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Semi-annual Progress Report
November 1, 1964 - May 1, 1965

AN INVESTIGATION OF COMPUTER-COUPLED, AUTOMATIC, REMOTE ACTIVATION ANALYSIS FOR LUNAR SURFACE ANALYSIS

Conducted Through the Texas A&M Research Foundation

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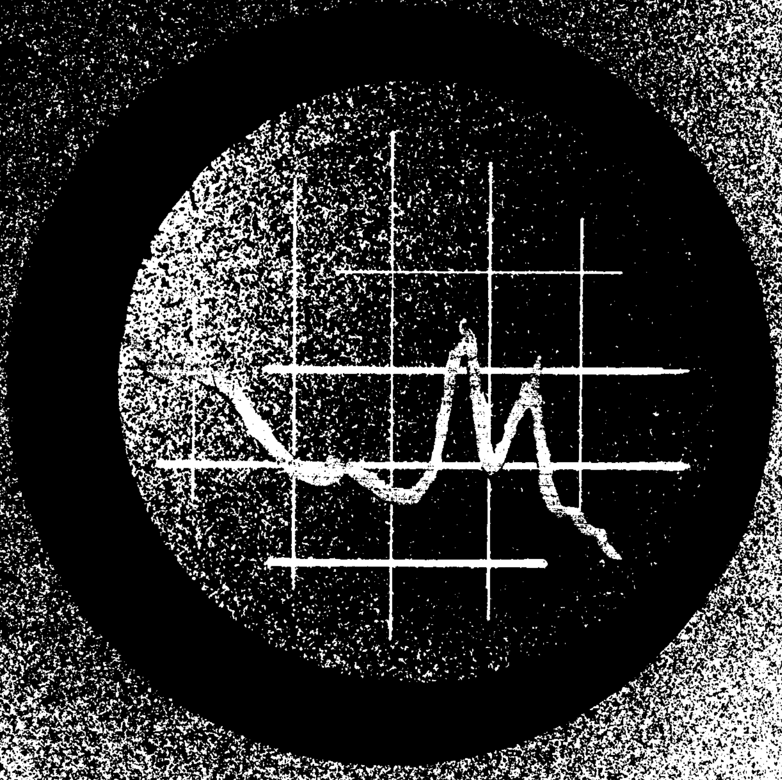

Activation Analysis Research Laboratory
Texas A&M University

AN INVESTIGATION OF COMPUTER-COUPLED, AUTOMATIC, REMOTE ACTIVATION ANALYSIS FOR LUNAR SURFACE ANALYSIS

Semi-annual Report
May 1, 1965

A report of research being performed under
National Aeronautics and Space Administration

Through the
Texas A&M Research Foundation

Manchery P. Menon, Ph.D.
Editor

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3. M. P. Menon, Ph.D., Assistant Professor of Chemistry, A.A. R.L. and Editor of this report.

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I. Introduction

The present semi-annual report represents a summary of the work performed during the latter part of the contract year, November 1, 1964, to April 30, 1965. Major effort was directed, during the last six months, to improving the analytical techniques for the determination of the five major elements that might be measured on the lunar surface using 14 Mev neutron activation techniques with emphasis on the upper and lower limits of measurement. Other areas of related work some of which are in progress are also described in this report.

Although several authors including Prof. Urey have suggested that the chemical composition of lunar surface material may be similar to that of chondritic meteorites, recent ranger shots that revealed close-up pictures of the moon have led many scientists to ponder whether it may have an entirely different composition. It is hard to prove or disprove entirely any hypothesis concerning the nature of the lunar surface unless an actual analysis is carried out; but the analyst should be prepared to analyze any kind of material his instruments may have access to. It is therefore important to determine how small a quantity of various elements can be measured in any matrix using the present techniques and what type of interference one can expect from other expected major components of the sample. For a specified volume or mass of the material to be analyzed there is also an upper limit of measurement of a particular element under a pre-chosen set of conditions due to such factors as the dead time loss encountered in the analyzer and this also has to be investigated before optimizing the procedural steps. Answers to the above questions were sought by analyzing "simulated moon matrix" samples with four of the five major elements fixed and different proportions of the fifth using pre-defined parameters for activation and counting. This work has been described in detail in Chapter II.

Development of a dual crystal NaI(Tl) - CsI(Tl) scintillation detector, in our laboratory, for the reduction of Compton contribution in gamma-ray spectrometry has been reported in the previous semi-annual report. The principle of operation of this detector system is essentially based on the reduction of the rise and decay times of the photomultiplier output pulse produced by a Compton interaction with the NaI crystal by the addition of another light pulse resulting from the interaction of the same Compton scattered gamma with the CsI crystal. This detector system has been used extensively to test and evaluate the degree of the Compton reduction that may be achieved in gamma-ray spectrometry. The limitations of the presently used system and the modifications that are necessary to get better results have been summarized in Chapter III.

It has been an important objective of this Laboratory in the past to seek or design systems which produce the best results from the analysis, for the five major elements, of any geological material with minimum restrictions. It was thought that coupling five single channel analyzers with a detector and choosing the proper window width for the principal gamma-ray peaks of these elements may help to perform the analysis in a minimum amount of time with simplified equipment. If the laboratory experiments prove to be successful such a system, which is expected to be lighter than the multichannel analyzer, might be recommended for substitution for the latter in future probe. Preliminary work performed so far along these lines is reported in Chapter IV.

All the samples so far analyzed by 14 Mev neutron activation techniques are terrestrial samples. It was felt desirable to apply similar techniques for the analysis of an extra-terrestrial sample, meteorites for instance, which may be quite similar in composition to lunar surface material. Although much work has been done to measure the abundances of most of the constituents of the chondritic meteorites the majority of the analyses for the major elements were performed using conventional gravimetric methods. Activation analytical techniques which might give better results on the abundances of these elements are now being employed to measure the concentrations in chondritic meteorites and also in two terrestrial samples, siderolite and tektite. Preliminary work on the measurement of these elements in the above samples together with a method for measuring the individual activities of Mn^{56} and Mg^{27}, both of which have virtually identical gamma-ray peaks, are described in the last chapter.

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II. Tolerance Levels of Interference from one Element to the Other in 14 Mev Neutron Activation Analysis of Simulated Moon Matrix - by M. Y. Cuypers, Ph.D.

Introduction: A lunar surface composition similar to that of chondritic meteorites or igneous rocks has been suggested by several authors (Urey, 1960; Fite et al, 1962). The major components of these rocks, which seem to be feasible for analysis by 14 Mev neutron activation technique are oxygen, iron, aluminum, silicon and magnesium. Other major components, such as, carbon, calcium, sulphur, titanium and nickel do not have good nuclear characteristics to be analyzed with this technique. Sodium and potassium will produce radioisotopes whose gamma-ray spectra are completely overlaped by those due to the first five elements.

The study of the feasibility of the analysis of oxygen, iron, aluminum, silicon and magnesium has been completed and described previously (Fite et al, 1964). Considering the possibility that the lunar surface may contain these five major elements but in different ratios than were observed in stone meteorites or igneous rocks, it was felt worthwhile to investigate the upper and lower limits of measurement of the above elements. In this study, answers to two specific questions were sought:

a) What is the minimum detectable concentration for each of the five elements in the presence of a normal moon matrix? A "normal moon matrix" will be defined later.

b) What is the upper tolerance limit for each major element present in a simulated moon matrix?

Because this general study is especially concerned with the dead time of the analyzer and also the shape of the gamma-ray spectrum obtained after activation of a mixture with 14 Mev neutrons, and that the size of the sample has no major influence on the answers for the two previous questions, this experimental work was done with a Cockcroft-Walton accelerator using 1 g samples.

<u>Experimental Procedure</u>: A "simulated moon matrix" was defined as a mixture of the oxides of iron, silicon, aluminum and magnesium in a proportion that corresponds to an average percent of each of these elements in igneous rocks or meteorites. Each oxide has been dried carefully in the oven. Table I shows the proposed lowest and highest per cent abundance for each of the five major components and in the last column the average value. A mixture containing Mg, Si, O, Al and Fe in a proportion

defined by this average value, was used, in these experiments as a simulated moon matrix.

To answer the first question as to what the minimum detectable concentration is for an element in a normal moon matrix and the second question, how much of one element is needed to make the analysis of another element impossible, a mixture containing four out of the five elements was prepared. These four elements were mixed in a proportion of the simulated moon matrix. In most cases 200 mg of these mixtures was taken and mixed with different amounts of the fifth element going from a very low amount, hardly detectable, to a very high, where it may possibly interfere with the determination of the other four elements. All the samples were packed in polyethylene vials.

The sample containing the mixtures of different compositions is transferred by a pneumatic system to the irradiation position. After an irradiation time of 5 minutes, the samples are counted several times, with a waiting time depending on the type of the mixture considered. The transfer time for the samples was 3 seconds and the flux of 14 Mev neutrons used was about 10^8 n/sec-cm^2.

Results: Four different sets of mixtures of oxides of Al, Fe, Mg and Si were prepared. In each set the concentration of one of the components was varied, while keeping the other elements in the same ratio as was chosen for the simulated moon matrix.

In each mixture K_2CO_3 was added, because potassium is one of the major elements that could be present. The precise determination of this element seems to us to be difficult, because of the many interferences. The K^{38} formed by $K^{39}(n,2n)K^{38}$ reaction emits positrons, whose annihilation produces two .51 Mev gamma rays. Many other elements such as, bromine, copper, chlorine, etc., on reaction with 14 Mev neutrons also produce positron emitters; therfore, the interferences are multiple.

Mixture I: Each sample of these mixtures is composed of the five elements including potassium with the following proportions by weight:

24.7 mg	of Mg
56 mg	of Si
6.7 mg	of K
18.6 mg	of Fe
87.6 mg	of O_2

The amount of Al added was variable. Except for potassium, oxides were used to make the mixtures.

a) Measurement of Silicon: The determination of the silicon, by the Al^{28} activity, shows a reproducibility within 6%. Table II shows the number of counts per mg of silicon obtained under the photopeak of 1.78 Mev.

b) Measurement of Aluminum: The determination of Al by the gamma peak coming from Mg^{27} is given in Figure 1. This calibration curve shows that aluminum can easily be determined in a matrix with the above composition. When the concentration of Al starts to be low, the activity due to Mn^{56} make the analysis less precise. The curve is not linear anymore under our experimental conditions when Al/Fe ratio in the mixture is $> 1/3$. This gives our detection limit for the determination of aluminum.

The results from the measurement of Fe content of this mixture, using the .84 Mev gamma ray coming from Mn^{56} are shown in Table III.

The waiting time was 80 minutes after the end of the irradiation. The reproducibility (difference between the mean value and the upper and lower values) is in this case, within 6%.

In this case where 105.7 mg of Al was present, 10% of the activity after 80 minutes waiting time was still due to the Mg^{27} activity. But in this case the Al/Fe ratio is 5.5. In a simulated moon matrix the ratio of Al/Fe is 0.7 and then the activity due to Mg^{27} is not higher than about 2% that of Mn^{56}. The Na^{24} activity coming from Mg and Al was not high enough to be calculated precisely.

Mixture II: Each sample of these mixtures is composed of the five elements including potassium with the following proportions by weight:

47.2 mg of Mg
26.4 mg of Al
35 mg of Fe
12.9 mg of K
76.2 mg of O_2

The amount of SiO_2 added was variable.

a) Measurement of Silicon: The Al^{28} activity due to silicon has been measured. Figure 2 shows the calibration curve of silicon in this particular mixture. The lowest limit of sensitivity for silicon in this mixture is about 1 mg. This is .5% of Si in the 200 mg mixture. Figure 3 shows a typical gamma-ray spectrum of this mixture containing .98 mg of silicon. Silicon was not added in lower amounts than 0.98 mg, because the homogeneity was not good. Also when lower amounts of silicon are present, we can suspect a relatively higher contribution in this energy region coming from the 1.81 Mev gamma ray of Mn^{56} produced by the $Fe^{56}(n,p)Mn^{56}$ reaction. The upper limit for the silicon concentration was drastically reduced by the fact that the dead time in the analyzer became too high to have significant counting losses when too much silicon was added. When 140 mg of silicon was present this factor influenced the quantitative results considerably. For 110 mg of silicon, which is the quantity present in the simulated moon matrix, however, the activity due to Al^{28} has disappeared after 20 minutes waiting time.

b) Measurement of Aluminum: The photopeak counts for the .84 Mev gamma ray due to Mn^{56} and Mg^{27} in this mixture, after 1 minute waiting time stays constant within 15%. Table IV shows the results of the .84 Mev activity as a function of the mass of silicon present and percent dead time in the analyzer. If 0-20% dead time is used for the analyzer, the reproducibility is within 7%.

Figure 4(a) shows a spectrum where only 100 mg of mixture has been used, instead of 200 mg. The .84 Mev gamma-ray activity is too low to be estimated precisely. However, after 11 minutes waiting time [see Figure 4(b)] this activity can be easily estimated. The Na^{24} activity due to Mg and Al has not been measured in this mixture, but the different masses of silicon should not vary this activity because, a waiting time of 30 minutes is always allowed before counting the 1.36 Mev gamma ray peak of Na^{24}. The same is valuable for the .84 Mev peak after 30 minutes waiting time, giving the determination of iron. Indeed in Mixture I the reproducibility for the iron measurement was within 6%. In Mixture II the Al^{28} activity cannot interfere after 30 minutes waiting time because of the complete decay of Al^{28} at that time.

Mixture III: Each sample of these mixtures is composed of the five elements including potassium with the following proportions by weight:

24.8 mg of Mg
14.1 mg of Al
6.7 mg of K
56 mg of Si
97.1 mg of O_2

The amount of iron oxide added is variable.

a) Measurement of Silicon: The determination of silicon in this mixture was made after a 1 minute waiting time. Table V shows the activity of Al^{28} as a function of different size mixtures.

The reproducibility in this case is about 6% and the table shows that the iron, even if present in a high amount, does not affect the determination of silicon in this case. The silicon has to be present in a submilligram quantity to have serious interference from iron.

b) Measurement of Aluminum: The determination of aluminum in the presence of iron is difficult if the Mg^{27}/Mn^{56} activity is low because both isotopes emit radiations with the same energy. To be sure of the Fe and Al determination, a very precise decay curve has to be made in each case. When the aluminum concentration, however, is about 100 times less than the iron concentration, the analysis of aluminum becomes difficult. A method is being developed in this laboratory, using computer techniques, to calculate accurately the Al and Fe content in a mixture of both components and this with a minimum number of recounts in a minimum period of time.

c) Measurement of Iron: We have calculated from our experimental results that after 40 minutes waiting time, 16 mg of Fe can still be detected in a simulated moon matrix. A high excess of aluminum does not interfere because the waiting time can be made longer, so that the Mg^{27} decays out.

The determination of iron by the 1.81 Mev gamma peak is feasible but the sensitivity is lower by a factor of $\sim$ 10 than by using the .84 Mev gamma peak.

Mixture IV: Each sample of these mixtures is composed of the five elements including potassium with the following proportions by weight:

15 mg	of Al
7.3 mg	of K
60.6 mg	of Si
20 mg	of Fe
95.6 mg	of O_2

The amount of magnesium oxide added is variable.

a) Measurement of Silicon: The results from the determination of silicon by the Al^{28} activity are given in Table VI.

The reproducibility is within 11% so that one can conclude that the difference in magnesium amounts do not change the analysis of silicon.

b) Measurement of Aluminum: The determination of the Mg^{27} + Mn^{56} activity from .84 Mev energy peak also is not affected by the different amounts of magnesium. Table VII shows this fact for samples counted after 13 minutes waiting time. The first value seems to be too high, otherwise the reproducibility obtained is about 10%.

c) Measurement of Magnesium: Eighteen mg of Mg can be detected in this mixture by the 1.36 Mev gamma-ray peak of Na^{24} after a 40-minute waiting time (see Figure 5).

When 400 mg of Mg is present the Compton effect from the 1.36 Mev peak can interfere in the determination of the aluminum and iron in the mixture. This much amount of Mg, however, could not be added to the mixture because of the limited vial capacity. The study on the analysis of the magnesium on the 2.75 Mev gamma-energy peak could not be done for the same reason.

Conclusion: In a simulated moon matrix it was found that all the five major components; namely, Fe, Mg, Al, Si, and O_2, can be analyzed easily. Only the Mg content will not be very precise because of the rather low Na^{24} activity induced, but no real interference is present. This is true when small samples (200 mg mixtures) are used. Using the moon probe Mg could be easily determined because of the larger size of the semi-infinite samples used in this preliminary measurement.

As long as the five major elements are present in the percentage range and in different ratios as used in these experiments, they can be analyzed for and no important interferences are observed. The only analysis that is sometimes difficult is that of aluminum when the content is too high. The oxygen was not measured because no interference due to the other four elements is possible and the analysis is identical to that which has been done previously.

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Fite, L. E., Steele, E. L. and Wainerdi, R. E., Quarterly Progress Report, February 1, 1963, Activation Analysis Research Laboratory, Texas Engineering Experiment Station, A&M College of Texas, College Station, Texas.

Urey, H. C., Proc. First Intern. Space Sci. Symposium, North Holland Publishing Company, 1114-1122, 1960.

TABLE I

Element	Lowest Abundance (%)	Highest Abundance (%)	Average (%)
O	4	48	26
Mg	.2	22	11
Si	15	35	25
Al	.5	12	6.2
Fe	1.5	15	8.2

TABLE II

	1.78 Mev Photopeak Counts/mg Of Si	Al Added mg	Amount of Mixture Used
	1399	1.6	200 mg
	1277	5.48	200 mg
	1350	10.59	200 mg
	1252	26.6	200 mg
	1381	106.1	50 mg
	1329	105.7	25 mg
Average =	1331		

TABLE III

	.84 Mev Photopeak Counts Corrected For Mg^{27} Activity	Al Added mg
	2277	1.6
	2164	5.48
	2300	26.6
	2343	52.9
	2451	105.7
Average =	2307	

TABLE IV

.84 Mev Photopeak Counts/Minute	Mass Si	Percent Dead Time Start	End
8,706	186.78	60	34
8,163	93.4	44	24
8,792	46.8	28	16
9,546	23.5	22	12
10,459	4.7	16	10
11,069	2.7	9	6

TABLE V

	1.78 Mev Photopeak Counts/Min/mg Si	Mass Iron (mg)	Amount Of Mixture Used
	1492	50.9	200 mg
	1645	199	200 mg
	1619	351	200 mg
	1580	418	100 mg
	1581	350	100 mg
	1602	50	100 mg
Average =	1586		

TABLE VI

	1.78 Mev Photopeak Counts	Mass Mg ∼ mg
	130,252	6.0
	125,072	18.1
	117,924	30
	105,230	60
	110,731	244
Average =	117,841	

TABLE VII

	.84 Mev Photopeak Counts	Mass Of Magnesium ~ mg
	5847	6.05
	5249	18.09
	5015	30.05
	4688	59.8
	4436	244
Average =	5037	

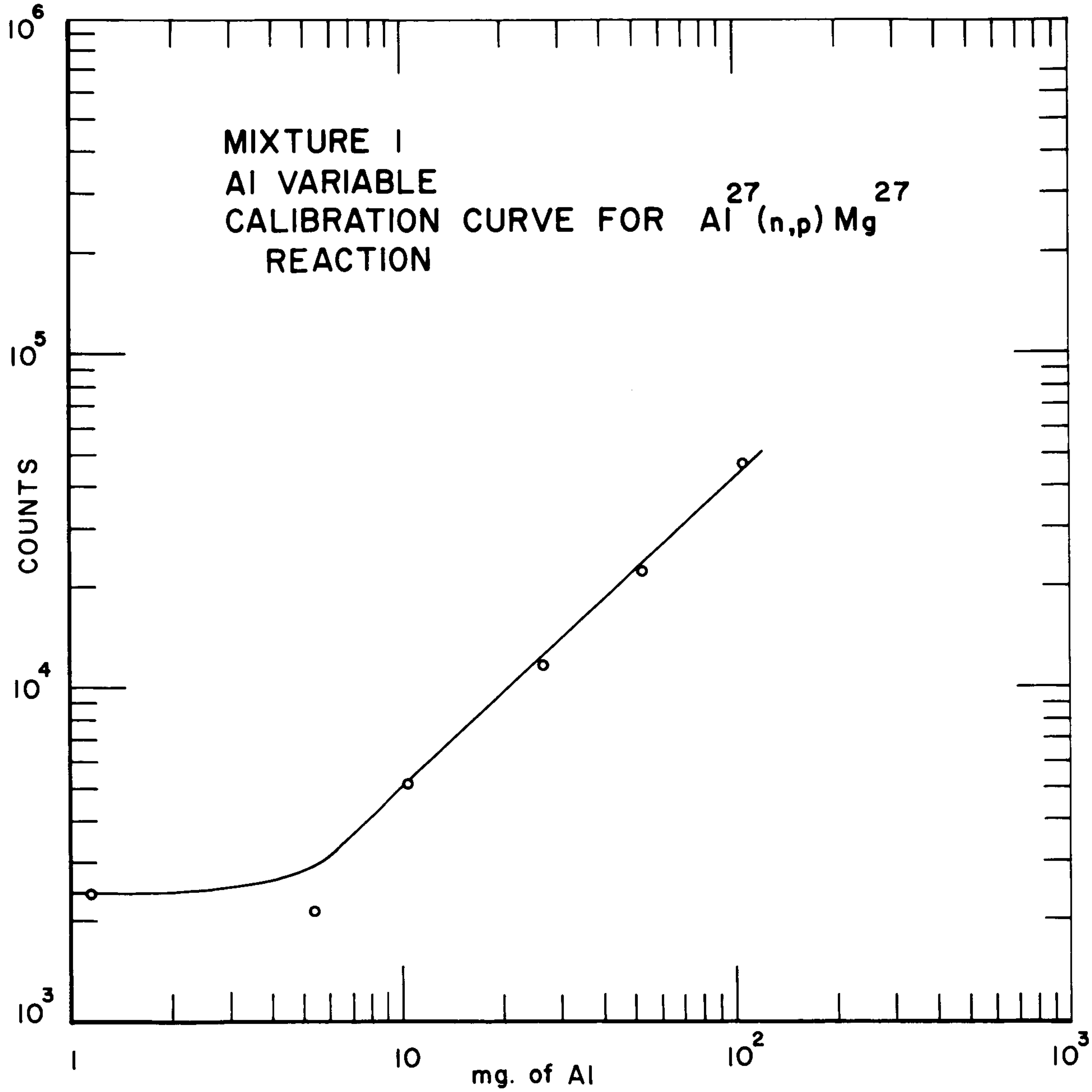

Figure 1. Calibration Curve for the Determination of Aluminum Using Mixture 1

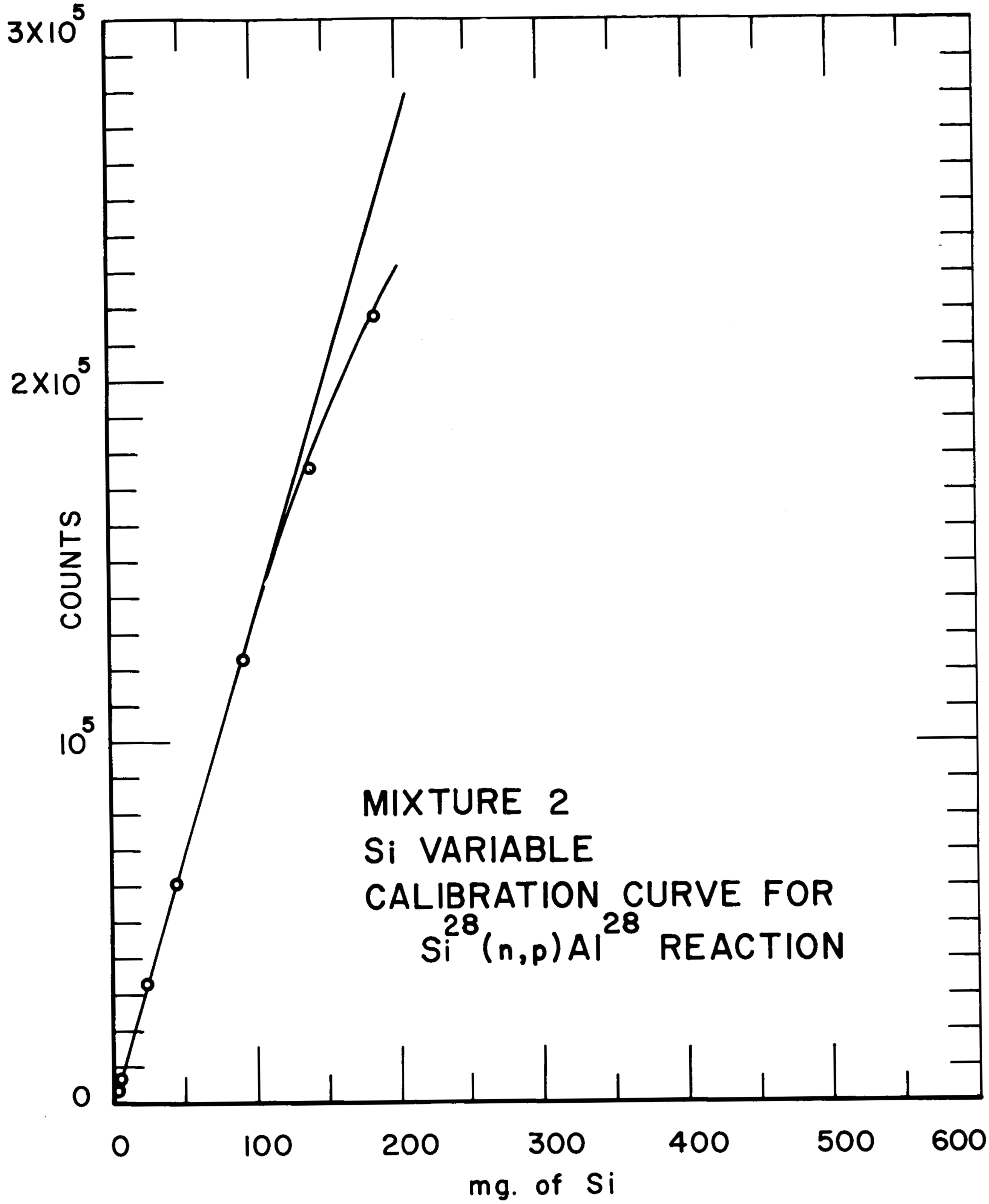

Figure 2. Calibration Curve for the Determination of Silicon Using Mixture 2

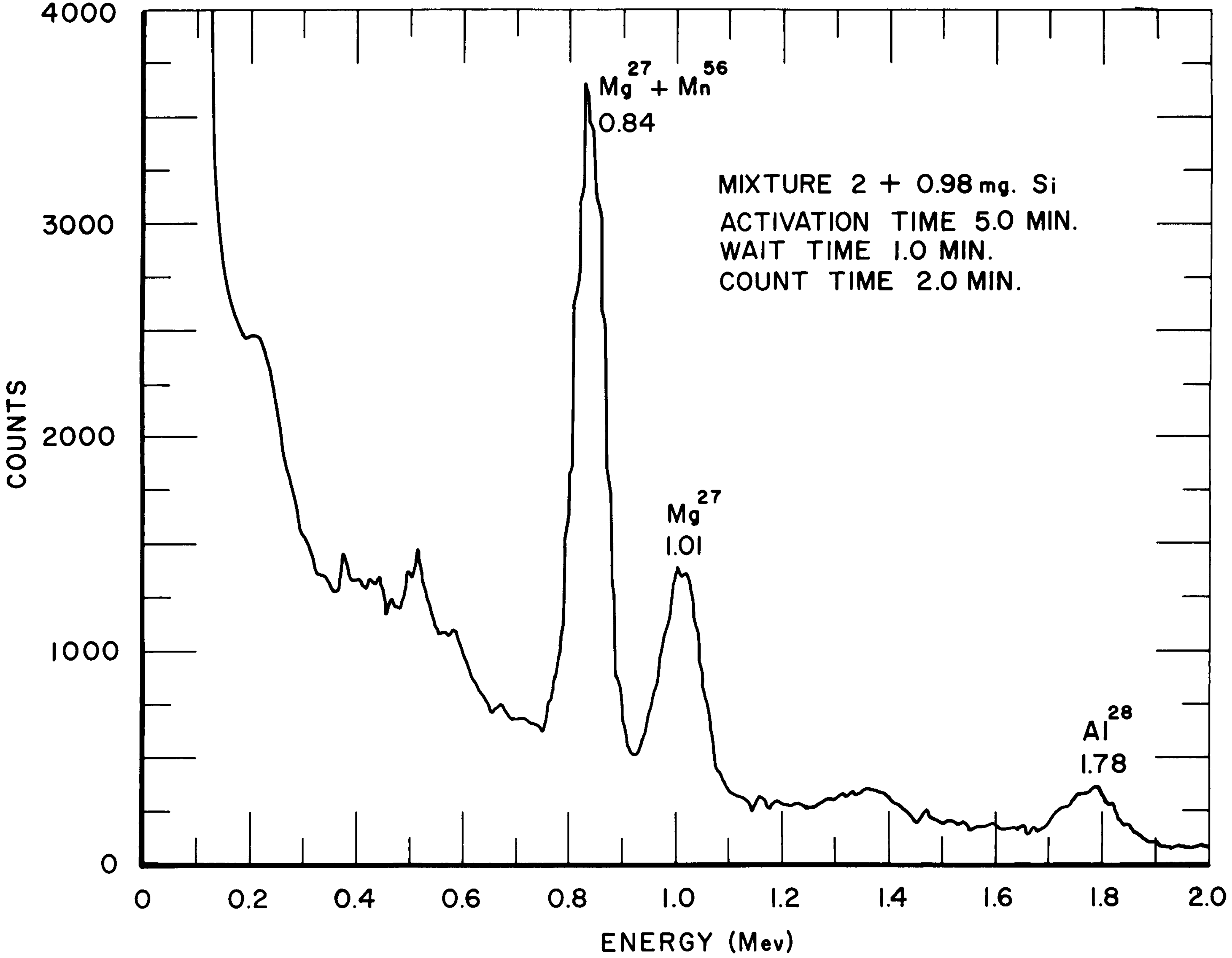

Figure 3. Gamma-ray Spectrum Showing the Lowest Limit of Detection of Silicon

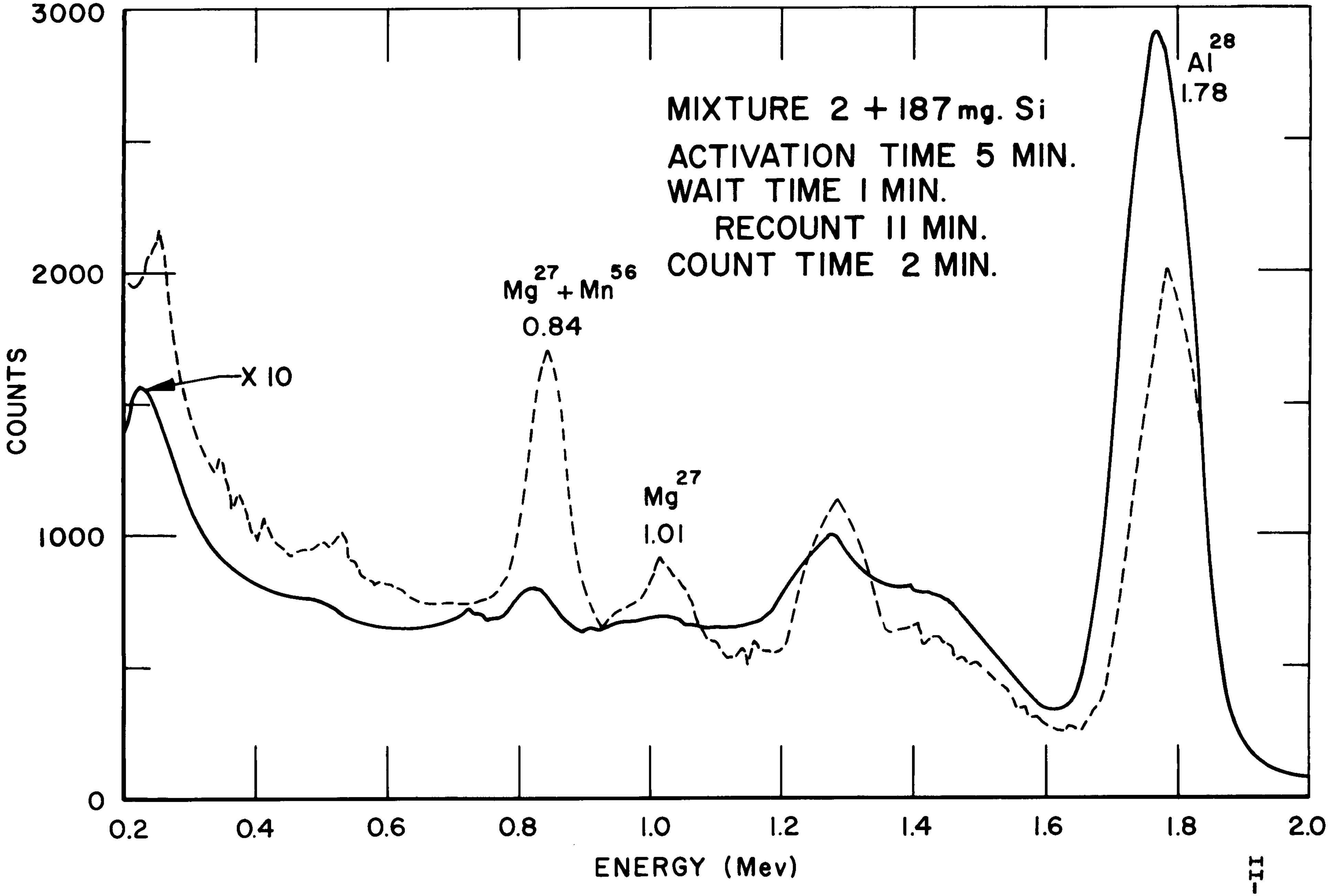

Figure 4: (a)_____,Gamma-ray Spectrum with Hardly Measurable 0.84 Mev Peak

(b)----,Recount Spectrum of the Same Sample

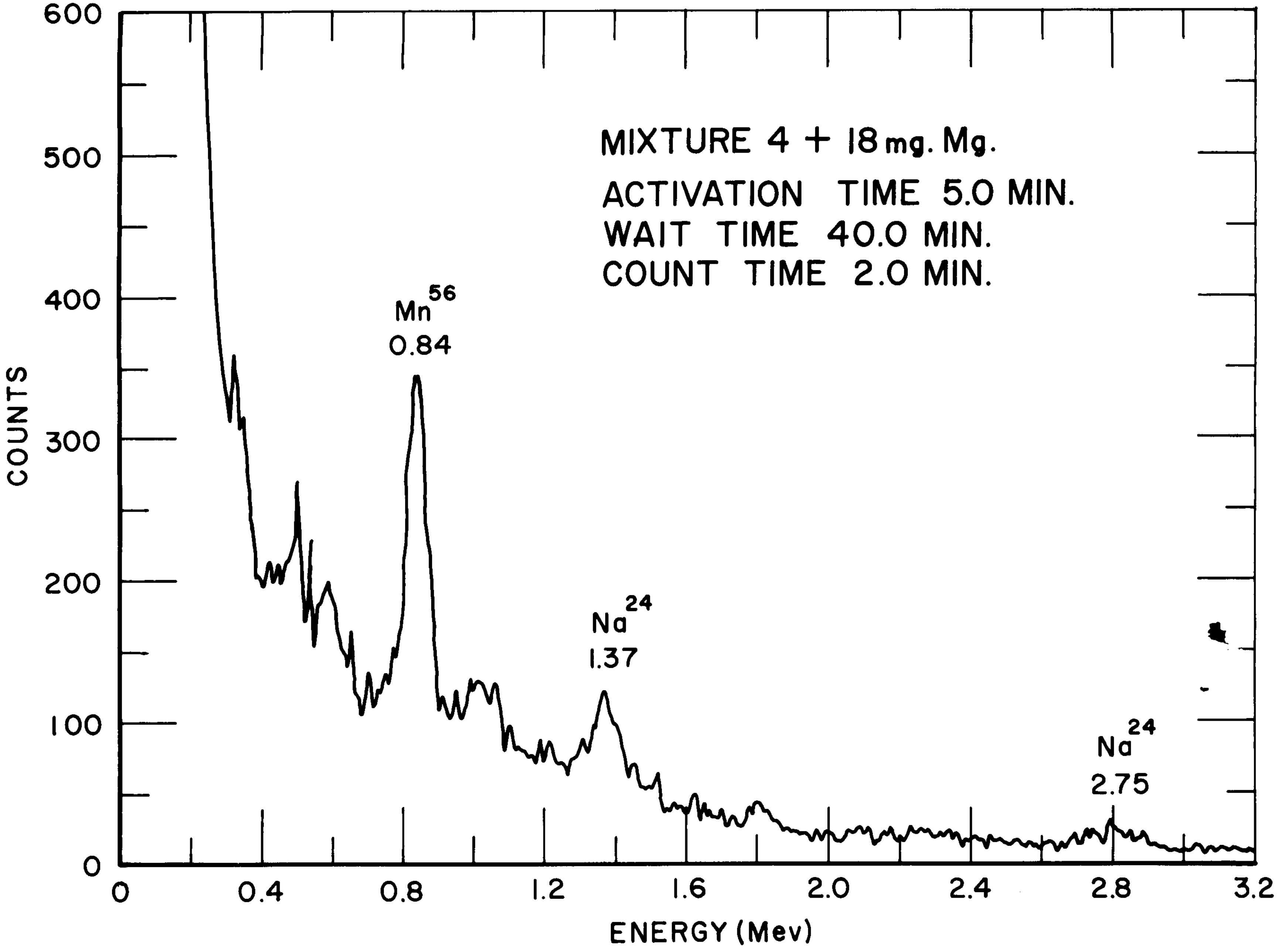

Figure 5. Gamma-ray Spectrum Showing the Lowest Limit of Detection of Magnesium Using Mixture 4

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III. Detector Research - by Carroll F. Lam

Introduction: Further research is being conducted on perfecting the pulse shape discrimination circuitry for use with the dual-crystal scintillation detector for Compton reduction described in the last report (Menon, 1964), and the progress achieved so far is summarized in this report.

This work has been directed to two major areas: (1) optimizing the relatively simple pulse shape discriminator described in the previous report, and (2) investigating other possible methods of pulse shape discrimination. The overall research objective is to provide important Compton reduction advantages for potential extra-terrestrial gamma-ray spectrometry.

Experimental System: Figure 1 shows the dual crystal detector system. A 3" x 3" NaI(Tl) primary detector crystal is surrounded on the side and top by a 5" x 5" CsI(Tl) secondary crystal. A 5" photomultiplier tube is optically coupled to the top of the CsI crystal to detect light pulses emanating from both crystals.

The principle of operation is as follows: When a gamma ray has a photoelectric interaction in the NaI crystal, the resulting fast rise light pulse is detected by the photomultiplier through the CsI crystal; however, when a gamma ray undergoes a Compton interaction in the NaI crystal and the scattered gamma ray, with reduced energy, escapes from the NaI crystal and interacts in the CsI crystal, the shape of the photomultiplier output pulse is altered. The light pulse from the interaction in the CsI crystal will add a slow component to the light pulse produced in the NaI crystal so that the voltage pulse produced by the photomultiplier will have a slower rise and decay time than it would for a NaI photoelectric interaction only.

A block diagram of the electronics system is shown in Figure 2.

The discriminator circuit presently in use is shown in Figure 3. It is similar to the circuit by Rethmeir et al (Rethmeir _et_ _al_, 1961) which was used to discriminate between different types of radiation in a single crystal. A similar circuit was used in a satellite-borne detector system by Kraushaar (Kraushaar, 1962) at MIT. This circuit is supposed to give a negative output pulse only when the fast-rise (NaI) pulses occur.

Results: Figure 4 illustrates typical voltage pulses produced at the output of the photomultiplier preamplifier by equal-energy photoelectric interactions in the NaI and CsI crystals. The upper curve, which is for a NaI photoelectric event reaches its maximum value at about one microsecond, while the lower curve, which is for a CsI photoelectric event of the same energy as the upper curve, reaches its maximum at about two microseconds. It can be seen that the luminescent efficiency of the CsI crystal is about one-third that of the NaI crystal.

Figure 5 illustrates typical results obtained with the presently used discriminator circuit. The sample was a combination of Co^{60} and Sb^{125}. The upper curve is without pulse shape gating and the lower curve is with pulse shape gating. It can be seen that the areas of the 0.427 and 0.6 Mev Sb^{125} peaks have been enhanced. The 0.175 Mev Sb^{125} peak is attenuated but not completely lost. The large peak on the left of the upper curve are the twin photoelectric peaks of Co^{60} in the CsI crystal. It can be seen that two peaks are not resolved by the CsI crystal. This lack of resolution has not been fully investigated although it is strongly suspected that it can be attributed to the unusual shape of the CsI crystal and the multiple reflections that light photons produced in the lower portion of the crystal may undergo before reaching the photomultiplier. However, this lact of resolution does not seem to create any problems in the system since the essential characteristics of the CsI pulse shape are maintained.

Figure 5 also indicates that there is very little, if any, reduction in the Compton continuum at the Compton edge. The amount of Compton reduction, however, seems to increase linearly with decrease in energy below the Compton edge.

The increase in Compton reduction as the energy decreases below the Compton edge can be attributed to two effects. First, at energies corresponding to the Compton edge, the Compton scattered gamma ray is scattered at an angle of about 180^{o} with respect to the incoming gamma ray. With the sample/crystal geometry used in this detector system, gamma rays scattered at these angles will not pass through the CsI crystal.

Secondly, due to the large NaI/CsI energy ratios at the Compton edge and the difference in luminescent efficiencies of the two crsytals, the relative magnitudes of the light pulses produced in the two crystals may be different by a factor of 15. Because of this large difference in pulse heights, the CsI component will have very little effect on the resultant photomultiplier pulse and the present pulse shape discriminator circuit is not sensitive enough to recognize this slow component and reject the pulse.

Conclusions: The detector in conjunction with the presently used discriminatory circuit is capable of reducing the area of the Compton background by about a factor of two. This appears to be about the limit of Compton reduction with the presently used discriminator circuit.

An alternate method of pulse shape discrimination is presently being studied to offer possibilities of considerbly increased Compton reductions. This method was elsewhere used (Bass et al, 1964) to discriminate between different types of particles in a single crystal.

In this method the pulses from the photomultiplier are passed through a double delay line amplifier. This type of amplifier produces a bipolar output pulse. It was found by Bass that the zero cross-over time of the output pulse was a function of the rise time of the input pulse. Since particles of different specific ionization densities produce pulses of different rise time, in a NaI crystal, Bass was able to use the difference in zero cross-over times to discriminate between the different interacting particles.

It is believed that a similar approach can be used to provide better Compton reduction with the dual crystal detector. Figure 6 shows a typical set of double delay line amplifier output pulses from equal energy NaI and CsI interactions. The large pulse is the NaI photoelectric event and the smaller pulse is the CsI photoelectric event. It can be seen that there is about 0.2 microseconds difference in zero cross-over times. The pulses that result from Compton interactions cross-over at times between those of the CsI and NaI photoelectric pulse cross-over times, Bass was able to obtain cross-over time resolutions of less than 10 nanoseconds.

A double delay line amplifier is on order and circuits are being designed that can discriminate the different zero cross-over points.

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Rethmeir, J. et al., Nuclear Instruments and Methods, 13, 153 (1961).

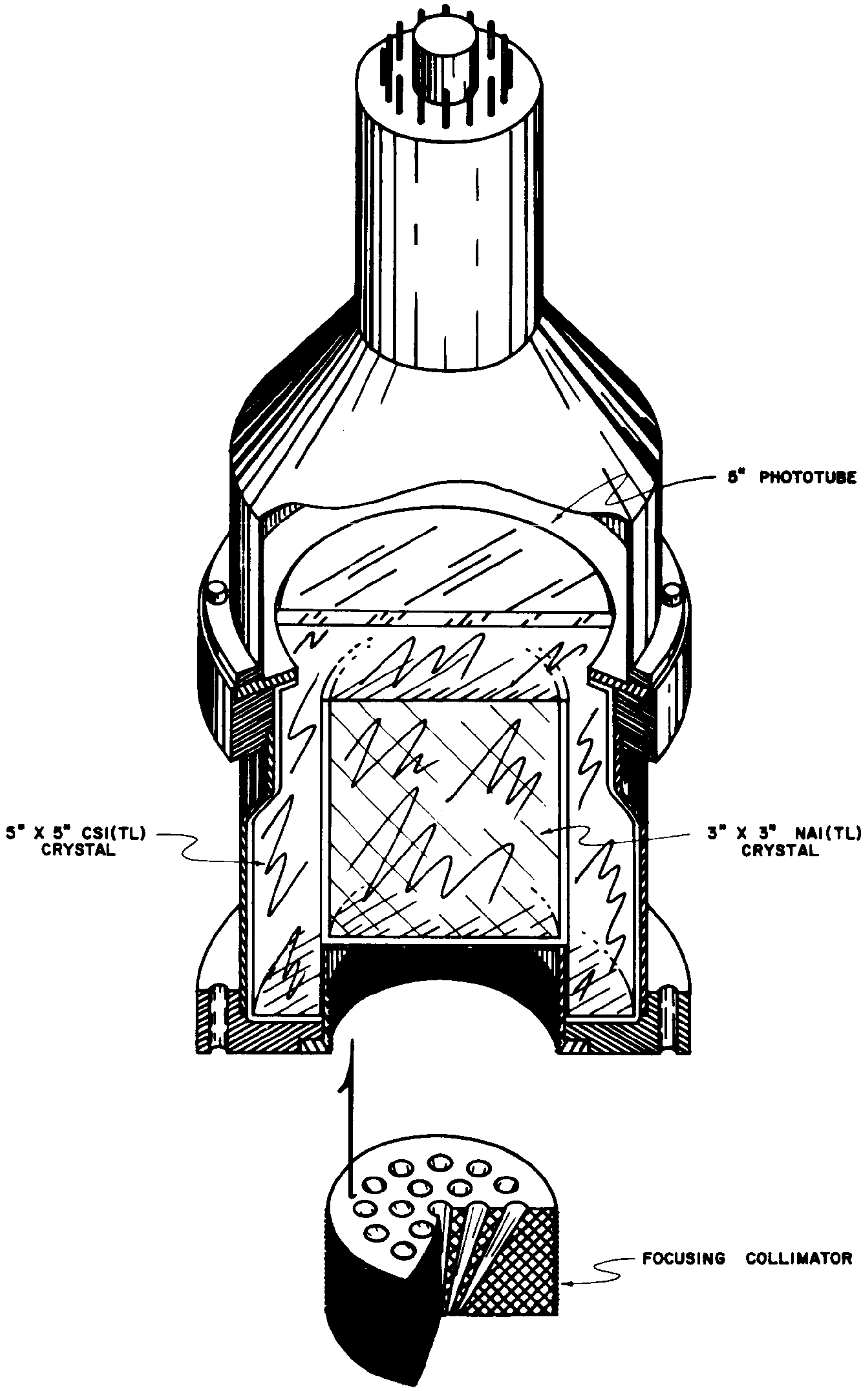

Figure 1. Dual Crystal Detector Assembly

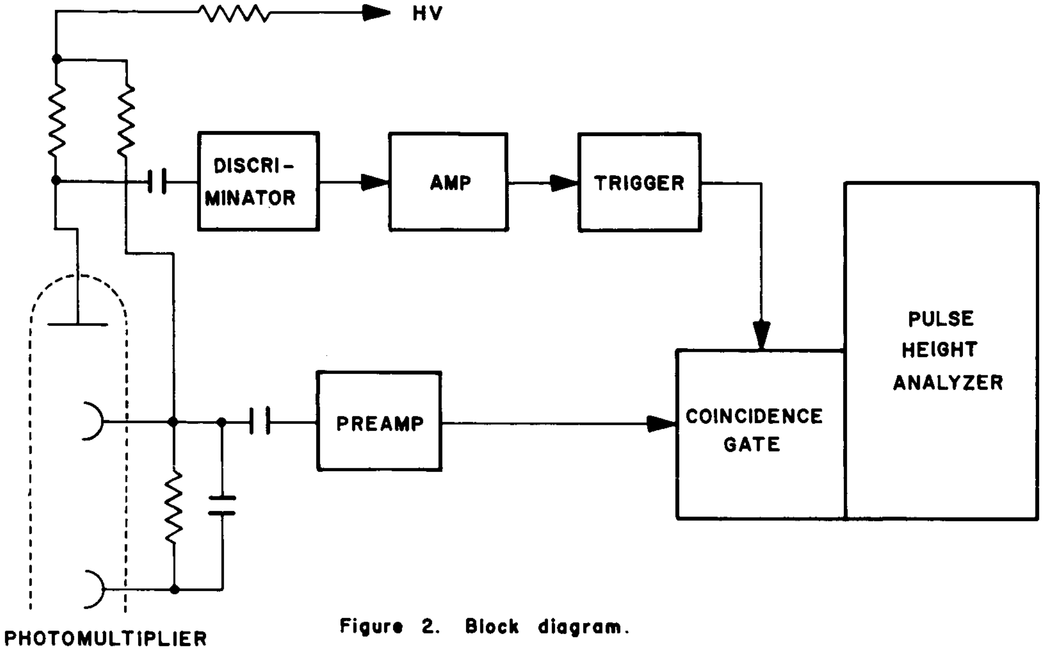

Figure 2. Block diagram.

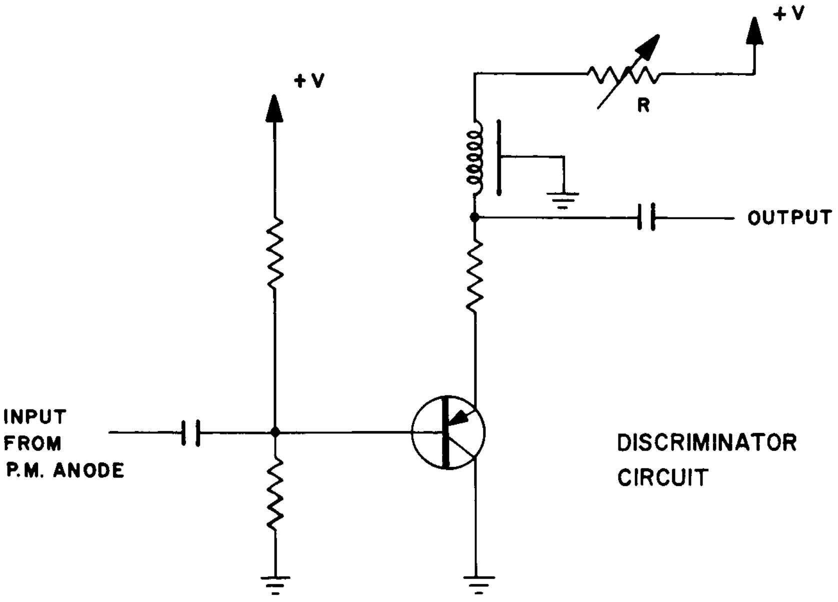

Figure 3. Discriminator circuit.

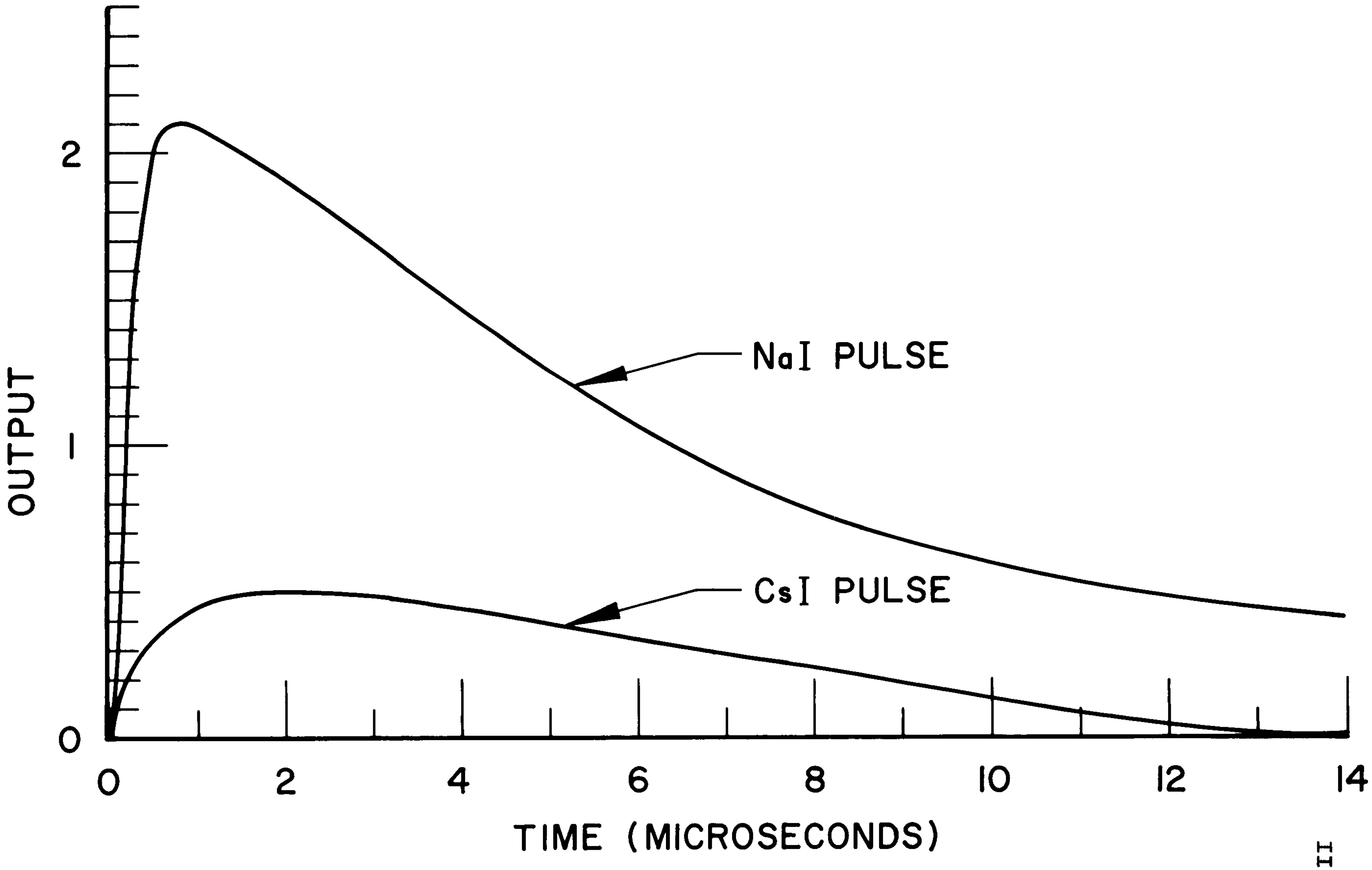

Figure 4. Detector Output Pulse Shapes

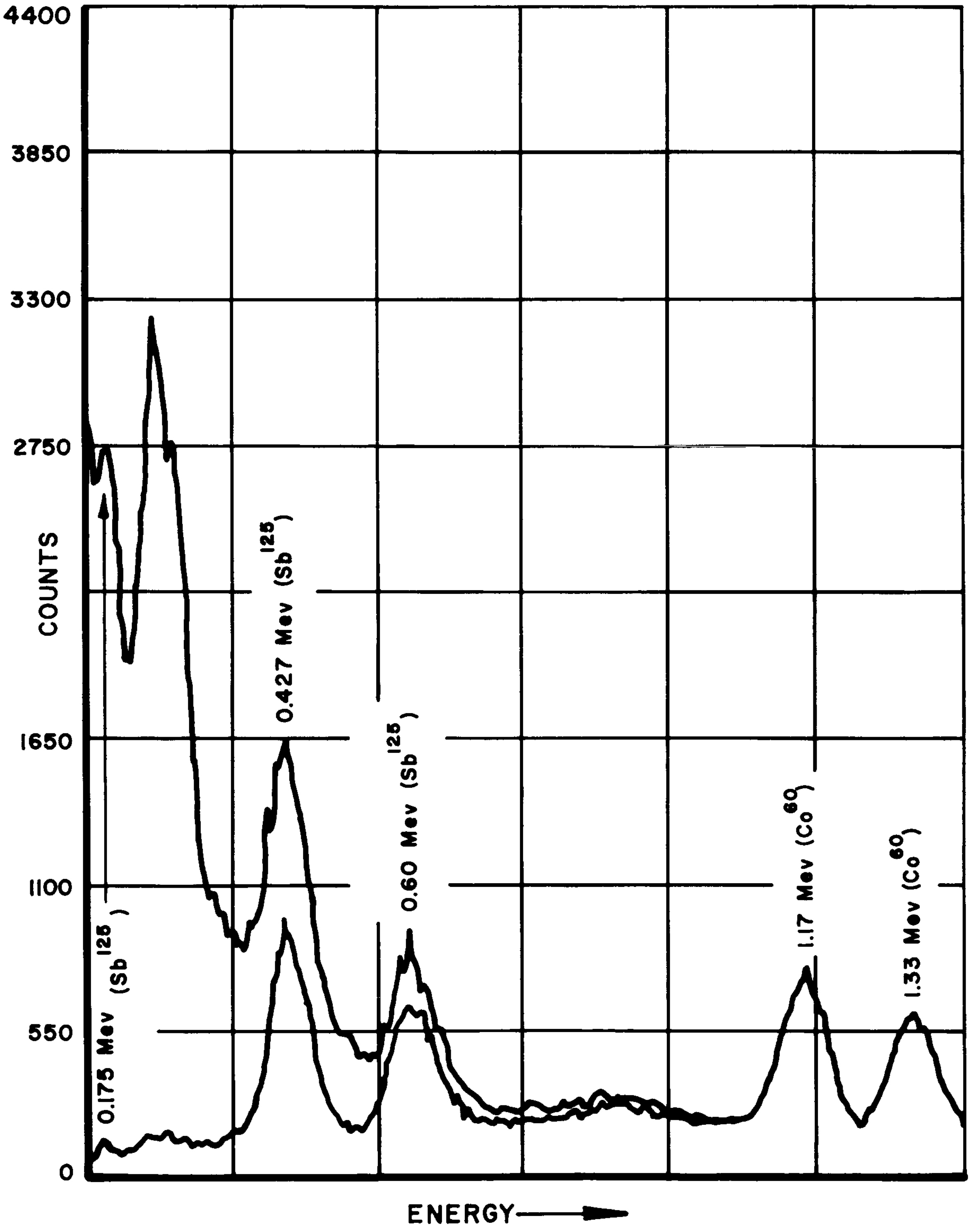

Figure 5. Gated and Ungated Spectra

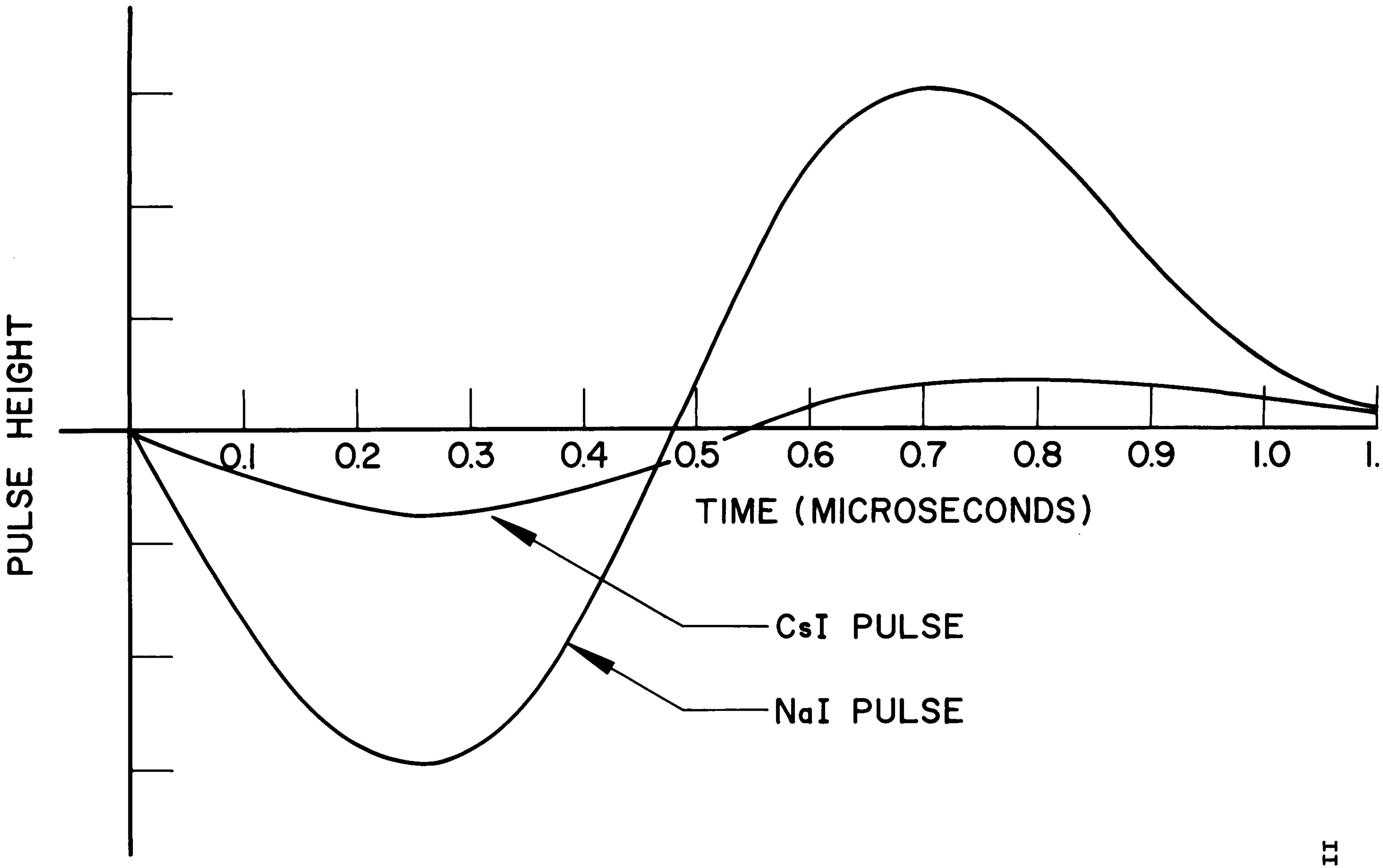

Figure 6. Double Delay Line Amplifier Output

N65-28092

IV. Preliminary Studies On The Possible Use Of Single Channel Analyzers For The 14 Mev Neutron Activation Analysis Of The Major Elements In Rock Samples - Pedro Jimenez and Lloyd E. Fite

Introduction: The utility of multichannel analyzers in the scintillation spectrometry is widely recognized and they are increasingly used in the activation analysis of various materials. However, in some selected cases of analysis, single-channel analyzers may have the advantages of light weight, simplicity, reduced cost, and less dead time to produce activation analysis data on an irradiated sample. An important advantage of a single channel analyzer is its low resolving time which is about two microseconds per pulse. In a commonly used multichannel analyzer, the dead time is governed by the circuitry of the analog to digital converter and is a function of the channel number into which a particular pulse is being stored. If the matrix is such that, on activation, the major isotopes produced have gamma-ray energies and/or half-lives widely different from one another, single channel analyzers may be better substituted for multichannel analyzers. Single channel analyzers have been successfully used by other authors for specific analyses (Wood and Roper, 1965, Van Wyk et al, 1965).

The present application involves the analysis of rock samples for five major elements, O_2, Mg, Al, Si and Fe all of which give radionuclides upon activation with 14 Mev neutrons with different gamma-ray energies and half-lives (See Table I). It was felt desirable to examine the possibility of using single channel analyzers for the above analysis for possible extra-terrestrial applications. This chapter summarizes the progress that has been made toward this end.

Experimental Procedure: Figure 1 shows a block diagram of the system used for this study. The detection assembly consists of two matched 3" X 3" NaI(Tl) scintillation crystals. Output pulses from the crystals are amplified by an amplifier common to both the multichannel analyzer and the single channel analyzers. The multichannel analyzer is calibrated to count gamma rays from 0 to 4 Mev in energy. The three single channel analyzers were calibrated so that the windows are set at .84 Mev, 1.78 Mev and 2.75 Mev, respectively, corresponding to about 8 channels for each peak. Data for a set of similar samples from the multichannel analyzer was used as a check on the relative counts obtained from the single channel analyzers. Thus, it can be determined whether there is appreciable channel drift or background addition to the counts from the single channel analyzers.

In this experiment, the samples were activated for five minutes and counted for two minutes after waiting times of 5, 17, 75 and 133 minutes.

Results and Discussions: Known samples of Mg, Al, Si and Fe have been activated and counted to study this reliability of single channel analyzers for measuring the respective activities produced by 14 Mev neutrons.

To evaluate the reproducibility of the data, five repetitive runs on a series of silicon samples were made. These data are presented in Table II. These data show that analyses performed with single channel analyzers are as precise as those performed with a multichannel analyzer, at least when there is only one major activity present in the irradiated sample.

For a matrix containing all four elements, one has to take into consideration the following equations for the treatment of the data:

$$A_{.84}\ (t_w = 5\ \text{min}) = A_{Mg-27} + A_{Mn-56} + A_{Na-24}\ (\text{Compt.}) + A_{Al-28}\ (\text{Compt.}) \quad \ldots\ldots\ldots\ldots (1)$$

$$A_{.84}\ (t_w = 17\ \text{min}) = A_{Mg-27} + A_{Mn-56} + A_{Na-24}\ (\text{Compt.}) + A_{Al-28}\ (\text{Compt.}) \quad \ldots\ldots\ldots\ldots (2)$$

$A_{.84}$ (t_w = 75 min) = A_{Mg-27} + A_{Mn-56} + A_{Na-24} (Compt.)$\cdot\cdot$ (3)

$A_{.84}$ (t_w = 133 min) = A_{Mn-56} + A_{Na-24} (Compt.)$\cdot$ . . . . . (4)

$A_{1.78}$ (t_w = 5 min) = A_{Al-28} + A_{Mn-56} + A_{Na-24} (Compt.)$\cdot$ (5)

$A_{1.78}$ (t_w = 17 min) = A_{Al-28} + A_{Mn-56} + A_{Na-24} (Compt.)$\cdot$ (6)

$A_{1.78}$ (t_w = 75 min) = A_{Mn-56} + A_{Na-24} (Compt.)$\cdot$ . . . . . (7)

$A_{1.78}$ (t_w = 133 min) A_{Mn-56} + A_{Na-24} (Compt.)$\cdot$ . . . . . . (8)

$A_{2.75}$ (t_w = 5, 17, 75 & 133 min) = $A_{Na-24}$$\cdot$ . . . . (9)

where $A_{.84}$ is the total activity corresponding to the 0.84 Mev peak, $A_{1.78}$ that due to the 1.78 Mev peak and $A_{2.75}$ is the activity due to 2.75 Mev peak. The problem of interference resulting from other nuclear reactions on some of these elements (See Table I) has not been taken into account in the above equations.

Careful measurements of the gamma spectrum of each element will be made to evaluate the extent of the Compton continuum and its reproducibility. After evaluation of all factors involved in this method, a group of standard rock samples will be analyzed and the results compared with multichannel analysis results to ascertain whether or not the single channel analyzers are practical for the analysis of the rock samples.

References:

Van Wyk, J. M. et al, "A Study of Macroscopic Distribution of Oxygen in a Steel Rod by Neutron Activation and Vacuum Fussion Techniques", paper submitted to the Symposium on Radioisotopes Instruments in Industry and Geophysic, Warsaw, Poland, Oct. 18-22, 1965.

Wood, D. E. and Roper, N. J., Kaman Nuclear, Colorado Springs, Colorado, Report KN-65-140(R), April 15, 1965.

TABLE I

Reaction	Reaction Cross-section (Barns)	Product Half-Life	E_γ Mev	Interferences
$O^{16}(n,p)N^{16}$		7.35 sec.		$F^{19}(n,\alpha)N^{16}$
$Si^{28}(n,p)Al^{28}$	.22	2.27 min.	1.78	$Al^{27}(n,\gamma)Al^{28}$ $P^{31}(n,\alpha)Al^{28}$
$Mg^{24}(n,p)Na^{24}$	.19	900 min.	1.37 2.76	$Al^{27}(n,\alpha)Na^{24}$ $Na^{23}(n,\gamma)Na^{24}$
$Al^{27}(n,p)Mg^{27}$	.07	9.45 min.	.84	$Si^{30}(n,\alpha)Mg^{27}$ $Mg^{26}(n,\gamma)Mg^{27}$ $Fe^{56}(n,p)Mn^{56}$
$Fe^{56}(n,p)Mn^{56}$	.110	154.8 min.	.84, 1.81 2.13	$Mn^{55}(n,\gamma)Mn^{56}$

TABLE II

Comparison Of Data From Single And
Multichannel Analyzers For Al^{28} Measurement

Silicon Sample Weight (mg)	Run	Relative Neutron Flux	Single Channel Analyzer (Photopeak Counts)*	% Dev.	Multichannel Analyzer (Photopeak Counts)*	% Dev.
50	1	360,227	11,327		16,774	
	2	365,230	12,449		17,930	
	3	380,612	12,285		17,612	
	4	345,135	12,286		16,558	
	5	362,037	12,556		17,142	
			Avg. 12,180 ± 491	± 4.03	17,203 ± 570	± 3.31
200	1	381,708	50,861		65,293	
	2	393,211	52,933		68,491	
	3	410,977	55,719		68,949	
	4	360,701	53,551		65,113	
	5	361,778	53,209		67,293	
			Avg. 53,255 ± 1742	± 3.27	67,029 ± 1774	± 2.64
362	1	374,123	88,829		108,701	
	2	384,876	80,528		107,809	
	3	359,563	86,661		107,192	
	4	361,653	89,286		108,703	
	5	361,976	73,459		95,937	
			Avg. 83,753 ± 6720	± 8.02	105,669 ± 5475	± 5.18

TABLE II (Cont'd)

Silicon Sample Weight (mg)	Run	Relative Neutron Flux	Single Channel Analyzer (Photopeak Counts)*	% Dev.	Multichannel Analyzer (Photopeak Counts)*	% Dev.
200	1	1,840,349	47,739		61,981	
	2	1,998,600	43,221		59,443	
	3	1,823,622	46,704		60,382	
	4	1,841,179	45,147		57,110	
	5	1,835,784	44,788		56,410	
		Avg.	45,520 ± 1750	± 3.84	59,065 ± 2131	± 3.61

* Corrected for neutron flux

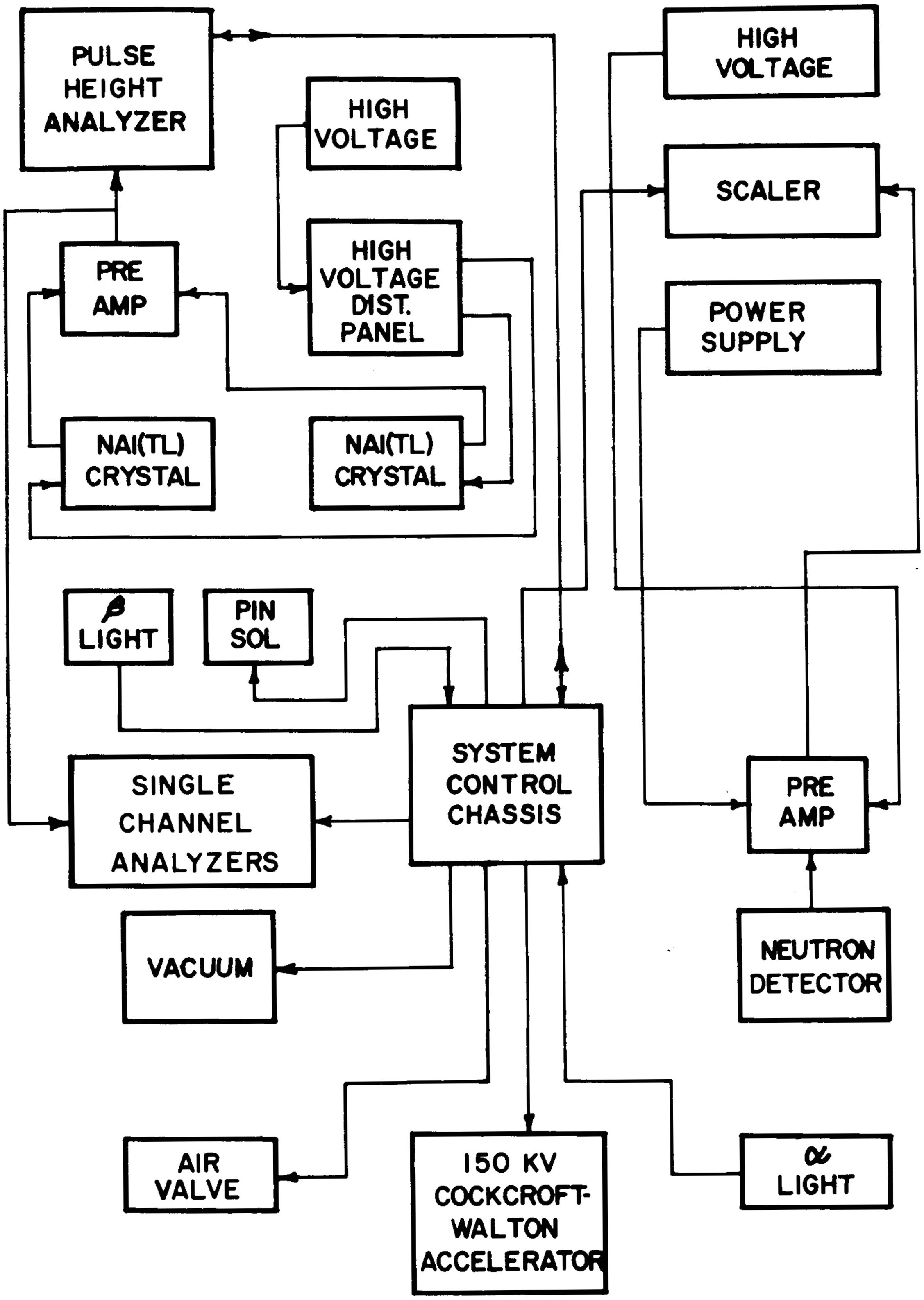

Figure 1. A Block Diagram of System Used for the Analysis of Rock Samples with Single Channel Analyzers

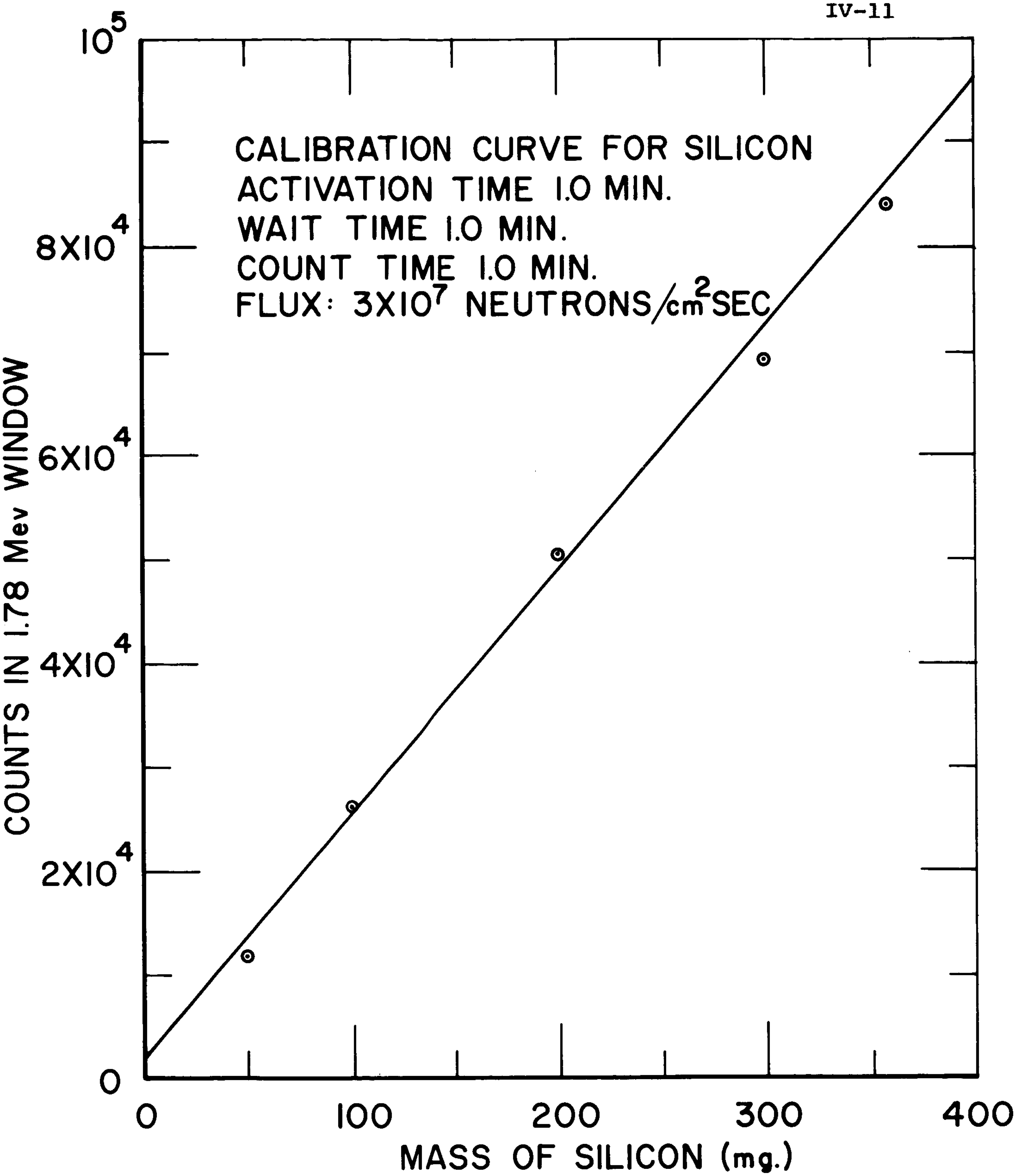

Figure 2. Calibration Curve for Silicon Analysis

N65 28093

V. Instrumental Activation Analysis of Meteorites, Siderolite and Tektite Using 14 Mev Neutrons - by M. P. Menon, Ph.D.

Introduction: There is a vast amount of data available in literature concerning the elemental composition of stone meteorites which are believed to represent the original solar matter (Urey and Craig, 1953; Ahrens et al, 1960). Chondritic meteorites were selected to measure the cosmic abundance of the elements because they were found to be reasonably uniform in their composition at least with respect to the lithophile elements, such as, Na, K, Si, Al, Mg and so on, and any deviation from this uniformity observed in a chondrite is indicative of some sort of fractionation. Most of the measurements on the abundance of major elements in meteorites have, however, been made using the so-called classical gravimetric procedures (Ahrens, Loc. cit.) and the coefficient of variance among these results seems to be larger than the usual systematic error. It has also been reported that, based on the available data, the lithophile elements, Si and Mg, are significally fractionated in chondritic meteorites (Ahrens, 1964) and there is an inverse relationship between the concentrations of the two most abundant constituents, Mg and Fe, in chondrites. Information regarding the abundance of some of these major elements and their ratios will help in learning more about the history and probably the origin of these meteorites. Apart from the fact that 14 Mev neutron activation analysis of meteorites may give more reliable data on the cosmic abundances of some of these elements, it will also help to optimize the conditions for the analysis of extra-terrestrial samples which may be similar in composition to the lunar surface material (Urey, 1959). Another motivation behind the meteoritic study is the expectation that some recovered lunar-surface material may be available in the future for a systematic laboratory analysis. It seems desirable to use only instrumental techniques for the analysis of such a "precious' sample so as to make it available to other laboratories for interlaboratory comparison of the final results (Fisher and Curni, 1964). Reactor neutrons, with a high flux, may have to be employed to irradiate the sample for the determination of such trace elements like Sc, Mn, Co and others. The meteorite, a representative of extra-terrestrial samples, may serve as a good specimen on which analytical techniques can be perfected. Not many analyses have been done on siderolite and tektites using neutron activation techniques; therfore, it was felt worthwhile to attempt to measure the concentration of the major elements using similar techniques.

Only preliminary results of the silicon measurement are included in this report. A method for measuring the individual activities of Mn^{56} produced by the $Fe^{56}(n,p)Mn^{56}$ reaction and Mg^{27} produced by the $Al^{27}(n,p)Mg^{27}$ reaction both of which have identical gamma-ray peaks is described below.

Experimental Procedures: The experimental set-up that was used for the analysis was the same as was described in the previous report (Menon, 1964). The sample sizes used for the silicon measurement were all 500 mg. All these samples were activated for 1 minute and counted for the same period after a delay of 1 minute and 3.3 minutes, respectively. A typical time dependent gamma-spectrum of one of the samples is shown in Figure 1. The photopeak activity was computed by Covell's method (Covell, 1959), corrected for the neutron dose and compared with the activity of the silicon standard to measure the silicon content.

Two or three sets of samples with weights ranging from 1 g to 2.5 g were activated with 14 Mev neutrons for five minutes to determine the concentration of Al and Fe in them. Each of these samples was counted five times with different waiting intervals in-between so that the components, Mg^{27} and Mn^{56}, of the total 0.84 Mev photopeak activity may be resolved.

Typical time dependent gamma-spectra of a chondrite and a siderolite sample resulting from the first and last count are shown as Figure 2 and 3, respectively.

Results and Discussions: The results of the silicon analyses are shown in Table I. It is hoped that the precision of the analysis can be improved by repeating some of these measurements taking counts at different waiting times and extrapolating the activity to zero waiting time.

The method used in the past for measuring the Mg^{27} and Mn^{56} activities individually was to count the activated sample twice, one at a waiting time of about 10 minutes and the other at a waiting time of about 130 minutes. The 0.84 Mev photopeak activity obtained from the second count (which is mostly due to Mn^{56}) is extrapolated back to the first waiting time and subtracted from the activity from the first count to get the Mg^{27} activity. The results from such measurements are based on one single count for each element which is not as precise as possible. An alternate method to save time for the analysis of a sample and probably to get better precision in the measurement is to count the activated sample at least 5 times within 30-40 minutes after irradiation and determine the individual activities from a Biller plot (Biller, 1953). If A_T is the total activity due to Mn^{56} and Mg^{27} at any time after the end of irradiation,

$$A_T = A_1^o \cdot e^{-\lambda_1 t} + A_2^o \cdot e^{-\lambda_2 t} \quad \ldots\ldots\ldots\ldots\ldots \quad (1)$$

where A_1^o and A_2^o are the activities due to Mn^{56} and Mg^{27} at the end of irradiation and λ_1 and λ_2 are the respective decay constants.

This equation may be rearranged as follows:

$$A_T \cdot e^{\lambda_1 t} = A_1^o + A_2^o \cdot e^{-(\lambda_2 - \lambda_1)t} \quad \ldots\ldots\ldots\ldots\ldots \quad (2)$$

The plot of $A_T \cdot e^{\lambda_1 t}$ against $e^{-(\lambda_2 - \lambda_1)t}$ in a linear graph paper will give a straight line the slope of which gives the value for A_2^o and the intercept with the ordinate gives that for A_1^o. Figure 4 shows two such Biller plots used to measure the Mg^{27} and Mn^{56} activities in a chondrite and a siderolite sample. This has both aluminum and iron present in it while the siderolite contains no appreciable amount of aluminum.

The precise measurement of the aluminum and iron content of the above samples will be made in the future using Biller plots.

References:

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Urey, H. C. and Craig, H., "The Composition of Stone Meteorites and the Origin of Meteorites", Geochimi et, Cosmochim Acta 4, 36 (1953).

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TABLE I

Preliminary Results From 14 Mev Neutron Activation Analysis Of Meteorites, Siderolite and Tektite For Silicon

Sample	Wt of Sample Used	Silicon Content (mg/g)	Mean Value (mg/g) With Standard Deviation
Chondrite Nebraska	500 mg	185.0	223 ± 28
	500 mg	231.0	
	500 mg	224.0	
	500 mg	253.0	
Chondrite Kansas	500 mg	171.8	
	500 mg	212.0	191.0 ± 20
	500 mg	189.0	
Siderolite Kansas	500 mg	157.0	
	500 mg	154.0	151 ± 12
	500 mg	160.8	
	500 mg	133.4	
Tektite Bohemia	500 mg	430.0	
	500 mg	374.0	419 ± 43
	500 mg	414.0	
	500 mg	456.0	

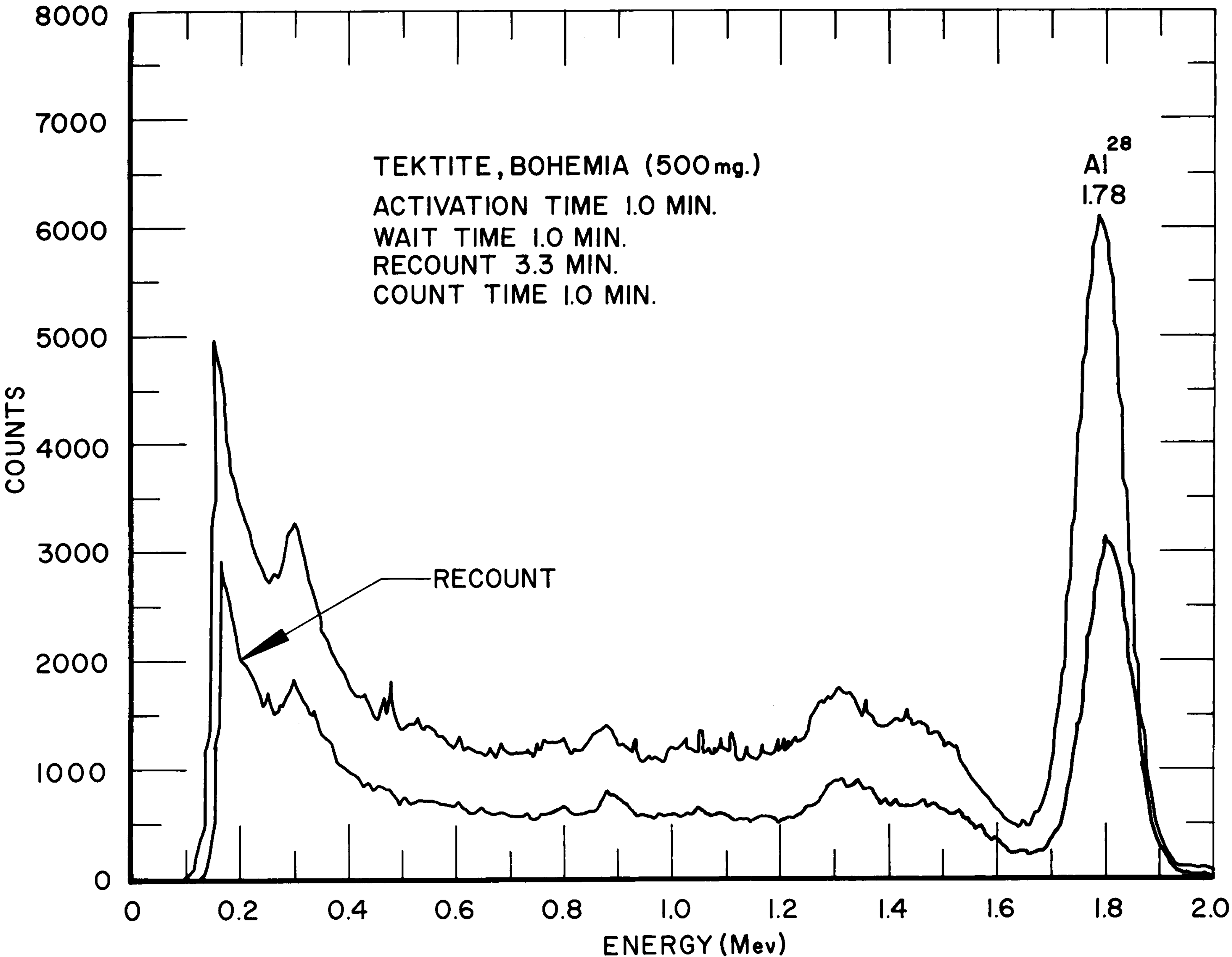

Figure 1. Typical Time-dependent Spectra of an Activated Tektite Sample

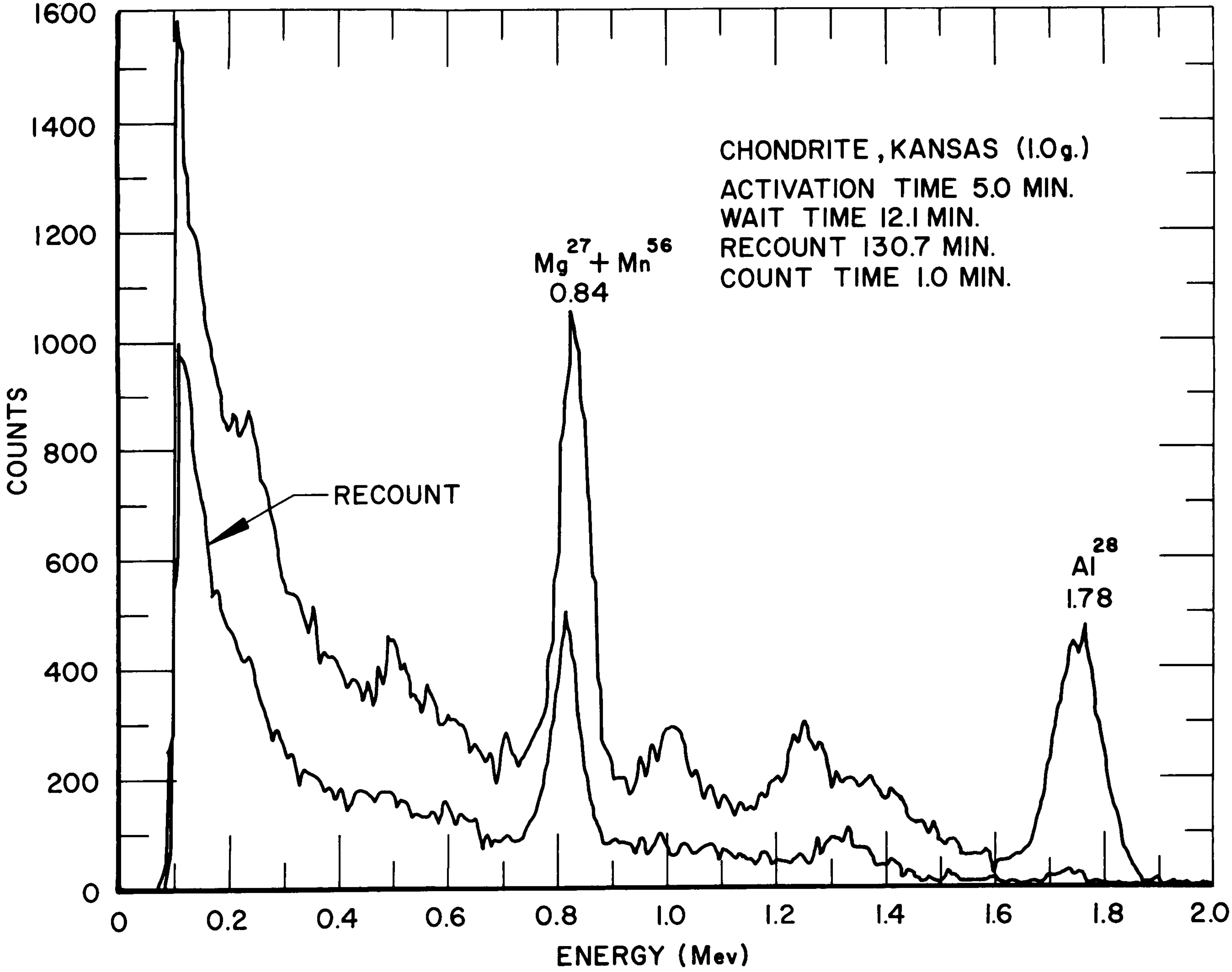

Figure 2. Typical Time-dependent Spectra of an Activated Chondrite Sample

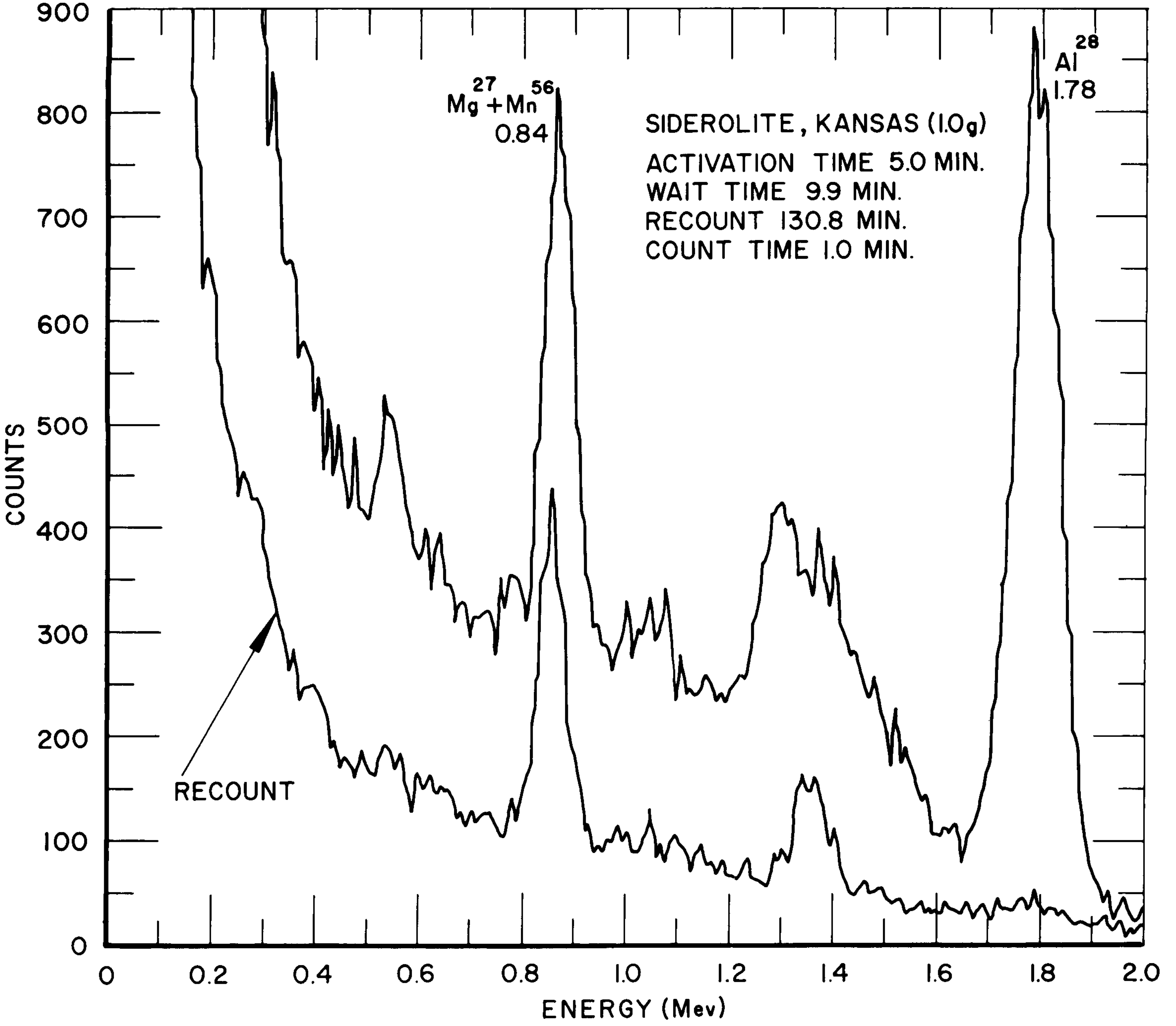

Figure 3. Typical Time-dependent Spectra of an Activated Siderolite Sample

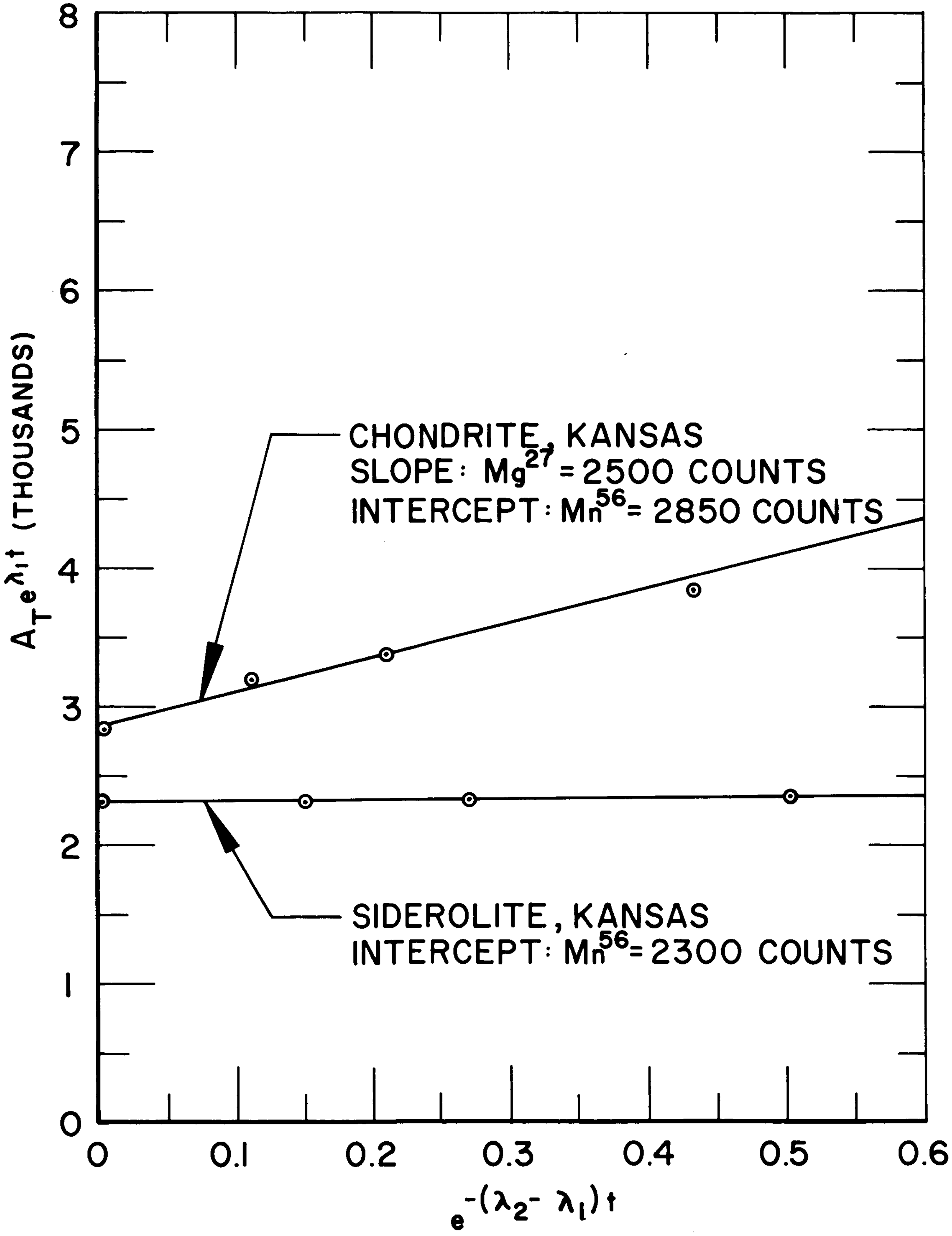

Figure 4. Measurement of Mn^{56} and Mg^{27} Activities by a Biller Plot

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