UNIVERSITY OF MICHIGAN

Department of Chemistry

PROGRESS REPORT 5

November 1955 - October 1956

NUCLEAR CHEMICAL RESEARCH

RADIOCHEMICAL SEPARATIONS
and
ACTIVATION ANALYSIS

W. W. Meinke, Project Director

November 1, 1956

Project No. 7 Contract No. AT(11-1)-70

U. S. ATOMIC ENERGY COMMISSION

Project No. 7, Contract No. AT(11-1)-70 between the University of Michigan and the U. S. Atomic Energy Commission was initiated on February 1, 1952. On November 1, 1952, Progress Report No. 1 was submitted to the Atomic Energy Commission. Progress Report No. 2 was submitted on November 1, 1953, Progress Report No. 3 was submitted on November 1, 1954, and Progress Report No. 4 was submitted on November 1, 1955.

The following is a report of the work which has been completed on the project during the year of November 1, 1955 to October 31, 1956.

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I FACILITIES

A. Michigan Phoenix-Memorial Laboratory

The hot chemistry laboratory facilities of this building described in the previous progress report (1) have been utilized throughout the year. At the present time installation of three pneumatic tube systems has just been completed to bring samples from the reactor face to the laboratory. These 1-3/16 inch diameter tubes terminate in each of three hoods in the chemistry laboratory and are controlled by automatic timing devices outside the hood. Predetermined activation cycles can be set in the laboratory and samples sent and received at the left rear corner of each hood.

B. Michigan Reactor

The Michigan Research Reactor adjoining the Phoenix Building has almost been completed and is scheduled to "go critical" in November of this year. Preliminary tests will occupy much of December and January but it is anticipated that the reactor will be in routine operation eight hours a day, five days a week by February 1, 1957.

The reactor is a " swimming pool" or "Geneva" type that will operate initially at 100 kilowatts and a maximum flux of about 10^{12} n/cm^2 sec but has been designed for 1 megawatt and a flux of 10^{13} n/cm^2 sec at full power.

C. Chemistry Building

One additional 6-foot radioisotope hood has been installed in what used to be a buffer room between the "hot laboratory" and the counting room. It replaces an old "chemical" hood which had never been suitable for radioisotope work. This increases to six the number of 6-foot hoods available for work with radioisotopes in the Chemistry Building.

II INSTRUMENTATION

A. Bombardment Chamber and Current Integrator

It became possible for the first time in two years to inspect the inside of the bombardment chamber (1,2) when the cyclotron staff dismantled certain parts of their analyzer system early in the summer. It was found that deposits of metallic dust and sulfur had formed on the Kovar seal of the suppressor ring cable inside the chamber as the result of burning up several targets. These deposits had caused shorting of the connection which in turn had burned off, leaving the suppressor ring disconnected from its power supply and hence inoperative. Fortunately no accurate absolute runs were taken during the time the suppressor ring was disconnected. New, more flexible cables have been installed to connect the Kovar seals with the suppressor ring and the Faraday cage, and the chamber has been carefully cleaned to ensure trouble-free performance in the future. In addition during subsequent bombardments special care is being taken to avoid evaporation of the sample targets.

The new current integrator (1) constructed during the summer and fall of 1955 was put into operation during the past year. Initial calibration and a recalibration check after six months operation gave a least squares standard deviation of 0.6% from perfect linearity for all the points. Part of this calibration curve is shown in Fig. 1. Since this deviation is a random phenomenon, currents up to 5 microamperes can be integrated with errors less than 1% with this instrument. In order to avoid burning the mylar substrate of the targets only currents up to 1 microampere have been used with the current integrator. Drift characteristics were found to remain within this 1% error over measurement periods of two hours and more.

The negative high voltage supply for the Faraday cup mentioned in the last report (1) proved very dependable, especially when operated with regulated line voltage in conjunction with the new current integrator. (O. U. Anders)

B. X-ray Proportional Counter

The 4-inch diameter krypton-filled x-ray proportional counter described in detail in previous reports (1,2) was finally put into operation during the past year and its performance proved satisfactory through many tests. The counter itself which was made from a Brookhaven design with slight modifications is pictured in Figs. 2 and 3. Its attendant power supply and non-overloading linear amplifier are shown with it in Fig. 4 and a block diagram of the equipment is given in Fig. 5.

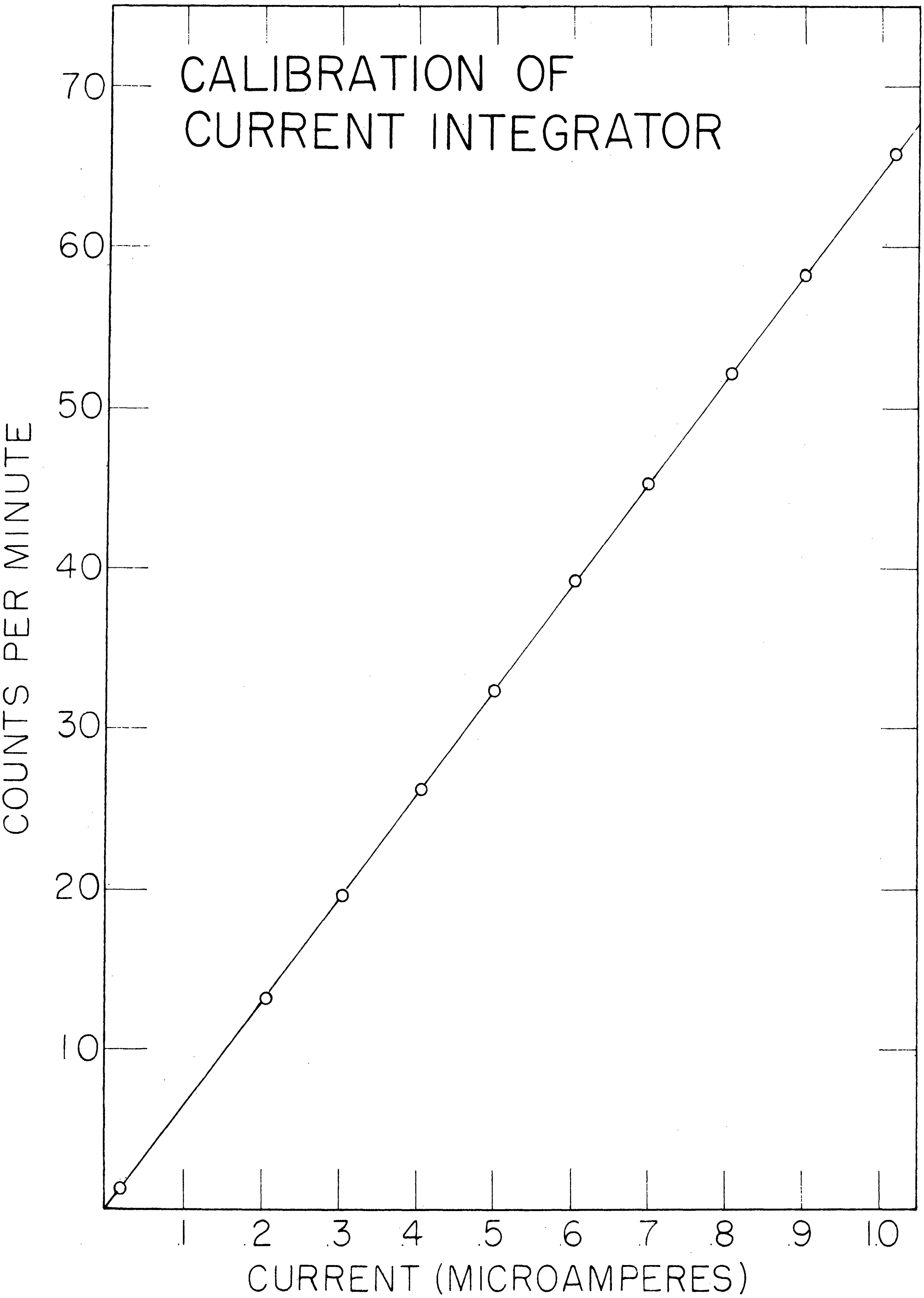


Fig. 1. Calibration curve for new current integrator.

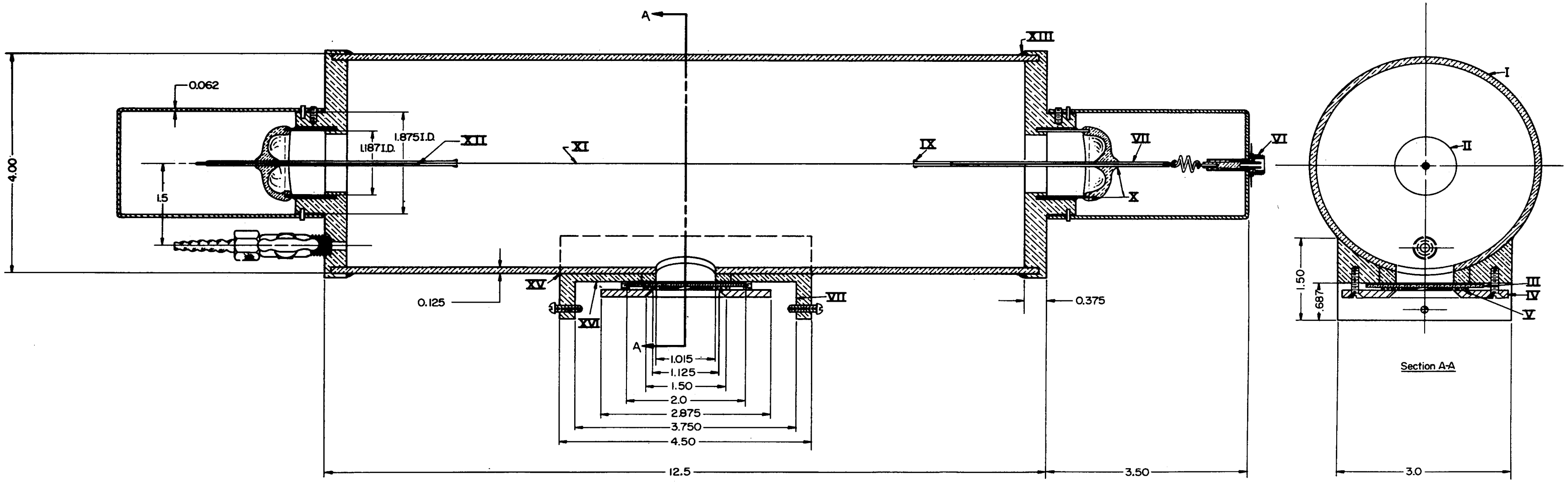


Fig. 2. Diagram of X-ray proportional counter.

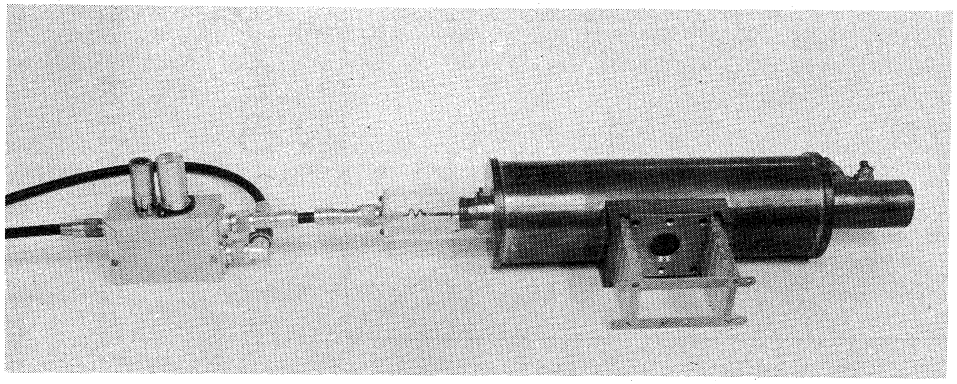


Fig. 3. X-ray proportional counter and preamplifier.

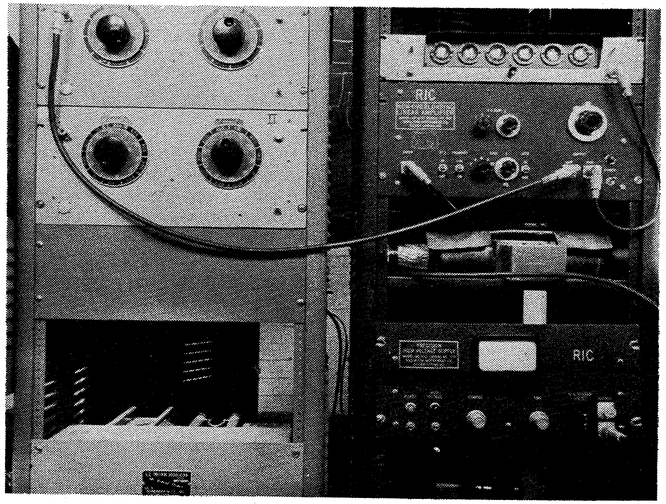


Fig. 4. X-ray proportional counter in operation.

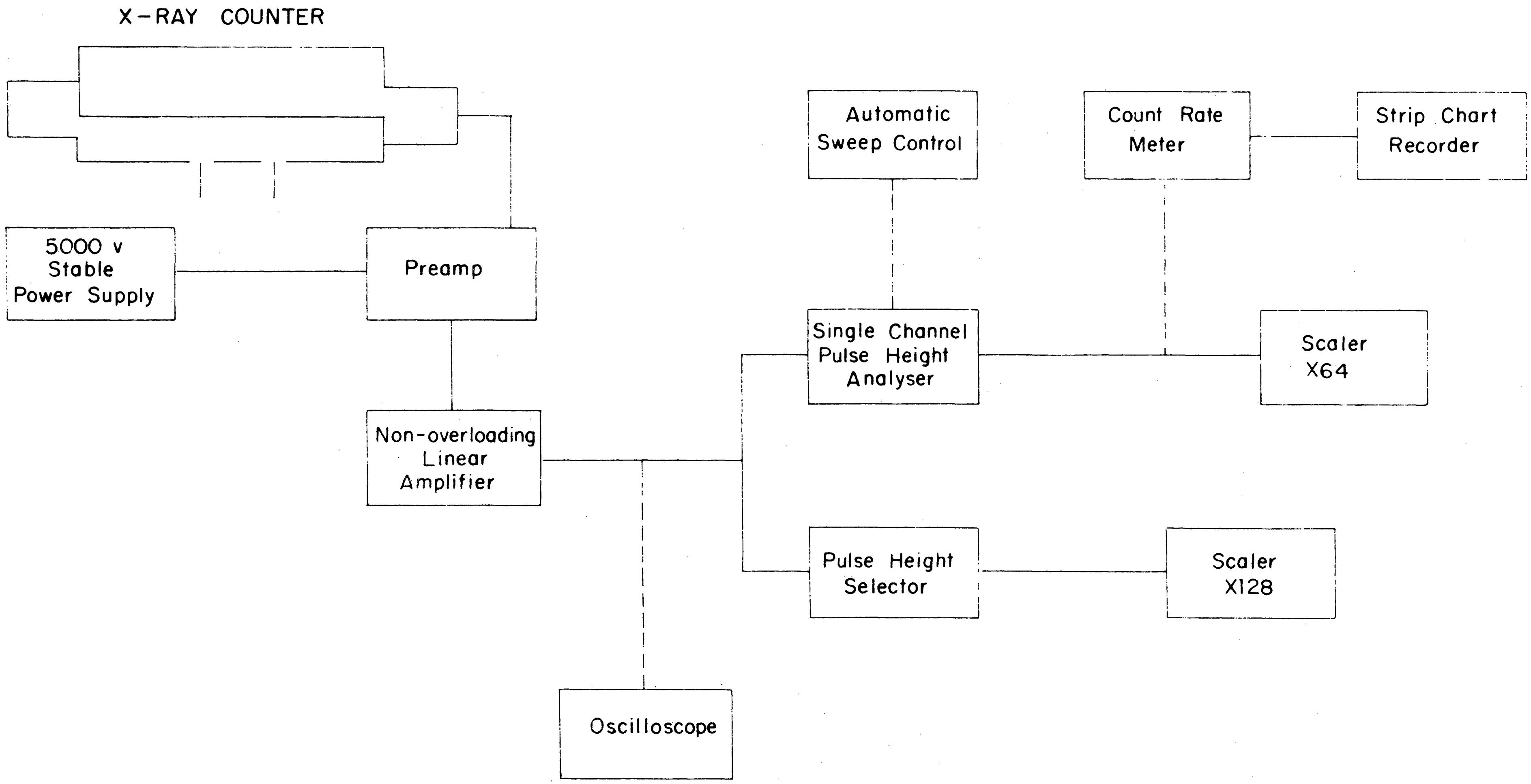


Fig. 5. Block diagram of X-ray proportional counter.

X-ray spectra of Cr^{51}, Mn^{54}, Zn^{65}, Y^{88}, Cd^{109} and Sn^{113} have been studied with this equipment. Three typical spectra are shown in Fig. 6--the resolution (half width at half maximum) of the Y^{88} K x-rays being approximately 16%.

In order to count total x-rays of a certain energy it was originally planned to scan the spectra by recording counts manually or semi-automatically with a step mechanism in each of 35 to 50 equal intervals and to integrate the counts under the peaks at the desired energy. This was found to be an extremely laborious undertaking, especially for samples of low activity. Bad statistics caused by the small number of counts and small drifts in the equipment over the long time intervals necessary to count low levels of activity, limited this method.

To overcome these difficulties a procedure was evolved whereby that part of the x-ray spectrum falling on the desired peak could be obtained in a single measurement. The peak was first located by a rapid sweep of the spectrum which was drawn out on a Speedomax recorder. By judicious setting of the pulse height analyser window edge on the high energy edge of the peak and the pulse height selector on the low energy edge it was possible to obtain two simultaneous counts, the difference of which gave the counting rate of the peak.

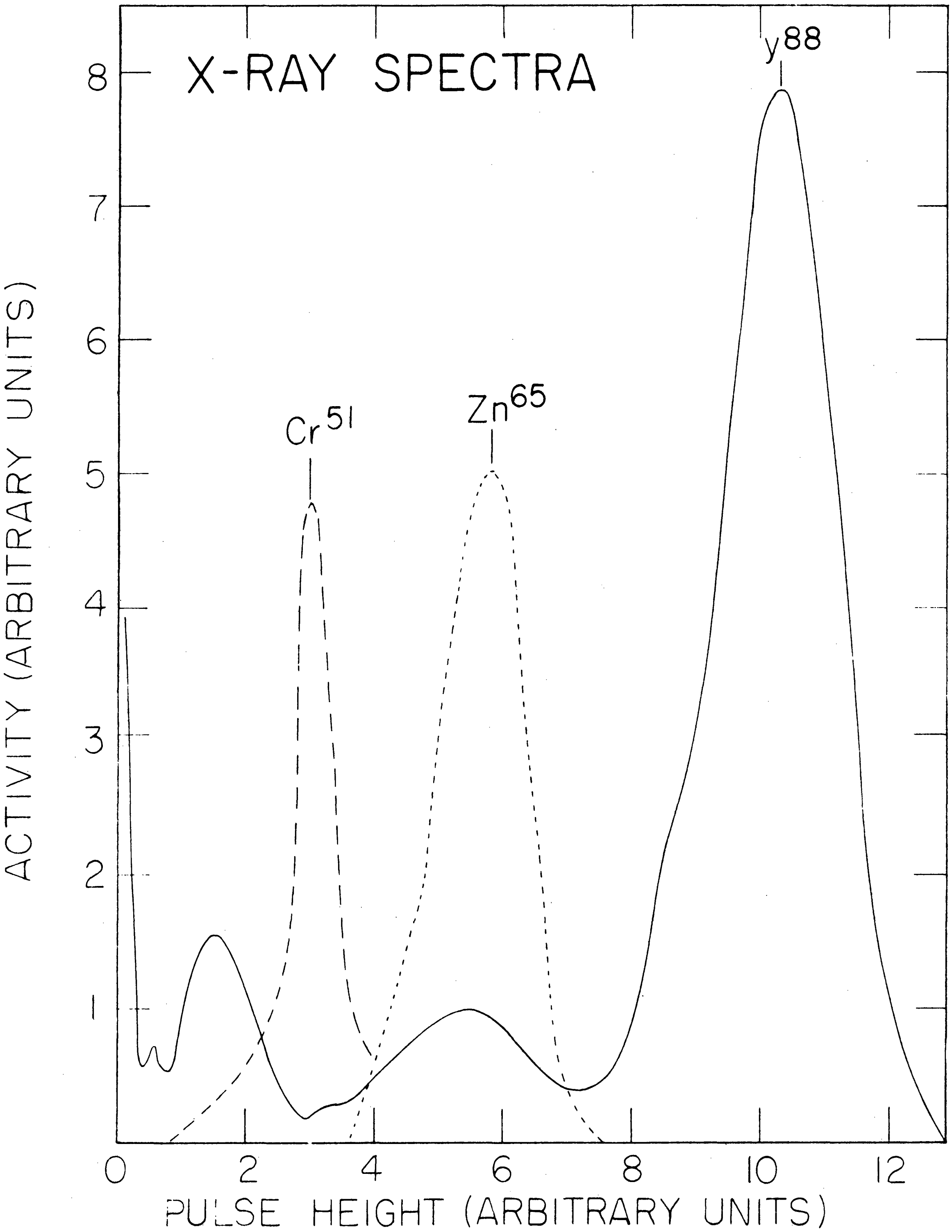


Fig. 6. Typical X-ray spectra taken with proportional counter.

A background count with the same setting but without the sample is also required since the background of this large counter is of the order of 300 counts per minute.

When care is taken to keep the counting geometry constant, the counting rates are reproducible by this method to within statistical limits. Slight drifts in the equipment will introduce errors only in the valley regions but these errors are generally much below the statistical errors of the counts. Attempts to establish "absolute" counting rates for x-rays have not been made as yet, but it is felt that "absolute" counting with errors not exceeding 10% should be possible with this equipment. (O. U. Anders)

C. 4 pi Counters and Absolute Counting

It has always been felt necessary to have at least two 4 pi proportional counters in working condition for the absolute cross section determinations. The spherical 4 pi chamber which has been in intermittent operation for the past two years (1,2) has never given as constant or reproducible results as the Borkowski type counter. Therefore a second counter was constructed according to the Borkowski design (3) and placed in operation early in the summer.

To facilitate data taking on these counters two new Ultrascalers were purchased from the Nuclear Instrument and Chemical Corporation of Chicago, replacing older scalers that were giving electronic troubles. At first the performance of these Ultrascalers was outstanding with these proportional counters. Plateaus of 1200 volts with slopes less than 0.2% per 100 volt could be obtained when operating above 3800 volts with almost any sensitivity setting. When the humid summer weather set in, however, the amplifier circuits began causing trouble and persistent difficulties impeded the work with the 4 pi counters for almost three months. At the present time it is again possible to obtain 1000 volt plateaus of negligible slope for both counters. Typical plateau curves taken with Sr-Y-90 samples are shown in Fig. 7.

The present arrangement of these counters in the counting room is shown in Fig. 8. The counter to the far left is a side window proportional counter of Los Alamos design (4) which is used for relative decay measurements. It has been found to work very satisfactorily (400 volt plateau with a slope of 0.5% per 100 volts) in conjunction with a slightly modified SC-3 scaler and PA-2 amplifier, custom made according to Los Alamos design (5).

a. Gold Plating of 4 pi Samples. The question of whether samples must be gold plated for accurate internal

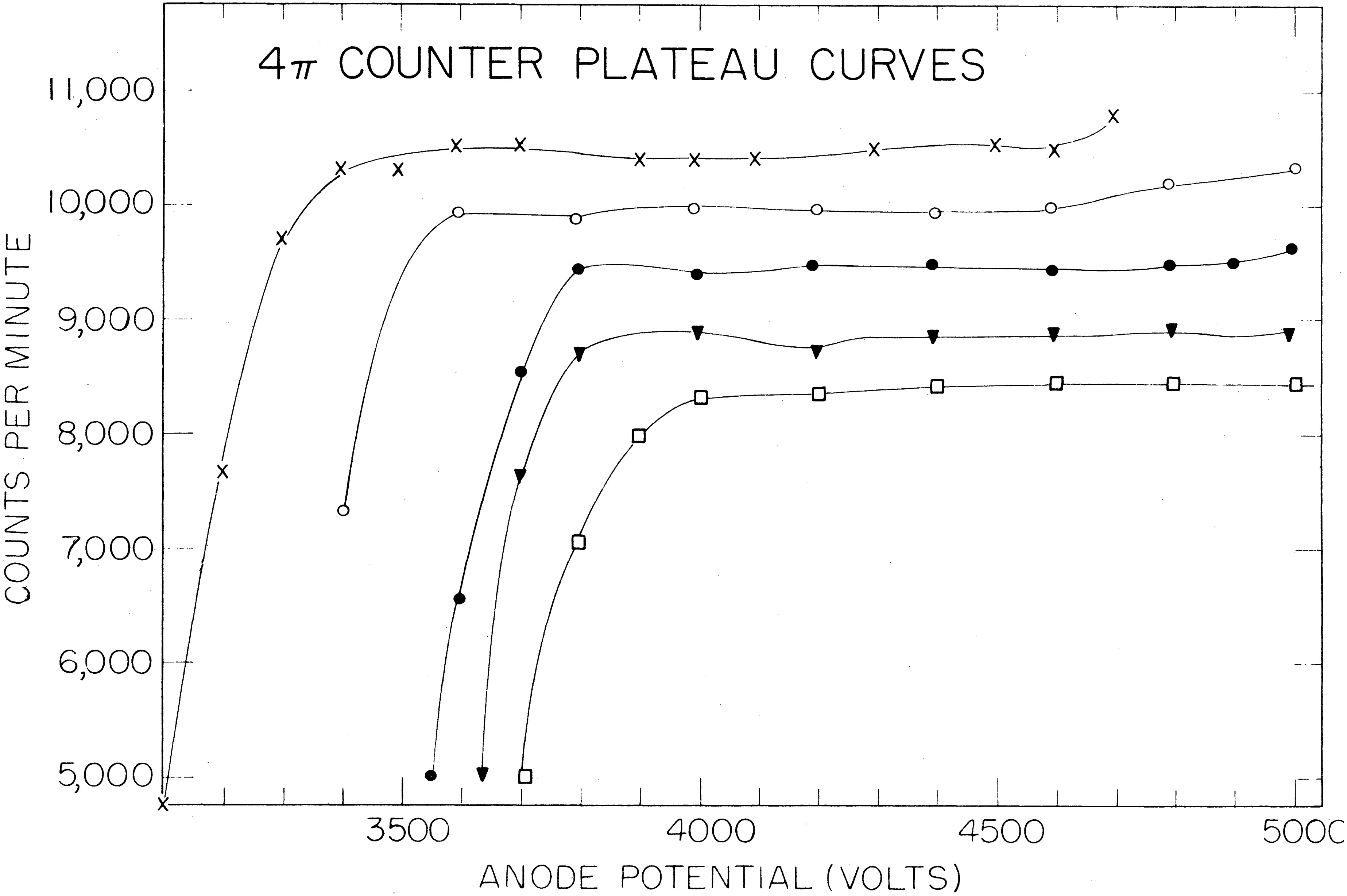


Fig. 7. Plateau curves taken at different sensitivities on Borkowski counter and Ultrascaler using a Sr-Y-90 sample.

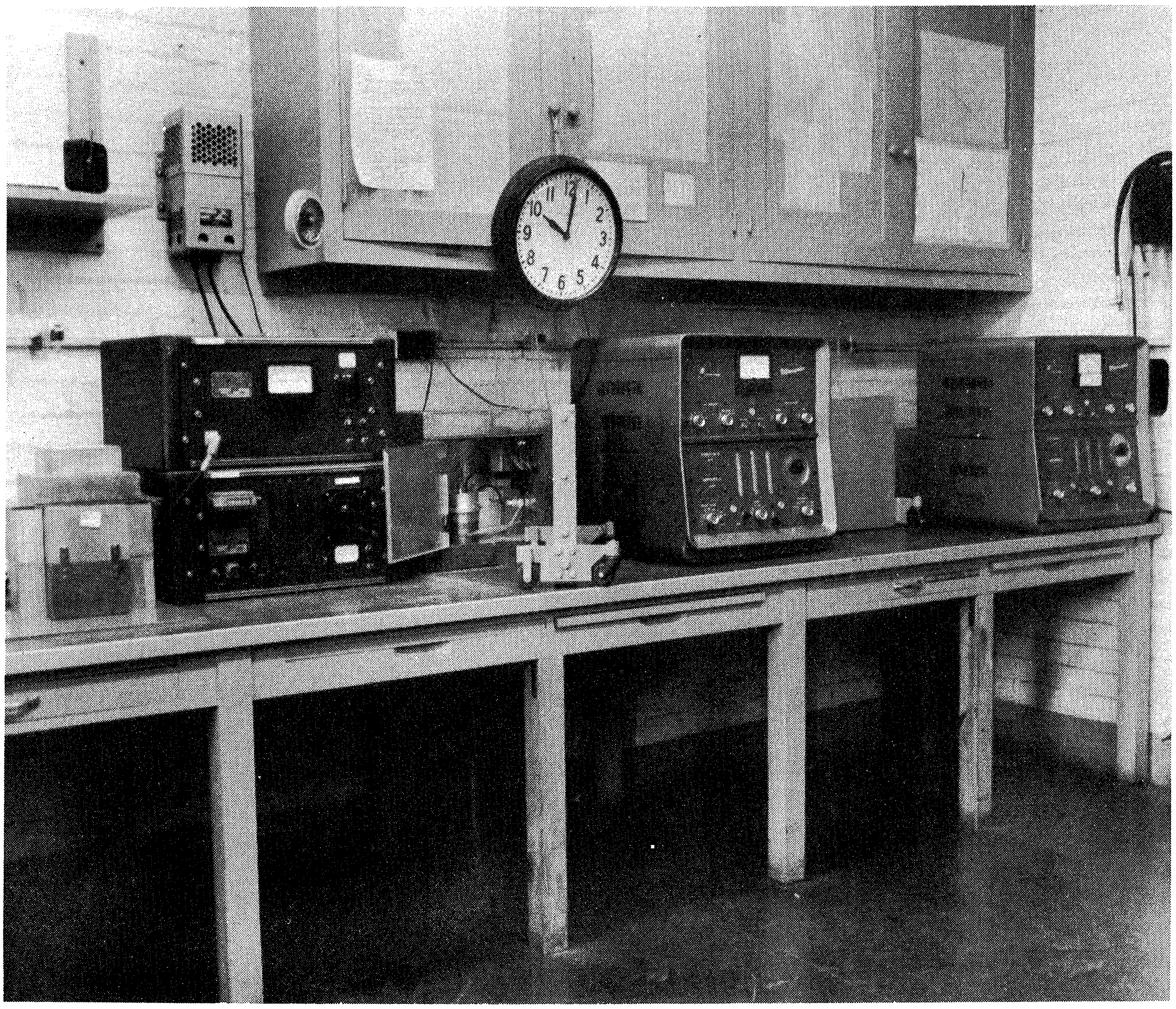


Fig. 8. Counting room arrangement.

4 pi counting has been debated by many workers. Results from a number of laboratories seem to indicate that much depends upon the size of the aperture across which the thin film bearing the sample is stretched. Sample plates used in this laboratory have openings 1/2" in diameter. Previous work (1) with Sr-Y-90 and even Co-60 samples has indicated that perfectly true and absolute results could be obtained without gold coating.

Utilization of the new scalers for 4 pi counting in conjunction with the two Borkowski counters however brought out certain sporadic inconsistencies and troubles in the counting of the very weak beta-ray emitters and x-ray emitters. A study of the effects of gold plating was then made for samples of this type.

When a very weak beta emitter such as Ni^{63} or an electron capturing isotope emitting x-rays such as Y^{88} is deposited on a non-conducting film and introduced into the 4 pi chamber in the above set-up, a plateau with a maximum is obtained and an abrupt rise in counting rate (discharge) occurs at a relatively low voltage. This maximum was encountered when one-minute counts were taken in succession at 100 volt intervals. If the plateau curve is repeated without interruption and before the discharge potential was reached on the previous curve, a curve with a maximum was again obtained, but the count rates were consistently far below those of the first curve. A third curve taken without interruption again lay below the second. Successive curves each lay below the other but approached a limiting curve as shown in Fig. 9. Interruption of the successive counts when the high voltage was at the upper end of the range resulted in a much lower following curve, while hesitation at a lower voltage value for one minute resulted in a higher plateau curve. When the voltage was turned off for several hours the original curve could be reproduced. This time and voltage dependent hysteresis effect can be explained by charge build-up on the sample.

Another phenomenon was traced to the same cause. When the voltage on the counter reached the breakdown potential, a discharge started, then acce lerated, until the scaler jammed. Reduction of the high voltage several hundred volts below the "breakdown" potential would not terminate the discharge. When the high voltage was turned off completely after a discharge and then turned on again, discharge started at a much lower potential then the first time. Longer waiting after the voltage was turned off raised the "breakdown" potential.

A sample of Y^{88} giving the peaked plateau curve described above was gold plated on one side without improving the discharge characteristics. When the

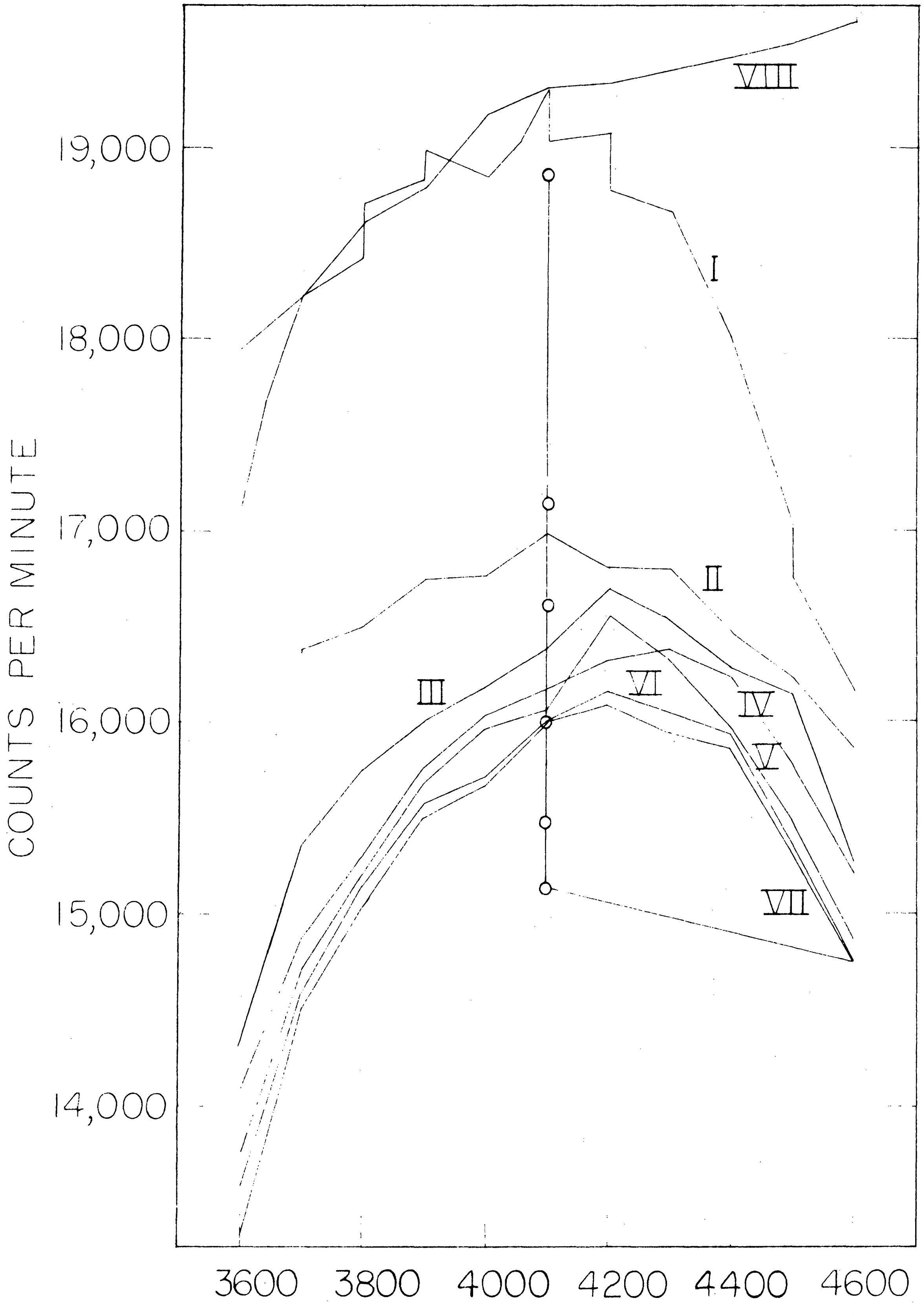


Fig. 9. Plateau curves taken in succession on a 4 pi sample of Y-88 emitting principally X-rays. Curve VIII was taken after both sides of the sample was gold plated.

other side was also gold-plated, however, a reproducible plateau was obtained. No maximum was observable in this plateau and no discharge occurred up to 5000 volts. The Ni^{63} samples behaved similarly.

As a result of these experiments all 4 pi counting samples are being gold-plated on both sides for absolute counting measurements.

An inordinately large amount of time has been spent during the last year in getting the two 4 pi counting set-ups to work consistently. Many of the difficulties which at first appeared to require dismantling and cleaning of the counter turned out to be strictly electronic. Often the trouble lay in the inability of the amplifier to handle oversize pulses or in excessive electronic noise.

b. <u>Gold Evaporator</u>. A new induction heated evaporator somewhat similar to that described by Pate (6) was built to reduce the time required to gold-plate the 4 pi sample plates from that required with the more elaborate metal target evaporator on hand. Fig. 10 shows a diagram of this evaporator and Fig. 11 pictures the apparatus.

The system consists of an evaporation chamber with ground glass lid for introduction of the sample plates. The neck of this chamber receives a hemispherical platinum crucible which can be heated by the

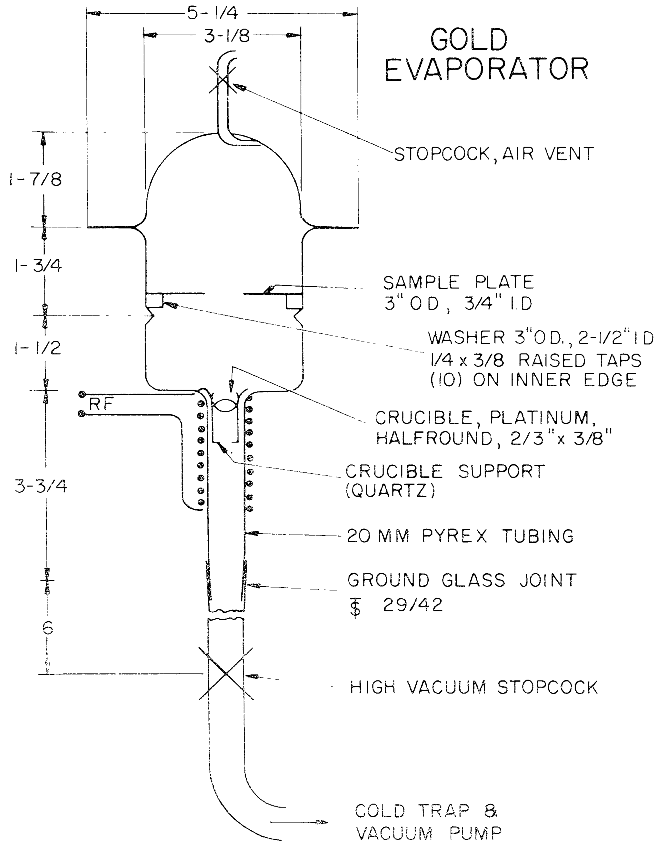


Fig. 10. Diagram of gold evaporator.

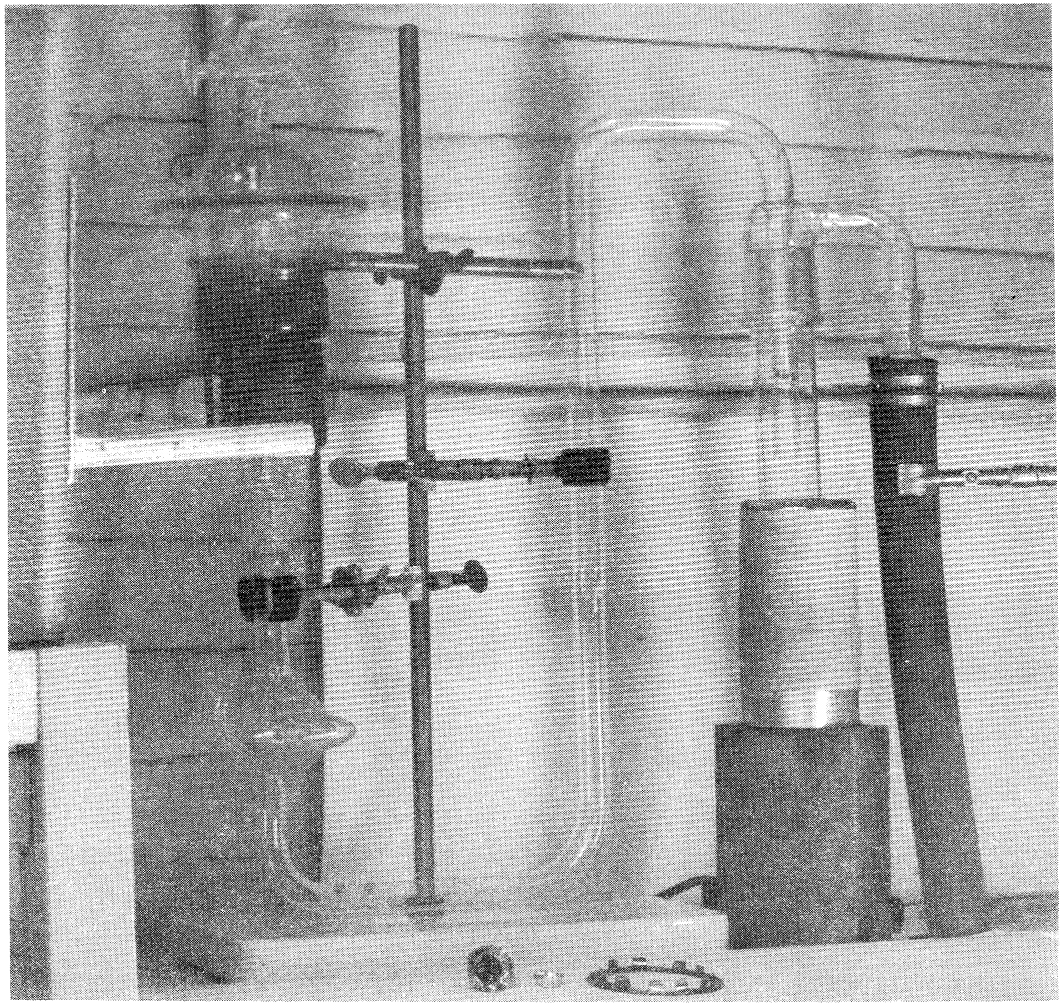


Fig. 11. Gold evaporation apparatus.

external induction coil made of 1/4 inch copper tubing. The chamber fits with its ground glass joint into a 3/4 inch diameter glass tube leading into a liquid air trap and then to a Welch Duo Seal High Vacuum pump that is able to pump the whole system down to 0.1 micron in two minutes. By using an all-glass system no leak problems were encountered. The induction heater employed is a rebuilt unit that was available in the laboratory; the induction coil can be water cooled if necessary. (O. U. Anders)

D. Metal Evaporator and Thin Film Gage

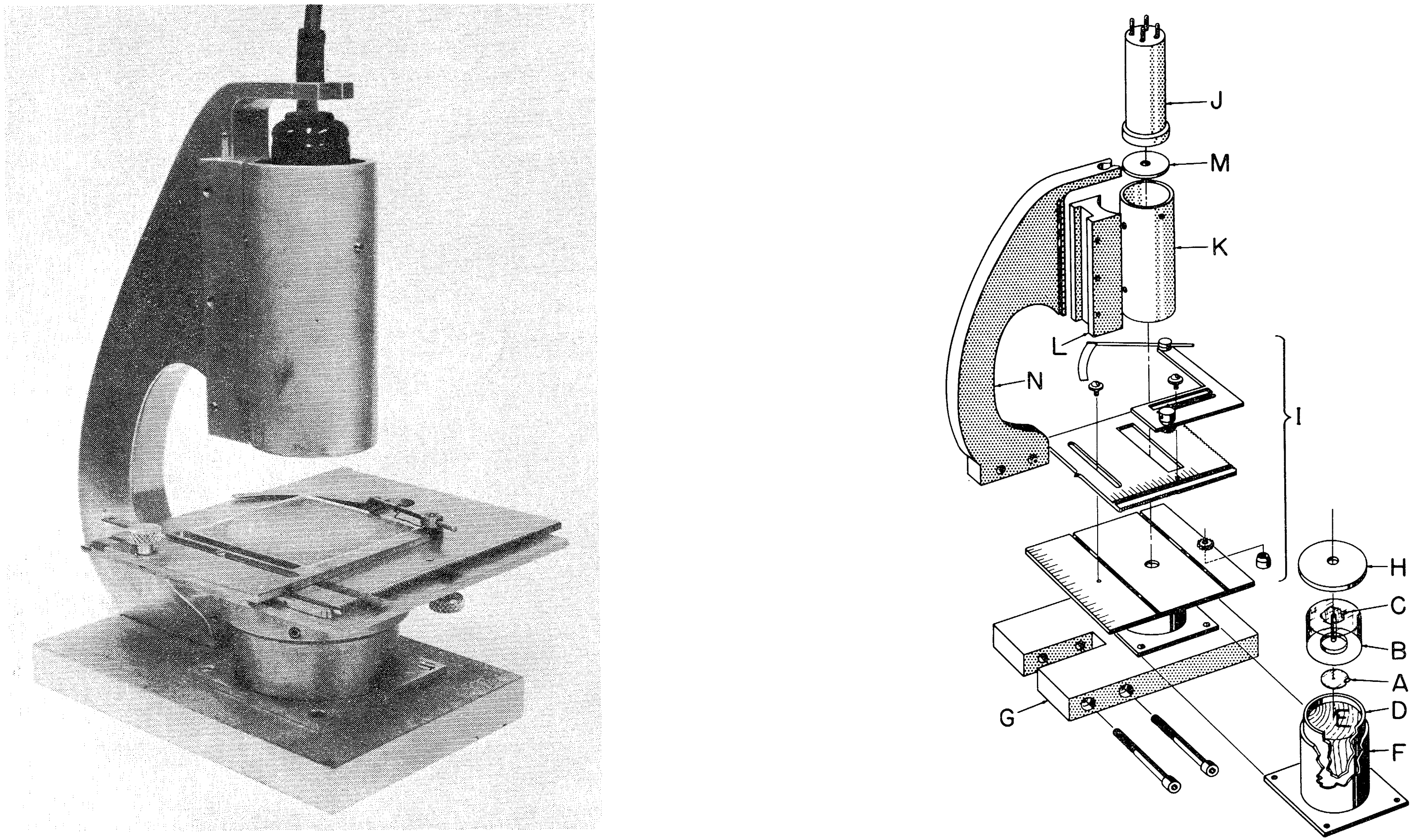
The metal evaporator mentioned in the previous reports (1,2) was used extensively during the past year for the preparation of thin, evenly distributed mylar-backed cyclotron targets. It is imperative, however, that the thickness variation of these thin targets be known accurately for absolute cross section measurements. Thus a thickness gage employing a collimated pencil of Pm^{147} beta particles was developed to measure the target evenness.

In this gage a 12 millicurie source of Pm^{147} was positioned inside a lead and brass-lined lucite collimator beneath a thin-window GM tube. A movable stage allows reproducible positioning of a sample whose absorption of the beta rays can be measured over a circular portion of the target, 3/32" in diameter. Test runs proved that 1% reproducibility of the points is easily obtainable and that thickness variations equivalent to less than 0.1 mg/cm^2 aluminum can be detected. The gage is described in more detail elsewhere (7) and is pictured in Fig. 12 and 13.

One example of the use of this gage was in the determination of the evenness of a cyclotron target consisting of ZnS evaporated on mylar. Fifteen measurements were taken on the mylar substrate at points distributed over the 25 cm^2 area. ZnS was then evaporated on the film and counts again taken at the identical 15 positions. The difference of the counting rates were taken for each position and the data evaluated by the least squares method. A standard deviation from perfect evenness of less than 1% was obtained for several ZnS deposits by this procedure. (O. U. Anders)

E. Gamma-Ray Spectrometer and Coincidence Apparatus

Essentially all of the electronic difficulties described in the previous progress report (1) have now been rectified. A Hickok Model 539B tube tester was purchased and has proven to be of great value in connection with instrument maintenance. One new single channel Atomic Pulse Height Analyzer, Model 510, has been purchased, the other two Model 510 analysers



Figs. 12 and 13. Thin film gage or target scanner.

overhauled, and the old Atomic Coincidence Analyzer, Model 502A, previously used has been exchanged for a new one. Thus, three single channel pulse height analyzers are now in use, one with the proportional x-ray counter and the other two with the two channel coincidence gamma-ray spectrometer. The physical arrangement of the equipment is shown in Fig. 14.

A considerable proportion of the resolution difficulties connected with the gamma-ray spectrometer was traced to the pre-amplifiers. When these were suitably modified and filters installed in the high voltage power supply, resolutions of 6.5 - 7.0% were obtained for the Cs^{137} gamma ray.

A modification of the automatic sweep arrangement for the single channel spectrometer has been designed and partially constructed. With this arrangement a spectrum may be continually swept, back and forth, while the data is recorded on a Speedomax Chart recorder. The sweep limits will be adjustable so that the entire spectrum, or any smaller portion of it, may be scanned at any of five different sweep speeds.

Another instrumental modification which will be of great value to the decay scheme program has just been completed. This permits coincidence studies to be carried out photographically by recording the oscilloscope presentation of only those pulses occuring

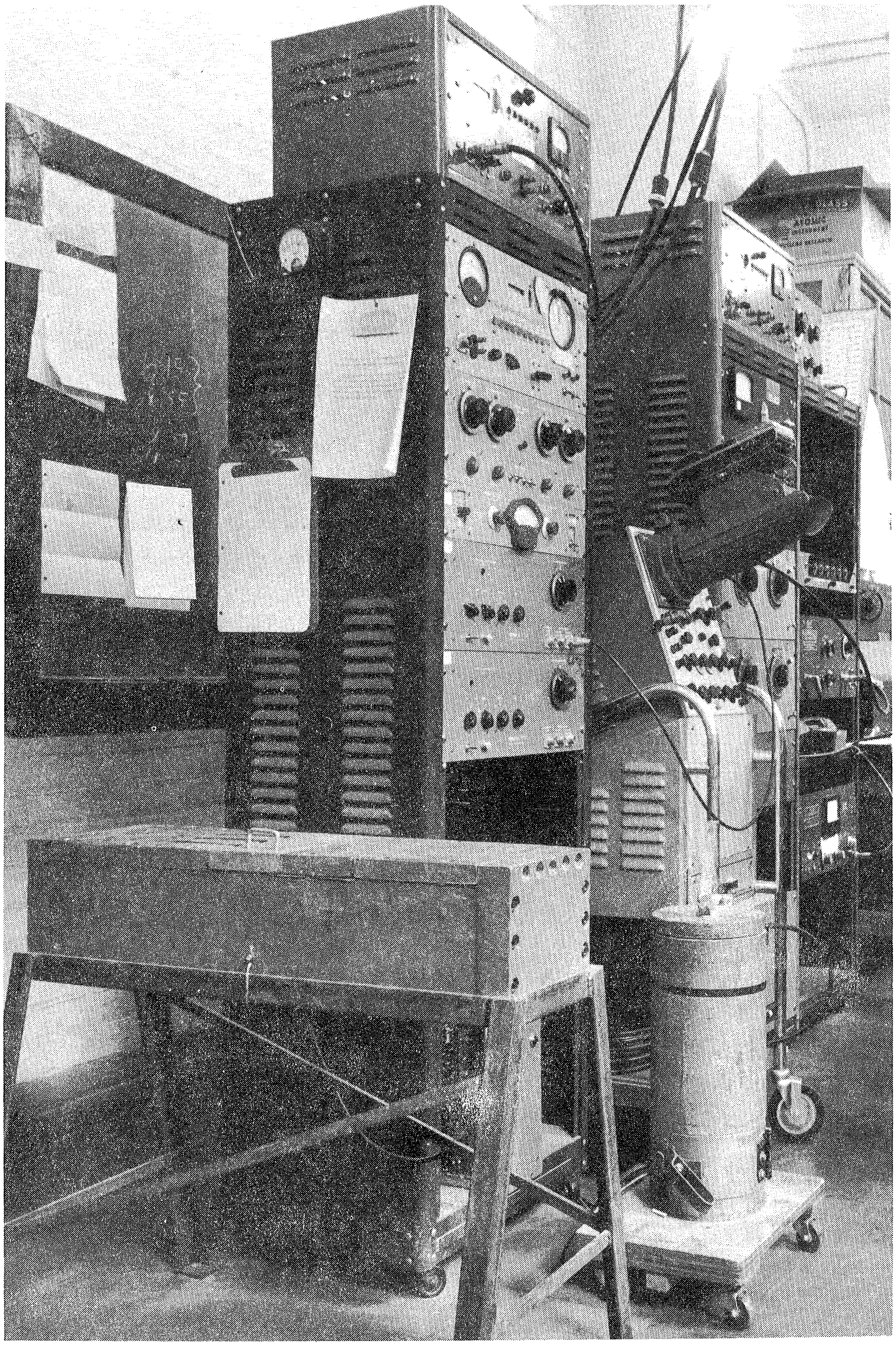


Fig. 14. Scintillation and proportional spectrometer; coincidence apparatus.

in one channel of the spectrometer that are in coincidence with selected pulses occuring in the second channel. Hence, photographs can be made of only those gamma rays in a complex decay scheme that are in coincidence with any predetermined gamma ray. In a similar way beta-gamma coincidence studies may be carried out.

The advantages of this arrangement over a manual coincidence sweep are many. No correction for sample decay is necessary, the results are obtained more rapidly and many more events are recorded since the entire spectrum is viewed at the same time rather than just a portion of it through a small window width. This last factor is important when dealing with short half-lived isotopes. The method is also easily adaptable to "Gray Wedge" recording techniques. Furthermore, the method required that only minor additions be made to the already existing electronic equipment. A more complete description of the equipment and results will be reported in the near future. (D. Gardner and R. Shideler)

F. Hollow Scintillator Beta-Ray Spectrometer

Preliminary investigations of the use of a hollow plastic scintillator as a detector in a beta-ray spectrometer were reported in the previous progress report (1). This investigation has been continued during the past year and brought to a successful conclusion.

Three phosphors of increasing size are now on hand. They are used, respectively, with maximum beta energies of ~1.5 Mev, ~2.2 Mev, and ~3.5 Mev (left to right in Fig. 15). Each phosphor was constructed in two parts consisting of a flat, solid, right circular cylinder base, and a top hollowed-out cylinder or truncated cone. A cross sectional drawing of the ~2.2 Mev phosphor is shown in Fig. 16.

An aluminum phototube support and lead shield were constructed to house the spectrometer and are shown in Fig. 17. The black neoprene cylinder at the right of the photograph has a dual purpose. First, it provides a light tight cover over the phosphor assembly. This is necessary since the entrance hole in the phosphor is not covered with foil as is the rest of the detector. Secondly it provides an absorber to stop electrons generated in the lead from striking the phosphor.

To assemble, the two parts of the phosphor are optically coupled together with Dow Corning 200 Silicone oil. The phosphor is then wrapped with aluminum foil which serves as a light reflector. The aluminum foil covering the entrance hole is cut away, and the phosphor mounted on a Du Mont 6292 phototube with the silicone oil. The phototube is connected to the

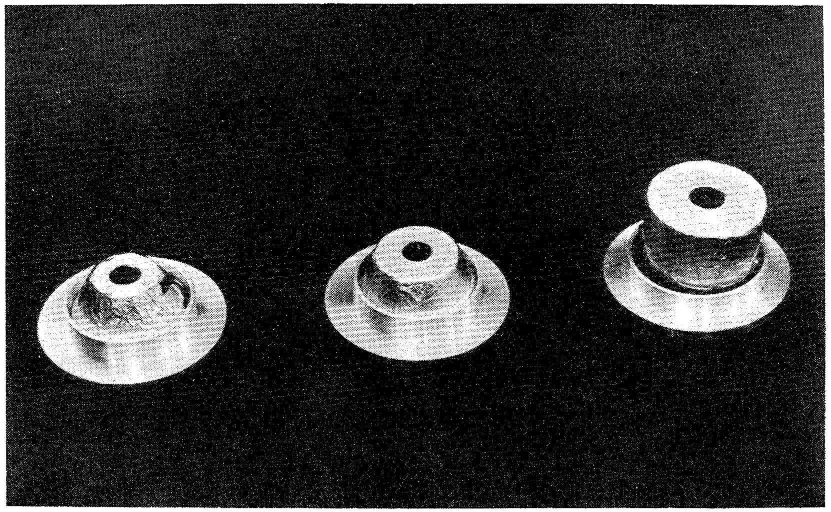


Fig. 15. Plastic scintillation detectors.

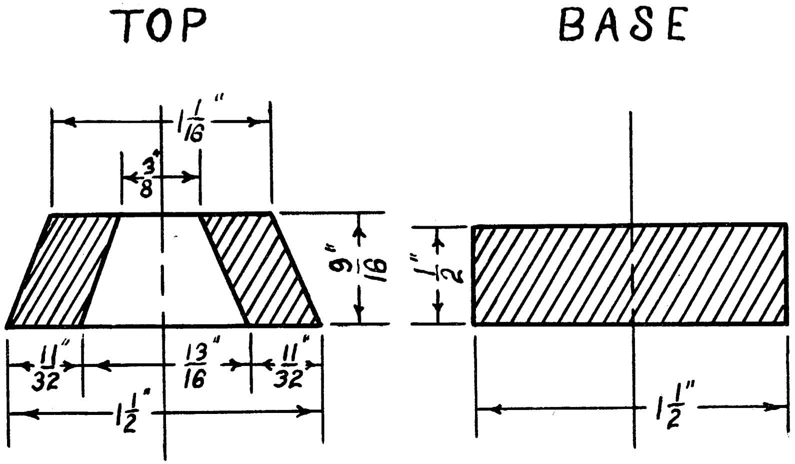


Fig. 16. Cross section of plastic detector.

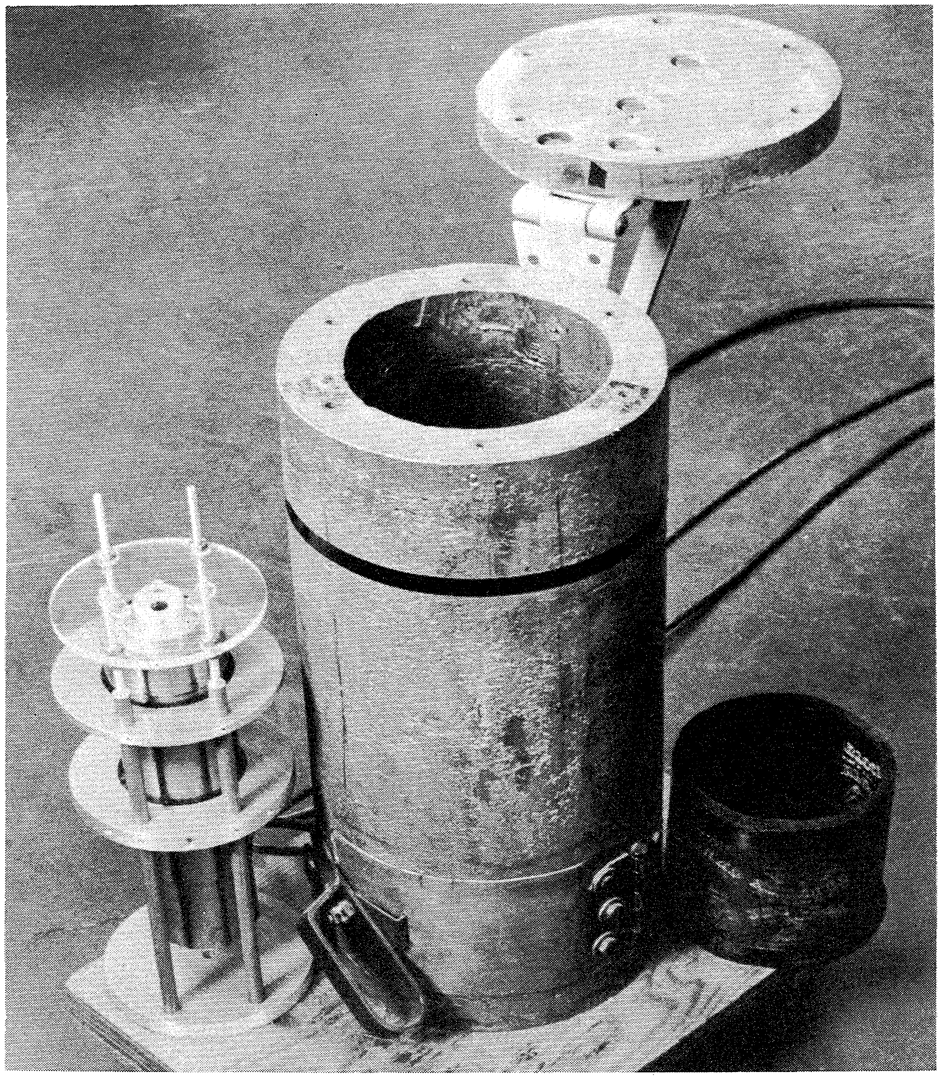


Fig. 17. Hollow scintillator spectrometer assembly.

preamplifier and the assembly mounted in the phototube support. The radioactive sample is placed in its lucite sample holder and pressed tightly against the opening in the phosphor. The light-tight cap is put in place and the unit placed inside the lead shield. Fig. 18 illustrates the performance of the spectrometer showing the allowed beta spectrum of In^{114} along with the resulting Kurie plot.

Compared to a lens type spectrometer, a scintillation device of this type requires only a relatively small amount of sample activity since the geometry is essentially 2 pi. The resolution, however, is appreciably poorer than the lens spectrometer, being about 12-14% for the Cs^{137} conversion line. Therefore, a correction for the resolution must be made. The correction is important only near the endpoint of the spectrum, and is made according to the directions of Palmer and Laslett (8). Fig. 19 shows the effect of this resolution correction on Kurie plots for Cs^{137} and Pm^{147} having beta-ray endpoints of 0.51 and 0.227 Mev, respectively.

In order to be of much value in resolving complex beta spectra, the response of a spectrometer must be sufficiently free of distortion to be able to distinguish a forbidden spectrum from an allowed one. For example, Y^{91} has a "unique" forbidden beta spectrum

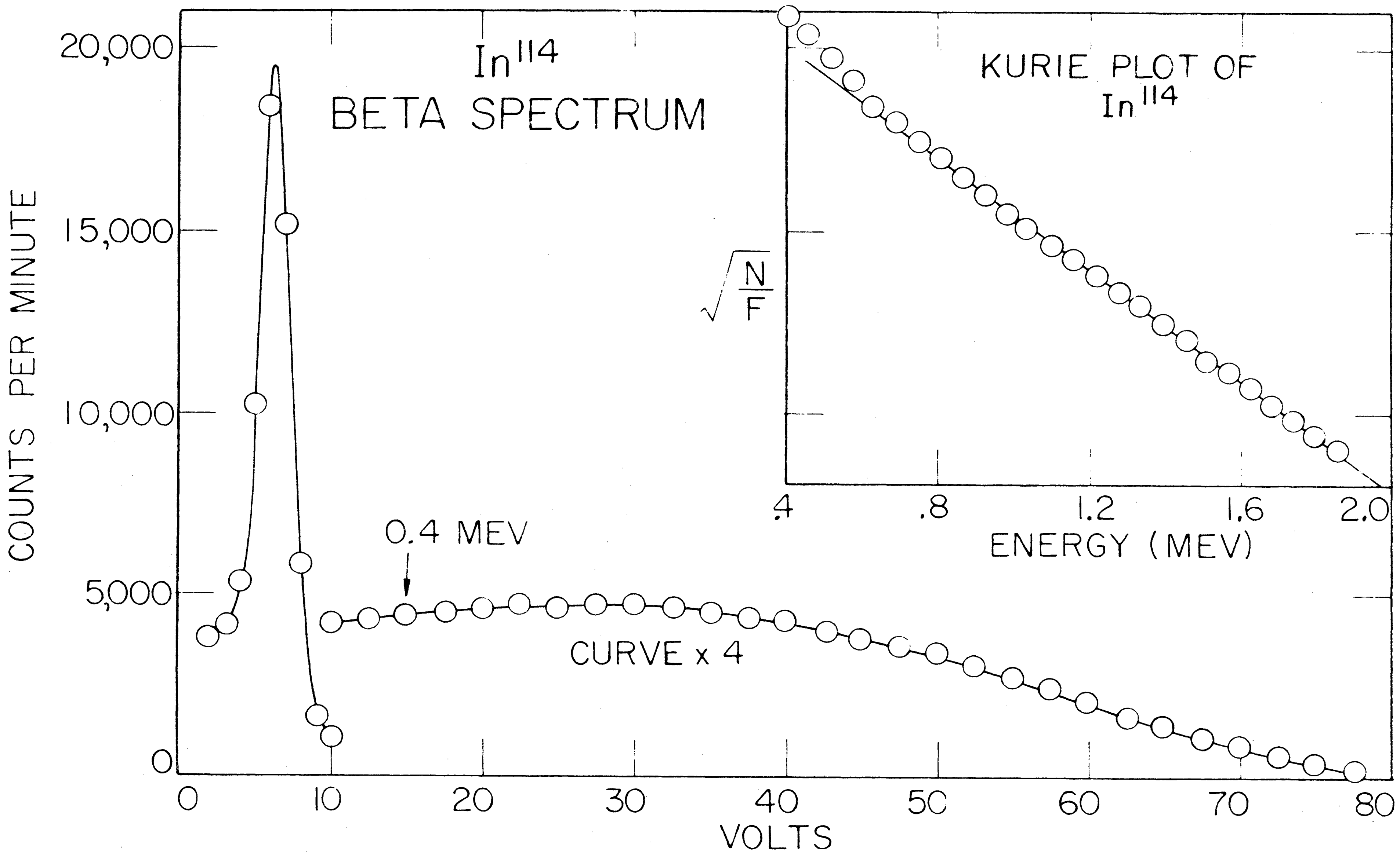


Fig. 18. Typical data taken with hollow scintillator beta-ray spectrometer.

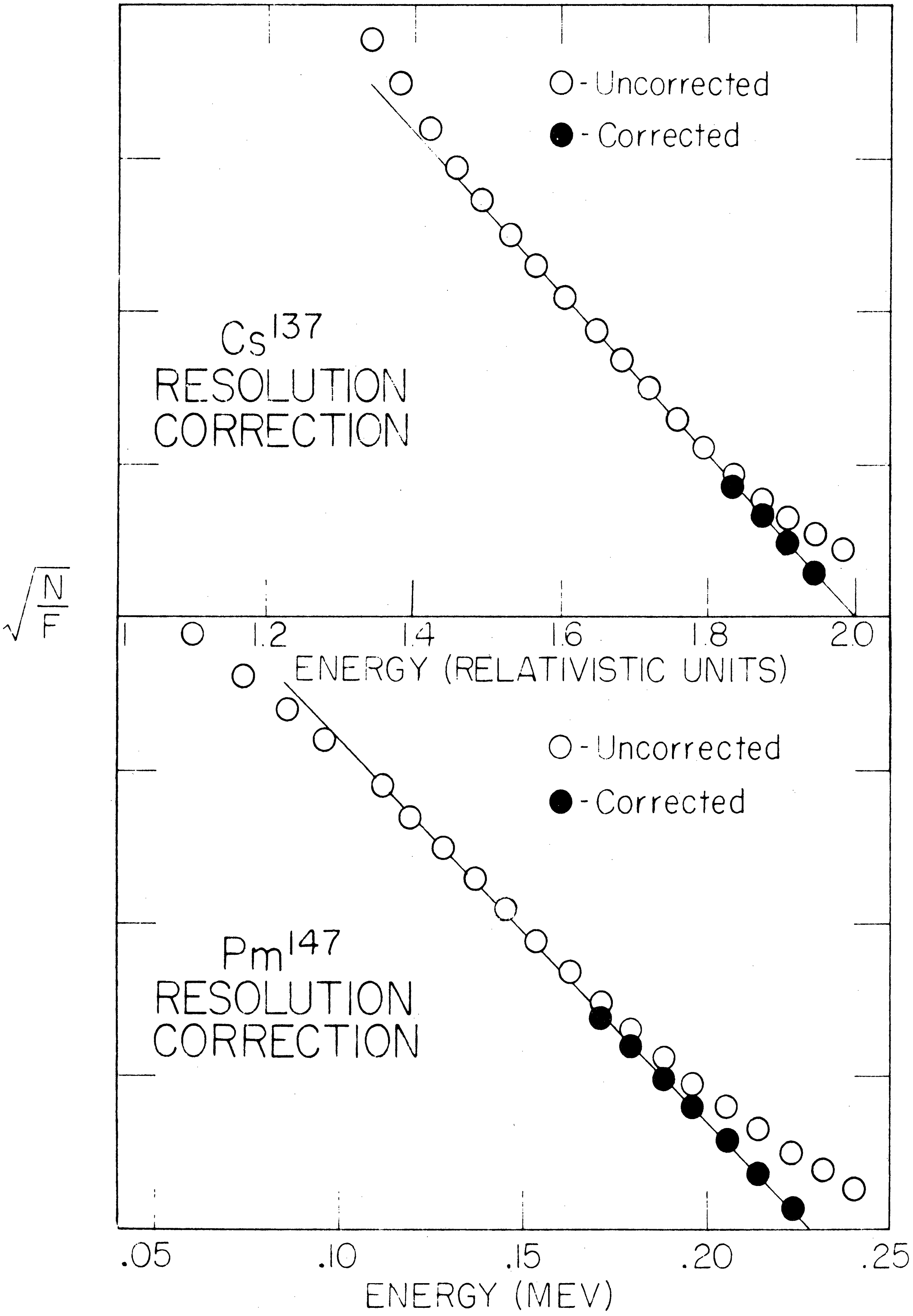


Fig. 19. Effects of resolution correction on spectra taken with hollow scintillator beta-ray spectrometer.

with an endpoint of 1.55 Mev. In Fig. 20 the Kurie plot for Y^{91} is shown, both with and without correction for forbiddenness. It can be seen that with the correction the Kurie plot is straightened out down to about 0.25 Mev.

To test out the ability of the hollow scintillation spectrometer to resolve complex spectra, a mixture of Cs^{137} with its forbidden spectra and Pm^{147} with an allowed spectra was run. Fig. 21 shows the resulting Kurie plot for the mixture, corrected for the forbiddenness of Cs^{137}. The straight line section corresponding to Cs^{137} was extrapolated back and subtracted from the total curve. The resulting curve was treated so as to remove the Cs^{137} forbidden correction, and the result calculated back to give the original Pm^{147} counting rate. A Kurie plot was then made from the Pm^{147} counting rate and is presented in Fig. 22. From the relative areas under the Kurie plots the ratio of Cs^{137} to Pm^{147} was calculated and found to agree with the known value to within 4%.

In this investigation the following items were studied:

1) Size and shape of detector.
2) Effect of sample backing.
3) Backscattering from sample holder.
4) Backscattering shields and collimators.
5) Gamma attenuation in absorber.
6) Effect of thick samples.

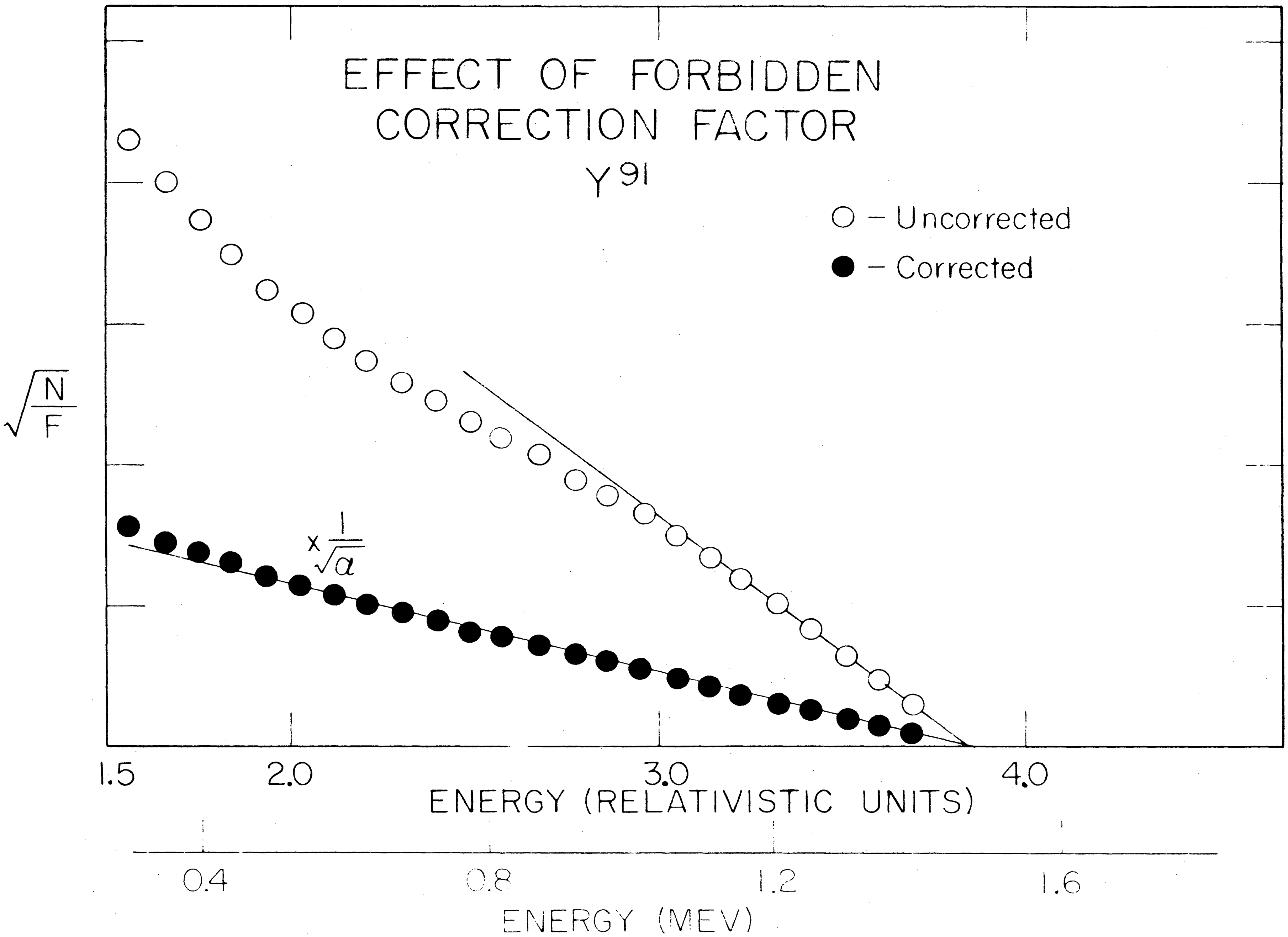


Fig. 20. Effects of forbiddenness correction on spectra taken with hollow scintillation beta-ray spectrometer.

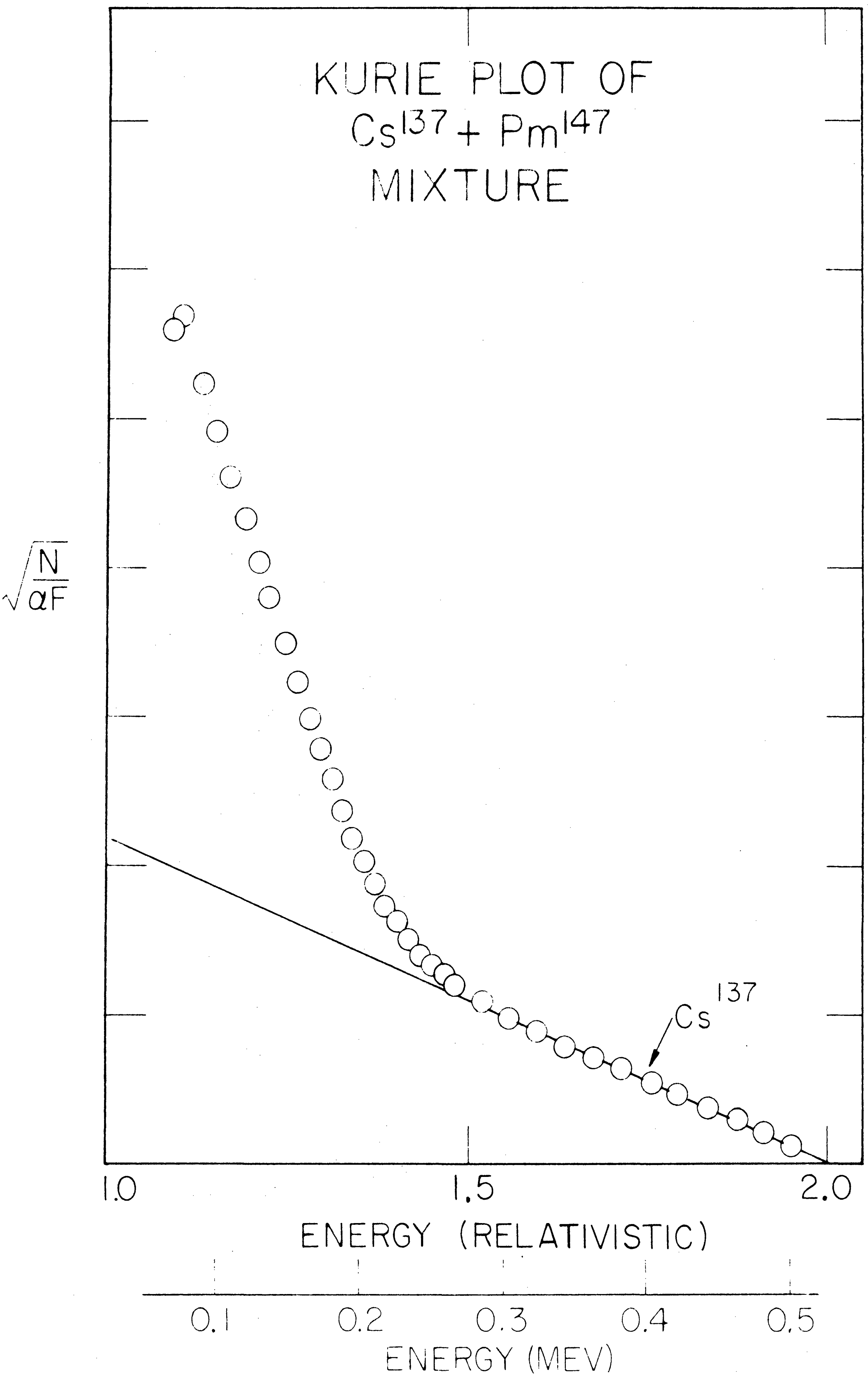


Fig. 21. Spectra taken with hollow scintillator beta-ray spectrometer.

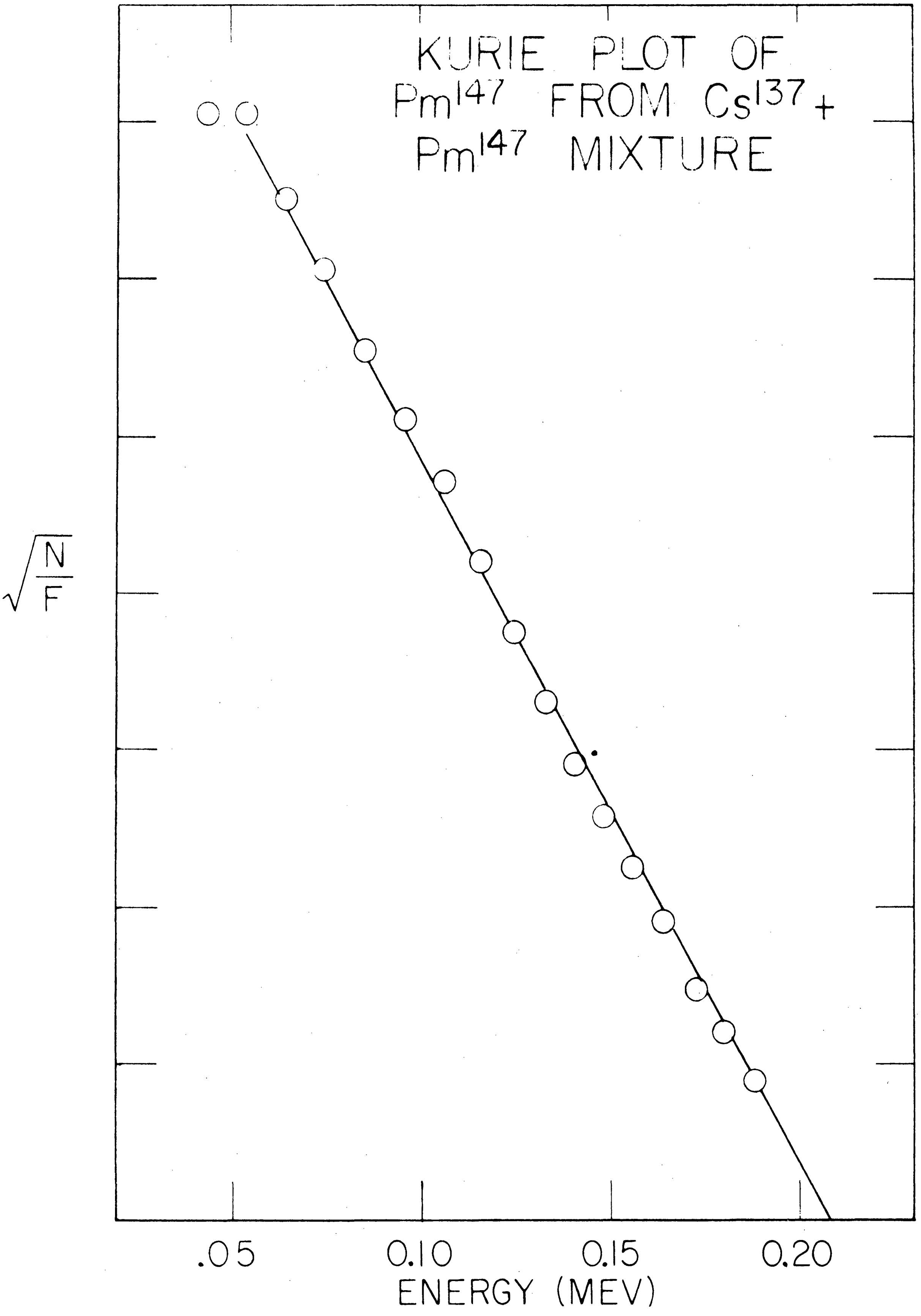


Fig. 22. Pm-147 component resolved from Figure 21.

7) Comparison of hollow with flat scintillators.
8) Amplifier overload and drift.
9) Useful energy range of spectrometer.
10) Corrections for instrument resolution.
11) Forbidden spectra.
12) Complex spectra.
13) Conversion coefficients.

The complete results will be written up for publication in the near future. (D. Gardner)

III EXPERIMENTAL

The past year has seen considerable progress in the nuclear chemical and separation phases of the program as well as an initiation of work on activation analysis. Twenty three bombardments have been made on the University of Michigan cyclotron during this time to support the nuclear chemical program. Unfortunately the program was slowed down during the first half of the year by considerable electronic difficulty both in the absolute counting equipment for the cross section work and in the components of the coincidence spectrometers.

The program for developing optimum radiochemical separations has temporarily halted with the completion by Mr. Sunderman of his doctoral work. Other students will, however, carry on this work in the near future. In addition much remains to be done to write up for publication the work already completed.

A. Nuclear Chemistry

1. Determination of (d,alpha) Reaction Cross Sections and Excitation Functions

The program initiated by K. L. Hall of investigation of absolute reaction yields of the (d,alpha) reaction has been continued. Thin targets were bombarded in the measured external cyclotron beam and then subjected to chemical separation. The desired product was separated carrier-free with known chemical yield and finally counted "absolutely" either in a 4 pi counter or in an x-ray proportional counter.

In considering the problem it was felt that any correlation of absolute cross section values should be made only in terms of normalization to the threshold energy of the reaction rather than in terms of specific energy values. Thus the experimental program has been expanded to include the determination for several (d, alpha) reactions of an excitation function composed of a number of relative cross section values in addition to one or two absolute points to establish the scale.

At the present time measurements are being made for the (d,alpha) reaction on zirconium to give yttrium isotopes, molybdenum to give niobium isotopes, sulfur to give phosphorus, titanium to give scandium, and iron to give manganese. Much of the decay data for these systems has already been accumulated but the analysis of the decay curves and the calculation of the actual cross section values have not yet been completed. It is expected that this work will be completed within the next six months and will be presented in detail at some later time in the form of a special report. (O. U. Anders)

2. Excitation Functions of Charged Particle Reactions (Literature)

The punched card compilation of photo copies of excitation functions which has been revised and made available each of the last two years as a supplement to the progress report has not been enlarged during the past year. Instead the cards were better categorized and put in more logical order, and have been accepted as supplementary material to a paper entitled "High Energy Excitation Functions in the Heavy Region" by W. W. Meinke, G. C. Wick and G. T. Seaborg which will appear in the Journal of Inorganic and Nuclear Chemistry in the near future.

This supplementary material has been "deposited as Document number 4999 with the ADI Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington, D. C. A copy may be secured by citing the Document number and by remitting $149.25 for photoprints, or $6.00 for 35 mm. microfilm. Advance payment is required. Make checks or money orders payable to: Chief, Photoduplication Service, Library of Congress."

3. New Determination of the P^{32} Half-Life

In the course of the work on absolute cross sections of the S(d,alpha)P reaction it was found necessary to know the P^{32} half-life with good accuracy. Values (shown below) (9) found in the literature vary from 14.6 to 14.1 days with errors too large for this program.

Sinclair and Holloway	14.59 ± 0.03	days
Lockett and Thomas	14.50 ± 0.04	
Klema and Hanson	14.35 ± 0.05	
Bayly	14.30 ± 0.09	
Cacciapuoti	14.30 ± 0.03	
Mulder, Hoecksema and Sizoo	14.07	no error quoted

Most authors worked with P^{32} samples that were not pure and corrections for the presence of S^{35} or P^{33} had to be applied to their curves. Furthermore, most of these determinations were made by following the P^{32} decay for less than six half-lives and they were made with instruments for which the reproducibility of the counting geometry, change of atmospheric conditions in the optical path and linearity of detection posed problems. These adversities can easily account for the discrepancies encountered and errors quoted.

It followed from these findings that a better method had to be used which circumvented or minimized the above sources of error in order to arrive at a significantly better value of the P^{32} half-life. The points that could be improved were isotopic purity of sample, reproducibility of counting conditions, and the number of half-lives during which the decay was followed.

A sample of high purity was obtained by producing the P^{32} by deuteron bombardment of pure ZnS. The 7.8 Mev deuterons employed for this purpose are not energetic enough to produce any long lived phosphorus isotope other than P^{32}.

The phosphorus activity was chemically purified in the following way. All cations were retained by passing the dissolved sample through a Dowex-50 column in its hydrogen form. The P^{32} was then selectively adsorbed on a Dowex-50 $Fe(OH)_3$ column in an adaptation of the method described by McIsaac and Voigt (10) and eluted with NaOH. The eluate was again passed through a Dowex-50 column to retain all of the sodium and yield the P^{32} in a carrier-free form.

The pure P^{32} thus obtained was mounted on a 30 microgram per square centimeter Zapon film, covered by a similar film and counted in a 4 pi beta proportional counter having a plateau of more than 800 volts of neglegible slope (less than 1% over plateau range) and excellent linearity. The decay of the sample was followed for eleven half-lives and 66 points taken during this time (Fig. 23). Calculation of the P^{32} half-life from these points (corrected for background and coincidence losses) yielded a new value of 14.221 ± 0.005 days, the quoted error being the standard deviation. Since this value is slightly smaller than most values reported, it would appear that systematic errors involving impurities have indeed been minimized

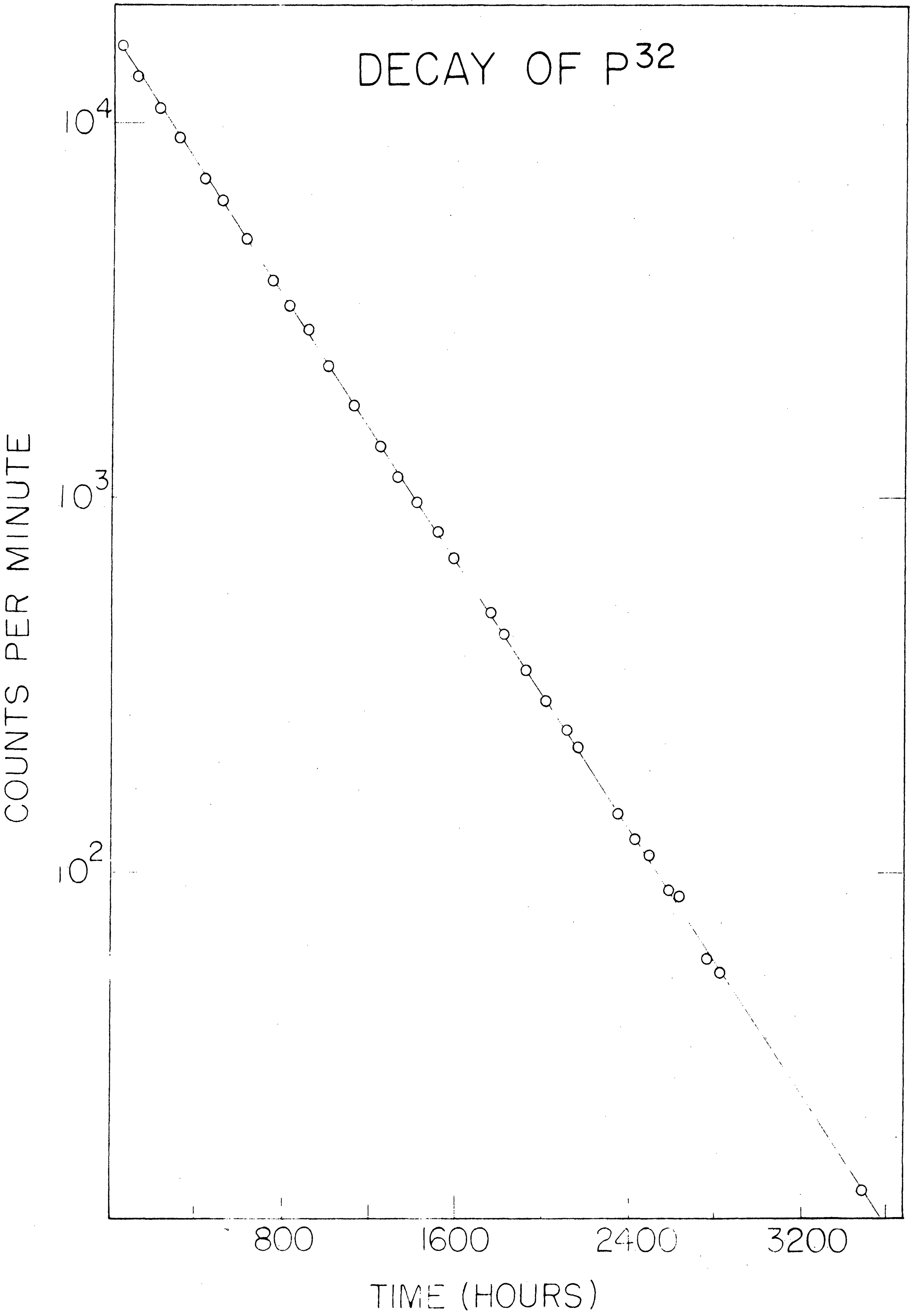


Fig. 23. Decay of highly purified P-32 from a (d,alpha) reaction on sulfur.

if not entirely eliminated. (O. Anders)

4. Decay Scheme Studies

a) Yttrium-92

An extension of the work by Cassatt and Meinke (11) on this decay scheme is underway. The beta emission, in particular, will be carefully examined by means of the Hollow Scintillation Spectrometer described earlier in this report. The photographic coincidence technique also described elsewhere in this report will be used to determine β-γ coincidences, and also to finally establish the position of certain low energy gamma rays in this decay scheme.

b) Yttrium-94

An isotopically enriched sample of Zr^{96} has been obtained. In this sample the abundance of Zr^{96} has been increased from 2.8% to 89.5%. Deuteron bombardment of this sample will produce Y^{94} by a (d,alpha) reaction. The rapid chemical separation described later in this report will permit this ~17-minute isotope to be studied.

c) Other Nuclides -- As explained previously, (1,2) the decay scheme work undertaken as part of this program is aimed primarily at products of the (d,alpha) reaction which are short-lived and come from low abundant target nuclides. For much of this work isotopically enriched targets are essential and present a chemical challenge in processing and recovery.

Small samples (of the order of 100 mg) of several enriched stable isotope mixtures have been procured and will be bombarded in the near future as part of this program. The samples include:

enriched zirconium-96 to yield the 17-minute yttrium-94;

enriched nickel-64 to yield the 14-minute cobalt-62;

enriched iron-54 to yield the 21-minute/6-day manganese-52;

enriched platinum-198 to yield the ~8-day iridium-196 all by (d,alpha) reactions ; and

enriched titanium-46 to yield the 33-minute vanadium-47 by a (d,n) reaction . (D. Gardner)

5. Use of Automatic Digital Computers in the Nuclear Program

An active interest has been developed in the applications of digital computers to problems in nuclear science. Experience has been gained on two machines: the Michigan Digital Automatic Computer, called MIDAC, and the IBM 650. Future computer work will be done on the IBM 650, primarily because of its ready availability.

Many experiments have been performed on the hollow scintillator beta-ray spectrometer discussed in an earlier section. During the evaluation of this instrument over sixty beta spectra were obtained; and each had to be evaluated by a Kurie Plot Analysis. Since it would have taken months to evaluate the data on a desk calculator, it was decided to make the computation on the IBM 650. A program was then prepared for this computer and has been in routine use for the last six months. Once the necessary data (counts per minute corrected for background versus energy) have been read into the machine, the computational time required to calculate a Kurie Plot for a beta spectrum consisting of 30 points is less than one-half minute.

In order to analyze a beta spectrum the following information must be supplied to the computer in the form of punched cards:

(a) The Program

The set of instructions used by the machine to compute the desired result is called a program. This is read into the machine by means of punched cards, and is then stored in the "memory". The program was first written in symbolic notation in which no definite memory locations or "addresses" were used. This was then assembled and optimized by means of a program known as S.O.A.P. or Symbolic Optimal Assembly Program. By means of this program the computer itself assigns the addresses for the instructions in such a way that a minimum of time is required to perform the program.

(b) Table A

This group of data cards contains information regarding the disintegration rate for each point in the beta spectrum. A maximum of 51 points may be used.

(c) Table B

These cards contain the energy in Mev associated with each point in the spectrum.

(d) Table C

This is the list of energies that serves as the argument for Table D. Table C is the same for every element.

(e) Table D

This group of cards is the list of the

Fermi functions used to calculate the Kurie Plot, and was obtained from the Bureau of Standards compilations (12). A separate table must be made for each element to be analyzed. There are on hand at present tables for S, P, Pm, Sr, Y, Rb, In, Cs, and Hf.

(f) Tables E and F

These contain the information necessary to correct the Fermi function for screening effects in the case of beta minus emission.

(g) Table G

Here the energy argument for tables E and F is listed.

(h) V1 and Z1

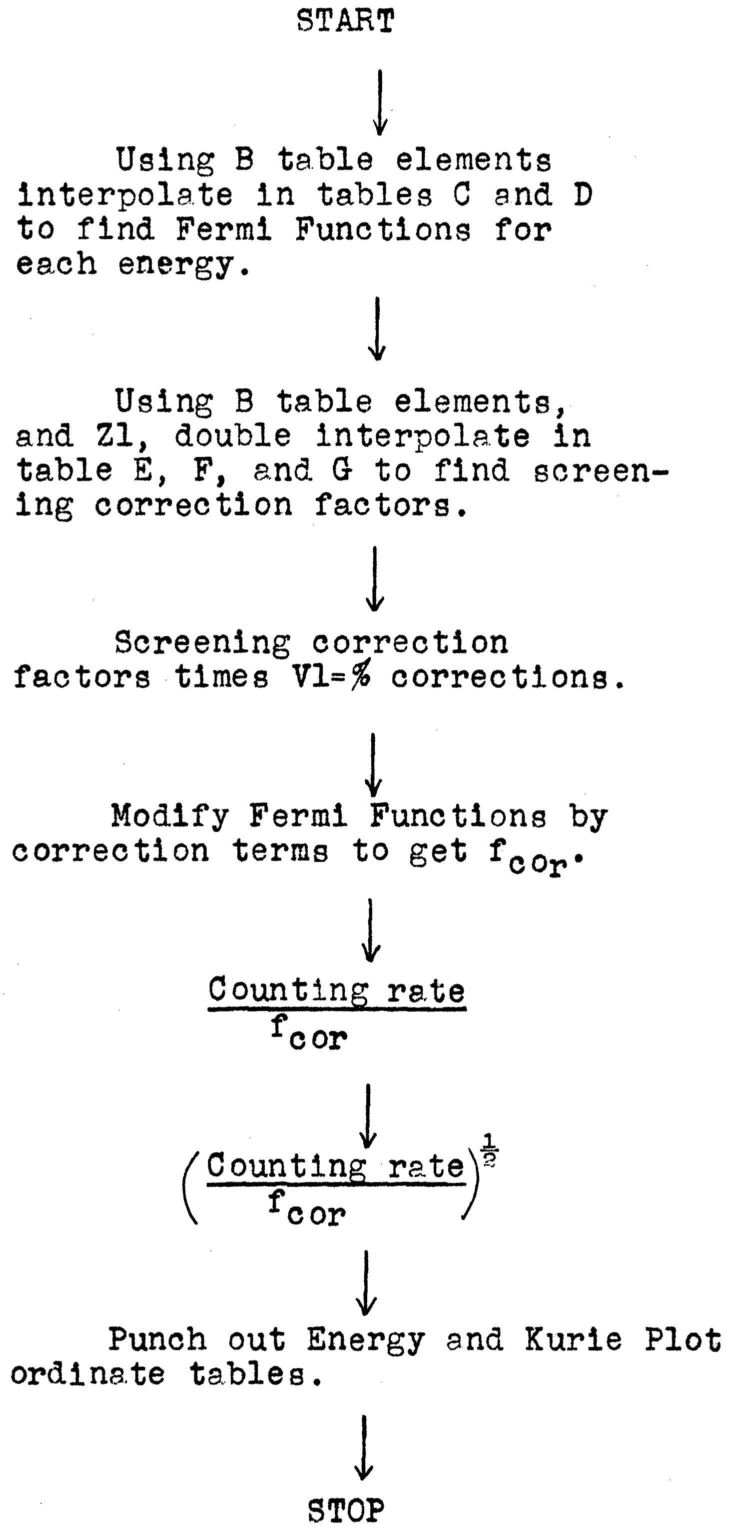
This is a single card, different for each element, that contains two pieces of data. V1 is the "outer screening potential" listed in the Bureau of Standards reference. Z1 is a number used to interpolate in tables E and F.

The results of the Kurie Analysis finally appear on punched cards. These cards are read by another machine which then prints out the results in two columns. The first column is the list of energy values, directly opposite which, in the second column, appear the appropriate ordinates for the Kurie Plots. Since most of the spectra to be studied are of the allowed shape, forbidden spectra are not corrected in the machine but by hand at present. A flow diagram showing the overall processes involved in the Kurie Plot Calculation by the IBM 650 is shown in Fig. 24.

Since facility has been gained in programing for computers, it is expected that other frequently encountered problems, such as decay curve fitting and resolution will be worked on in the future. (D. Gardner)

6. Chemical Procedures

Specific procedures used in the laboratory are given to supplement those already available in previous reports. Three more of the procedures included with the last progress report (1), those by Hall for the carrier-free separation of sodium, phosphorus and scandium, have been distributed in a slightly modified form by H. L. Finston in his compilation of Radiochemical Procedures sponsored by the National Research Council (13).



START

Using B table elements interpolate in tables C and D to find Fermi Functions for each energy.

Using B table elements, and Z1, double interpolate in table E, F, and G to find screening correction factors.

Screening correction factors times V1=% corrections.

Modify Fermi Functions by correction terms to get f_{cor}.

$$\frac{\text{Counting rate}}{f_{cor}}$$

$$\left(\frac{\text{Counting rate}}{f_{cor}}\right)^{\frac{1}{2}}$$

Punch out Energy and Kurie Plot ordinate tables.

STOP

Fig. 24. Flow diagram of Kurie Plot Calculation by the IBM 650 Computer.

CHEMICAL SEPARATIONS

Element separated: Yttrium	Procedure by: Gardner
Target Material: Zirconium	Time for sep'n: ~1 hr.
Type of bbdt: ~7.8 Mev deuterons	Equipment required: Lusteroid centrifuge tubes, two ion exchange columns of AG 2-X8, 200-400 mesh resin obtained from Bio-Rad. Lab. Resin beds were 8 mm x 140 mm.

Yield: ~90%

Degree of purification: Estimated $>10^6$ from Nb and Zr

Advantages: Essentially carrier-free separation with high decontamination. Can be shortened to ~5 min. if carrier can be tolerated.

Procedure:

(1) Place target foil in a lusteroid centrifuge tube containing 5 ml of 6 N HF, and 10 mg of Nb hold-back carrier. (Sample dissolved immediately.)

(2) Add 10 mgs of Pb^{++}, stir precipitate, and centrifuge. (See remark 1).

(3) Wash precipitate twice with ~5 ml of ~1 N HF, centrifuging in between. Wash once with a minimum amount of H_2O.

(4) Transfer precipitate to a glass beaker. Dissolve in several ml. of conc. HCl and evaporate to dryness with a few drops of HNO_3. Take up in conc. HCl and evaporate to dryness again.

(5) Take up in several drops of conc. HCl and place on column previously washed with conc. HCl.

(6) Elute with conc. HCl. at rate of ~1-2 drops/sec. The Y will begin to come off after 3-4 mls of eluent; most will appear in another 4-6 mls. (See remarks 2 and 3.)

(7) If desired, the Y may be separated from the lead as follows. Concentrate the eluent from step 6 to < 1 ml, dilute with H_2O until ~5-7 N in HCl, and place on column prepared by washing with

6 N HCl. Elute with 6 N HCl. (See remark 4.)

(8) If the Pb carrier can be tolerated, along with an almost negligible amount of Nb and Zr, the procedure may be shortened to ~5 minutes if the sample is mounted after step 3. Filtering, using non-glass equipment, will reduce the time even further.

Remarks:

(1) Essentially all of the Y follows the Pb.

(2) A large part of the Pb will also appear in Y fraction.

(3) Any small amounts of Zr and Nb possibly present will remain on column.

(4) Pb will remain on column.

(5) General reference for this type procedure: Hicks, H. G., et al., "The Qualitative Anionic Behavior of a Number of Metals with an Ion Exchange Resin, 'Dowex 2'", Livermore Research Laboratory Report, LRL-65, Dec. 1953.

CHEMICAL SEPARATIONS

Element separated: Yttrium

Procedure by: Anders

Target Material: Zirconium foil

Time for sep'n: ~3 hours

Type of bbdt: 7.8 Mev deuterons

Equipment required: 10 and 50 ml beakers, 15 and 50 ml centrifuge cones, lustroid tubes, Dowex 2 resin (200-400 mesh), HCl form, two 10 cm x 0.4 cm columns.

Yield: ~80%

Degree of purification: ~10^4

Advantages: carrier-free separation for 4 pi absolute counting.

Procedure:

(1) Place yttrium tracer (for determining chemical yield) and target in 50 ml beaker; add ~3 ml conc. H_2SO_4 and 2 drops 30% H_2O_2; heat until dissolved, adding H_2O_2 until solution is clear.

(2) Transfer to 50 ml centrifuge cone, dilute to 30 ml, stir, add 1 mg niobium carrier.

(3) Add NH_4OH to precipitate $Zr(OH)_4$(carrying the Y), centrifuge and wash.

(4) Dissolve in minimum conc. HCl, dilute and reprecipitate, wash twice.

(5) Dissolve in minimum conc. HCl, add 3 mg Ca^{++} carrier, transfer to 15 ml lustroid cone.

(6) Add 2 ml H_2O and precipitate CaF_2 with 1 ml conc. HF, wash. (Y carries on the precipitate.)

(7) Transfer the slurried residue to 15 ml centrifuge cone, centrifuge and decant.

(8) Add 1/2 ml conc. H_2SO_4 and heat to fumes, dilute to 12 ml, heat in water bath for ~3 minutes, centrifuge.

(9) Add 2 mg Zr carrier to supernate, stir, make ammoniacal and centrifuge. Wash three times with conductivity water.

(10) Dissolve precipitate in minimum conc. HCl (~1/2 ml). Saturate with HCl gas.

(11) Transfer to Dowex-2 column. Adsorb and start collecting eluate after one column volume-- ~ 8 drops (1 drop/~7 sec).

(12) Soak in with 10 drops conc. HCl, elute Y^{+++} with additional 2 ml conc. HCl.

Remarks:

(1) General reference on anion exchange procedures: H. J. Hicks et al, Livermore Research Laboratory Report LRL-65, December 1953.

CHEMICAL SEPARATIONS

Element separated: Indium

Procedure by: Sunderman

Target Material: Misc. activities

Time for sep'n: 30-45 min.

Equipment required: Extraction funnel, (see remark 1). Other standard equipment.

Yield: ~90%

Degree of purification: 2 to 10^4 per cycle depending upon elements present.

Advantages: Good decontamination available from many elements.

Procedure:

(1) Prepare the solution containing 10 mg of indium and tracer for the contaminating element. Make up to 10 ml with a final concentration of 4.5 M hydrobromic acid.

(2) Contact this solution with 30 ml of diethyl ether in the cylindrical separatory funnel with rapid stirring for a period of two minutes.

(3) Allow the phases to separate and remove the lower aqueous phase, taking care that a very small amount of organic phase is also removed. (This is done by allowing the aqueous phase to drain from the vessel and one drop of organic phase to appear at the tip of the outlet tube.)

(4) Add 10 ml of 4.5 M hydrobromic acid solution to the organic solution in the vessel and stir rapidly for two minutes. Separate the phases as described in step (3).

(5) Repeat step (4) if greater decontamination is required.

(6) Add 10 ml of 6 M hydrochloric acid to the organic solution in the extraction vessel and stir rapidly for two minutes.

(7) Separate the phases and withdraw the aqueous phase but in this case allow one drop of aqueous solution to remain in the tip of the outlet tube.

(8) Take a portion of this aqueous solution and transfer to a culture tube for counting in the scintillation well counter or evaporate on plate and count.

Remarks:

(1) The separatory funnel used is cylindrical with an open top. The bottom of the cylinder is tapered very sharply (in about 1 cm of length) to a small bore, heavy-wall, ground-glass stopcock. The outlet of this stopcock is about 1 cm long and ground to a 45° taper to the tip (14).

(2) Contamination of foreign ions in this procedure (one cycle):

Antimony	2.2%	Ruthenium	0.015%
Cerium	0.01%	Selenium	23.0%
Cesium	0.015%	Silver	0.055%
Chromium	0.012%	Strontium	0.006%
Cobalt	0.009%	Tantalum	0.08%
Iodine	6.7%	Tin	48.0%
Iridium	0.017%	Zirconium	0.018%

For further information see pg. 139 of ref. (14).

CHEMICAL SEPARATIONS

Element separated: Indium

Procedure by: Sunderman

Target Material: Misc. activities

Time for sep'n: 1 hour

Equipment required:
Column 9 cm x 4 mm dia.
Dowex 2 x 8, 200-400 mesh.

Yield: 95%

Degree of purification: 2 to 10^3 depending upon the elements present.

Advantages: Carrier-free

Procedure:

(1) Add the sample in 6 M hydrochloric acid to the top of an 8.5 cm column of resin (Remark 1) and allow it to seep into the resin bed. (Remark 2.)

(2) Elute with 6 M hydrochloric acid using a flow rate of 1 ml in three minutes and saving the 6 through 20 ml portion which contains a majority of the indium.

Remarks:

(1) The sample must be added directly to the top of the resin and not allowed to contaminate the reservoir above the resin. It must then be allowed to seep into the resin bed until the solution level reaches the top of the resin.

(2) No air is allowed within the resin bed. If the sample is "pushed" into the resin bed by air or otherwise forced into the resin it will spread itself over the entire column and separation will be negated.

(3) Contamination of foreign ions in this procedure:

Antimony	2.0%	Ruthenium	22.0%
Cerium	0.13%	Selenium	0.3%
Cesium	0.11%	Silver	25.0%
Chromium	0.6%	Strontium	0.1%

Cobalt	43.0%	Tantalum	6.2%
Iodine	1.1%	Tin	0.4%
Iridium	3.7%	Zirconium	31.0%

(4) Further information on this separation is given in reference 14.

(5) General information on this type of ion exchange separation is given by Hicks and coworkers (15).

B. Radiochemical Separations

1. The Development and Evaluation of Radiochemical Separation Procedures for Barium, Calcium, Strontium, Silver and Indium

The objective of this research was to conduct a program of development and critical evaluation of radiochemical separation procedures. Procedures for individual elements reported in the literature were collected and subdivided into individual separation steps. Those steps which were found unique and possessing general applicability were studied experimentally to determine optimum conditions (of both yield and decontamination) for separation. These steps were then further evaluated under optimum conditions to determine the effects on the separation of a number of diverse but representative elements and materials.

The alkaline earth elements barium, calcium and strontium were the first elements studied in this manner. It was found that the conditions for nitrate, chromate and chloride precipitations must vary widely from commonly accepted analytical methods due to the demands of such factors as non-equilibrium operation, necessity for rapid precipitation, character of the precipitate and manipulatory techniques. Yield data were obtained for barium, calcium and strontium in the above separations under conditions varied to show the effects of excess or deficiency of reagents, quantitative or nonquantitative precipitation and methods of adding precipitating reagents. Decontamination factors were determined with tracers of a large number of typical elements (Ag, Ba, Ca, Ce, Co, Cr, Cs, I, Ir, Ru, Sb, Se, Sn, Sr, Ta, Zr). An optimum procedure was determined for the systematic separation of these three ions in tracer solutions.

The precipitation of the hydrous oxides of iron (III) and lanthanum (III) for scavenging purposes has been studied and their action and efficiency have been evaluated. Representative tracers have been used to determine the portion of each specie which would be carried either in the presence or absence of macro amounts of the contaminating activity. It was found that higher decontamination may be accomplished by scavenging in the presence of carrier amounts of the contaminating species. Lanthanum precipitation is more specific with respect to the activities carried, while the more bulky iron hydroxide may give higher decontamination factors for certain elements.

The principle of isotopic exchange has been adapted to a novel separation and determination of radioactive silver in a mixture of radioactive species, and a quantitative procedure has been developed. The separation of radioactive silver by isotopic exchange with a silver chloride surface was found to be quantitative in fifteen minutes or less, even in the presence of high concentrations of many inorganic salts and organic solvents. The method is highly specific for silver, with decontamination factors between 10^3 and 10^7 for eighteen representative radioactive species. The technique was also applied to the separation of silver from fission product mixtures and bombardment samples and found satisfactory in both cases.

The standard procedures for the separation of radioactive silver, by chloride precipitation and electro-deposition, as well as a new method, that of precipitation with benzotriazole from ammoniacal versene solution, were evaluated. The precipitation reactions produce decontamination factors from 40 to 200 for many elements, those made in acid solution being more specific than those in basic solutions. The electrodeposition of silver affords decontamination factors from 100 to 500 for many elements. A sample procedure is given which illustrates the high decontamination of which these methods are capable.

The separation of indium by sulfide precipitation, by solvent extraction of the bromide with diethyl ether, and by anion exchange in hydrochloric acid solution has been evaluated. A comparison of the efficiency of separation from the representative radioactive tracers showed that the sulfide precipitation lacked specificity as a decontamination step. The use of solvent extraction was found to give decontamination factors of 10^4 for dissimilar elements when conditions were well standarized. The technique of ion exchange loses much of its specificity when the flow rate is high as is required when time is an important factor. When operating far from equilibrium with a short column, however, decontamination factors of about 10^3 were possible for many elements with a procedure that requires one hour.

Experimental results for the barium, strontium calcium and silver separations were reported in the last progress report (1). The same type of yield and contamination data for indium separated both by H_2S precipitation, by solvent extraction of the bromide into ether solution and by ion exchange are given in the table below. These results are discussed in much more detail in the report by Sunderman (14). (D. N. Sunderman)

Method	H_2S Pptn.	Extraction	Ion Exchange
Element	Percentage carried with the indium		
Antimony	96.	2.2, 0.25*	2.0
Cerium	20-70 (var.)	0.01	0.13
Cesium	2.0	0.015	0.11
Chromium	1.0	0.012	0.6
Cobalt	1.7	0.009	43.
Indium	91.8±2.5	93.0±2.0	95.5±1.2
Iodine	4.3	6.7, 6.8*	1.1
Iridium	4.9	0.017	3.7
Ruthenium	96.	0.015	22.
Selenium	80.	23., 7.*	0.3
Silver	100.	0.055	25.
Strontium	100.	0.006	0.1
Tantalum	1.6	0.08	6.2
Tin	97.	48., 40.*	0.4
Zirconium-Niobium	8.7	0.018	31.

* These values were obtained with a procedure employing two 4.5 M HBr washes.

2. Temperature Dependence of Silver-Silver Chloride Exchange

The temperature dependence of the silver-silver chloride exchange has been studied at 2°, 26°, 38°, 53°, 64°, 74°, 84° and 96° Centigrade using radioactive silver-110. The silver chloride is present as a thin coating on a platinum gauze electrode. The preparation of the electrode, the exchange procedure, the results of the experiments, and the errors inherent in the methods used are discussed in detail in a separate paper (16) accompanying this report. This work was undertaken in conjunction with the program of separation of radioactive silver by isotopic exchange described by Sunderman (14). (T. W. Olcott)

3. Other Separations by Isotopic Exchange

Since the separation of radioactive silver by the isotopic exchange method was so outstanding the possibility of using the same method with other elements and compounds was explored. A search of the literature revealed a number of compounds which exchanged rapidly with their metal ions. The problem then was to try to mount this compound on the platinum gauze in a manner similar to the silver chloride.

Several compounds of cobalt, including the sulfide, phosphate, and oxalate were tried; in addition manganese dioxide, several compounds of tin, ruthenium sulfide, and some rare earth hydrous oxides were also tried. The number of possible compounds available for this separation was reduced considerably when the difficulty of electroplating the metal onto the electrode and the additional difficulty of changing the metal to the compound by back electrolysis were considered.

Of the elements and compounds tried, only cobalt sulfide and manganese dioxide gave encouraging results but even these could scarcely be called satisfactory. For cobalt sulfide a high temperature exchange for 5 minutes gave a yield of 56% of the cobalt-60 with contaminations of 86% for ruthenium-rhodium-106, 11% for silver-110, 12% for zinc-65 and 6% for cesium-134. For the manganese dioxide at 20°, a 5-minute exchange gave a yield of 30% for manganese-54, with contaminations of 20% for cobalt-60, 16% for zirconium-niobium-95 and tantalum-182, 13% for ruthenium-rhodium-106, 10% for cerium-144, 6% for cesium-134 and antimony-124, 4% for silver-110 and less than 1% for chromium-51 and selenium-75.

The results were discouraging for the other elements and compounds tried since it either was not possible to make a suitable electrode of the compound which would stick or the contamination by other elements was so great or the yield of the desired element so small that it offered no advantage whatsoever.

It is interesting to note that in the case of certain compounds such as manganese dioxide that the crystal form that exchanges readily is different from the crystal form that is obtained on the electrode. Thus it is not always possible to extrapolate data given in the literature for heterogeneous exchange reactions to use in an exchange separation.

This cursory report of the exchange separation will be written up in more detail in the near future. It does seem to indicate, however, that the separation of radioactive silver by isotopic exchange is rather unique in separation technology and thus that the general method of separation by isotopic exchange has a rather limited application.

(H. W. Wissenberg, W. W. Meinke)

4. Literature Search and Punch Card Compilation

Little additional work has been done during the past year on the punch card compilation of chemical procedures and consequently a revision of the microfilm has not been made. It is still felt that this compilation is important and work will continue on it in the future as suitable personnel become available.

C. Activation Analysis

1. Study of the Use of Antimony-Beryllium Sources in Activation Analysis

For the past year a program of research has been carried out to further explore the possible uses of portable neutron sources in activation analysis. Several years ago the use of radium-beryllium sources was explored and it was found that while they did not give highly sensitive methods, it was possible with their use to discriminate against a large number of elements in a mixture which would not give any activity with the small neutron flux available. Actually it was found that of all the elements of the periodic table only silver, rhodium, and indium would give radioisotopes whose radiations were penetrating, enough to be detected in a thick sample after an irradiation of several minutes. Procedures for analysing for milligram amounts of these elements by activation with a portable radium-beryllium source were included in an earlier publication (17).

Since antimony-beryllium sources which would give a neutron flux some 20 to 50 times greater than that of a 25 mg. radium-beryllium source were available from Oak Ridge, it was decided to explore the uses of this type of portable source. The primary disadvantage of the antimony-beryllium source is the bulky shielding which is required to reduce personnel hazards. The equivalent of 6 to 8 inches of lead is required around the source. An inexpensive yet satisfactory irradiation chamber was set up for this program and is shown in Figs. 25 and 26. When not in use the antimony-beryllium source was stored in the lead pig at the right of Fig 25. For an irradiation it was transferred by tongs to the hole in the paraffin block in Fig. 26 and the paraffin and lead cover slid over the top with tongs. Even though the source is very active (several curies of antimony-124) it has been possible to work routinely with this simple arrangement making as many as 50 - 75 irradiations a week without the operator picking up more than 1/10 tolerance dosage.

After irradiation the analyses were carried out by measuring the gamma radiations of the radioisotopes using three different detection devices, a scintillation well counter, a manual sweep spectrometer (Atomic Instrument Company) and an automatic sweep spectrometer (RCL Spectrogammeometer). The relative advantages and disadvantages of these instruments and methods were

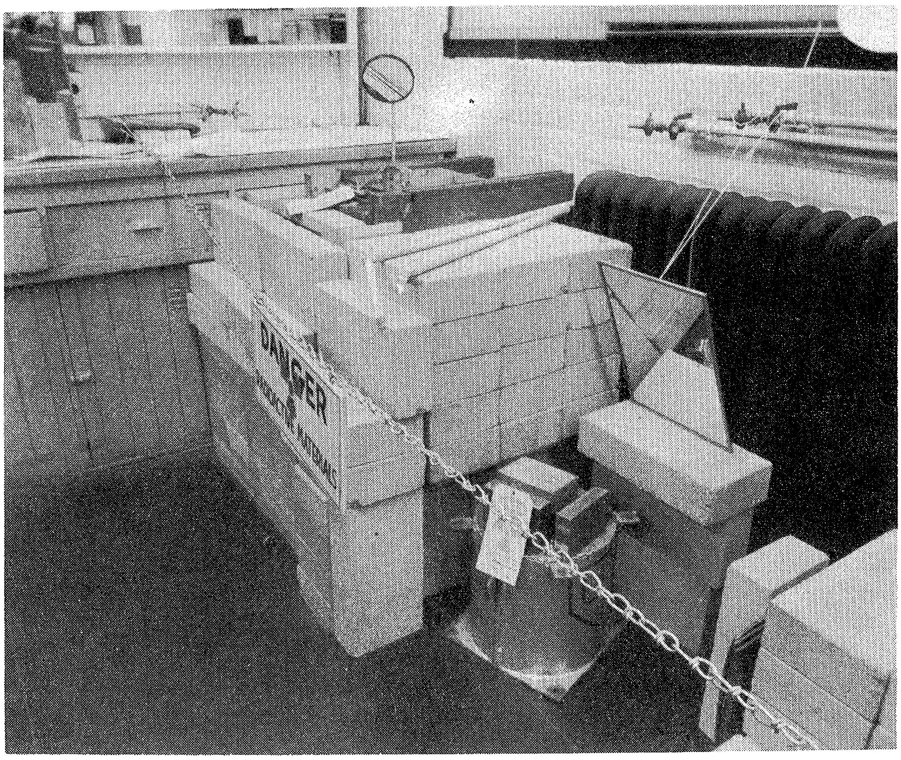


Fig. 25. Shielding for antimony-beryllium source.

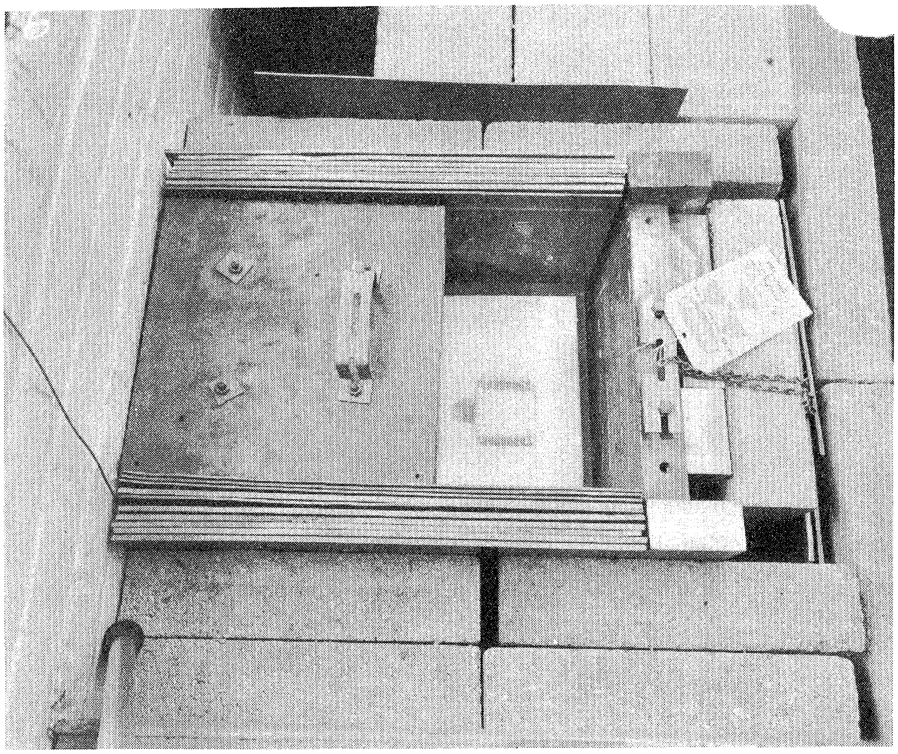


Fig. 26. Antimony-beryllium source set-up, top view.

determined.

In addition synthetic solid mixtures of a number of elements or their appropriate salts or compounds were irradiated for a time, varying from 30 minutes to 2 hours, depending upon the desired amount of induced activity. Each irradiated sample was allowed to stand for three minutes in order that the short-lived daughter isotopes, if any, decay and then was measured. Comparative standards of a pure element were employed for quantitative measurements. Several analyses were also done on mixtures using one-step chemical separation procedures to isolate or partially concentrate the desired isotope which, after purification, was counted in the well counter.

The procedures were run on certain National Bureau of Standards alloys as well as minerals of known composition. The program using the well counter included the analysis of silver and cobaltic oxide, and silver and lead iodide mixtures; the analysis of manganese and ferromanganese alloys and pyrolusite; the analysis of copper and aluminum in the presence of other elements; and the analysis of iodine in mixtures after a one-step separation. Determinations of manganese in mixtures were also studied with the manual sweep scintillation spectrometer. With the automatic sweep spectrometer studies were made of the analysis of manganese in mixtures as well as in ferromanganese alloys and in pyrolusite.

The sensitivity of activation analysis using the antimony-beryllium neutron source was also explored. Calculations were made and checked by some experimental work to indicate the elements which would give certain levels of activity after a specific radiation time in the source. Plots have been made of a number of these groupings of elements with the same sensitivity values. Manganese was taken as a reference standard for the calculations.

This work has been completed under the sponsorship of the Michigan Memorial Phoenix Project and is being written up for publication at the present time. (A. K. De, W. W. Meinke)

2. Neutron Activation Cross Section Graphs

Activation cross section values for thermal neutrons are important primary data for the analytical chemist since they indicate the relative probability with which a given radioisotope will be formed when a substance is exposed to a source of neutrons. Thus the detection sensitivity for an element in activation analysis as well as the yield of tracers in an irradiation depend upon these values.

Highes and the Neutron Cross Section Group at Brookhaven National Laboratory have for a number of years been compiling these thermal neutron cross section values as well as values for higher energy neutrons and first made their compilation available in a report in 1952 (18). Meinke and Anderson (17) found that these cross section values could readily be summarized by plotting them against the half-life of the daughter radioisotope produced. This type of graph has proven very useful in discussions of the use of low level portable neutron sources (17) in activation analysis as well as in considerations of the sensitivity of activation analysis (19).

Recently Hüghes and coworkers have revised their cross section values (20). These new "best" cross sections have now been plotted against the half-lives as in the previous graphs -- and in addition the standard error quoted by Hüghes for each value is included on the graph. These graphs are being submitted for publication at the present time. (R. Maddock, W. W. Meinke)

3. Activation Analysis with the Michigan Reactor

The University of Michigan reactor and the Michigan Phoenix Memorial Building present an excellent integrated facility for work with short half-lived radioisotopes. The reactor will be connected with the hot chemistry laboratories by a pneumatic tube system which will deliver samples into the hoods six seconds after the end of irradiation. The sample can then be utilized in nuclear chemical characteristizations of the short-lived radioisotopes induced, or it can become part of a scheme of trace analysis by activation.

The current research program carried out in nuclear and radiochemistry has included many studies of this type with radioisotopes of longer half-lives. Decay schemes and absolute cross section values of radioisotopes produced in the Michigan cyclotron have been determined with spectrometers and absolute counting equipment built up during the past few years. Similarly, research on activation analysis using portable sources has been carried on for several years to develop techniques and gain insight into the use of neutrons for this method of analysis - preparatory to using the Michigan reactor.

After irradiation in the reactor, the sample contains many different contaminating activities which must be eliminated or discriminated against before the desired constituent can be accurately measured. This discrimination can be done by either chemical or physical means (or both). At present elaborate chemical methods are utilized. Unfortunately, very few chemical procedures giving good decontamination can be completed in less than a few minutes. Hence physical discrimination in the measurement of the shorter-lived radioisotopes must be relied upon. This takes the form of gamma-, beta-, and x-ray spectrometry. Since the energy spectrum of one isotope is, in general, quite different from that of another, it is often possible to measure a part of the spectrum which maximizes detection of the desired constituent and greatly reduces the effects of contaminating materials.

At the present time spectrometers for gamma-, beta- and x-rays are available in the nuclear chemistry program. The spectra from these instruments are obtained by use of a single-channel pulse height analyser which feeds a Speedomax pen recorder. This equipment is entirely satisfactory for radioisotopes whose half-lives are long compared to the

10, 40 or 100-minute sweep times of the analyser. It, however, limits to 10 - 20 minutes the half-lives of activities what can now be studied.

When multi-channel pulse height analyser equipment can be coupled with the pneumatic tube system of the reactor it will lower this half-life limit to a few seconds. This integrated "rapid delivery" irradiation facility of the Phoenix Project is practically unique, even in government laboratories, since the sample is delivered directly to a chemical hood for rapid processing and measurement.

Several typical examples will illustrate the great extension of the range of work possible with the pneumatic-tube system (in some cases with the multi-channel analyser proposed).

a) Silver when irradiated with neutrons gives two short-lived activites of 24.5 sec. and 2.33 min. with very high yield. With rapid chemical separations now available, it should be possible to make a complete silver analysis on any type of sample within five minutes with this equipment. (This time presupposes some work preliminary to the irradiations for preparing certain difficult samples such as ores for the analysis). At present it is not possible to work with these silver activities in the spectrometers since more than 95% of them would decay during one sweep of the single-channel equipment. Oak Ridge in their activation analysis service makes use of the 270-day silver isotope preduced in 100-times smaller yield during a one month's bombardment for their silver determination.

A number of other elements such as aluminum, vanadium, rhodium, germanium and zinc give short-lived isotopes with half-lives of the order of one minute for which similar saturation bombardments could easily be made.

b) Certain of the elements of low atomic number are very important in physical and biological systems but neutron activation of these elements gives no isotope with a half-life greater than a few minutes. For example by (n,γ) reactions fluorine gives a 12-second fluorine-20 radioisotope, oxygen-18 a 29-second oxygen-19, nitrogen a 7.3-second nitrogen-16, while fluorine gives a 29-second oxygen-19 by an (n,p) reaction. It will be possible with this equipment to analyse for small amounts of these elements by neutron activation without chemical separation, by delivering the sample from the pneumatic tube directly into the counting chamber of the spectrometer.

d) A look at an up-to-date isotope chart shows many short-lived radioisotopes which are poorly characterized and whose mass assignment is in doubt. An example is the 2.4-second isotope tentatively assigned to carbon-15. Irradiations of carbon-14 in this facility would permit characterization of this carbon isotope. Decay schemes for many radioisotopes with half-lives from a few seconds to a few minutes could be studied -- thus extending tremendously the range of the present spectrometry program. In some cases, such as the carbon-15 where physical discrimination is sufficient, no chemistry would be required, In other cases, rapid chemical separations would be coupled with spectrometry to give sufficient discrimination. (W. W. Meinke)

IV PERSONNEL, PUBLICATIONS, TALKS

A. Personnel Listing

Staff Meinke, W. W.

Graduate Students Anders, O.*

Gardner, D.* (Eastman Kodak Fellowship, Fall, 1956)

Sunderman, D. N.*(') (Predoctoral Fellow, Michigan Memorial Phoenix Project, Fall, 1955)

Undergraduate Students Burton, A.**

Corteg, W.**(')

Nagler, B.**(')

Non Staff Ackermann, I.**(')

De, A.K.(') (Michigan Memorial Phoenix Project No. 95)

Maddock, R. S.

Wahlgren, M.*

Warren, G.(') (Michigan Memorial Phoenix Project No. 121)

Wissenberg, H. (Summer only) (')

Typing Anders, E.**(')

Warren, J.**(')

Electronics Shideler, R. W.*

* Half time

** Hourly

(') Terminated

B. Papers and Reports Published

1. A Versatile Hot Lab for University Research. W. W. Meinke, A. H. Emmons and H. J. Gomberg. Nucleonics 13, No. 11, 76 (1955). 4 pages, 7 figs.
2. Silver-111 Beta-Ray Sources. W. Wayne Meinke and D. N. Sunderman. Nucleonics 13, No. 12, 58 (1955). 2 pages, 1 fig. (AECU-3070).
3. Decay Scheme of In-107. Wayne A. Cassatt, Jr. and W. Wayne Meinke. Phys. Rev. 100, 1372 (1955). 2 pages, 2 figs.
4. Utilization of Isotopic Exchange for Radiochemical Separations. D. N. Sunderman and W. W. Meinke. Preprinted and presented at the Nuclear Engineering and Science Congress, Cleveland, Ohio, December 14, 1955.
5. The Development and Evaluation of Radiochemical Separation Procedures for Barium, Calcium, Strontium, Silver and Indium. Duane N. Sunderman. Nuclear Chemical Research Project Report, University of Michigan, February 1956. 197 pages, 25 figs. (AECU-3159).
6. Annual Reviews of Fundamental Analytical Chemistry: Nucleonics. W. Wayne Meinke. Anal. Chem. 28, 736 (1956). 20 pages.

7. Beta Gage for Localized Measurements of Thin Films. Oswald U. Anders and W. Wayne Meinke. Rev. Sci. Inst. 27, 416 (1956). 1 page.
8. Temperature Dependence of Silver-Silver Chloride Exchange. T. W. Olcott. Nuclear Chemical Research Project Report, University of Michigan October, 1956. 13 pages, 8 figs.
9. Radiochemical Separations: I Barium, Strontium and Calcium. Duane N. Sunderman and W. Wayne Meinke. (In process of revision along lines of referee's comments for Anal. Chem.)
10. Separation of Radioactive Silver by Isotopic Exchange. Duane N. Sunderman and W. Wayne Meinke. To be published in the Proceedings of the XVth International Congress of Pure and Applied Chemistry (Analytical), Lisbon, Portugal, September 9-16, 1956.
11. High Energy Excitation Functions in the Heavy Region. W. W. Meinke, G. C. Wick and G. T. Seaborg. Journal Inorg. and Nucl. Chem. (In press.)

C. Talks

1. W. W. Meinke, Discussion leader on Instrumentation at Symposium on Trace Analysis, The New York Academy of Medicine, New York City, November 2-4, 1955.
2. W. W. Meinke, "Activation Analysis", Symposium Program, Western New York Section, American Chemical Society, Buffalo, New York, November 19, 1955.
3. D. N. Sunderman (with W. W. Meinke), "Utilization of Isotopic Exchange for Radiochemical Separations", presented by D. N. Sunderman at the Nuclear Engineering and Science Congress, Cleveland, Ohio, December 14, 1955.
4. D. G. Gardner, "Nuclear Bombardment of Semiconductors", U. of M. Chemistry Department Seminar, Ann Arbor, January 5, 1956.
5. W. W. Meinke, "Fundamentals and Applications of Activation Analysis", The St. Louis Society of Analysts, St. Louis, Missouri, February 28, 1956.
6. W. W. Meinke, "Activation Analysis", Association of Analytical Chemists (Anachems), Detroit, Michigan, April 2, 1956.

7. W. W. Meinke, "Chemistry (Nuclear and Analytical) Program for the University of Michigan Reactor", U. of M. Chemistry Department Seminar, Ann Arbor, May 10, 1956.
8. W. W. Meinke, "Gross Fission Products as Gamma Ray Sources" presented at the First Ann Arbor Industry - Education Symposium on "Sources of Radiation for Industry", Ann Arbor, May 11, 1956.
9. W. W. Meinke (with D. N. Sunderman), "Separation of Radioactive Silver by Isotopic Exchange" presented by W. W. Meinke at the XVth International Congress of Pure and Applied Chemistry (Analytical), Lisbon, Portugal, September 14, 1956.
10. W. W. Meinke, "Fundamentals of Radioactivity" presented in the Michigan Memorial Phoenix Project Seminar, "An Introduction to the Use of Isotopes in Biology", Ann Arbor, October 2, 9, and 16, 1956.

V ACKNOWLEDGEMENTS

The kind cooperation of Professor W. C. Parkinson and the crew of the University of Michigan cyclotron in furnishing bombardments is greatly appreciated.

The help of the Isotopes Division of the Atomic Energy Commission in supplying both enriched and radioactive isotopes for use in this work is acknowledged.

We also appreciate the assistance of the Michigan Phoenix Memorial Project in generously supporting part of this program of research under Phoenix Projects Nos. 95 and 121, and in making available additional laboratory space for this research in the Phoenix Building.

VI LIST OF REFERENCES

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5. E. L. Kemp, "Scaler Model SC-3", Los Alamos Laboratory Report LA-1207, (July 1950). V. G. Rexroth, "Model PA-2 Pulse Amplifier", Los Alamos Laboratory Report LA-1239, (May 1951).
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7. O. U. Anders and W. W. Meinke, Rev. Sci. Inst. 27, 416 (1956).

8. J. P. Palmer and L. J. Laslett, Atomic Energy Commission Report, AECU-1220, (March 1951).

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