ion pair are essentially at the same point. As the field moves the charges apart and the electrons approach the collectrode, the induction effect due to the electrons increases. The induction effect of the positive ions on the collectrode is opposite in sign to that of the electrons, and hence tends to partially cancel the electronic effect. Since the positive ions remain practically stationary during the period of electron collection, their cancellation effect remains essentially constant. Thus, if only the electronic part of the pulse is used, the total charge collected in the ordinary ionization chamber is smaller than would be the case if both positive and negative charges were completely collected.

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*Collecting electrode.

The magnitude of such cancellation can be easily calculated if the electrodes have suitable symmetry,(47) such as infinite plane electrodes, infinite coaxial cylinders or concentric spheres. Each charge is considered separately, by treating it as though it were uniformly smeared out on a surface lying symmetrically between the two electrodes with the position of the actual charge determining the position of the surface. For example, a charge Q lying between two concentric sphere electrodes at a distance R from the center would be considered spread uniformly over a concentric sphere of radius R. The magnitude of the total charge induced on both electrodes by the charge Q is equal to Q. The partition of the total induction between the two electrodes is determined by the ratio of the electrostatic capacities of the artificial surface to each of the electrodes. The closer the charge Q is to one electrode, the larger the capacity of the artificial surface to this electrode and hence the larger the induced charge.* For plane parallel electrodes, the effect is linear since the capacity varies directly as the distance between the plates. For the spherical and cylindrical case, the linear relation does not hold.

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* In the case of two concentric sphere electrodes, where the inner one is the collectrode, the magnitude of the charge induced on the collectrode is $Q \frac{a}{r} \frac{(b-r)}{(b-a)}$, where a = radius of collectrode, b = radius of outer electrode and r is the distance of Q from the center. Note that the charge is Q when r = a, and is zero when r = b. For the case where b >> a, the induced charge is $Q \frac{a}{r} (1-\frac{r}{b})$, i.e., it drops off hyperbolicly with r (for b >> r).

If the ion chamber is being used for energy determinations, the "positive ion effect" is annoying, since the magnitude of the cancellation depends upon the geometrical position of the positive ion in the chamber. For alphas emitted in various directions, the variation in the cancellation effect can cause quite serious differences in pulse heights.[45] This difficulty may be minimized by several methods. First, the collectrode may be placed at some distance from the ionization path, which decreases the induction effect. Second, advantage[9,11,49] may be taken of the fact that the capacity between concentric spherical and cylindrical surfaces does not vary linearly with distance from the central surface. The collectrode may be made with so small a radius of curvature that its capacity to any surface even a little distance away is small, and hence practically no induction can take place unless the charges are very close to it. This method has an added advantage in that the small size of the collectrode serves to decrease its capacity to the high voltage electrode and to the rest of the chamber, thus tending to increase the pulse height. It has the disadvantage that the electric field drops off with distance from the collectrode even more rapidly than does the capacity of the artificial surface. Thus, the field is quite non-uniform and relatively weak at the high voltage electrode, on which the sample is often placed. Fortunately, electron mobilities in free electron gases are so high that even in weak fields, the velocities are high and saturation easy to attain.

A third method of reducing the positive ion effect involves the use of a grid(18,19,21,23,50,51,52) in front of the collectrode in a parallel plate ion chamber. The grid serves to shield the collectrode from induction effects due to charges between grid and high voltage electrode. It has no shielding effect, however, on induction from charges which have passed through the grid into the region between grid and collectrode. Since the positive ions never enter this region, the positive ion effect is removed. Collection of electrons on the grid is prevented by the use of a high negative potential, but one not as high as that on the high voltage electrode. From experiments in this and other laboratories, it seems that the grid-type chamber is the best kind to use if great uniformity of pulse size is desired under a variety of conditions. The present analyzer, however, has been built about a chamber designed to reduce the positive ion effect by the first two methods.

The ionization chamber (Fig. 3) is mounted in a large steel vacuum-tight enclosure. The high voltage electrode consists of a covered bucket supported on insulators large enough to allow the use of up to 5000 volts (negative). Into the bottom of this electrode the sample support is fitted by means of a breech thread, a projecting pin being used to simplify the alignment of the thread as the support is pushed upward. The sample support may be removed through the bottom door which pivets on a single loose hinge and is

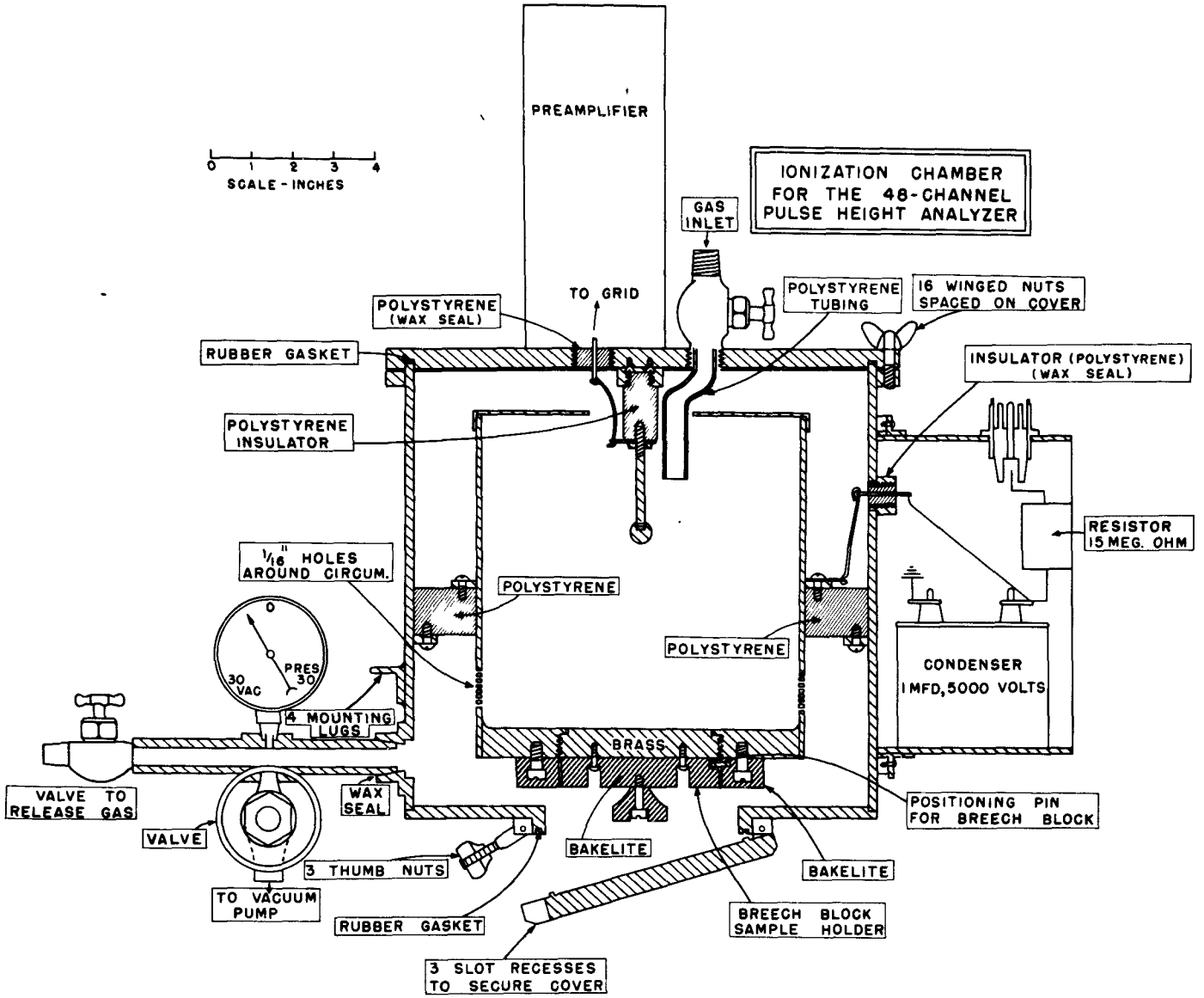

PREAMPLIFIER
0 1 2 3 4
SCALE - INCHES
IONIZATION CHAMBER FOR THE 48-CHANNEL PULSE HEIGHT ANALYZER
GAS INLET
TO GRID
POLYSTYRENE (WAX SEAL)
POLYSTYRENE TUBING
16 WINGED NUTS SPACED ON COVER
RUBBER GASKET
INSULATOR (POLYSTYRENE) (WAX SEAL)
POLYSTYRENE INSULATOR
RESISTOR 15 MEG. OHM
1/16" HOLES AROUND CIRCUM.
POLYSTYRENE
POLYSTYRENE
CONDENSER 1 MFD, 5000 VOLTS
0
PRES
30
30 VAC
4 MOUNTING LUGS
BRASS
VALVE TO RELEASE GAS
WAX SEAL
POSITIONING PIN FOR BREECH BLOCK
VALVE
BAKELITE
BAKELITE
3 THUMB NUTS
TO VACUUM PUMP
RUBBER GASKET
BREECH BLOCK SAMPLE HOLDER
3 SLOT RECESSES TO SECURE COVER

FIGURE 3

clamped against the rubber gasket by three thumb-screws. To facilitate rapid clamping these screws are hinged and slide into slots in the door. The bottoms of the sample support and high voltage electrode are insulated to protect the instrument operator.

The collectrode consists of a half-inch brass sphere at the end of a narrow rod. To reduce the capacity to ground, the rod is supported by a long insulator, and a fine wire leading from the rod through the insulator in the top of the can is used to carry the signal. The wire and the polystyrene insulator through which it passes are vacuum sealed with Apiezon W wax, and the insulator is also mechanically supported by screw threads. The high voltage lead comes through a similar insulator, with a one mfd. (5000 v, oil) condenser connected outside the vacuum chamber. The condenser is in a shielding brass box, through the wall of which passes a grounding key for discharging the condenser. The preamplifier is mounted on top of the vacuum can directly above the lead from the collectrode.

After a sample has been introduced, the chamber is evacuated with a high-capacity mechanical pump to a pressure of about .07 mm, and then filled with argon. Repetition of this process has been tried, but seems not to have made any difference in the results so that one pumping is generally considered sufficient. Since the time required for sample changing is short, only a small amount of air is introduced

during this operation and as a result the partial pressure of O_2 after one pumping is tolerably low. Oxygen interferes by combining with the free electrons, forming slow-moving negative ions.

The argon used is purchased in large tanks and is said by the manufacturer to be 99.8% pure. It is not prohibitively expensive, being commercially manufactured for use in welding. For a while, the argon was purified by passage passed over hot copper, but it was found that no difference could be noticed when this step was omitted provided the gas was led directly from the tank via copper tubing.

Since no difference has been found in operation characteristics as a result either of purification procedures (for oxygen removal) or of flushing the chamber once with argon, it seems likely that the oxygen content in the chamber is so small as to have little effect. According to the results of Clark, Spencer-Palmer, and Woodward,[18] this means that the oxygen content is probably less than 100 parts per million (or $<$ .07 mm at one atmosphere). Purified nitrogen has been used, but is less satisfactory than argon in a number of respects, among them being the higher voltage required for saturation.

After the chamber has been evacuated, it is filled to a pressure of 1 to 2 atmospheres, and is then used either statically or with gas flowing through at the rate of a few-tenths ml. per second. It has been found that the characteristics of the pulses change more rapidly when the gas is

merely allowed to stand in the chamber rather than being allowed to flow through. Presumably this phenomenon is due to vapors arising from the organic matter in the chamber (insulators and rubber gaskets) or to air entering through very small leaks. The drift with static gas expresses itself as a decrease in pulse size for alphas of a given energy as well as in a decrease of resolution, i.e., increased spread of pulse heights corresponding to a given energy. When the chamber is adequately vacuum-tight, the drifts occurring with flowing gas are smaller than with static gas and even these steady down to quite small effects after an initial flowing period. The importance of such drifts as do occur is minimized by the use of alpha-emitting standards as described below. The potential on the high voltage is -1500V to -2500V for 1 atmosphere and -3000V to -4000V for two atmospheres.

Clark, Spencer-Palmer and Woodward[18] have found, by using a system with practically no rubber and with wax joints, by pumping down to a high vacuum and by very elaborately purifying the argon with hot copper and drying agents, that no drifts due to gas deterioration would occur over some days when the static method was used. At Los Alamos and elsewhere[14,21,23,42,53] it has been found that continuous circulation of the argon over hot calcium achieved the same purpose after a preparation period taking several hours. However, for large numbers of runs, such elaborate preparations are prohibitively time-consuming, so that the drifts which may be ascribable to insufficient gas purity have been

accepted and their effects minimized, insofar as possible, by the use of standards.

Although the use of a small collectrode located at some distance (12 cm) from the sample tends to minimize the positive ion effect, it does not eliminate it entirely. The effect may be calculated using some simplifying assumptions and the mathematical device referred to above.(47) If the electrodes are approximated by two concentric spheres of radii 1/4 inch and 4 3/4 inch and the alpha-particle specific ionization taken as a constant, then the maximum positive ion effect (vertically-emitted alpha) at one-atmosphere pressure results in a reduction of pulse height of 1.6% for an alpha of 5.0 cm. range, 1.2% for 4.0 cm range and 0.8% for 3.0 cm. An alpha emitted at small angles to the sample plate undergoes practically no positive ion effect at all, so that these percentages represent the maximum pulse height deviation due to the positive ion effect alone. If the pulse height distribution without positive ion effect can be represented by a Gaussian curve, then the reduction in the average pulse height is less than the maximum deviation, being 0.7% for the 5.0 cm alpha. The effect on the resolution is much less, a straggling parameter* of 1.0% increasing only to 1.2%.

*If the distribution curve is approximately representable by a Gaussian error curve, the straggling parameter (α) is one-half of the width at $\frac{1}{e}$ the height. (See Sec. 6.)

The positive ion effect is evidently non-linear, being larger for more energetic alphas. This factor does not introduce any added complications because the slight non-linearity of the amplifier necessitates the use of alpha standards in any case.

The mild reduction of resolution by the positive ion effect is of importance only when samples are counted at 50% geometry. When, by the use of collimators, only vertically emitted alphas are counted, all alphas of a given energy are affected approximately alike. However, the non-linearity with energy still remains. The collimation technique is described below.

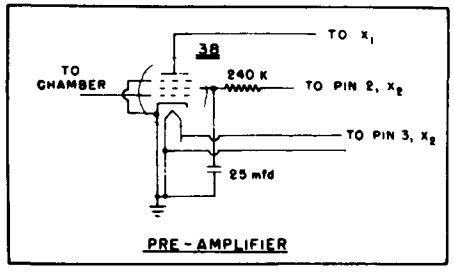
Besides the positive ion effect, the angular distribution of the alpha particles gives rise to another source of difficulty. The pulse shape and "rise time" from a vertically emitted alpha is not the same as that of a horizontally emitted alpha, because of the different induction effects from the various alpha tracks. This effect need cause no great difficulty if the amplifier is adequately designed.

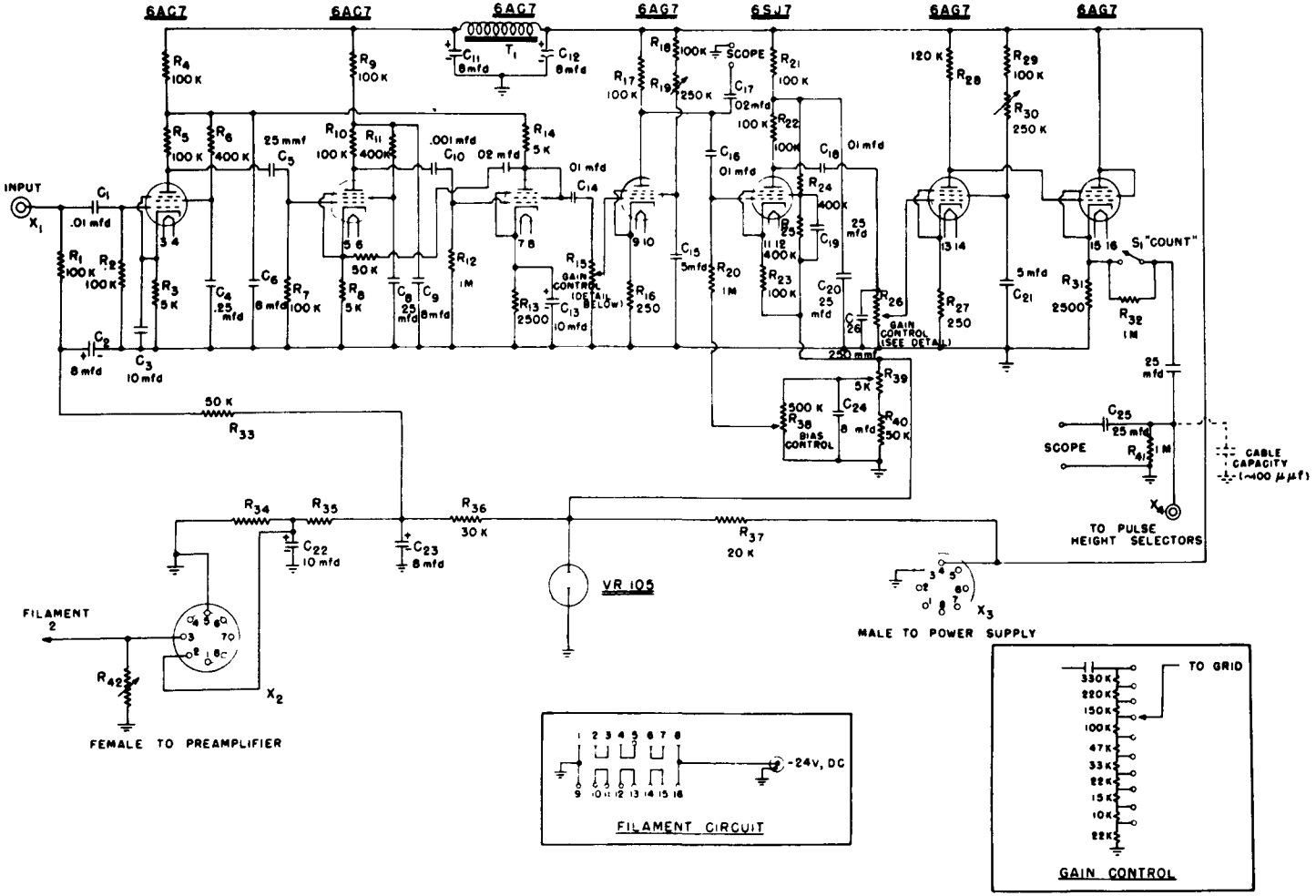
4.3. Amplifier.-- The circuits are shown in Fig. 4. The entire amplifier has a moderately low frequency response (Fig. 5). This frequency characteristic was determined empirically to give the best resolution with the chamber of Fig. 3.

PULSE ANALYZER

AMPLIFIER

FIGURE 4





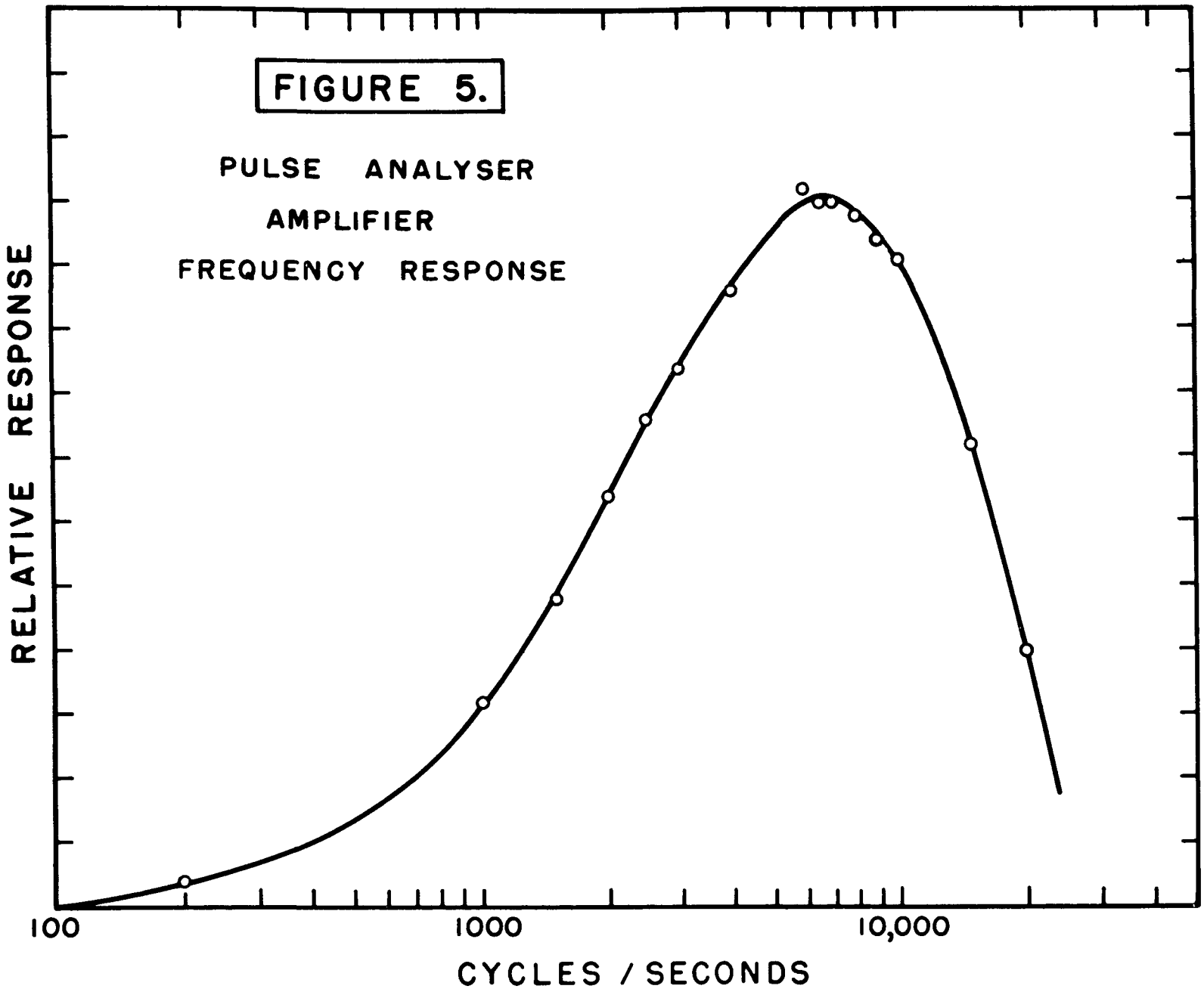


FIGURE 5.
PULSE ANALYSER
AMPLIFIER
FREQUENCY RESPONSE
RELATIVE RESPONSE
100
1000
10,000
CYCLES / SECONDS

The preamplifier is mounted directly on the chamber. The filament of the 38 tube is operated in series with the filaments of the first three amplifier tubes from a 24 volt D.C. supply* piped into the counting rooms; one side of the 38 filament is grounded. The use of a regulated D.C. filament supply prevents hum-pickup through the filament and prevents gain variations due to filament voltage changes.

The first amplifier consists of four stages followed by the pre-discriminator. The second amplifier consists of two stages, the last one being a cathode follower. Some of the problems in designing the amplifiers and prediscriminator were: (1) to achieve a large signal, as close to linear as practicable, (2) to design an output circuit with an output impedance sufficiently low so that the signal would not be attenuated when all the selector units were conducting, (3) to obtain a constant gain and discrimination level, and (4) to use a frequency response that would allow pulses of various rise times to come through with the same output heights.

It is desirable to have a large signal for the pre-discriminator input and for the input to the 48 selector units, to minimize the effect of drifts in the discriminator sensitivities. A linear signal is desirable so that the pulse heights may be linearly related to the alpha energies. However, small departures from linearity can be easily allowed for by calibration. There was no difficulty in obtaining

* Regulated batteryless supply (Nobatron: Sorenson and Co.)

linear amplification from the first three stages of the amplifier since the outputs of these tubes are at a low signal level. The fourth stage, however, and the amplifying stage following the pre-discriminator, are both identical, giving a 160-170 volt pulse output. Each has a 250 ohm unbypassed cathode resistor to help increase linearity, and a variable screen resistor to adjust for linearity. This adjustment is discussed later in the section on tuning the analyzer.

When the 48 thyratrons in the analyzer strike simultaneously, the grid to ground impedance is lowered sufficiently to decrease the signal voltage, unless the impedances of the thyratron grid circuits are isolated from the plate circuit of the last amplifying stage. Due to the finite rise-time of the pulse, thyratrons with a larger bias strike later than do those with smallest bias,(40) so that it is important to avoid changing the pulse height by loading the output. To reduce the output impedance, the output amplifier is coupled to a cathode follower on the amplifier chassis which is in turn connected to each of eight cathode followers on each of the eight selector chassis. The low output impedance of the cathode follower also reduces capacitative losses in the cable. Each of the cathode followers uses a 6AG7 tube, chosen for its high trans-conductance, and each of the cathode followers on the selector chassis has a 10,000 ohm stopping resistor in series with the grid, in order to prevent oscillations. There is no measurable attenuation of the signal when all the thyratrons are conducting.

The pre-discriminator is a 6SJ7 operated as a phase inverter. Its cathode is connected through a 0.1 Meg resistor to a 105 volt source stabilized by a VR-105 tube. The grid bias is also derived from the stabilized 105 volt source, and may be varied with the potentiometer R-38 (Bias Control). The adjustment of this control in conjunction with the adjustment of the first amplifier gain control (R-15) shifts the position of a particular alpha peak among the channels. R-39 is a tapped resistor, which is adjusted so that the zero position of the Bias Control corresponds to the cutoff voltage of the tube.

Since the pulses entering the prediscriminator are of the order of 100 volts in height, a change of 0.1 volt in the cutoff voltage of the 6SJ7 introduces an error of only 0.1%. The variation in the tube cutoff is ordinarily no greater than 0.1 volt, although fluctuations in the VR tube may result in tripping height variation of as much as 0.3 volt.

The pre-discriminator has a gain of one since its unbypassed cathode resistor is equal to the plate load resistor. The current feedback resulting from the unbypassed cathode resistor serves to give a linear response for large pulses. The output of the prediscriminator is amplified in a stage identical with the one preceding the 6SJ7 and fed through the cathode follower output to the 48 channels as described above.

The frequency response shown in Fig. 5 was determined empirically to give the best resolution with the chamber and gas used. The high frequency response is determined by R-26 and C-26, while the low frequency response is fixed by R-7 and C-5. Other combinations would undoubtedly work as well; these were left as they were when it was found that they gave good results. The necessity for adjusting the frequency response in this manner occurred primarily when 50% geometry was used. In this case, as discussed above, the pulse rise-time depends upon the angle of emission of the alpha-particle. When the alphas are collimated, the effect of the frequency response upon pulses of various rise-times is relatively unimportant, since collimated alphas give pulses of about the same rise-time.

An analysis of the circuit provides an explanation for the existence of an optimum frequency response band for the chamber and gas used and also for the fact that the pulse height and resolution decreases when a certain optimum high voltage value is exceeded. The analysis[54] of a circuit whose upper frequency cut-off is determined by one RC constant and whose low frequency cutoff is determined by another RC constant, shows that a plot of pulse height versus pulse rise-time goes through a maximum. The breadth of this maximum is determined by the breadth of the frequency response curve. The moderately narrow band of the present amplifier is adjusted for the rise-times occurring with the chamber and gas used. Increase of high voltage beyond a certain point

increases the electron velocity, decreasing the rise-time of some pulses beyond the maximum. These pulses are diminished in size thus worsening the resolution by broadening the pulse distribution curve.

At times, when slight leaks or other sources of gas impurities developed, the resolution at 50% geometry has been seriously impaired, presumably because the rise-times changed sufficiently to move away from the maximum in the pulse height vs. rise-time curve. In these cases operation with collimated sources was feasible. When the leaks became more serious, even collimated sources showed poor resolution, presumably because of negative oxygen ion formation.

The straggling contributed by the amplifier noise and other variations in the electronic circuits was tested by the use of artificial pulses. The output of a pulse generator, suitably attenuated, was delivered to the collecting electrode by capacitative coupling through the high voltage electrode. Since the driving frequency was derived from the 60 cycle supply, the output pulses were synchronized with 60 cycles, thus minimizing the variations due to the slight hum in the pulse generator output. The pulse input was adjusted so that the output corresponded to that due to a 5 Mev alpha particle. By roughly approximating the pulse distribution with a Gaussian curve, the straggling parameter was found to be about 12 Kev (.24%). This value gives an upper limit to the straggling due to such electronic factors as amplifier noise.

Since, as will be discussed later, this value is less than that found for alpha-particles, the resolution of the present instrument has not been limited by amplifier noise. The relatively low noise level is at least partially due to the moderately narrow frequency response band of the amplifier.

4.4. Multichannel Selector Units.-- The multichannel analyzers mentioned above as well as the one described here have in common the fact that each channel contains three essential components: (1) a discriminator whose tripping action is set by an externally supplied bias and whose output pulse is independent of the tripping pulse, (2) a cancellation circuit, which cancels the output pulse in all those channels tripped by a pulse except in the channel with highest bias and (3) a recording circuit. The discriminator outputs should all be the same, in order to provide trouble-free cancellation action. The discriminators used have been either thyratrons or trigger-pairs, either of which give constant-output pulses. Trigger-pair circuits can be made somewhat more stable in tripping level than thyratrons and can be used at higher counting rates, but require more circuit components. In order to simplify the circuit construction, thyratrons (Type 2050) are used as the discriminators in the analyzer described here. Other factors keep the useful counting rates low enough so that the thratron is no limitation. The drift of the thratrons, however, is important and will be discussed below.

The cancellation circuits must allow only the channel whose bias voltage corresponds to the peak voltage of the pulse to record that pulse. For example, if a pulse trips the discriminators of channels 1-5, the output pulse from the channel 2 discriminator cancels the output pulse from the channel 1 discriminator, channel 3 cancels channel 2, etc. Only channel 5 remains uncancelled and thus is the only one to record.

Several types of cancellation have been used. In one general type, the pulses from the discriminators are used to cancel each other. In one such scheme,[38,40] pulses from adjoining channels are fed into a multigrid mixer tube in opposite phase; a signal emerges into the recorder only if but one pulse enters the mixer. In another method of this type, the pulses from adjacent channels are fed into the opposite ends of an impedance, the total output being the algebraic sum of the two. Cancellation occurs unless only one pulse enters this impedance. One of the British analyzers[18] used a resistive cancellation circuit of this type, while the analyzer described here uses an interstage transformer. Because of the finite rise-time of the pulse emerging from the amplifier, direct cancellation of the discriminator output pulses can be used only with relatively slow pulses[40].

For cancellation at high counting rates with circuits utilizing fast pulses, gating circuits have been used.[37,39] A pulse from the discriminator activates a gating tube, which may also be deactivated by a pulse from the neighboring channel.

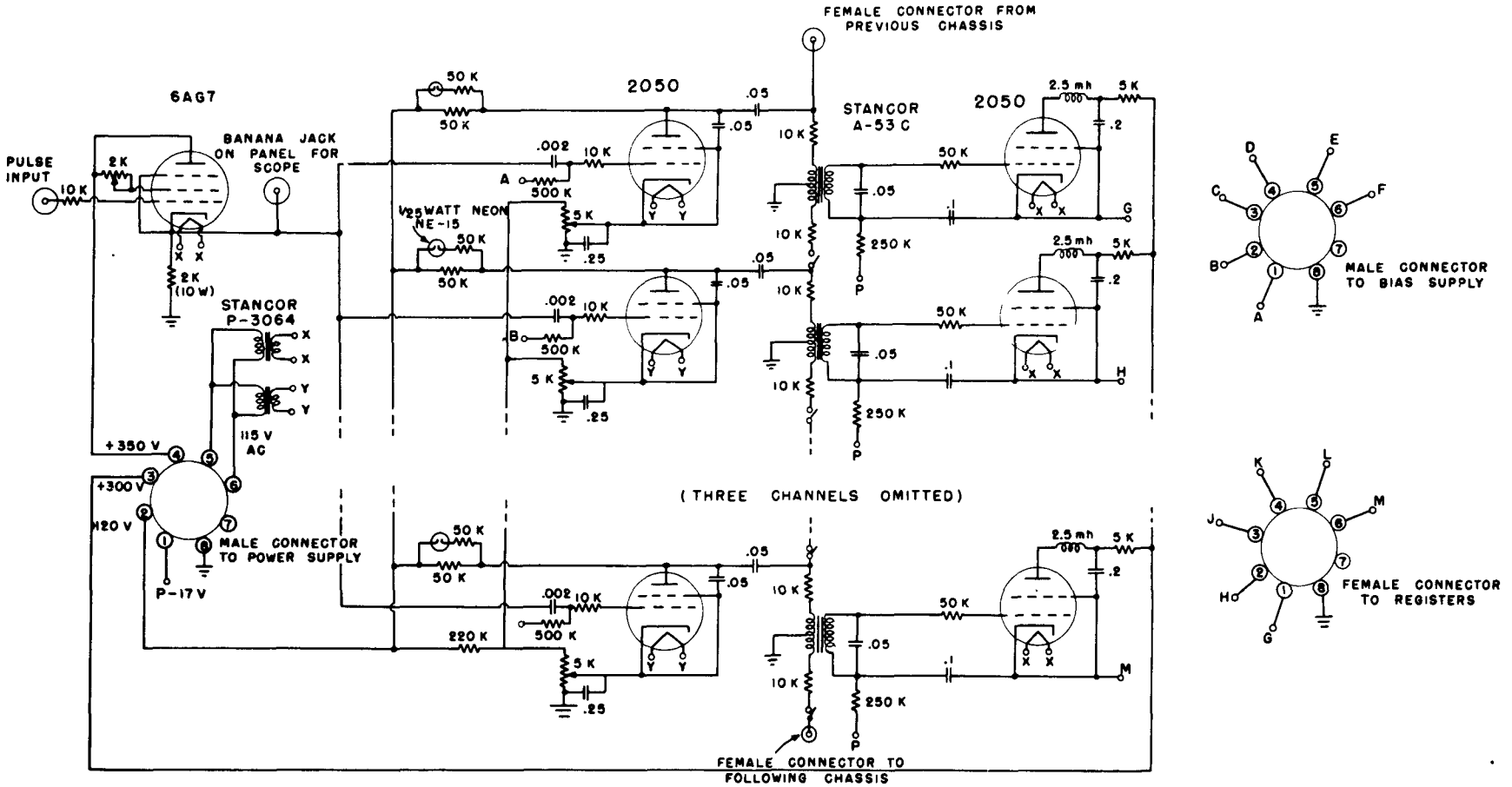
A timing circuit, activated by the pulse output of the amplifier, sets off the gating circuits of all the channels at the same time. Only that gating circuit which has been activated by its own channel and has not been de-activated by its neighboring channel allows the count to go through. Since the activating and de-activating pulses can be made to span the rise time, even very fast circuits can be made to cancel correctly with pulses of finite rise-time. This type of cancellation circuit is quite complicated, involving many circuit components so that it would be of little advantage with the present analyzer, where the amplifier frequency response and the recorders limit the pulse speed, and hence the counting rate.

In the present analyzer, the thyratron pulse height selectors (Fig. 6) consists of eight identical banks. Each bank contains a 6AG7 cathode follower, six selector thyratrons, six cancellation circuits and six recorder thyratrons. The latter are connected by cable to the recorders which are mounted in an adjacent relay rack. The eight pulse selector banks together with the amplifier occupy the other relay rack. Each of the eight banks is connected to the preceding one through a shielded cable to provide cancellation between the first tube of one bank and the last tube of the preceding bank. The last selector on the last bank counts all pulses whose voltages are greater than its bias setting, whereas each of the other selectors counts only those pulses whose voltages are within its three volt band.

PULSE ANALYZER

PULSE SELECTORS

FIGURE 6



4° The cathode follower in each chassis feeds the pulses to six units. Each unit contains a thyratron pulse height selector, a transformer cancellation circuit connected to the preceding and succeeding units, and a thyratron recording circuit. The bias for each of the 48 selectors comes from an external bias supply. The channel width is variable, but if, as is usual, it is set at 3 volts, then the external bias supplied to each thratron is 3 volts more negative than that on the previous tube. Variations in the cut-off characteristic of each thyratron may be corrected for by means of a potentiometer in the thyratron cathode circuit, which can be adjusted from the front panel. A small (~ 2 1/2 volts) positive voltage is applied to the potentiometer and the cathode tapped off of it. The methods of adjusting the 48 potentiometers will be discussed in section 4.7.

In the plate circuit of each 2050 discriminator tube, there is a neon light, mounted on the front panel, which flashes when the tube conducts. This enables visual observation of each pulse height as well as aiding in the tuning process. The thyratron quenching is due to the drop of plate voltage resulting from the discharge of the .05 mfd. plate to cathode condenser. Plate voltage is restored by charging thru the 50,000 ohm plate load resistor. In each thyratron grid circuit is a 10,000 ohm resistor, which serves to keep the grid impedance during discharge relatively high, so that the discharge of the first thyratron does not short the cathode follower.

The cancellation circuit is a modification of one described by Roberts,[35] and involves the use of a push-pull transformer for mixing the cancelling pulses. The pulse output of the 6AG7 cathode follower is positive, and if it exceeds the bias on the first selector, the tube trips, giving a negative pulse of about 100 V, which passes through one side of the transformer. If the cathode follower pulse is too small to trip the second selector, there is no cancelling voltage, and the negative pulse to the input of the transformer appears as a positive pulse of about 100 volts on the secondary output. Since the recording thyratron is biased to only -17 volts, it is tripped by the output pulse, thus activating the register and recording the count. If the cathode follower pulse is large enough to trip the second discriminator, its negative output pulse is applied to the opposite end of the transformer primary in the first channel, and no pulse emerges from the transformer secondary. Since the recording thyratron bias is -17 volts, considerable inequality in the discriminator outputs and the primary windings can be tolerated, because incomplete cancellation does not result in recording unless the difference exceeds 17 volts. In practice, no difficulty has been encountered even with aging tubes. According to Fig. 6, the first thyratron selector in each chassis does not have the same plate load as the other thyratrons, except by connection to the previous chassis. To make the plate load for the first thyratron in the first chassis the same as that of the other

selectors, a dummy transformer is used. To allow all the chasses to remain interchangeable, this transformer is mounted on a plug which fits into the female normally used to connect to the previous selector bank.

The recorders are of the Cyclotron Specialties type, which can be made to run at a rate of 60/sec on uniform pulses. To conserve space and for ease of mounting, these were removed from their cases and mounted on panels. It was found that when the recorders were used in the plate circuit of the recording thyratrons, grounding of the recorder shell with the coils at high potential resulted in occasional shorting of the relay coils. Since the D.C. voltage on the cathode is zero when the tube is non-conducting, the recorders were placed in the cathode circuits, thus removing the shorting danger.

In each of the cancellation circuits is a switch, which is normally closed to allow cancellation. When these switches are open, an integral rather than a differential distribution is measured, since each register then records all of the pulses that trip its discriminator. Such integral curves are sometimes used to measure the total counts in a certain number of channels (corresponding perhaps to a particular nuclide) without the bother of adding up the counts in each channel. Usually differential curves are preferred, since they not only give the energy distribution directly, but also allow higher counting rates. When an integral curve is taken, the first few channels must record all of the pulses.

Since the recorders limit the useful counting rate per channel, the total counting rate must then be fairly low. When the counts are spread by the differential method over many channels, the counting rates can be considerably higher.

The last register records all of the pulses larger than the bias on the last channel.

4.5. Bias Supply.-- The bias supply (Fig. 7) consists of a voltage divider supplying 48 leads which go to the 48 thyratron pulse height selectors. The first lead can be supplied with any voltage from 0 to 159 volts, and the others with voltages increasing by constant steps (called the channel width), usually 3 volts, but which may be 2, 1, 1/4, 1/10 or zero volts. The channel-width is determined by switch S3. The various resistor values were accurately adjusted to give the proper voltages. The parallel and series resistances at the various settings of S3 determine the total voltage across the 48 precision bias resistors, and thus the voltage across each one. They also maintain the total resistance in the voltage dividing network constant, so that changes in the channel-width do not affect the range control.

The voltage on the first thyratron (range) is determined by switches S4 and S5. Since S4 is also arranged so that changes in its setting do not affect the total resistance in the voltage dividing network, changes in the range control do not affect the band width. S4 provides voltages in 20 volt steps from 0 to 140 volts, and S5 provides

1. This resistor is adjusted so that the parallel combination has a value of exactly 10,800 ohms.

2. All resistors are wire wound precision resistors, IRC type WM-4, except for the voltage control to the 6SJ7, which is a General Radio, type 314, potentiometer.

3. The channel widths (voltages obtained across each 250 ohm resistor) are 0, 0.1, 0.25, 1, 2, and 3 V. for positions 1, 2, 3, 4, 5, 6 respectively on switch S_3. This is a two pole, six position switch.

4. The galvanometer is mounted on the chassis behind the panel. The needle is viewed thru a 1" hole in the panel, and is illuminated with a 6.3 V. pilot lamp placed to one side of the scale.

5. These resistors are adjusted so that the total resistance is 1425 ohms.

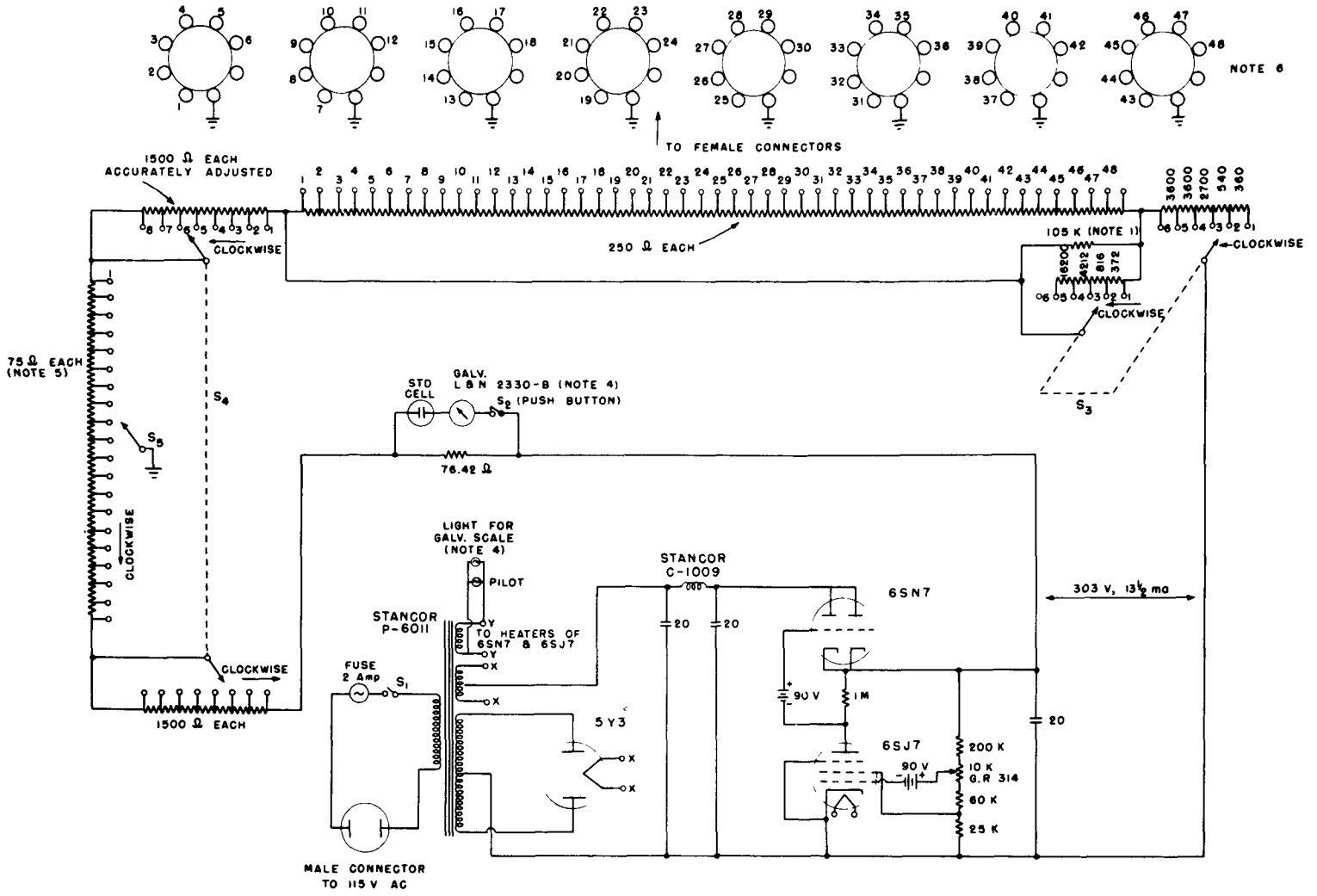
6. These connectors are octal sockets mounted on the back of the chassis.

NOTES (FIGURE 7)

PULSE ANALYZER

BIAS SUPPLY

FIGURE 7



voltages in one volt steps from 0 to 19 volts. A change in the range control, by changing the bias of the first thyratron selector, changes the bias of each of the selectors by the same amount, since the bias difference between successive thyratron selectors is constant.

The original purpose of incorporating variable range and channel-width controls was to allow the pulse distribution from a mixture of alpha emitters to be examined in greater or less detail, as desired. Increasing the range control setting brings the higher energy alpha particles into the recording region of the instrument. Decreasing the channel-width increases the sensitivity and detail with which a particular alpha energy region is examined. In actual practice neither control is varied, except during the tuning process. The range control is generally set at ten volts, and the channel-width at three volts. The maximum channel width is used, since using smaller channel widths accentuates the effect of small drifts in the thyratron cut-off characteristics. In practice, changes in the region or detail of examination are made by varying the gain controls in the amplifier and the bias setting of the pre-discriminator.

The voltage divider is supplied with 303 volts from a regulated power supply whose reference voltages are taken from dry batteries. The current through the divider is standardized against a standard cell by bucking the cell against the voltage generated across a 76.42 ohm resister. Divider current changes may be corrected by adjusting the

grid bias of the 6SJ7 regulating tube until the galvanometer is balanced at zero. The 48 bias voltages are carried through eight octal cables to each of the eight selector chasses.

4.6. Power Supplies.--The low voltage power supply (Fig. 8) furnishes the following voltages: regulated 350 V for the 6AG7 cathode followers on the pulse height selector chasses, regulated 300 volts for the amplifier B+ supply, unregulated 300 V for the register-driving thyratrons, regulated 120 V for the selector thyratrons and unregulated-17V for the bias on the register thyratrons. The power supplies are regulated by circuits similar to those used in the bias supply.

The high voltage supply(55) supplies up to -5000 V and is regulated by the use of an A.C. saturated transformer. The voltage is adjusted by means of a variable autotransformer in the primary.

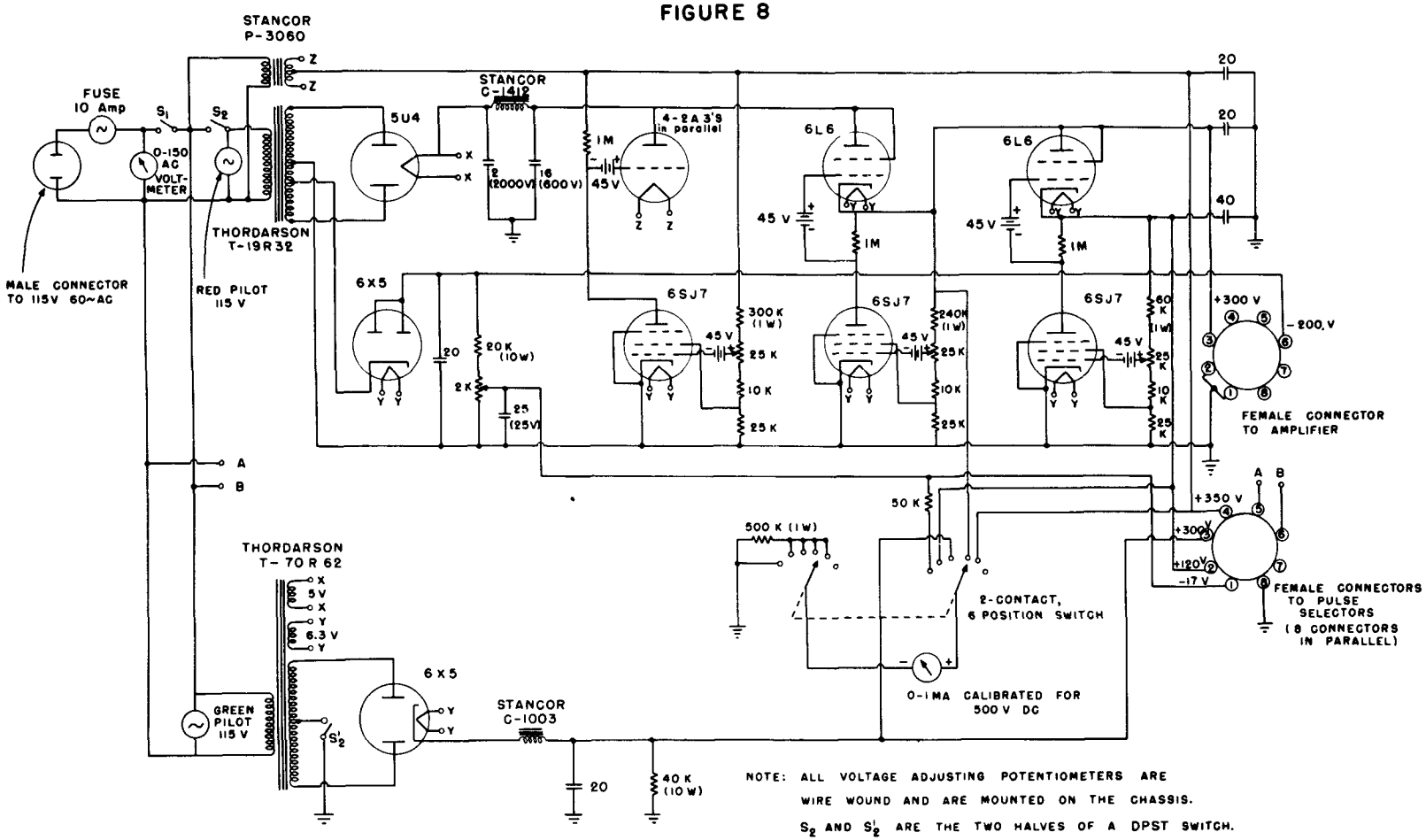
The entire 110 V A.C. supply is regulated by means of a Sola transformer.

4.7. Tuning the Analyzer:--The high level stages in the amplifier are adjusted for approximate linearity by varying the screen resistors. This adjustment is only approximate, but serves to avoid gross non-linearity. After the first lining-up, the drift of the circuits is seldom large enough to be detectable by the test used. A signal from a stabilized pulse generator is introduced into the grid of the fourth amplifying stage (6AG7). The output of this stage is connected to the horizontal amplifier of an oscilloscope

PULSE ANALYZER

POWER SUPPLY

FIGURE 8



NOTE: ALL VOLTAGE ADJUSTING POTENTIOMETERS ARE WIRE WOUND AND ARE MOUNTED ON THE CHASSIS.
S_2 AND S'_2 ARE THE TWO HALVES OF A DPST SWITCH.

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while the pulse generator is connected to the vertical amplifier. The screen resistor is varied until the scope pattern is a straight line. A similar adjustment is made on the other high level amplifying stage. Using the pulse generator, the output cathode follower is checked by observing the pulse output with all the thyratrons firing (range control at zero) and with none firing (range control set high). There should be no observable change in the oscilloscope.

Because the analyzer is in constant operation, the components age and drift sufficiently in about a month to require tuning of the selector circuits, although precise work may require more frequent tuning. At times the analyzer has been in operation for two to three months without retuning. The major portion of the drift is probably due to changes in the triggering points of the thyratrons and in the transconductances of the cathode followers. The process of tuning involves adjusting the potentiometers in the cathode follower screen circuits and in the thyratron selector cathode circuits until the selectors trip on pulses of the correct voltage.

The method for lining up the selector units involves the use of a pulse generator and the range and bias controls. The pulse generator is connected at some convenient point in the amplifier (generally the second gain control), the channel width is set at zero and the coarse range control set at about 130 V. The pulse generator is then set so the

selectors are just firing. The triggering of a channel can be easily observed by the flashing of its neon light on the front panel. With slow variation of either the pulse generator or range control the thyratrons which trip at voltages either above or below the norm are located. If they are not too far off, the tripping points may be adjusted with the cathode potentiometers; otherwise the tubes are replaced. For fine control, the pulse is fed in before the prediscriminator and its bias voltage varied.

If an entire bank of six selectors is off (either low or high), all (or most) of the channels in that bank will trip before or after the others. The banks may be adjusted relative to each other by adjusting the resistors in the cathode follower screen circuits, while slightly varying the output of the signal generator (or prediscriminator bias) so that the selectors are alternately tripped and not conducting. Variation between banks can be minimized by using matched 6AG7 cathode follower tubes. By selecting tubes, transconductances can be matched to about 5%.

The entire operation is repeated at several settings of the range control and pulse generator, adjusting thyratron and 6AG7 circuits. A method suggested by Kohman[56] for systematically repeating this operation enables the adjustment to be made fairly rapidly. Using zero channel-width and a large pulse (and correspondingly high voltage on the range control), the cathode follower screen resistors are

adjusted until all or almost all of the channels fire simultaneously. With a small pulse (and low range voltage), the thyratron potentiometers are adjusted until all fire simultaneously. However, the adjustment of the thyratrons at small pulse levels disturbs the alignment at high pulse height, so that the entire alignment procedure is repeated, until no discrepancies occur at both pulse heights. Only a few repetitions are necessary, since the discrepancies disappear rapidly.

The particular sequence used results from the fact that variations in the cathode follower gains are of little importance for small pulses but of considerable importance for large pulses. Thus, if e_i = pulse voltage from pulse generator, g_k = gain (< 1) of one cathode follower, and e_r = range control voltage, then, with zero channel width, the voltage put on the thyratron grids of one selector bank is $e_i g_k - e_r$. Suppose $e_i = 150$ V, and e_r is selected so that the net output voltage is zero for one selector bank. If the gain g'_k of another cathode follower differs by 5% from g_k, then the output voltage will differ from zero by 7 volts. If $e_i = 5$ V, then the output voltages differ only by 1/4 V. Thus the cathode followers are adjusted with large pulses where differences in their outputs are larger than differences due to misalignment of the thyratron selectors. Correspondingly, the selectors are adjusted with small pulses, where differences between the thyratron selectors are greater than differences due to cathode follower variation.

As a final test, the tripping of the channels is checked with the range control. First the channel width is set at one volt, the pulse generator is set so the last register trips, and the range control increased in one volt steps, so that each register trips in turn. Then the channel-width is set at 0.1 volt, and the pulse generator output varied slowly, so that each succeeding register trips in turn. Since the channel width bias used regularly is 3 volts, this insures that the error in the effective channel width is no greater than 3%. A single alpha peak usually is covered by at least four or five registers, so that errors tend to cancel, when the total count in one peak is taken, unless there is a consistent error in the selector settings. To check the possibility of such cumulative errors, the range control is advanced in one volt steps with the channel width set at 0.1 volt. The channels tripped at each range control setting should change in groups of ten. With a few different pulse height settings, the possibility of cumulative errors is eliminated.

Despite the tuning, some of the selector banks seem to get out of line more rapidly than would be expected. It is felt that these drifts are due to the thyratrons and the use of eight cathode followers. Future circuits will utilize more stable selector circuits and will avoid the use of multiple cathode followers.

The registers used have a number of adjustments which can slip or wear over a period of time. Part of the tuning process, therefore, involves checking and adjusting the registers. The method of adjusting Cyclotron Specialties registers is described elsewhere.(57) Several methods can be used to check them. One is to determine the counting rate of a pulse generator whose repetition frequency has been calibrated. The range control is used to bring in one register at a time, and its ability to follow the pulse generator is determined. Another method, although much noisier, is faster. The switches in the primary circuits of the cancellation transformers are opened. A pulse large enough to trip the last channel is used. All the registers fire away simultaneously as pulses come through, and it can soon be seen if any are counting ahead or behind the others.

After tuning, the instrument is usually recalibrated with a standard containing an artificial mixture of several alpha-emitters of known energy. This is a final check on the tuning process, since the counts should fall on smooth distribution curves.

5. Sample Preparation and Effect of Self-Absorption

To get good results in an alpha energy analysis, self-absorption in the sample must be quite small. For vertically-emitted alphas, the effect of sample thickness is not very serious, within limits, since a vertical alpha from a sample of surface density 40 $\mu g/cm^2$ loses an average of about .01 Mev.

However, the alphas emitted at an angle to the perpendicular lose considerably more energy, the amount lost increasing with angle. Self-absorption not only reduces the apparent energy, but also decreases the resolution by spreading the peak. A "thick" sample measured at 50% geometry has a very marked low energy tail extending down toward zero energy; the greater the thickness, the larger the fraction of alpha-particles appearing in this tail.

The best kind of sample for use in the analyzer consists of a thin, uniform film spread on a smooth surface. A simple method for making samples of this type involves electroplating on polished metal surfaces. (The details of technique in this and other methods have been discussed elsewhere.)[2,58,59] Another method which gives very good samples involves the evaporation of the active material onto a cooled plate in high vacuum. While the samples prepared in this fashion are more uniform than those made by any other technique, the labor and apparatus involved make the method impractical for most samples. The electroplating method, where applicable, has proven to be the best method. For the best samples, polished stainless steel or stellite have been used, although discs punched from bright surfaced platinum sheet have given fairly good results. In any of the methods of preparation, the sample plate is heated to a high temperature after preliminary drying, in order to remove acids, volatile salts and traces of moisture. If the platinum sheet is thin ($<$ .005")

and is strained, it may warp on heating, and an alpha colliding with the warped part of the plate gives rise to short pulses. The plate may be taped to a thick aluminum disc to flatten it out.

If the radioactivity to be investigated is in solution with a quantity of non-volatile salt, the activity must be chemically concentrated before it is practical to prepare samples. Such separation may be performed by solvent extraction, by resin column adsorption, by electrolysis or by precipitation with specific carriers. The solutions resulting from solvent extraction or resin column absorption may be essentially free of non-volatile salts and so can be directly evaporated to give a thin film.

It is often not feasible to use samples prepared with carriers at 50% geometry because of the difficulty in making a solid spread uniformly. Even if the carrier is redissolved and spread, it often tends to clump after evaporation of the solvent. The use of relatively non-volatile spreading agents (e.g., tetra-ethylene glycol, or TEG) serves to make the deposition more uniform by causing evaporation from a film rather than a droplet.

In cases where it is necessary to avoid separation of different chemical elements (e.g., uranium and plutonium), chemical concentration from non-volatile salts may not be feasible and the original material must be spread. The thickness may then be so great that the energy straggling may be excessive, even with the use of TEG. In this case and in cases

of large amounts of carriers, samples cannot be used at 50% geometry. The collimation methods used are described below.

6. Resolution

The ability to detect the presence of one peak when it lies very close to another is determined by the natural width of the peak. Under good conditions, the peak shape may be represented by a Gaussian curve of the form

$$P(E)dE = \frac{1}{\alpha\sqrt{\pi}} e^{-\frac{(E-E_0)^2}{\alpha^2}} dE$$

(E = energy corresponding to particular pulse height, P(E)dE = probability that a particular alpha gives rise to a pulse corresponding to energy between E and E+ dE, E_0 = energy corresponding to maximum of peak, α = straggling parameter = one-half the peak width at 1/e the peak height.) When the Gaussian shape applies, the peak width may be described by the straggling parameter, although some authors have used the "half-width" (total peak width at one-half the peak height). For a Gaussian curve, the "half-width" is 1.67 times the straggling parameter. Even though the peak shape deviates somewhat from a Gaussian, it is still useful to use the straggling parameter measure of peak width. The use of the straggling parameter rather than the half-width as a measure of resolution is arbitrary, since one can be calculated from the other if the pulse shapes are actually Gaussian. Because of asymmetric straggling, however, it is preferable to use a measure of peak width which is determined lower down on the

peak. Such a practice gives greater weight to the low energy tail. Although the choice of 1/e the peak height is certainly arbitrary, the authors believe it to be preferable to the use of the half-width.

The width of the approximate Gaussian curve gives a measure of the ease or the possibility of distinguishing another peak close in energy. From the Gaussian measure, however, the ability to find a second peak should be a function only of the straggling parameter and the energy difference (ΔE) of the two peaks, and should not depend upon whether the energy of the second peak is greater or less than that of the first. Actually, some of the straggling factors are asymmetric in their effect, resulting in greater straggling on the low energy side. They have the effect, not only of widening the peak, but also of shifting the peak towards lower energy, and in some cases, of creating a low energy tail which stretches towards zero energy. If the energy of the first peak is E, and that of the second peak is $E-\Delta E$, it will than be harder to find the second peak than if its energy were $E+\Delta E$. In fact, if the second peak is lower in energy and is present in very low concentration, it may be impossible to locate it. Despite the fact that the straggling parameter is not an unambiguous measure of the resolution, it is very useful for those cases where the second peak is present in a concentration sufficient to keep it from being lost in the low energy tail. This is true only when most of the alphas

(of the more energetic group) are in the main part of the peak. However, where the asymmetric effect is large (e.g., with thick samples), the straggling parameter has little significance, because a large fraction of the alphas are then in the low energy tail.

Since the natural spread (as emitted) of the alpha energies is much less than 1 ev, the observed straggling is an instrumental effect. From the emission of the alpha to the recording of the resulting pulse height, a number of steps are involved, each step independently introducing its own statistical variations. The most important sources of straggling are: (1) source straggling, due largely to variations in self-absorption (energy loss of alpha in solids on sample plate) and to alpha back-scattering; (2) ionization straggling, or variation in number of ion pairs formed by alphas of identical energy; (3) chamber straggling, due to such things as the positive ion effect or grid capture of electrons; and (4) amplifier noise. Because the straggling effects are independent of each other, the total straggling parameter is taken as the square root of the sum of the squares of the individual parameters:

$$\alpha^2 = \alpha_s^2 + \alpha_i^2 + \alpha_c^2 + \alpha_n^2$$

where α = total straggling parameter, and s = source, i = ionization, c = chamber, and n = amplifier noise.

As mentioned above (Sec. 5), a thick sample results in 59 energy degradation of many of the alphas, the energy losses being greatest for alphas emitted at small angles to the sample plate. At 50% geometry this results in a long low energy tail, but the tail may be decreased in magnitude by collimation (Sec. 7.4). α_s is of the order of magnitude of (1.5 to 2 times) the energy lost by an alpha emitted vertically through the entire sample thickness. Thus, for a heavy metal oxide (e.g., U_3O_8) uniformly spread on a smooth plate, α_s is very roughly 15 Kev for a 20 $\mu g/cm^2$ sample.

Chamber straggling for the type of chamber used in the present instrument is of several types. If a small amount of oxygen is present, there is a statistical variation in the number of electrons captured. At 50% geometry there is a variation in the positive ion effect. As mentioned above (Sec. 4.2), if the straggling of a 5.0 cm (6.3 Mev) alpha were 1.0% (63 Kev), then the total straggling is increased by the positive ion effect to 1.2%. That is $\alpha_{\text{pos. ion}} = 41$ Kev. If the rise times of the pulses corresponding to a particular alpha peak are not all the same (e.g., at 50% geometry), the amplifier frequency response may be such as to cause slight variations in the amplification of the pulses. The last two effects are decreased by collimation, since they arise from differences between pulses from vertically and horizontally emitted alphas.

It was pointed out (Sec. 4.3) that the amplifier noise was not a serious source of straggling, since pulse generator tests show α_n to be about 12 Kev.

Ionization straggling has not been investigated to any great extent. It is quite different from range straggling, which results from variations in the number of collisions per unit path length as well as in the energy lost by the alpha per collision. Ionization straggling arises from variations in the energy required to form an ion pair. Because a majority of the ion pairs are actually formed by δ-rays (fast secondary electrons knocked out by the alpha-particle), there are more actual ionizing events than alpha collisions. The statistical variation in the number of ion pairs formed would then be expected to be smaller than the statistical variations in range straggling. In the simplest type of calculation, assuming a Gaussian distribution, the standard deviation ($\sigma = \alpha/\sqrt{2}$) would be equal to the square root of the number of total events (i.e., number of ion pairs formed). For Po alphas (5.30 Mev), since 186,200 ion pairs are formed in argon,[15] the standard deviation is .0023% or 12.3 Kev and $\alpha_i = 17.4$ Kev. The problem has been theoretically analyzed by Fano[60], who showed that the standard deviation was $\sqrt{FJ_o}$, where J_o is the total number of ion pairs and F is a factor between 0 and 1, but generally lying between 1/3 and 1/2. For $F = 1$, $\sqrt{FJ_o}$ is the standard deviation for the Poisson distribution (which for large J_o is representable by a Gaussian). Thus, according

TABLE I

ALPHA PARTICLE STRAGGLING PARAMETERS* ACHIEVED IN A NUMBER OF TOTAL IONIZATION SYSTEMS

Authors	Average Straggling Parameters		Gas	Type of chamber	Type of charge collection
	50% geometry	collimated			
Bunemann, Cranshaw, and Harvey(51)		30 to 35 Kev	argon	grid	Electron
Schintlmeister,(12, Vol.146)and Schintlmeister & Rona(61)	250 to 280 Kev		air	Parallel plate	Ion
Jentschke(10)		137 Kev	air	"	Ion
Stetter(15)		27 Kev 78 Kev 86 Kev	H_2 air argon	"	Ion
Maeder(34)	78 Kev		air	"	Ion
Brown & Curtiss(27)		96 Kev	N_2	thin rod collect-rode	Electron
Martin Deutsch & M. Ramsey(21)	75 Kev		2% CO_2 in argon	grid	Electron
O.R. Frisch(19)		100 Kev	N_2	grid	Electron
Clark, Spencer-Palmer & Woodward(18)		60 to 75 Kev	argon	grid	Electron
Fowler & Rosen(23)	96 Kev		argon	grid	Electron
Parsons(28)	50 to 55 Kev		argon	grid	Electron
Ghiorso, Jaffey, Robinson and Weissbourd(62)	55 to 70 Kev	45 to 70 Kev	argon	small ball collect-rode	Electron
Jaffey & Connor(63)	45 to 65 Kev	45 to 65 Kev	argon	grid	Electron

*These have been measured from the published curves, and are only for alpha particles. The values represent the best results from each instrument, if more than one or two curves were published.

to Fano, the ionization straggling for Po should be less than 17.4 Kev; taking $F = 0.5$, $\alpha_i = 12.3$ Kev.

Of all the published papers on total ionization measurements, Bunemann, Cranshaw and Harvey[51] have achieved the lowest total straggling parameter (Table I). By the use of a pulse generator and by varying the chamber straggling through changing the grid structure and voltage, they were able to evaluate α_n and α_c. They found (by difference) that $\sqrt{\alpha_i^2 + \alpha_s^2}$ was about 20 Kev. Since their sources were made by simply evaporating Po solutions on backing plates, and since 50% geometry measurements showed a considerable low energy tail, it is likely that their samples gave rise to a sizeable amount of source straggling. It is quite possible, then, that thinner samples might have given a closer check with Fano's results.

If the lowest values for the various individual straggling parameters are taken ($\alpha_i = 12$ Kev, $\alpha_n = 12$ Kev, $\alpha_c = 3.1$ Kev,[51] $\alpha_s = 5$ Kev), then the best straggling parameter that could be expected with this type of instrument is about 18 Kev. Although this is greater than that of the best magnetic spectrographs,[2] it is not much greater. In Chang's[2,64] instrument, for example, $\alpha = 5.4$ Kev. Thus, if the total ionization method can be pushed to its limit, its resolution can be made comparable to that of the best magnetic spectrographs.

The present pulse analyzer has a larger straggling parameter than that found by Bunemann, Cranshaw and Harvey (See Table I). Since α_n was measured to be only 12 Kev, since the total α has been found to be approximately independent of of sample, and since Fano's results indicate that α_i is quite small, it seems apparent that most of the straggling must be due to α_c. It is probable that this is also true of many of the other systems listed in Table I.

7. Techniques of Operation

7.1 Counting rates, tolerable beta activity and gas pressure.-- Because of the relatively low frequency response of the amplifier and the long time constants of the selector and recording circuits, it is not possible to use high counting rates. At 50% geometry, the counting rates are generally held to 500-1000 c/m if all the activity is concentrated in one peak. If the activity is spread over three or four peaks, the counting rate can be as high as 2500 c/m. Even for a single peak, the counting rate may be increased if the amplifier gain is high, so that the peak is spread over many channels. If, for some reason, the sample submitted exceeds these values, the effective counting rate is reduced by masking or collimating. The primary limitation on the counting rate is the slowness of the registers. Since the counts occur randomly, the maximum counting rate per register must be kept considerably below the permissible rate for uniformly spaced pulses. However, when the activities are spread over many

channels, so that the counting rates per register are reasonably low, the maximum counting rate is then limited by the amplifier frequency response, which determines the pulse shape, and in particular, the pulse width. If a pulse enters the amplifier before the decaying tail of the previous one has died away, the height of the new pulse is changed. If this kind of event occurs frequently enough, the pulse distribution is distorted, the peak widths increased and the resolution thereby decreased. This type of partial coincidence is minimized by limitations on the maximum counting rate.

The resolution is also decreased due to peak widening when large numbers of betas are emitted from the sample. Due to the random fluctuation of beta-emission, the effect of the beta pulses is to greatly increase the apparent noise level. Generally, more than a few hundred thousand c/m of betas cause difficulty.

The chamber can be used at one atmosphere pressure, provided the energy is not much greater than 5 Mev. For higher energies, greater pressures are often used, in order to cut down the range and thereby reduce the positive ion effect. The voltage is then correspondingly increased.

7.2 Operating region.-- Although the gain can be lowered enough so that the 48 channels cover the entire energy range of alpha-emitters, the resolution becomes poorer because the channel width is then about equal to the peak width. Usually, preliminary considerations and tests make it possible to eliminate large portions of the energy range before a complete

curve is taken, and the gain is set at the point necessary to apply the 48 channels to the region of interest. The bombardment and chemical histories of the samples very often serve to give a fairly good idea as to what nuclides may be present and in what relative concentration. For most samples, the 48 channels are concentrated in the 4.6-6.0 Mev region. To insure that no activity is missed in the range 4.0 to 4.6 Mev, the pre-discriminator bias is lowered for a few minutes and the first few banks of registers checked. If no activity shows up, the bias is returned to its usual position and the pulse analysis made. The presence of alphas with energies greater than 6.0 Mev is easily detected, since these are registered by the 48th channel. If it is desired to see a particular energy range in greater detail, the energy increment per channel may be decreased by an increase in amplifier gain while the position is set by the pre-discriminator bias control.

7.2 Abundance determination.--The relative abundance of alpha activity at the various peaks is determined simply by adding up the counts in the channels involved at each peak. The comparison made in this manner of the areas under each peak is more reliable than comparison of peak heights, because of the possible variation of straggling parameter from peak to peak. A larger straggling parameter drops the peak height even though the area remains the same. If two peaks overlap, the intermediate region must be appropriately divided by drawing the overlapping high and low energy tails to correspond to

the shapes of the non-overlapping high and low energy tails. Thus, for example, if the two overlapping peaks occur at 5.14 and 5.30 Mev, the high energy side of the 5.14 Mev peak is drawn to be similar to the high energy side of the 5.30 Mev peak, since this side is unperturbed. In the same fashion, the low energy sides are made similar. This procedure involves the assumption that the effects of self-absorption, alpha back-scattering and positive ion effect are about the same for both peaks. The assumption is approximately true, since the energies of two overlapping peaks are fairly close. However, because of this approximation, the estimate of areas in closely overlapping peaks is difficult to do with good accuracy, especially where one nuclide is present in low concentration.

In general, the total counts found under the peaks is less than the counting rate determined in an alpha counter; for some samples it is as much as 10% lower. This phenomenon is primarily due to sample self-absorption and alpha back-scattering,[2] both of which cause a reduction in the alpha energy. The effect occurs even in well-prepared thin samples, but is more severe in thicker samples. The energy distribution of the degraded alphas stretches from the original energy to zero in a continuous fashion, the actual variation depending upon the atomic weight of the backing material and upon the atomic weight and thickness of the

sample material. (The back-scattering effect is greater on high atomic weight materials (e.g., Pt) than on light metals.) Thus, both the fraction of alphas which are degraded and their energy distribution are functions of the backing material and the sample thickness. Since these effects are approximately the same for various energies, the ratio of counts in the peaks gives a fairly close approximation to the relative concentrations of the alpha-emitters. This may not be accurate if the energies are quite different, since the degrading effects do vary somewhat with energy. Another source of difficulty is that some of the degraded alphas from the high energy peak will appear under the low energy peak and raise its apparent concentration. This may be particularly serious if the low energy peak is present in low concentration. One way to evaluate this effect is to prepare a pure standard sample of the high energy nuclide, attempting to duplicate the method of sample preparation, and to measure the number of alphas at the position of the low energy nuclide.

Because of the energy degradation, ratios of activity can be more accurately determined at 50% geometry than can absolute activities. For this reason, it is best to evaluate the absolute activity of a particular peak by first measuring the activity of that peak relative to that of an internal standard and then counting the standard in an alpha counter.

Because of such problems as back-scattering, sample self-absorption, overlapping of peaks and variation in peak shapes with energy, it has not proven possible to make relative activity determinations with high precision. The major usefulness of the analyzer has been in the following types of measurements: (1) establishing the presence or absence of a particular nuclide (already known), (2) establishing the existence of a new alpha-emitting nuclide, (3) determining the alpha energies of newly discovered nuclides and (4) making abundance measurements where highly precise quantitative values are not required.

7.4. Collimation.-- It is possible to decrease the relative concentration of degraded alphas by collimating the particles emitted from the sample. The method is effective since most of the back-scattered alphas emerge at small angles to the sample plate,[2,65] while self-absorption is also important only for alphas emitted at small angles. To cut out these alpha-particles, the geometry of the collimator need not be very low, since the degrading effects are concentrated in the small angle region. As mentioned above, collimation also has the effect of decreasing the straggling of the pulse height distribution by minimizing variations in the positive ion effect and in the collection time of the electrons. Thus, even with well-prepared samples, the best resolution is not obtainable at 50% geometry, but requires some collimation.

When the sample is rather thick, the low energy tail becomes 69 so large as to make measurements very difficult. Although in this case the accuracy and resolution cannot be made as good as that of a thin sample, they are improved by collimation.

As mentioned before, collimation is also used to cut the counting rate of the sample. Several different collimators are available to vary the fraction of alphas cut out.

The collimators consist of brass discs having 1/16 inch holes, in close packed array, over the area of a circle about two inches in diameter. The area occupied by holes is about 68% of the total area. The thicknesses of the discs used are 1/32, 1/16, 1/8, 3/16, 1/4, 3/8, and 1/2 inches and the extent of collimation is described by the collimation ratio: hole depth to hole diameter. Thus, the 3/16 inch thick disc is called the 3 to 1 collimator. In drilling the collimators, use was made of a hardened-steel jig which had been carefully made with a milling machine. In making the thicker collimators, care had to be taken to keep the drill from wandering and making a non-vertical hole. The collimator discs were made with a supporting ring (1/16" thick) around the lower edge, which served to lift them above the samples. Table II shows the geometry per hole for the various collimators, as well as the overall geometry (which includes the transparency). The geometry is given relative to 50% geometry. It is evident from the table that even mild collimation causes a considerable decrease in counting rate. The activities required to give reasonable statistics and counting rates greater than background become correspondingly higher.

If the concentration of one alpha-emitter is a very small fraction ($< 0.1\%$) of the total alpha-activity, it is quite difficult to detect and measure it. At 50% geometry, it is possible to detect such activity when it is higher in energy than the other alphas; if it is lower in energy, detection becomes impossible because of the low energy tail of the high concentration peak. Even with collimation, detection of low energy alphas present in low concentration is difficult because of the presence of a small but definite tail on the peak.

TABLE II

THE COLLIMATORS

Collimator	Geometry** per Hole	Overall Geometry	Energy Lost in Collimator Hole*
1/2 to 1	16.6%	11.3%	.074 Mev
1 to 1	9.35	6.39	.15
2 to 1	2.87	1.96	.30
3 to 1	1.33	.909	.45
4 to 1	.779	.532	.61
6 to 1	.344	.235	.93
8 to 1	.194	.133	1.27

* Energy lost by vertically-emitted Po alpha, assuming no ions in hole are collected. For the thin collimators, alphas emitted at an angle lose more energy.

**Relative to 50% (2π) geometry.

This tail is possibly due to energy degradation resulting from scattering off the collimator. Improved results may be attained by determining the low energy background with a pure sample of the high energy peak. When the low concentration activity is of higher energy than the main peak, either the 50% geometry or collimation methods may be used. The sample must be counted for a long time in order to accumulate a statistically significant number of counts. This is feasible only if the counter background is very low, at least as low as the activity to be measured. Long counting times may also give rise to errors if there is any tendency of the chamber and circuit characteristics to drift.

7.5. Mica-low geometry method.--Because the total counting rate which may be used is limited, it is not feasible with either the 50% or ordinary collimation methods to increase the absolute counting rate of the low concentration activity by enlarging the total sample activity. However, if it were possible to prevent the large numbers of alphas from the low energy peak from entering the ion chamber, the sample size could be increased. This may be done by collimating the sample and using a mica absorber thick enough to absorb all or almost all of the low energy alphas, but thin enough to allow the higher energy alphas to get through. The residual range may be quite small, however, in which case a high amplifier gain must be used. Because a major portion of the range is absorbed in the mica, care must be taken to avoid excessive

angle straggling, i.e., significant deviations in path length through the mica. For example, the ratio of maximum to minimum path lengths in the mica is 1.41 for the 1 to 1 collimator, 1.12 for the 2 to 1, 1.05 for the 3 to 1, and 1.03 for the 4 to 1. The average deviation is, of course, smaller. Excessive angle straggling tends to spread the peaks, so that the small one may not be observable.

If the high energy peak is quite close to the main peak, the highest resolution is necessary, requiring at least a 4 to 1 or a 6 to 1 collimator. The actual resolution required depends upon the relative amounts of the main activity and the trace activity, as well as upon the energy separation. If the concentration of the high energy activity is very low, the high energy straggling of the main peak may tend to hide the small peak unless the resolution is very good. On the other hand, if the concentration of the high energy peak is not too low, the high energy straggling of the other peak is of less importance. The effect of the high energy straggling of the main peak is also decreased if the peaks are fairly well separated. This is illustrated in Fig. 9.

If the resolving power is measured by the peak width, then the resolution of the mica-collimation method is generally somewhat poorer than that of the 50% geometry or ordinary collimation methods. This fact is due to the increased straggling arising from the variation in path length through the mica (angle straggling) and from the fact that straggling in

range is greater than straggling in the ionization yield. On the other hand, the mica method increases the stability, since most of the alpha range is cut off, and variations in the chamber and circuits affect only the small residual ionization. The low counting rates resulting from the high degree of collimation can be partially compensated by long counting times and because of the increased stability involved in the mica method, instrumental drifts do not cause much difficulty even over quite long counting periods.

However, to get significant results even with long counting times, a very low background is necessary. For the large chamber in Fig. 3, this is difficult to achieve under the best conditions, because of the large surface area. Because most of the range is lost in the mica, the resulting pulses are small, and these are about the same size as pulses due to natural alpha-emitters contained in the chamber walls. A smaller chamber, a modified form of an atmospheric parallel plate alpha counter,[2] serves as a low background chamber. The bottom electrode is lowered to admit the collimator and mica absorber and to allow a few centimeters of argon between the collectrode and mica absorber. The electrode areas are quite small, making a total background of less than 0.5 c/m possible. Despite the large collectrode, the variation in positive ion effect is not important, because the range of the alphas beyond the mica is usually rather small.

The mica absorbers were made by splitting thick mica sheets down to approximately the correct thickness and measuring their thickness by weighing and area determination. By selection, it was possible to build up a series of absorbers ranging from about 1.6 to 8.0 mg/cm^2 in steps of 0.1 to 0.05 mg/cm^2. These were mounted on thin metal supporting rings about two inches in diameter.

7.6. Energy Calibration of Analyzer.--Calibration of pulse height versus energy is carried out by the use of standard samples containing alpha-emitters of known energy. If the output pulse height versus energy relation could be assumed to be linear, and if the amplifier gain and cut-off bias were accurately known, only one alpha energy would be necessary to establish the relation. Actually, in the present instrument, the pulse-height versus energy relation has a small curvature, due to such things as the positive ion effect and the non-linearity of the high level stages in the amplifier. Hence, at least two alpha energies in the region of interest are necessary to fix the scale. Because the instrument drifts in calibration, it is checked with standards rather frequently.

The standard samples are mounted on thick stellite discs, cut into quadrants of a circle and optically polished to a mirror finish. The activities are electroplated on these quadrants. By combining the quadrants, a number of combinations of alpha energies may be made. In Table III is a list of the more commonly used standards. Of course, for particular experiments, special standards of pure nuclides are often prepared.

The four peak sample is the most popular standard. It serves to provide a rapid energy-pulse height calibration in the region of greatest interest. It also provides a very rapid check on the instrument's resolution, because high resolution gives rise to very deep valleys which almost go down to zero, and these can be seen just by watching the registers. For higher energies, the RdTh or the Ra series are usually used.

To make an accurate energy determination, it is necessary to use an internal standard (i.e., a standard sample placed in the chamber with the unknown sample), to avoid the possibility of drift. One method involves the use of one internal energy standard and the energy increment per channel previously evaluated with the four peak standard. In another method, a high-energy and a low-energy standard are used to bracket the unknown. The alpha energies of the new nuclides which have been determined with the pulse analyzer have been measured by these methods. With the present instrument, the accuracy has been generally found to be no better than .01-.02 Mev.

A quick method of identifying an unknown peak which is suspected of being a well-known nuclide is to put in a standard sample containing that nuclide. Perfect superposition serves as identification, provided the history of the sample eliminates the possibility of other nuclides with energies very close.

TABLE III

STANDARD ALPHA ENERGY SAMPLES

	Nuclides	Energy (Mev)	c/m (50% geometry)
4-peak standard	U^{233}, Pu^{239}, Pu^{238}, Pu^{236}	4.823, 5.14, 5.50, 5.75	One with 500 c/m per peak; one with 6000 c/m per peak
MsTh Standard (and daughters)	RdTh, ThX, Tn, ThA, ThC, ThC'	5.42, 5.33, 5.68, 6.28, 6.77, 6.05, and 8.78	Total of 1600 counts (equilibrium concentrations)
	Io^{230}	4.66	One with 500 c/m, one with 7700 c/m
	U^{234} and U^{235}	4.76 & 4.40 Mev	1000 c/m
	Np^{237}	4.73	5600 c/m
	Pa^{231}	5.01 (87%) 4.74 (13%)	1960 c/m and 6500 c/m
	Pu^{239}	5.14	One each with 500 c/m, 5000 c/m, 50,000 c/m and 300,000 c/m
	Po^{210}	5.30	300 c/m, 5000 c/m 7400 c/m
	U^{232}	5.31	1200 c/m and 2400 c/m
	Pu^{238}	5.50	800 c/m and 6600 c/m
	Am^{241} and Cm^{242}	5.46 & 6.10	One each with total counts 11,000 and 670
Radium Standard	Ra^{226}, Rn, RaA, RaC'	4.79, 5.49, 6.00 & 7.68	Total of 3800 c/m (equilibrium concentrations)

Calibrations made at 50% geometry do not apply when the collimators are used, because part of the range is absorbed in the collimator holes. The order of magnitude of this loss is shown in Table II. Each collimator requires its own calibration, and great care must be taken to make the distances of the sample and standard from the collimator bottom identical.

7.7. Backgrounds.--Because of the large surface area of the chamber, the natural alpha background (due to the traces of uranium and thorium series normally found in metals) gives rise to a minimum of about 5 c/m. These counts are spread over a wide energy range, so that the number of counts per channel is considerably less than 1 c/m. The background counting rate per channel determines the detectability of low concentrations of activity. To keep the sample support from becoming contaminated, the sample plate is placed on an aluminum disc. These are thin punched discs which are discarded after each measurement. Even with these, after some use with a number of samples, the chamber inevitably becomes contaminated. When the background becomes too high for use, the chamber is cleaned, first by washing with organic solvents and then by polishing with #1 polishing paper. As a final cleanup, the chamber is mounted in a lathe and strips of polishing paper used in a manner designed to prevent recontamination of cleaned areas. Each strip is used briefly and then discarded. By this method, the background is

reduced although never as low as that of a new chamber. The background is generally less than 0.1 c/m per channel*, except in the region 5.1 to 5.5 Mev (many of the samples occur in this range) where the background may be 0.2-0.3 c/m. The operating background, after some use, may run as high as 0.5 to 1.0 c/m per channel; above this level the chamber is cleaned. Contamination of the collimator occurs rather easily, because the bottom part of the collimator is brought quite close to the sample in order to minimize the gas absorption. The cleaning process is the same as that used on the chamber, except that greater care must be taken, to avoid transferring activity into the collimator holes.

8. Useful tables

As an aid in interpreting the pulse analyzer curves, especially in identifying unknown activities, it has proven useful to have several tables readily available. In Table IV is a list of many of the alpha-emitters in the heavy region, placed according to energy. This makes possible a ready determination of which nuclides have energies close to the observed one. Table V, which gives the members of each of the known disintegration chains, makes it easy to see which peaks are likely to occur together due to radioactive equilibrium. Table VI, which gives the alpha-emitting nuclides of each element, makes it easy to see which peaks will occur together throughout a series of chemical operations. Most of the energies have been taken from Seaborg's table.(66)

* With the 48 channels spread over the region 4.6 to 6.0 Mev.

TABLE IV. ALPHA-EMITTERS ARRANGED ACCORDING TO ENERGY

Nuclide	E(∝)	Nuclide	E(∝)	Nuclide	E(∝)
Th^{232}	3.98	$RdTh^{228}$	5.333 (17%)	Fr^{221} (?)	6.05 (25%)
U^{238}	4.180	$RdTh^{228}$	5.418 (83%)	ThC^{212}	6.081 (9%)
U^{235}	4.396	RaC^{214}	5.444 (.022%)	Cm^{242}	6.10
		Rn^{222}	5.486	Pu^{234}	6.2
Io^{230}	4.66	Am^{241}	5.46	AcC^{211}	6.273 (16%)
Np^{237}	4.73	Pu^{238}	5.50	Tn^{220}	6.282
Pa^{231}	4.736 (13%)	RaC^{214}	5.505 (.018%)	Fr^{221}	6.30 (75% ?)
U^{234}	4.763	AcX^{223}	5.606 (36%)	Th^{226}	6.30
Ra^{226}	4.791	Pa^{229}	5.66	Cm^{240}	6.3
U^{233}	4.825	ThX^{224}	5.681	Ra^{222}	6.51
Th^{229}	4.85 (~70%)	AcX	5.717 (55%)	AcC^{211}	6.619
Th^{229}	4.94 (~20%)	$RdAc^{227}$	5.717 (15%)	At^{218}	6.63
Ac^{227}	4.98 (1%)	Pu^{236}	5.75	ThA^{216}	6.774
Pa^{231}	5.012 (87%)	$RdAc^{227}$	5.764	An^{219}	6.824
Th^{229}	5.02 (~10%)	Ac^{225}	5.80	At^{217}	7.02
Po^{207}	5.1	U^{230}	5.85	Em^{218}	7.12
Pu^{240}	5.1	At^{211}	5.94	AcA^{215}	7.365
Pu^{239}	5.14	$RdAc^{227}$	5.988 (25%)	AcC'^{211}	7.434
Po^{208}	5.14	RaA^{218}	5.998	At^{216}	7.64
Po^{206}	5.2	Bi^{213}	6.0, 5.86	RaC'^{214}	7.68
Po^{210}	5.300	ThC^{212}	6.042 (24%)	Po^{213}	8.336
U^{231}	5.31	$RdAc^{227}$	6.049 (20%)	At^{215}	8.4
				ThC'^{212}	8.776

TABLE V.(A). HEAVY ELEMENTS ARRANGED BY RADIOACTIVE SERIES

Z	4n Series Nuclide	$T_{1/2}$	E(∝) and Branching Ratio	Z	4n + 1 Series Nuclide	$T_{1/2}$	E(∝) and Branching Ratio
94	Pu^{240}	~6000 yr	5.1	95	Am^{241}	498 yr	5.46
				93	Np^{237}	2.20×10^{6} yr	4.73
90	Th^{232}	1.39×10^{10} yr	3.98	91	Pa^{233}	27.4 d	β^-
88	$MsTh_I^{228}$	6.7 yr	β^-	92	U^{233}	1.62×10^{6} yr	4.825
89	$MsTh_2^{228}$	6.13 hr	4.5 (≤ 0.01%) ca. 100% β^-	90	Th^{229}	7000 yr	4.85 (~70%) 4.94 (~20%) 5.02 (~10%)
90	$RdTh^{228}$	1.90 yr	5.418 (83%) 5.333 (17%)	88	Ra^{225}	14.8 d	β^-
88	ThX^{224}	3.64 d	5.681	89	Ac^{225}	10.0 d	5.80
86	Tn^{220}	54.5 sec	6.282	87	Fr^{221}	4.8 m	6.30 (75% ?) 6.05 (?) (25% ?)
84	ThA^{216}	0.158 sec	6.774				
82	ThB^{212}	10.6 hr	β^-	85	At^{217}	0.018 sec	7.02
83	ThC^{212}	60.5 m	6.081 (9%) 6.042 (24%) β^- (66.3%)	83	Bi^{213}	47 m	6.0 (4%), or 5.86 (2%) β^-(96%)
84	ThC'^{212}	2.6×10^{-6} sec	8.776 (66.3%)	84	Po^{213}	3×10^{-6}	8.336 (96%) or (98%)
81	ThC''^{208}	3.1 m	β^- (33.7%)	81	Tl^{209}	1 hr	β^- (4%) or (2%)
82	Pb^{208}	Stable		82	Pb^{209}	3.32 hr	β^-
				83	Bi^{209}	Stable	
96	Cm^{240}	29 d	6.3	91	Pa^{229}	1.5 d	5.66
94	Pu^{236}	2.7 yr	5.75	89	Ac^{225}	10.0 d	5.80
92	U^{232}	70 yr	5.31				
90	$RdTh^{228}$	1.90 yr	5.418 (83%) 5.333 (17%)				
	etc. as above (descendants of RdTh)				etc. as above (descendants of Ac^{225})		

TABLE V.(B). HEAVY ELEMENTS ARRANGED BY RADIOACTIVE SERIES

Z	4n + 2 Series Nuclide	$T_{1/2}$	E(∝) and Branching Ratio	Z	4n + 3 Series Nuclide	$T_{1/2}$	E(∝) and Branching Ratio
92	U_I^{238}	4.498×10^9 yr	4.180	94	Pu^{239}	2.44×10^4 yr	5.15
90	UX_I^{234}	24.3 d	β⁻	92	AcU^{235}	8.91×10^8 yr	4.396
91	UX_2^{234}	1.14 m	99.85 % β⁻ 0.15% I.T.	90	UY^{231}	24.3 hr	β⁻
91	UZ^{234}	6.7 hr	β⁻	91	Pa^{231}	3.45×10^4 hr	5.012 (87%) 4.736 (13%)
92	U_{II}^{234}	2.35×10^5 yr	4.763	89	Ac^{227}	13.5 hr	4.98 (1%), β⁻ (99%)
90	Io^{230}	8.0×10^4 yr	4.66				
88	Ra^{226}	1622 yr	4.791	87	AcK^{223}	21 m	β⁻ (1%)
86	Rn^{222}	3.825 d	3.488	90	$RdAc^{227}$	18.9 d	6.049 (20%)
84	RaA^{218}	3.05 m	5.998				5.988 (25%)
82	RaB^{214}	26.8 m	β⁻				5.764 (20%)
83	RaC^{214}	19.7 m	β⁻				5.717 (10%)
84	RaC'^{214}	1.5×10^{-4} sec	7.680	88	AcX^{223}	11.2 d	5.717 (55%) 5.606 (36%)
82	RaD^{210}	22 yr	β⁻	86	An^{219}	3.92 sec	6.824 (82%)
83	RaE^{210}	5.0 d	β⁻	84	AcA^{215}	1.83×10^{-3} sec	7.365
84	Po^{210}	140 d	5.300	82	AcB^{211}	36.1 m	β⁻
82	Pb^{206}	Stable		83	AcC^{211}	2.16 m	6.619 (84%) 6.273 (16%) 0.32% β⁻
96	Cm^{242}	150 d	6.1	84	AcC'^{211}	5×10^{-3} sec	7.434 (0.32%)
94	Pu^{238}	92 yr	5.50	81	AcC''^{207}	4.76 m	β⁻
92	U_{II}^{234}	2.35×10^5 yr	4.763	82	Pb^{207}	Stable	
	etc. as above (descendants of U^{234})						
96	Pu^{234}	8.5 hr	6.2 (1%) 99% K	92	U^{230}	20.8 d	5.85
				90	Th^{226}	30.9 m	6.30
94	Np^{234}	4.44 d	K	88	Ra^{222}	38 sec	6.51
92	U_{II}^{234}	2.35×10^5 yr	4.763	86	Em^{218}	0.019 sec	7.12
	etc. as above (descendants of U^{234})			84	RaC'^{214}	1.5×10^{-4} sec	7.680
					etc. as above (descendants of RaC')		

TABLE VI. ALPHA EMITTERS ARRANGED BY ELEMENT

Z	Nuclide	$T_{1/2}$	E(∝) and Branching Ratio
83	RaE^{210}	5.0 d	4.8 (10^{-4} - 10^{-5} %)
	AcC^{211}	2.16 m	6.619 (84%) 6.273 (16%)
	ThC^{212}	60.5 m	6.081 (9%) 6.042 (24%)
	Bi^{213}	47 m	6.05 (4%)
	RaC^{214}	19.7 m	5.505 (0.018%) 5.444 (0.022%)
84	Po^{206}	9 d	5.2 (10%)
	Po^{207}	5.7 hr	5.1 (0.01%)
	Po^{208}	3 yr	5.14
	Po^{210}	140 d	5.300
	AcC'^{211}	5×10^{-3} sec	7.434
	ThC'^{212}	2.6×10^{-6} sec	8.776
	Po^{213}	3×10^{-6} sec	8.336
	RaC'^{214}	1.5×10^{-4} sec	7.680
	AcA^{215}	1.83×10^{-3} sec	7.365
	ThA^{216}	0.158 sec	6.774
	RaA^{218}	3.05 m	5.998
85	At^{211}	7.5 h	5.94 (40%)
	At^{215}	Short	8.4
	At^{216}	54 sec	7.64
	At^{217}	0.018 sec	7.02
	At^{218}	Several sec?	6.63
86	Em^{218}	0.019 sec	7.12
	An^{219}	3.92 sec	6.824 (82%)
	Tn^{220}	54.5 sec	6.282
	Rn^{222}	3.825 d	5.486
87	Fr^{221}	4.8 m	6.30 (75% ?) 6.05 (?) (25% ?)
88	Ra^{222}	38 sec	6.51
	AcX^{223}	11.2 d	5.717 (55%) 5.606 (36%)
	ThX^{223}	3.64 d	5.681
	Ra^{226}	1622 yr	4.791
89	Ac^{225}	10.0 d	5.80
	Ac^{227}	13.5 yr	4.98 (1%)
	$Ms\,Th^{228}$	6.13 hr	4.5 (0.01% ?)
90	Th^{226}	30.9 m	6.30
	$RdAc^{227}$	18.9 d	6.049 (20%) 5.988 (25%) 5.764 (20%) 5.717 (15%)
	$RdTh^{228}$	1.90 yr	5.418 (83%) 5.333 (17%)
	Th^{229}	7000 yr	4.85 (~70%) 4.94 (~20%) 5.02 (~10%)
	Io^{230}	8.0×10^{4} yr	4.66
	Th^{232}	1.39×10^{10} yr	3.98
91	Pa^{229}	1.5 d	5.66
	Pa^{231}	3.45×10^{5} yr	5.012 (87%) 4.736 (13%)
92	U^{230}	20.8 d	5.85
	U^{232}	70 yr	5.31
	U^{233}	1.62×10^{5} yr	4.825
	U^{234} II	2.35×10^{5} yr	4.763
	AcU^{235}	8.91×10^{8} yr	4.396
	U^{238} I	4.498×10^{9} yr	4.180
93	Np^{237}	2.20×10^{6} yr	4.73
94	Pu^{234}	8.5 hr	6.2
	Pu^{236}	2.7 yr	5.75
	Pu^{238}	92 yr	5.50
	Pu^{239}	2.44×10^{4} yr	5.14
	Pu^{240}	~6000 yr	5.1
	Pu^{241}	ca. 10 yr	(ca. 0.002%)
95	Am^{241}	498 yr	5.46
96	Cm^{240}	29 d	6.3
	Cm^{242}	150 d	6.10

9. Typical Experiments Performed with the Pulse Analyzer

9.1 Detection[67] of minute amounts of Cm^{242} in the presence of large amounts of Am^{241}.--In this experiment, an attempt was made, first, to show that Cm^{242} could be prepared by Am^{241} (n,γ) $Am^{242} \xrightarrow{\beta} Cm^{242}$ [67,68] and second, to measure the cross-section of the reaction. The method used was to bombard an americium sample in the pile and then to measure the growth of Cm^{242} from the Am^{242} beta-decay. Since the activity of the Cm^{242} was far less than that of the Am^{241}, this measurement was most conveniently made by cutting out the americium activity by the use of mica. The problem was simplified by the fairly large energy separation (Cm^{242} - 6.10 Mev and Am^{241} - 5.46 Mev). The 6 to 1 collimator was used (0.24% of 50% geometry). With a mica absorber most of the Am^{241} alphas were cut out, while the Cm^{242} alphas came through (tested with a Cm^{242} standard). Since the Am^{241} activity was 3.43×10^6 d/m, the Am activity passing through the collimater holes was 4030 c/m. The total background at the Cm^{242} position was about 0.18 c/m, so that concentrations of Cm^{242} as low as 25 parts in 10^6 (by activity) could be barely detected. Figure 9 shows the growth of the Cm^{242} activity following the neutron bombardment.

9.2 Detection of small concentrations of Pu^{236} in the presence of Pu^{238}.--This determination arose in a measurement of the half-life of Pu^{238} by direct decay measurements.[69] The Pu^{238} was formed in the bombardment of normal uranium by the reaction[70,71] U^{238} (d,2n) $Np^{238} \xrightarrow{\beta} Pu^{238}$. Samples

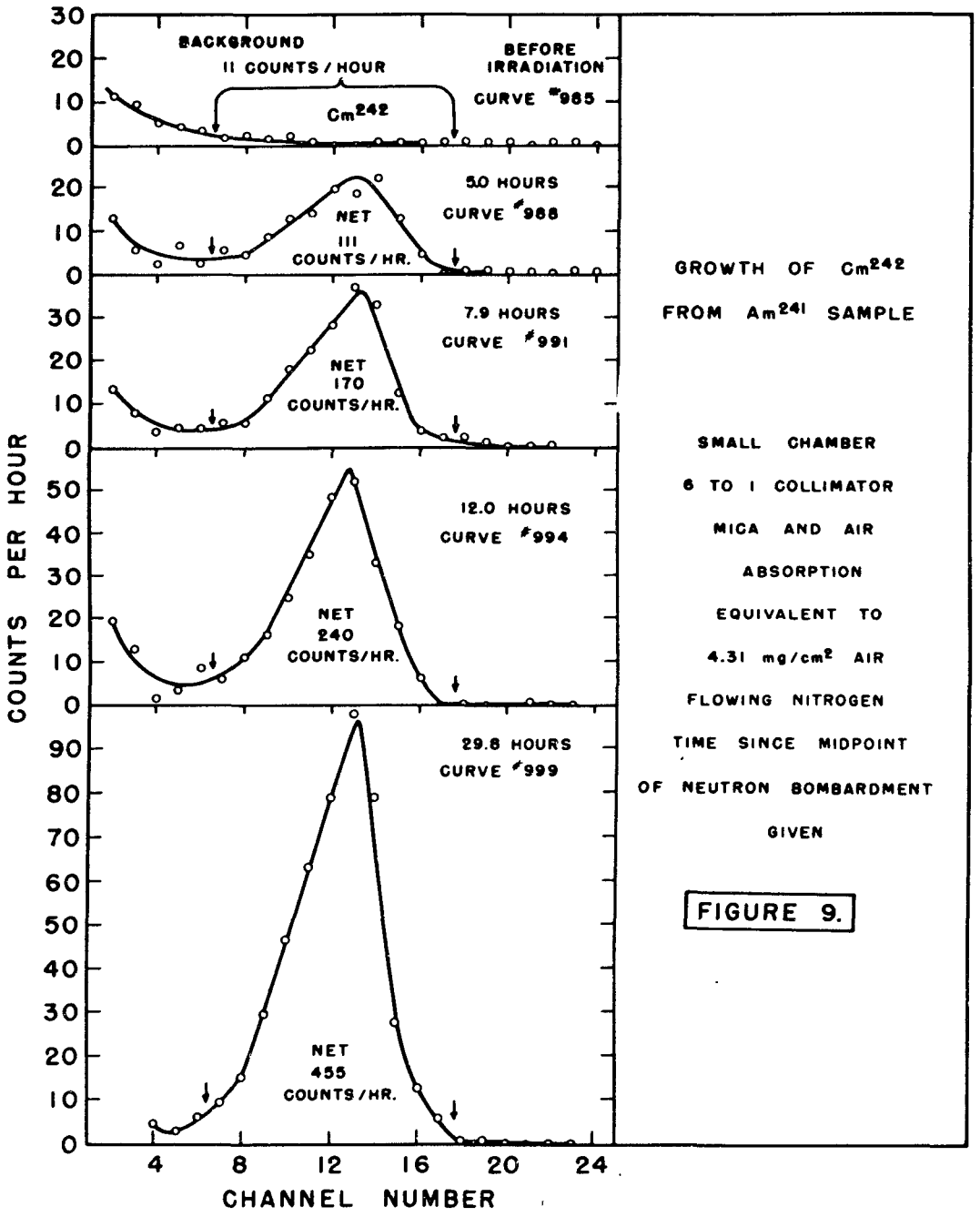

BACKGROUND
11 COUNTS / HOUR
BEFORE IRRADIATION
CURVE #985
Cm242
5.0 HOURS
CURVE #988
NET
111
COUNTS / HR.
7.9 HOURS
CURVE #991
NET
170
COUNTS/HR.
12.0 HOURS
CURVE #994
NET
240
COUNTS/HR.
29.8 HOURS
CURVE #999
NET
455
COUNTS/HR.
COUNTS PER HOUR
CHANNEL NUMBER
GROWTH OF Cm242
FROM Am241 SAMPLE
SMALL CHAMBER
6 TO 1 COLLIMATOR
MICA AND AIR
ABSORPTION
EQUIVALENT TO
4.31 mg/cm2 AIR
FLOWING NITROGEN
TIME SINCE MIDPOINT
OF NEUTRON BOMBARDMENT
GIVEN

FIGURE 9.

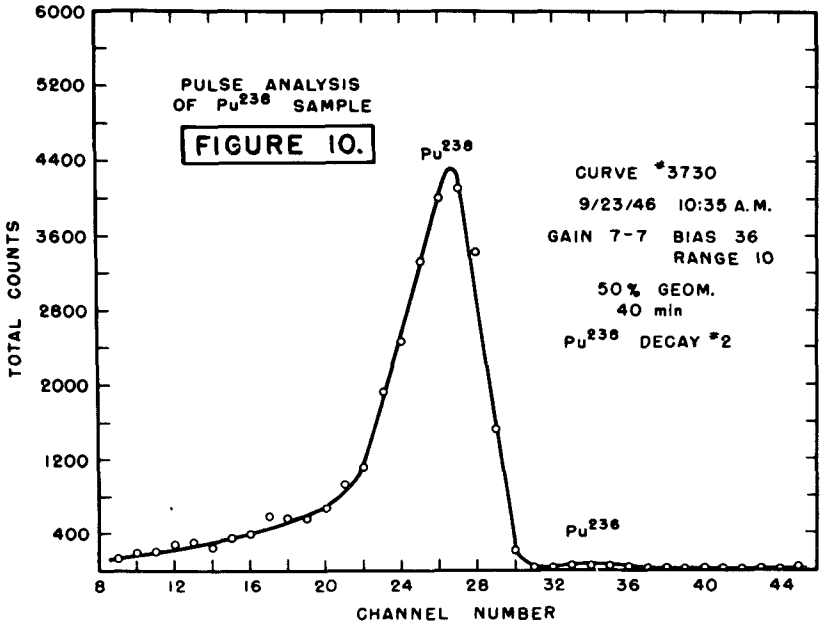


FIGURE 10.

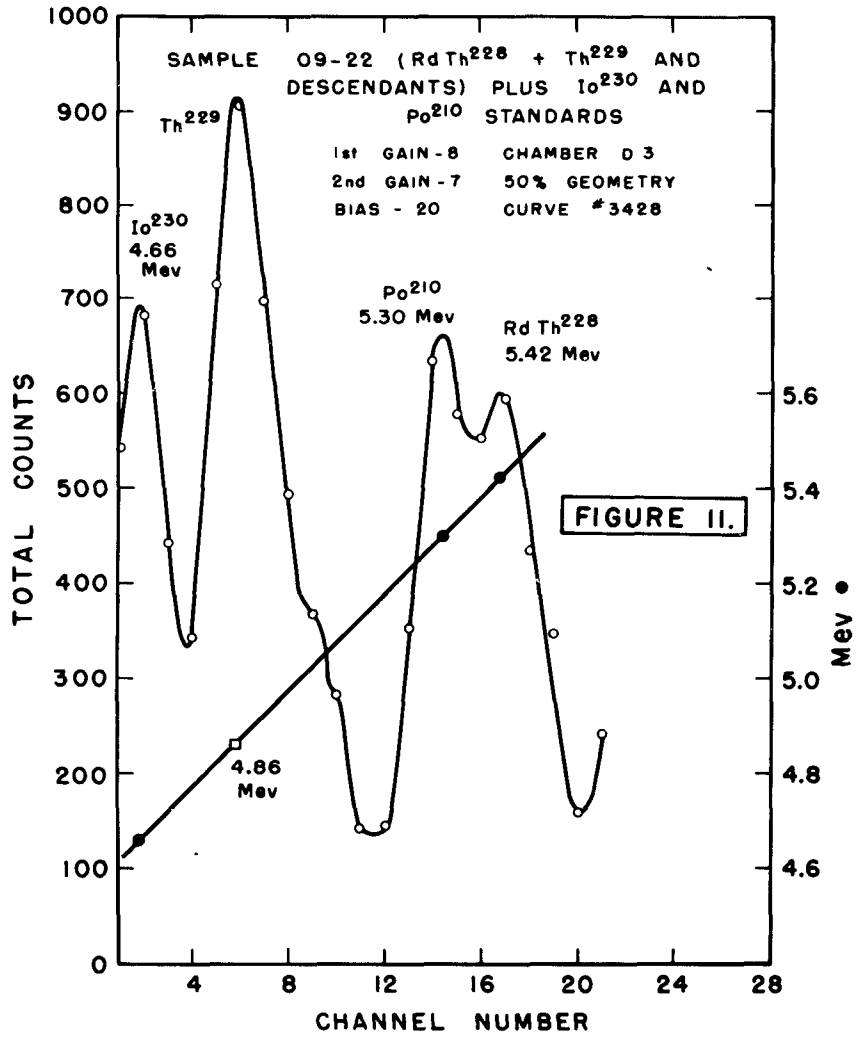


FIGURE 11.

containing some solid carrier were counted in a parallel plate ionization chamber and were followed for almost three years. It wasn't until near the end of this time that it was found that the Pu^{238} sample probably contained Pu^{236} as well, [72] probably formed by the reaction U^{235} (d,n) $Np^{236} \xrightarrow{\beta} Pu^{236}$. The relatively short half-life (2.7 years[72]) of the alpha-emitting Pu^{236} caused it to interfere seriously with the decay measurement, although it was present in low concentration. The measurement was salvaged by determining the Pu^{236} concentration with the pulse analyzer. Because of the large low energy tail due to sample thickness, this determination was made feasible only because the Pu^{236} energy (5.75 Mev) was greater than that of Pu^{238} (5.50 Mev). Figure 10 shows a typical pulse analysis curve. The analysis had to be made at 50% geometry, because the total activity of the sample was only 750 c/m. Since it was not possible to eliminate the low energy tails by collimation, the relative concentrations were determined by adding up the counts in the six or seven channels in the neighborhood of each peak, on the assumption that the low energy tails of both peaks were similar in shape. The results of a number of runs gave a Pu^{236} to Pu^{238} activity ratio of $0.0045 \pm .0007$ (probable error), which resulted in a Pu^{238} half-life measurement of 89 ± 9 years (most of the error arising from the uncertainty in the Pu^{236} concentration). This value agreed with the more accurate value 92 ± 2 years determined by another method.[73]

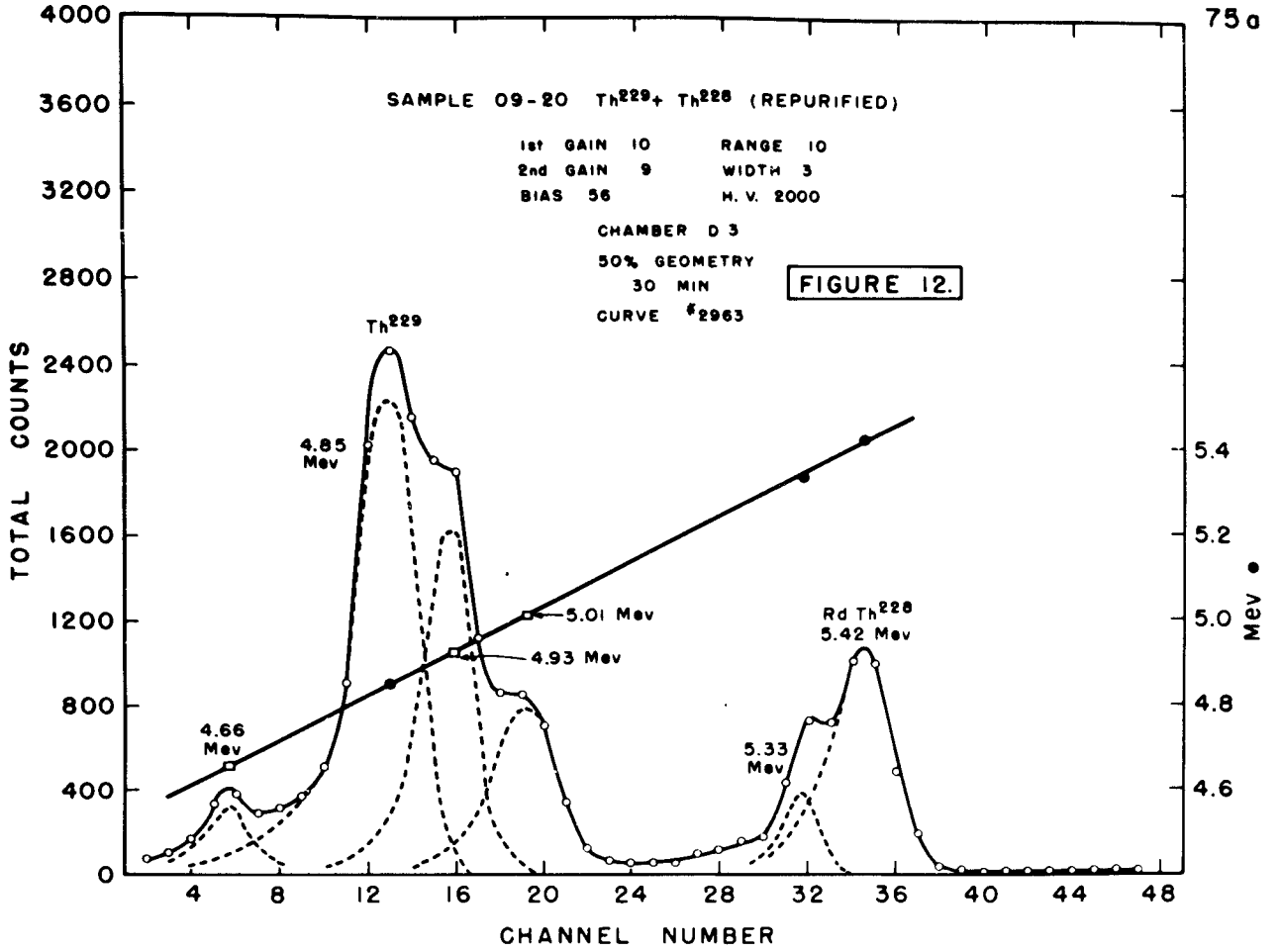


FIGURE 12.

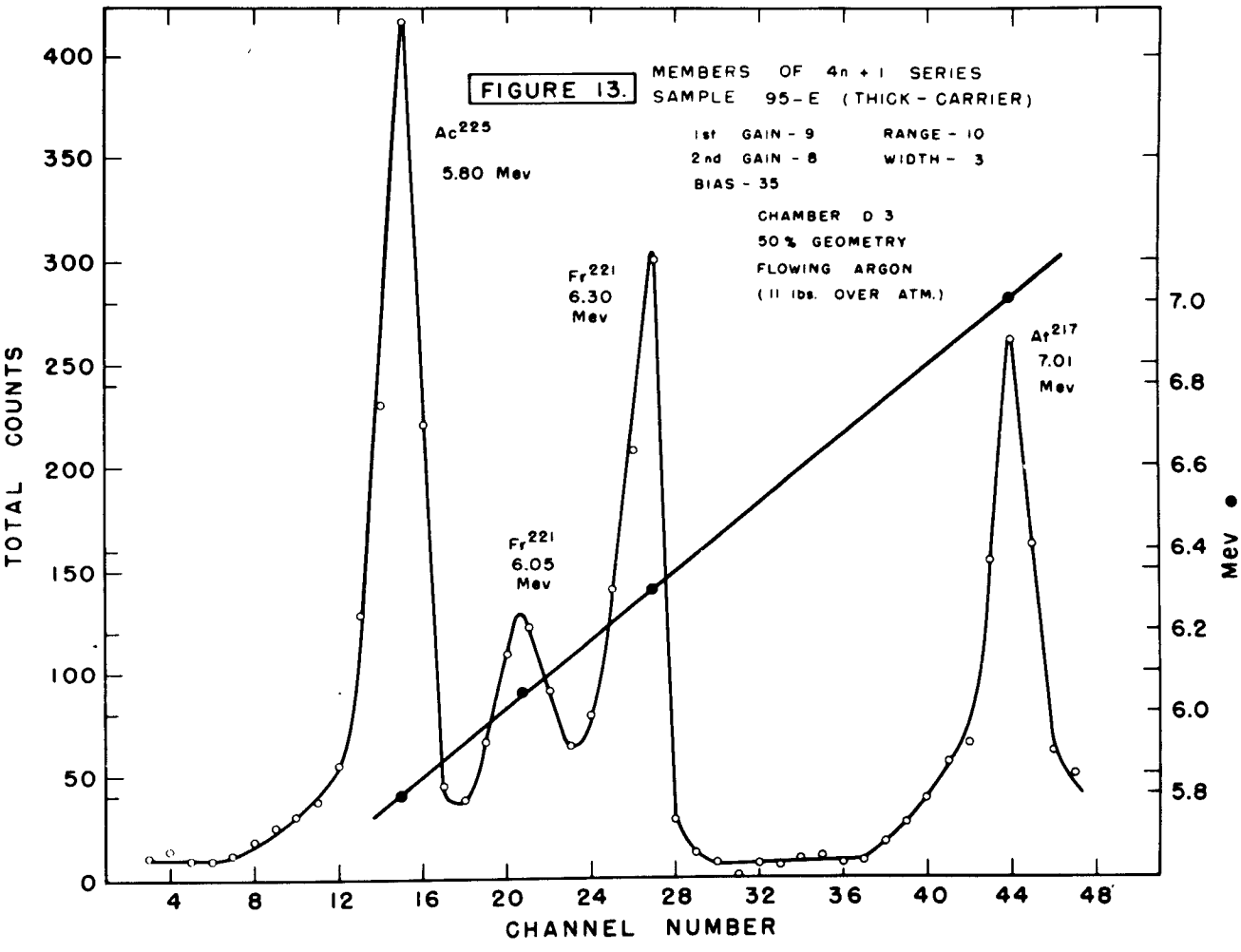


FIGURE 13.

9.3 Alpha energies in the 4n+ 1 Series.(74)--These energies are listed in Table V. They were measured by reference to alpha standards of known energy. In Fig. 11 is shown a typical pulse analysis in which the energy of the main peak of Th^{229} was measured by bracketting it with the standards: Io (Th^{230}), Po^{210} and RdTh (Th^{228}) Although this particular curve resulted in the value 4.86 Mev, the average of seven such curves was 4.85 ± .01* Mev.(89). The energies of the other Th^{229} peaks were determined (Fig. 12) using as standards the 4.85 Mev. main peak and the $RdTh^{228}$ peaks. The average of twelve curves gave(89) 4.94 ± .01*Mev, and 5.02 ± .01* Mev. The sample also showed a small peak at 4.66 Mev. which was not evident in later samples and may have been due to some Io^{230} contamination. Since Io is isotopic with Th^{229} and RdTh, it would not have separated during the chemical purification. Fig. 13 shows some of the other members of the series. The 6.05 Mev. peak has been assigned to Fr^{221} on the basis of abundance considerations. The energies corresponding to the peaks in Fig. 13 were measured relative to a MsTh series standard containing RdTh and daughters. The energies in Table V have been taken from these values and those measured at the Canadian laboratories.(75,76,77)

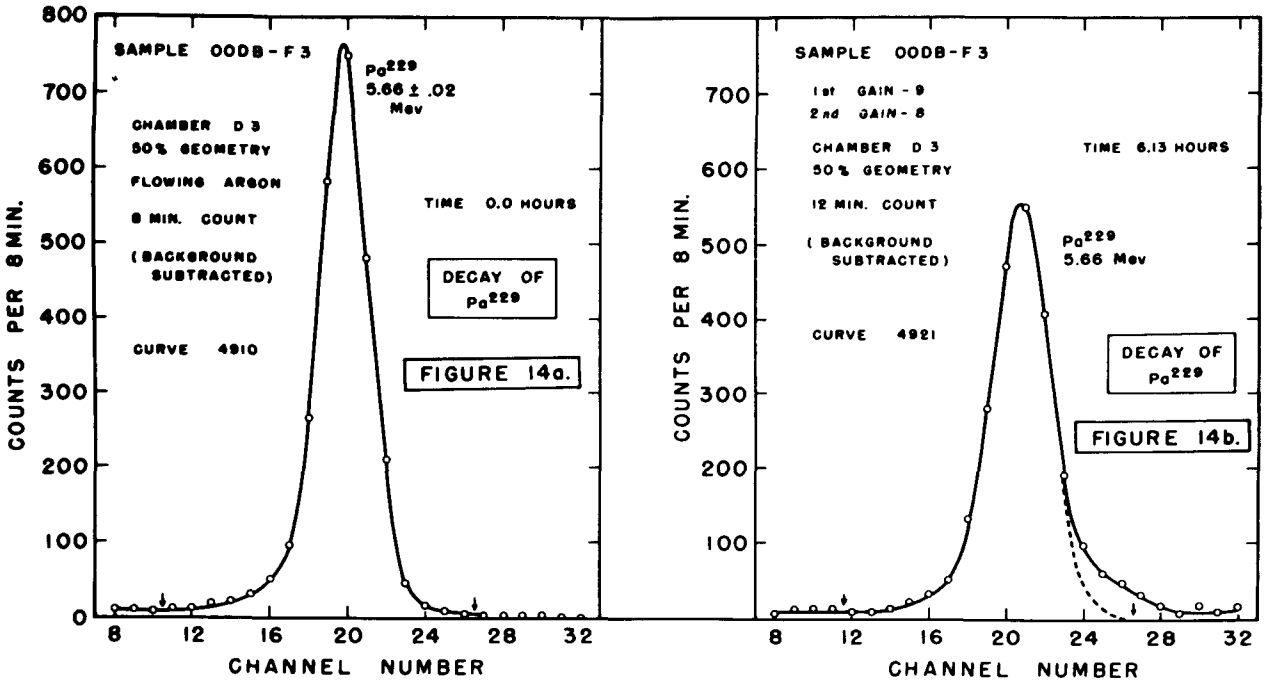
9.4 Measurement of decay. (Pa^{229})-- The pulse analyzer has proven to be quite useful in the determination of the half-life of a particular nuclide when it is present with

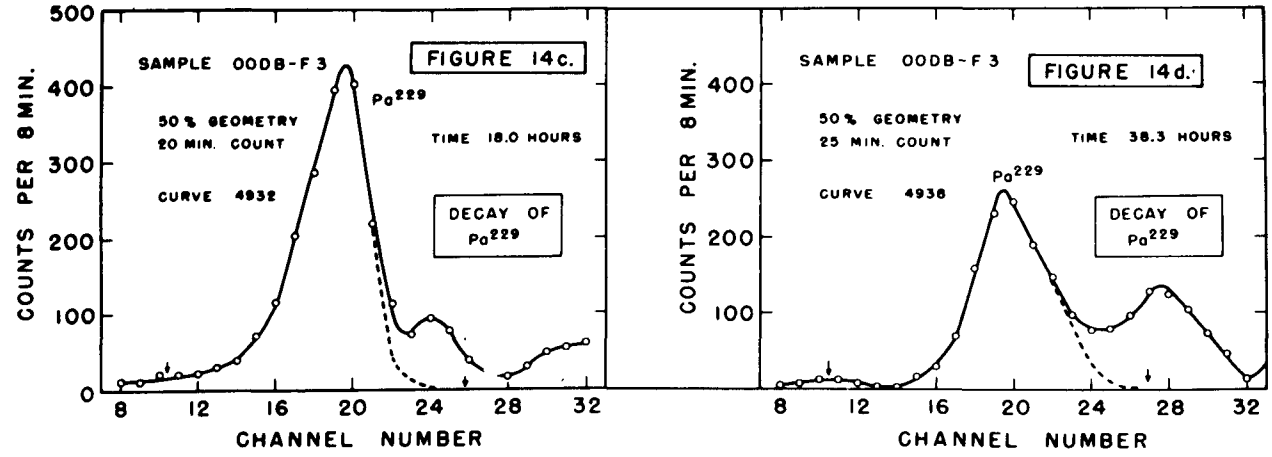
*Average deviation

a number of other growing and decaying alpha activities. Whereas, a gross decay curve would be almost worthless in such a case, the decay in the activity of a particular peak can be determined with reasonable accuracy. Such a determination was made[78,79] of the half-life of Pa^{229}. This nuclide was prepared by deuteron bombardment of an Io (Th^{230}) sample which contained about five times as much Th^{232} as Io. The products of this bombardment included all the protactinium isotopes which could be formed from Th^{232} and Io by (d, xn) reactions and by (d,p) reactions followed by beta-decay. With the 20 mev deuterons used, reactions up to (d,4n) occurred. Thus Pa^{233}, Pa^{232}, Pa^{231}, and Pa^{230} were formed[80,81] as well as Pa^{229}.

Immediately after the separation of the protactinium fraction from the bombardment mixture, the only significant alpha activity in the fraction was due to Pa^{229}, since Pa^{233} and Pa^{232} are beta-emitters, and the Pa^{231} activity was too low for observation due to its long half-life. However, the beta-active Pa isotopes, on decaying, gave rise to alpha-active descendants. The most serious difficulty was due to the formation of U^{230}, which immediately gave rise to the entire U^{230} series[81,82] (Table V). In addition, the decay of Pa^{229} resulted in the formation of a 10-day member of the (4n+1) series--Ac^{225} (See Table V), so that members of this series soon began to appear as well. Figs. 14a-d show the decay of the Pa^{229} peak. Due to the presence of overlapping peaks, the high energy side of the Pa^{229} peak had to be constructed. This procedure gave rise to an error which became more serious

9b


SAMPLE OODB-F3
Pa^{229}
5.66 ± .02
Mev
CHAMBER D 3
50% GEOMETRY
FLOWING ARGON
8 MIN. COUNT
(BACKGROUND SUBTRACTED)
CURVE 4910
TIME 0.0 HOURS
DECAY OF Pa^{229}
FIGURE 14a.
COUNTS PER 8 MIN.
CHANNEL NUMBER
SAMPLE OODB-F3
1st GAIN - 9
2nd GAIN - 8
CHAMBER D 3
50% GEOMETRY
12 MIN. COUNT
(BACKGROUND SUBTRACTED)
CURVE 4921
TIME 6.13 HOURS
Pa^{229}
5.66 Mev
DECAY OF Pa^{229}
FIGURE 14b.
COUNTS PER 8 MIN.
CHANNEL NUMBER

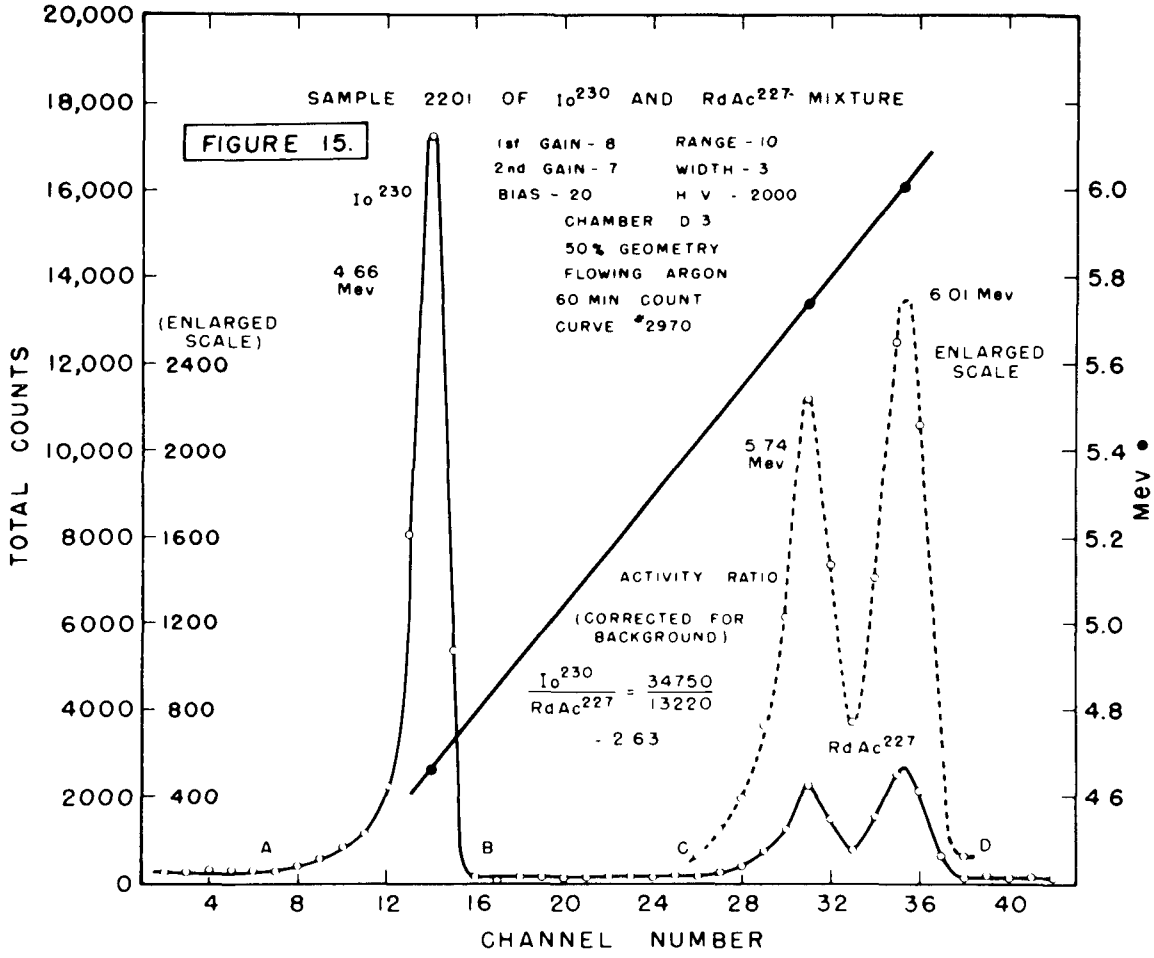

SAMPLE OODB-F3
FIGURE 14c.
Pa^{229}
50% GEOMETRY
20 MIN. COUNT
CURVE 4932
TIME 18.0 HOURS
DECAY OF Pa^{229}
COUNTS PER 8 MIN.
CHANNEL NUMBER
SAMPLE OODB-F3
FIGURE 14d.
50% GEOMETRY
25 MIN. COUNT
CURVE 4938
TIME 38.3 HOURS
Pa^{229}
DECAY OF Pa^{229}
COUNTS PER 8 MIN.
CHANNEL NUMBER

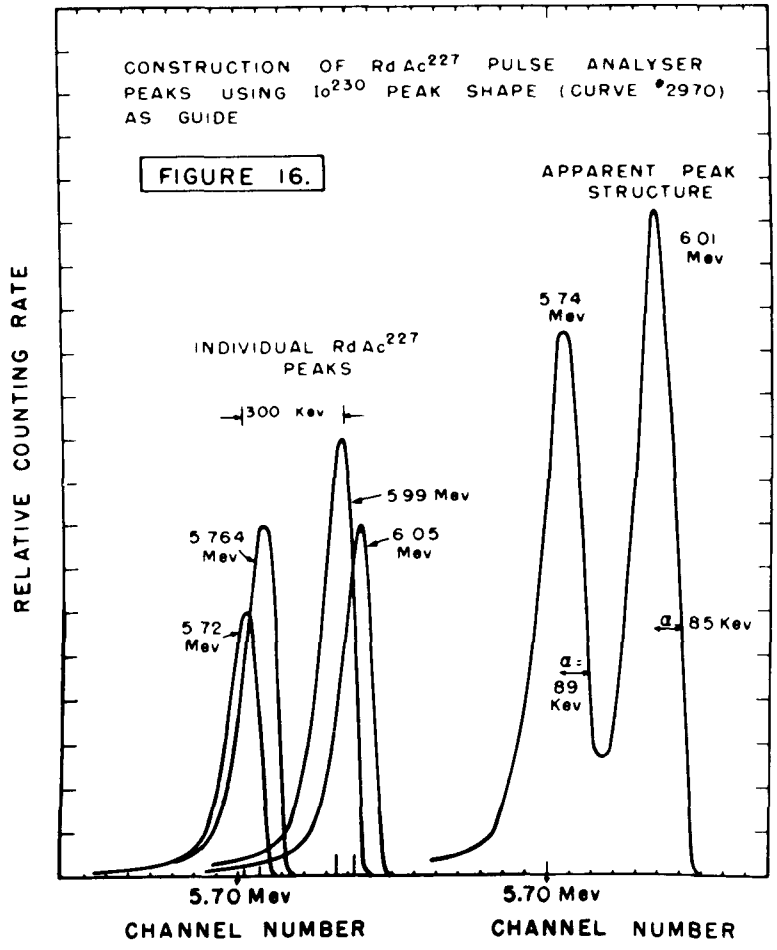
as time passed and the interfering peaks grew relatively larger. The arrows indicate the channels whose counts were added to give the Pa^{229} activity. The half-life was determined to be 1.50 days.(79) A number of energy measurements gave $5.66 \pm .02$ Mev.(79)

9.5 Determination of abundances by means of an internal standard. $RdAc^{227}$.--In the actinium decay series, the half-life of RdAc (Th^{227}) is difficult to determine, because its descendant AcX (Ra^{223}) has a comparable half-life (Table V). Previous half-life measurements had been made by evaluating the constants which would best fit the curves which represented the gross growth and decay of the total alpha activity.

The method by which the Pa^{229} decay was followed (Sec. 9.4), by measuring the decay of a single peak, could not be easily used here, because the daughter activity (AcX) overlaps the RdAc in alpha energy.

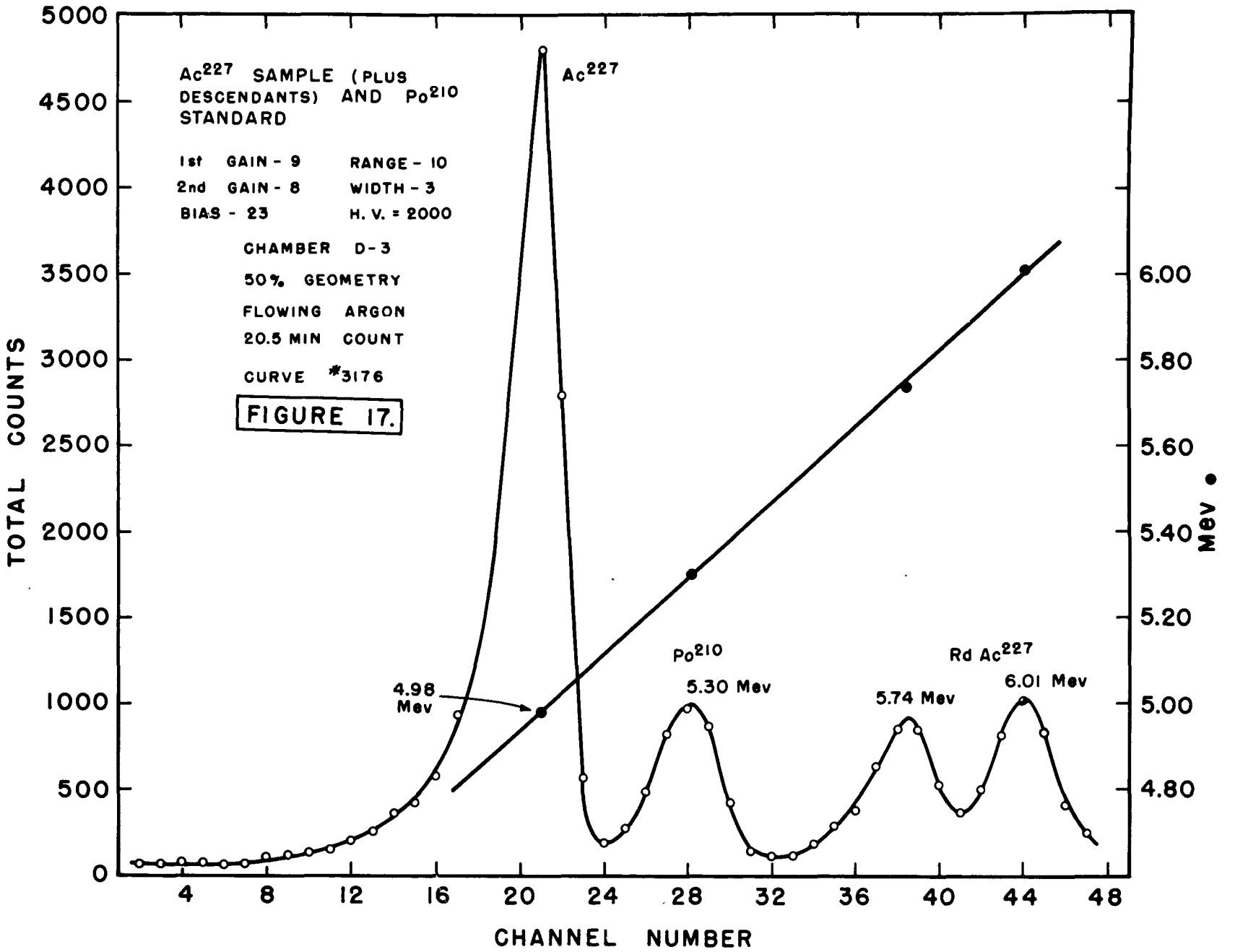
The method actually used(83) involved the periodic radiochemical isolation of RdAc from a solution containing a decaying sample, and the measurement of this RdAc(Th^{227}) activity with the pulse analyzer. To make automatic corrections for chemical losses in separation and purification, an internal standard (Io, or Th^{230}) was added to the decaying sample. Thus, to get the RdAc decay curve, it was only necessary to measure the ratio of RdAc to Io activity in each isolated thorium sample. Fig. 15 shows one of the curves used in this determination.


SAMPLE 2201 OF Io230 AND RdAc227 MIXTURE
FIGURE 15.
1st GAIN - 8
2nd GAIN - 7
BIAS - 20
RANGE - 10
WIDTH - 3
H V - 2000
CHAMBER D 3
50% GEOMETRY
FLOWING ARGON
60 MIN COUNT
CURVE #2970
Io230
4 66 Mev
(ENLARGED SCALE)
6 01 Mev
ENLARGED SCALE
5 74 Mev
ACTIVITY RATIO
(CORRECTED FOR BACKGROUND)
Io230 / RdAc227 = 34750 / 13220
- 2 63
RdAc227
A
B
C
D
TOTAL COUNTS
Mev
CHANNEL NUMBER


CONSTRUCTION OF RdAc227 PULSE ANALYSER PEAKS USING Io230 PEAK SHAPE (CURVE #2970) AS GUIDE
FIGURE 16.
APPARENT PEAK STRUCTURE
6 01 Mev
5 74 Mev
INDIVIDUAL RdAc227 PEAKS
300 Kev
5 99 Mev
6 05 Mev
5 764 Mev
5 72 Mev
α = 89 Kev
α = 85 Kev
RELATIVE COUNTING RATE
5.70 Mev
CHANNEL NUMBER
5.70 Mev
CHANNEL NUMBER

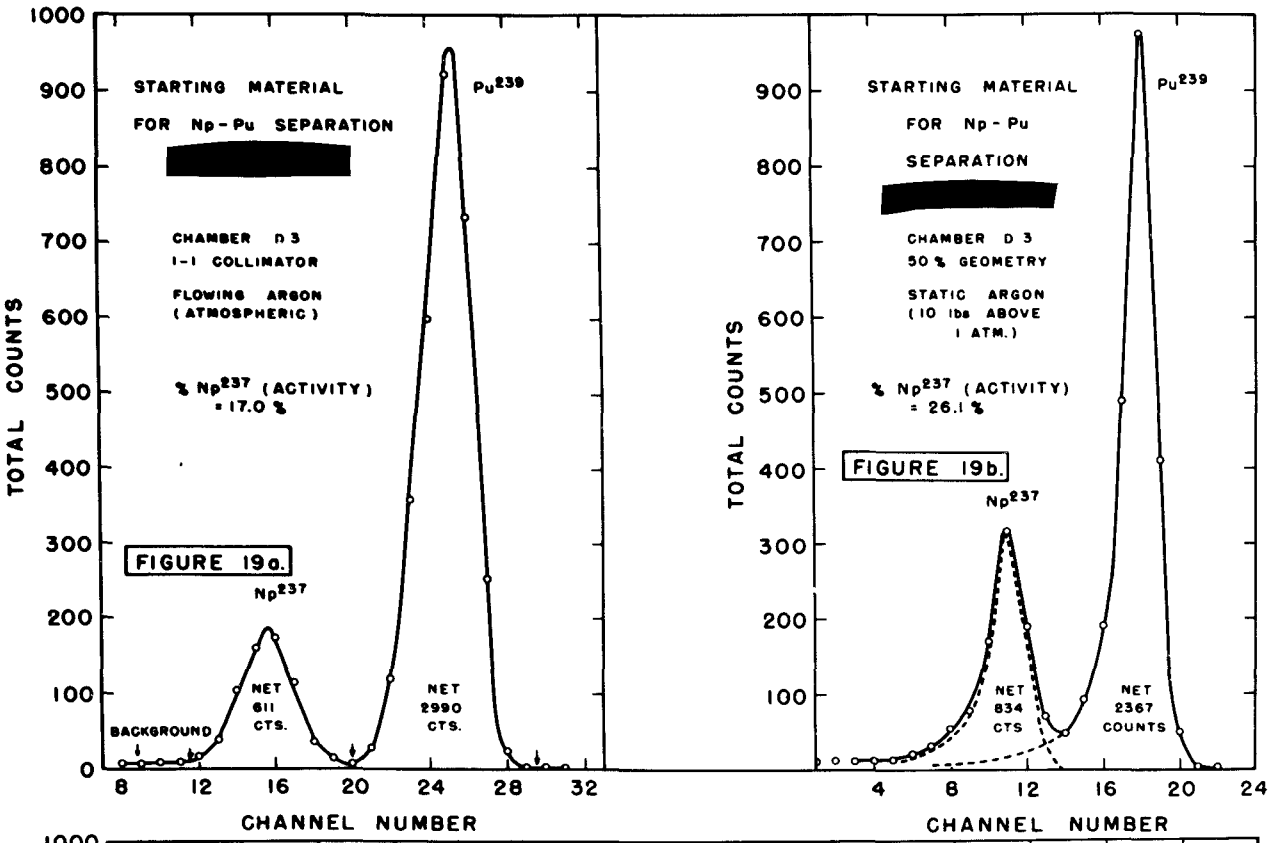
Although Table IV lists four energies for RdAc (measured by the magnetic spectrograph), only two peaks are evident in Fig. 15. Assuming that the straggling of the RdAc peaks was the same as that of the Io peak, Fig. 16 was constructed. The energies and relative abundances measured with the magnetic spectrograph were used to construct four individual peaks with the same shape as that of the Io peak (Fig. 15). Adding up the points of the four peaks, it can be seen that two peaks result which match quite well with those in Fig. 15. The RdAc peaks in Fig. 15 are drawn on an enlarged scale to make the likeness more evident. The constructed RdAc peaks (Fig. 16) seem to have better resolution than the actual ones (Fig. 15), since the valley between the two peaks is deeper in Fig. 16. This discrepancy indicates that the straggling of the RdAc alphas is not the same as that of the Io alphas, as was assumed, but is actually larger.

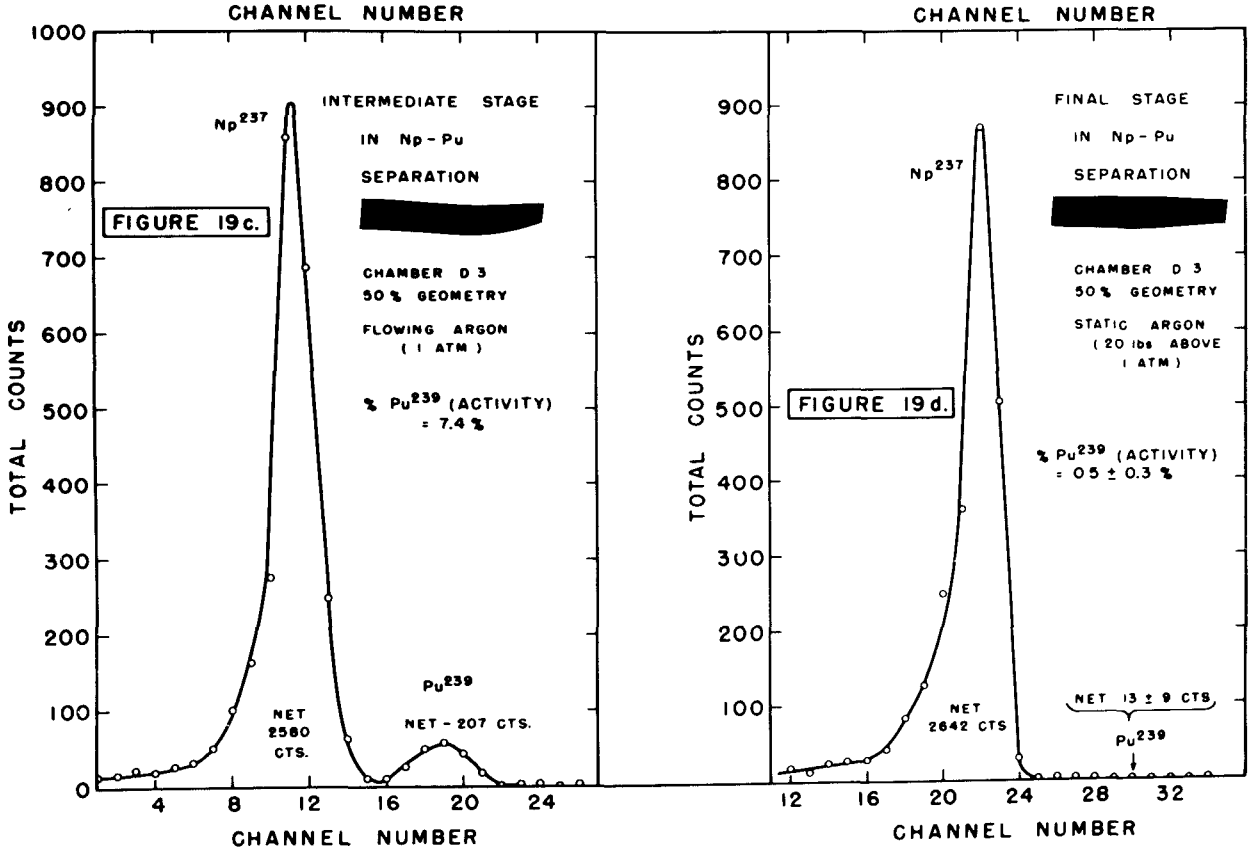
9.6 Energy of Ac^{227}.-- In an experiment in which the alpha-branching of Ac^{227} was determined,[84] the Ac alpha energy was also measured by the use of the standards Po^{210} and the daughter of the Ac^{227} beta-decay RdAc (Th^{227}). Fig. 17 shows the results of the measurement. Extensive sample straggling is quite evident in this figure, and is due to the use of a carrier-containing sample at 50% geometry.


Ac227 SAMPLE (PLUS DESCENDANTS) AND Po210 STANDARD
1st GAIN - 9
RANGE - 10
2nd GAIN - 8
WIDTH - 3
BIAS - 23
H. V. = 2000
CHAMBER D-3
50% GEOMETRY
FLOWING ARGON
20.5 MIN COUNT
CURVE #3176
FIGURE 17.
Ac227
4.98 Mev
Po210
5.30 Mev
5.74 Mev
Rd Ac227
6.01 Mev
TOTAL COUNTS
CHANNEL NUMBER
Mev
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
4
8
12
16
20
24
28
32
36
40
44
48
6.00
5.80
5.60
5.40
5.20
5.00
4.80

9.7 Tracing chemical separations. Am^{241} and Pu^{239}, Np^{237} and Pu^{239}.-- In working with the alpha-emitting heavy nuclides (e.g. Pu^{239}), the efficiency of particular chemical separation procedures is most conveniently tested by following the gross alpha-activity. Where two or more alpha-emitters are being separated, measurement of the gross alpha-activity cannot be used, and the pulse analyzer is used to follow the separation.

Figs. 19a-d show the concentration[87,88] of Np^{237} from considerably larger amounts (by activity) of Pu^{239}. Fig. 19a shows the peak separation when a sample of starting material is collimated, while Fig. 19b shows a sample of another batch of starting material measured at 50% geometry. Measurement of the activity of the low concentration Np^{237} is evidently easier with a collimated sample than at 50% geometry. Low concentrations of a higher energy constituent can be measured with or without collimation Fig. 19d).


STARTING MATERIAL
FOR Np - Pu SEPARATION
[redacted]
CHAMBER D 3
1-1 COLLIMATOR
FLOWING ARGON
(ATMOSPHERIC)
% Np237 (ACTIVITY)
= 17.0 %
FIGURE 19a.
Np237
Pu239
BACKGROUND
NET
611
CTS.
NET
2990
CTS.
TOTAL COUNTS
CHANNEL NUMBER
STARTING MATERIAL
FOR Np - Pu
SEPARATION
[redacted]
CHAMBER D 3
50 % GEOMETRY
STATIC ARGON
(10 lbs ABOVE
1 ATM.)
% Np237 (ACTIVITY)
= 26.1 %
FIGURE 19b.
Np237
Pu239
NET
834
CTS
NET
2367
COUNTS
TOTAL COUNTS
CHANNEL NUMBER


INTERMEDIATE STAGE
IN Np - Pu
SEPARATION
[redacted]
CHAMBER D 3
50 % GEOMETRY
FLOWING ARGON
(1 ATM)
% Pu239 (ACTIVITY)
= 7.4 %
FIGURE 19c.
Np237
Pu239
NET
2580
CTS.
NET - 207 CTS.
TOTAL COUNTS
CHANNEL NUMBER
FINAL STAGE
IN Np - Pu
SEPARATION
[redacted]
CHAMBER D 3
50 % GEOMETRY
STATIC ARGON
(20 lbs ABOVE
1 ATM)
% Pu239 (ACTIVITY)
= 0.5 ± 0.3 %
FIGURE 19d.
Np237
NET
2642 CTS
NET 13 ± 9 CTS.
Pu239
TOTAL COUNTS
CHANNEL NUMBER

9.8 Products of Alpha-bombardment of uranium isotopes.-- The nuclides formed by high energy (~40 Mev) helium-ion bombardment of a mixture of U^{235} and U^{238} include[72] the alpha-emitters Pu^{239}, Pu^{238} and Pu^{236}. These are formed by (α, χn) reactions on both U^{235} and U^{238} (χ varies from 1 to 6). Since these nuclides are isotopic, identification and measurements of concentration could be carried out most conveniently by an instrument like the pulse analyzer. The results of bombarding a sample enriched in U^{235}, but containing a considerable amount of U^{238}, are shown in Fig. 20.

9.9 The Protactinium series.[81]--Pa^{230} is formed in the deuteron bombardment of Th^{232} by the reaction[81] Th^{232}(d,4n), in the bombardment of Io (Th^{230}) by the reaction[78] Th^{230} (d, 2n), and in the bombardment of Pa^{231} by the reaction[82] Pa^{231} (d,p2n). U^{230} is formed by the beta-decay of Pa^{230}, and in a series of alpha-decay steps, Th^{226}, Ra^{222}, Em^{218} and RaC' (Po^{214}) are formed. Since all of the alpha-emitters but U^{230} are short-lived, a sample of U^{230} soon has all five of the alphas in equilibrium. The U^{230} sample (and daughters in equilibrium) shown in Fig. 21 were separated from Pa^{230} formed in the deuteron bombardment of Pa^{231}. The energy of U^{230} was measured relative to the energies of Pa^{231} and U^{232} (also formed in the Pa^{231} bombardment), and also by the use of a RdTh (and daughters) internal standard. It was found[89] to be 5.85 $\pm$ .01 Mev.* Since the alpha with highest energy is

*Average deviation of nine determinations.

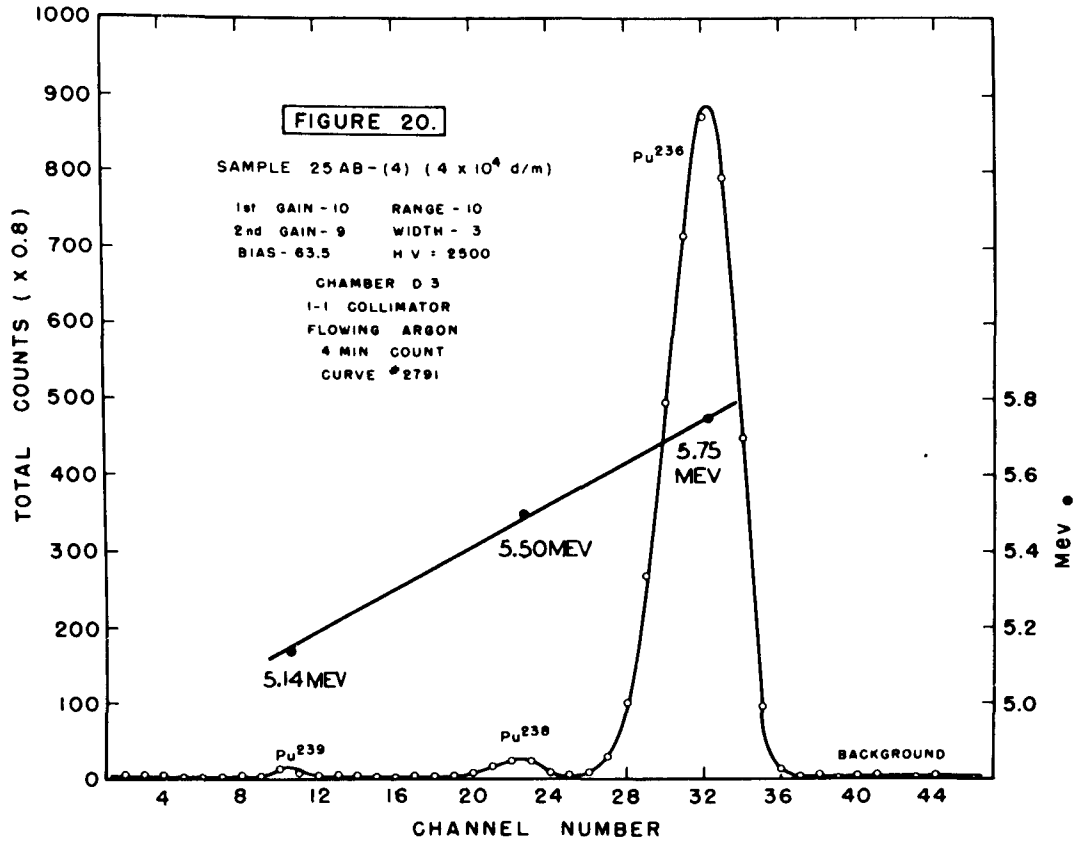


FIGURE 20.
SAMPLE 25 AB-(4) (4 x 10⁴ d/m)
1st GAIN - 10 RANGE - 10
2nd GAIN - 9 WIDTH - 3
BIAS - 63.5 H V = 2500
CHAMBER D 3
1-1 COLLIMATOR
FLOWING ARGON
4 MIN COUNT
CURVE #2791
TOTAL COUNTS (X 0.8)
CHANNEL NUMBER
Mev
Pu²³⁶
Pu²³⁹
Pu²³⁸
BACKGROUND
5.14 MEV
5.50 MEV
5.75 MEV

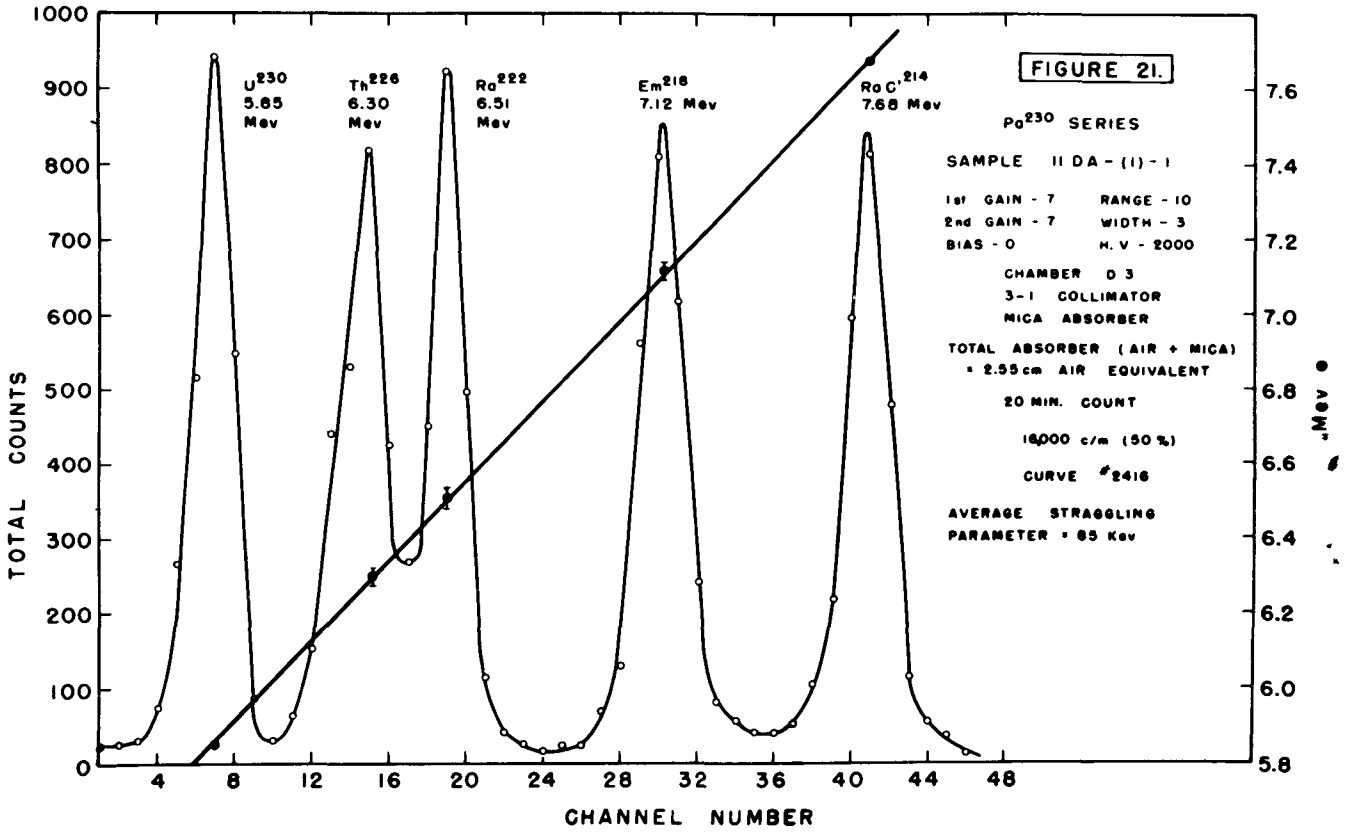


FIGURE 21.
Po²³⁰ SERIES
SAMPLE II DA-(1)-1
1st GAIN - 7 RANGE - 10
2nd GAIN - 7 WIDTH - 3
BIAS - 0 H. V - 2000
CHAMBER D 3
3-1 COLLIMATOR
MICA ABSORBER
TOTAL ABSORBER (AIR + MICA)
= 2.55 cm AIR EQUIVALENT
20 MIN. COUNT
16,000 c/m (50 %)
CURVE #2416
AVERAGE STRAGGLING
PARAMETER = 85 Kev
TOTAL COUNTS
CHANNEL NUMBER
Mev
U²³⁰ 5.65 Mev
Th²²⁶ 6.30 Mev
Ra²²² 6.51 Mev
Em²¹⁸ 7.12 Mev
RaC'²¹⁴ 7.68 Mev

due to RaC', whose energy (7.68 Mev) has been accurately measured with the magnetic spectrograph, it was used as an internal standard. The energies of Th^{226}, Ra^{222} and Em^{218} were determined using U^{230} and RaC' as standards; Ra^{222} and Em^{218} were also measured using RdTh (and daughters) as an internal standard. The energies determined[89] were Th^{226}, $6.30 \pm .025$ Mev*; Ra^{222}, $6.51 \pm .03$ Mev*; Em^{218}, $7.12 \pm .025$ Mev*. The assignment of these peaks to the particular nuclides was based upon the Geiger-Nuttall law.

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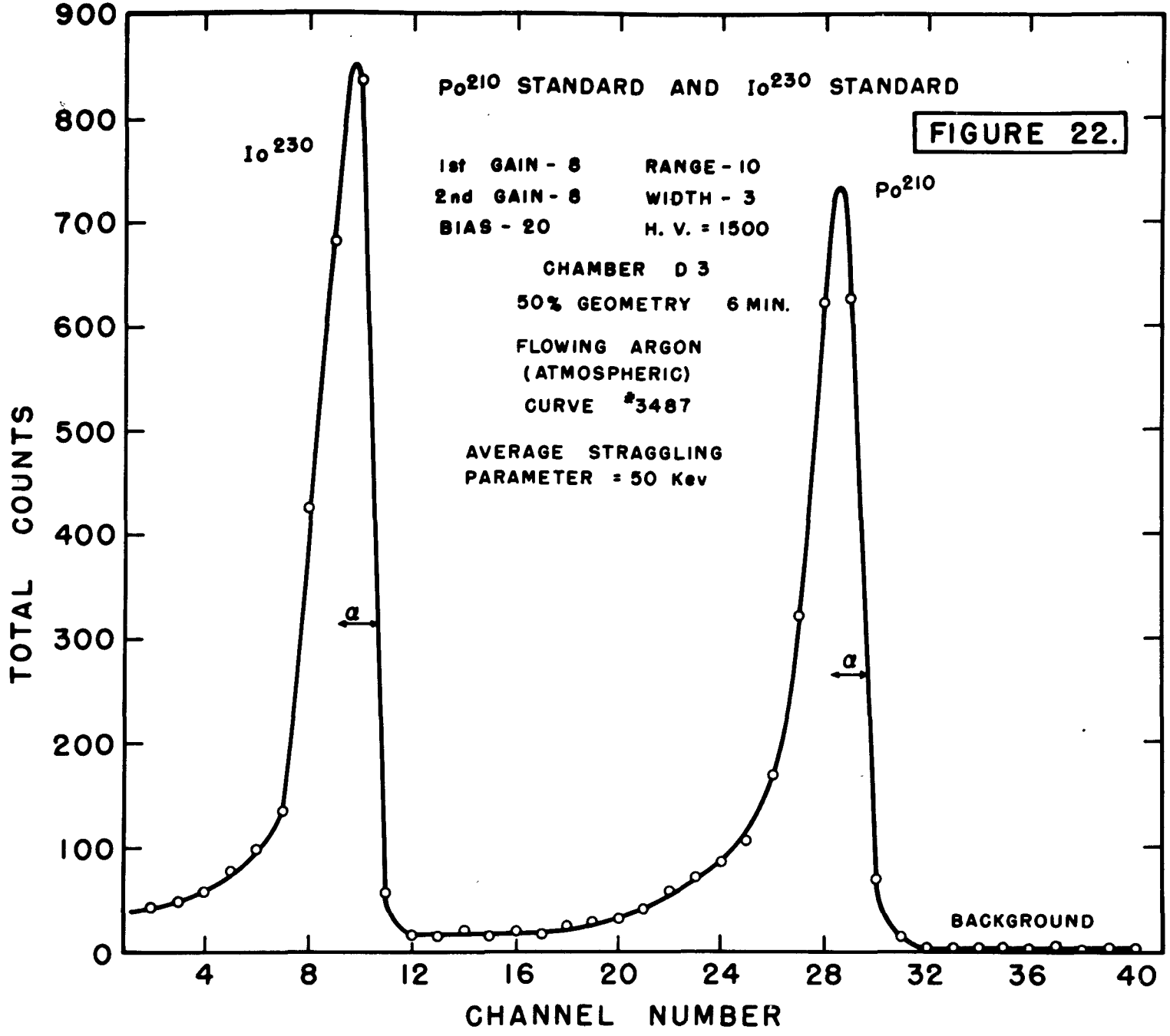
The relatively large average straggling parameter (85 Kev) is due partly to the range straggling (and hence straggling in the residual ionization) introduced by the mica and partly to the relatively low collimation (3 to 1) which allows angle straggling (variation in mica absorption with angle of emission) to occur.

9.10 Some Standard Samples.-- An Io (Th^{230}) and a Po^{210} standard are shown in Fig. 22. While the straggling parameters for the two peaks are approximately the same, the low energy straggling for the Po sample is worse than that of the Io^{230} sample. This phenomenon may be due to the same sort of gradual deterioration which has been reported by many other users of Po sources.

Figs. 23a-c show the effect of varying the number of channels per peak on the resolution. The straggling parameter is the same for Figs. 23a and 23b but is increased in Fig. 23c, where the peaks are crowded together. The spread of the peaks

*Average deviation of 20 determinations

100


Po210 STANDARD AND Io230 STANDARD
FIGURE 22.
Io230
Po210
1st GAIN - 8
2nd GAIN - 8
BIAS - 20
RANGE - 10
WIDTH - 3
H. V. = 1500
CHAMBER D 3
50% GEOMETRY 6 MIN.
FLOWING ARGON
(ATMOSPHERIC)
CURVE #3487
AVERAGE STRAGGLING
PARAMETER = 50 Kev
α
α
BACKGROUND
TOTAL COUNTS
CHANNEL NUMBER
0
100
200
300
400
500
600
700
800
900
4
8
12
16
20
24
28
32
36
40

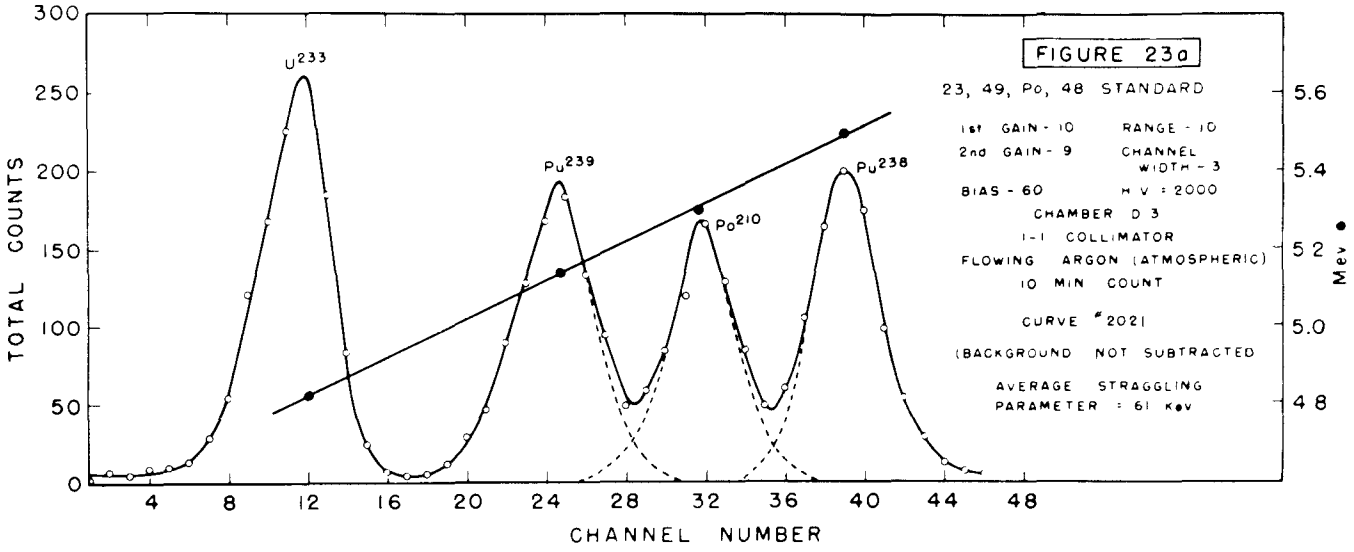


FIGURE 23a
23, 49, Po, 48 STANDARD
1st GAIN - 10
RANGE - 10
2nd GAIN - 9
CHANNEL WIDTH - 3
BIAS - 60
H V = 2000
CHAMBER D 3
1-1 COLLIMATOR
FLOWING ARGON (ATMOSPHERIC)
10 MIN COUNT
CURVE #2021
(BACKGROUND NOT SUBTRACTED
AVERAGE STRAGGLING PARAMETER = 61 Kev
U233
Pu239
Po210
Pu238
TOTAL COUNTS
CHANNEL NUMBER
Mev

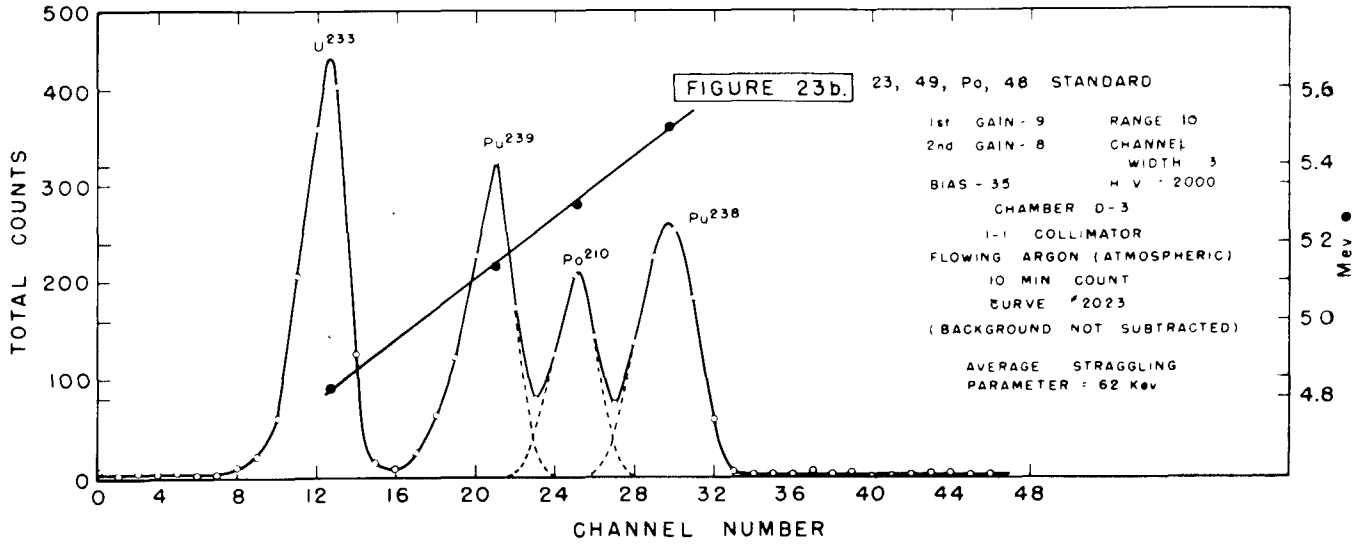


FIGURE 23b.
23, 49, Po, 48 STANDARD
1st GAIN - 9
RANGE 10
2nd GAIN - 8
CHANNEL WIDTH 3
BIAS - 35
H V - 2000
CHAMBER D-3
1-1 COLLIMATOR
FLOWING ARGON (ATMOSPHERIC)
10 MIN COUNT
CURVE #2023
(BACKGROUND NOT SUBTRACTED)
AVERAGE STRAGGLING PARAMETER = 62 Kev
U233
Pu239
Po210
Pu238
TOTAL COUNTS
CHANNEL NUMBER
Mev

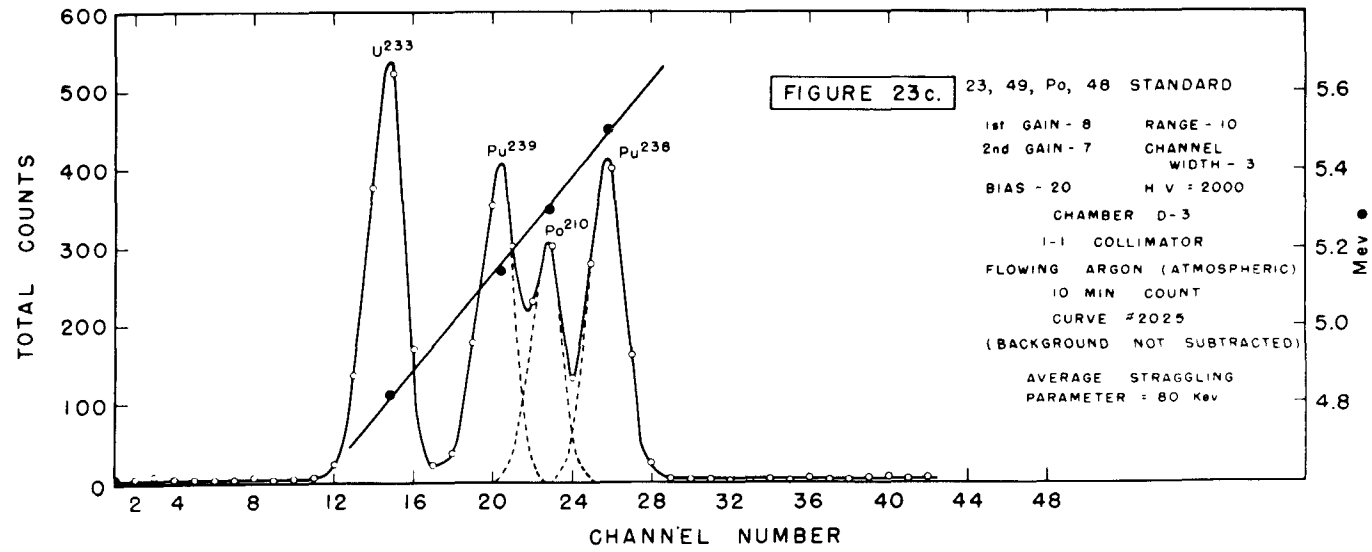


FIGURE 23c.
23, 49, Po, 48 STANDARD
1st GAIN - 8
RANGE - 10
2nd GAIN - 7
CHANNEL WIDTH - 3
BIAS - 20
H V = 2000
CHAMBER D-3
1-1 COLLIMATOR
FLOWING ARGON (ATMOSPHERIC)
10 MIN COUNT
CURVE #2025
(BACKGROUND NOT SUBTRACTED)
AVERAGE STRAGGLING PARAMETER = 80 Kev
U233
Pu239
Po210
Pu238
TOTAL COUNTS
CHANNEL NUMBER
Mev

over the channels was varied by shifting the positions of the gain controls (see Fig. 4).

Figs. 24a-c show the effect of collimation. The straggling parameters are about the same for 50% geometry and for 1 to 1 and 2 to 1 collimation. However, the low energy tails in the 50% geometry measurement decrease the resolution on the low energy side of each peak, as is evident from the fact that the valleys between the peaks are lower in Figs. 24b and 24c than in Fig. 24a. No significant difference seems evident between the results obtained with 1 to 1 or 2 to 1 collimation. This fact is also shown in Figs. 25a and 25b.

The peaks from a RdTh (and daughters) standard are shown in Fig. 26. The non-linearity of the energy-pulse height relation becomes significant only for the highest energy peak (ThC'). This non-linearity is probably due both to the positive ion effect and to amplifier non-linearity at high signal levels.

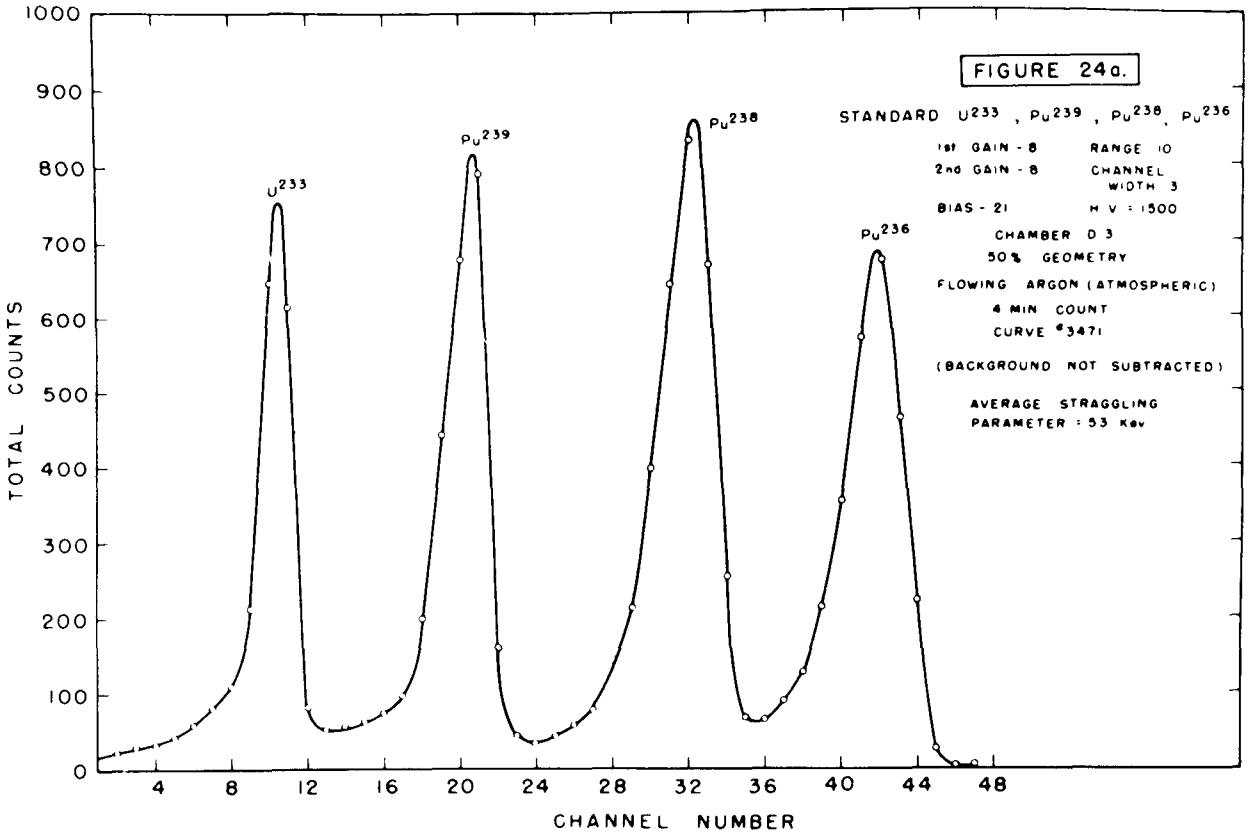


FIGURE 24a.
STANDARD U233 , Pu239 , Pu238, Pu236
1st GAIN - 8 RANGE 10
2nd GAIN - 8 CHANNEL WIDTH 3
BIAS - 21 H V = 1500
CHAMBER D 3
50% GEOMETRY
FLOWING ARGON (ATMOSPHERIC)
4 MIN COUNT
CURVE #3471
(BACKGROUND NOT SUBTRACTED)
AVERAGE STRAGGLING PARAMETER = 53 Kev
U233
Pu239
Pu238
Pu236
TOTAL COUNTS
CHANNEL NUMBER

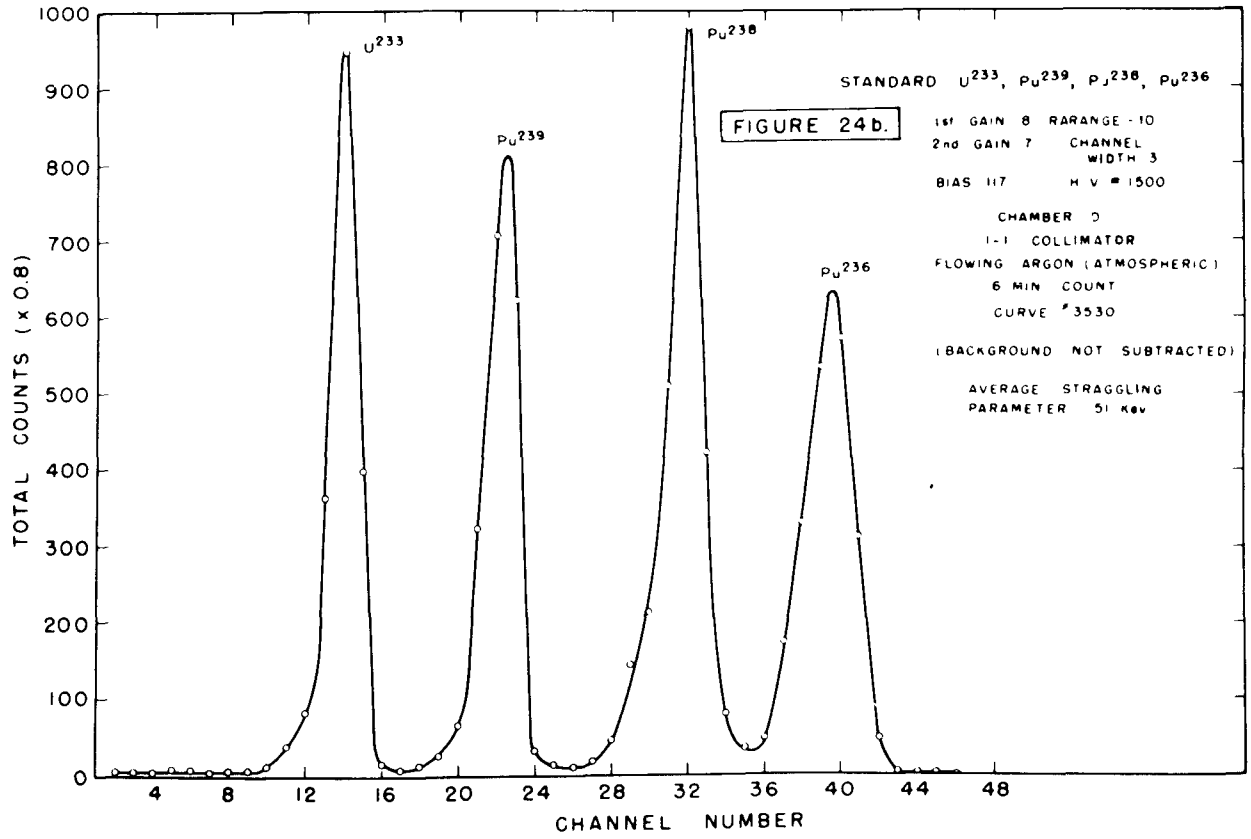
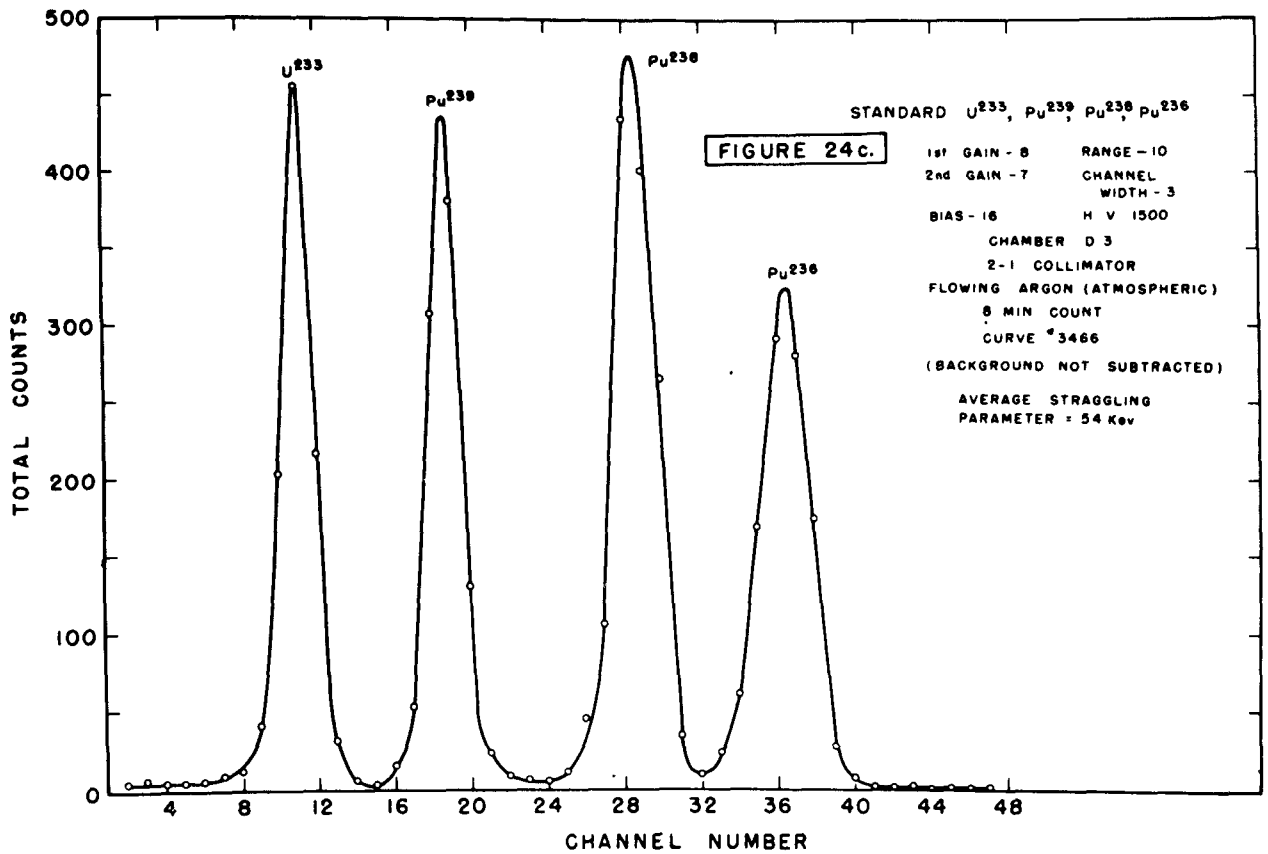


FIGURE 24b.
STANDARD U233, Pu239, Pu238, Pu236
1st GAIN 8 RARANGE - 10
2nd GAIN 7 CHANNEL WIDTH 3
BIAS 117 H V = 1500
CHAMBER D
1-1 COLLIMATOR
FLOWING ARGON (ATMOSPHERIC)
6 MIN COUNT
CURVE #3530
(BACKGROUND NOT SUBTRACTED)
AVERAGE STRAGGLING PARAMETER 51 Kev
U233
Pu239
Pu238
Pu236
TOTAL COUNTS (x 0.8)
CHANNEL NUMBER


500
400
300
200
100
0
TOTAL COUNTS
U233
Pu239
Pu238
Pu236
4
8
12
16
20
24
28
32
36
40
44
48
CHANNEL NUMBER
STANDARD U233, Pu239, Pu238, Pu236
FIGURE 24c.
1st GAIN - 8
RANGE - 10
2nd GAIN - 7
CHANNEL WIDTH - 3
BIAS - 16
H V 1500
CHAMBER D 3
2-1 COLLIMATOR
FLOWING ARGON (ATMOSPHERIC)
8 MIN COUNT
CURVE #3466
(BACKGROUND NOT SUBTRACTED)
AVERAGE STRAGGLING PARAMETER = 54 Kev

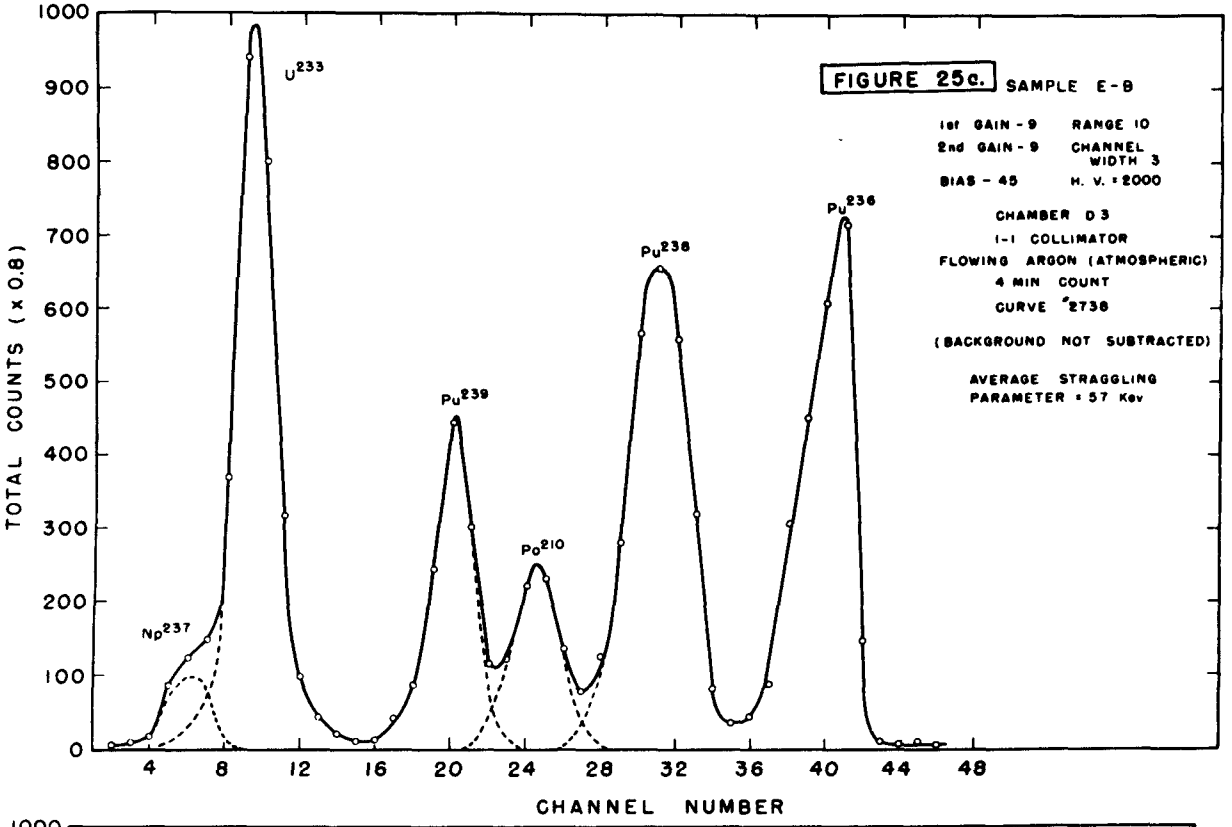


FIGURE 25c. SAMPLE E-8
1st GAIN - 9 RANGE 10
2nd GAIN - 9 CHANNEL WIDTH 3
BIAS - 45 H. V. = 2000
CHAMBER D 3
1-1 COLLIMATOR
FLOWING ARGON (ATMOSPHERIC)
4 MIN COUNT
CURVE #2736
(BACKGROUND NOT SUBTRACTED)
AVERAGE STRAGGLING PARAMETER = 57 Kev
TOTAL COUNTS (x 0.8)
CHANNEL NUMBER
U233
Np237
Pu239
Po210
Pu238
Pu236

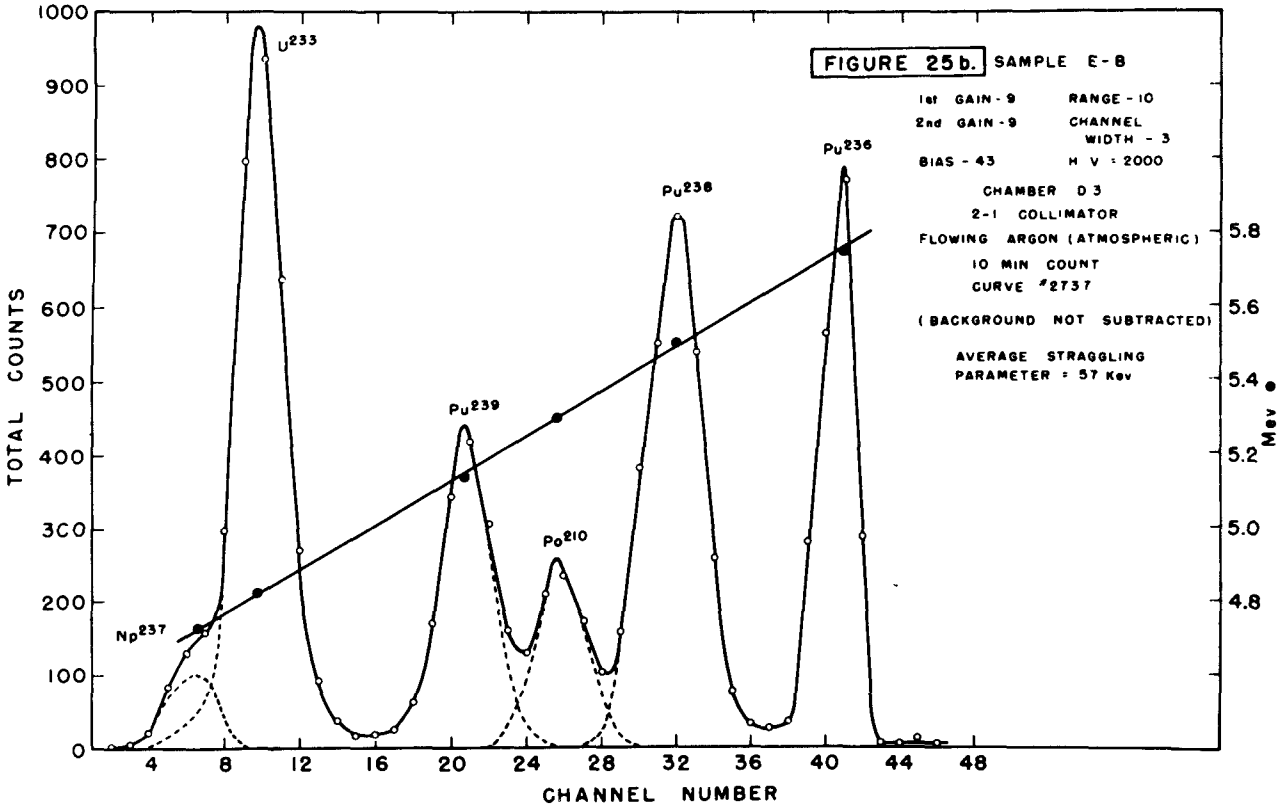


FIGURE 25b. SAMPLE E-8
1st GAIN - 9 RANGE - 10
2nd GAIN - 9 CHANNEL WIDTH - 3
BIAS - 43 H V = 2000
CHAMBER D 3
2-1 COLLIMATOR
FLOWING ARGON (ATMOSPHERIC)
10 MIN COUNT
CURVE #2737
(BACKGROUND NOT SUBTRACTED)
AVERAGE STRAGGLING PARAMETER = 57 Kev
TOTAL COUNTS
CHANNEL NUMBER
Mev
U233
Np237
Pu239
Po210
Pu238
Pu236

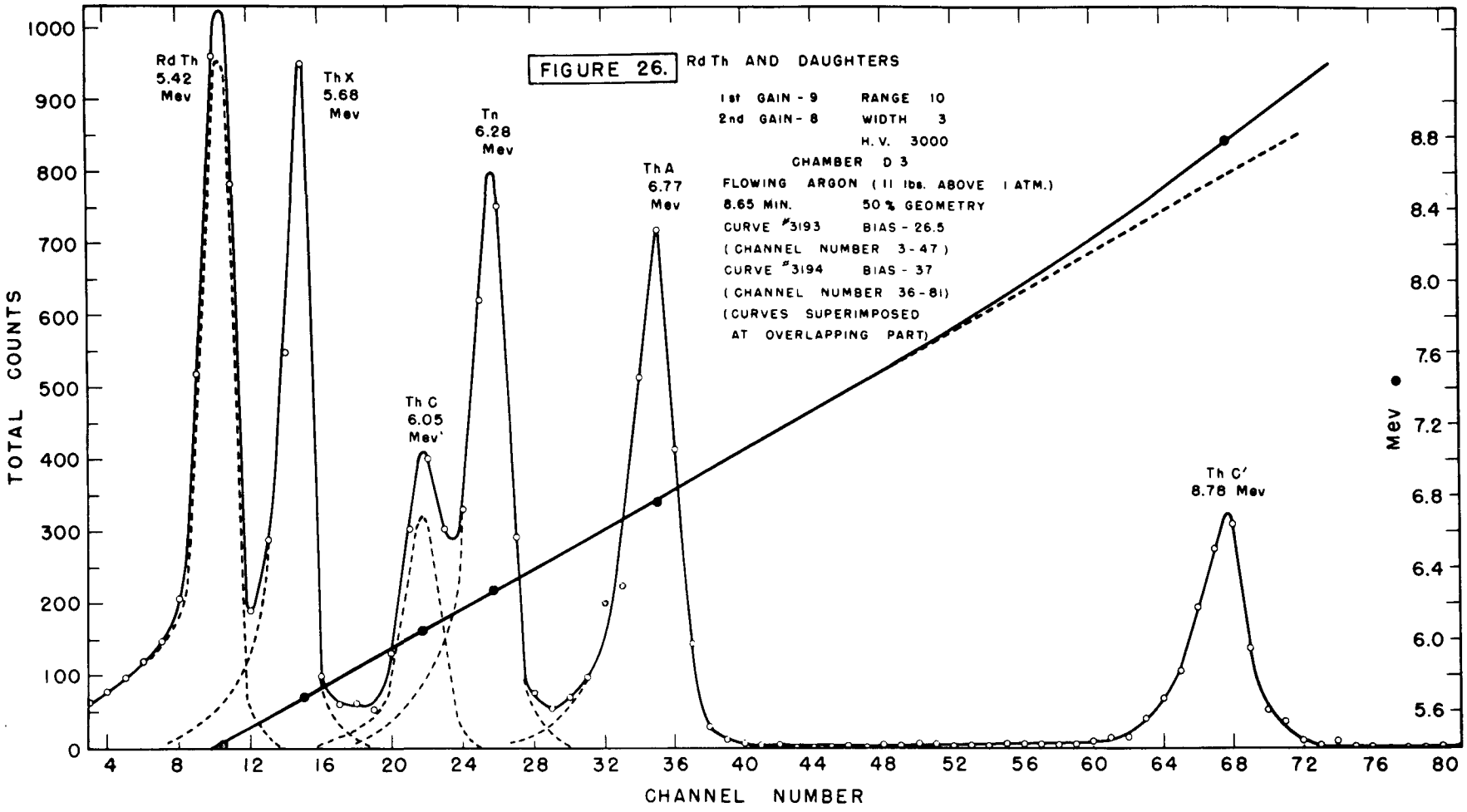


FIGURE 26. Rd Th AND DAUGHTERS
1st GAIN - 9 RANGE 10
2nd GAIN - 8 WIDTH 3
H.V. 3000
CHAMBER D 3
FLOWING ARGON (11 lbs. ABOVE 1 ATM.)
8.65 MIN. 50 % GEOMETRY
CURVE #3193 BIAS - 26.5
(CHANNEL NUMBER 3 - 47)
CURVE #3194 BIAS - 37
(CHANNEL NUMBER 36 - 81)
(CURVES SUPERIMPOSED
AT OVERLAPPING PART)
Rd Th 5.42 Mev
Th X 5.68 Mev
Tn 6.28 Mev
Th A 6.77 Mev
Th C 6.05 Mev'
Th C' 8.78 Mev
TOTAL COUNTS
CHANNEL NUMBER
Mev
0
100
200
300
400
500
600
700
800
900
1000
4
8
12
16
20
24
28
32
36
40
44
48
52
56
60
64
68
72
76
80
5.6
6.0
6.4
6.8
7.2
7.6
8.0
8.4
8.8

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~~SECRET~~

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