DETERMINATION OF SODIUM IN SILICATE MINERALS

BY NEUTRON ACTIVATION ANALYSIS

by

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S.B., Massachusetts Institute of Technology

(1959)

SUBMITTED IN PARTIAL FULFILLMENT

OF THE REQUIREMENTS FOR THE

DEGREE OF MASTER OF

SCIENCE

at the

MASSACHUSETTS INSTITUTE OF

TECHNOLOGY

February, 1961

Signature of Author . . . .
    Department of Geology and Geophysics, December   , 1960

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ABSTRACT

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BY NEUTRON ACTIVATION ANALYSIS

By Gerald Lawrence Schroeder

Submitted to the Department of Geology and
Geophysics on December   , 1960 in partial fulfillment
of the requirements for the degree of Master of Science.

A procedure for the neutron activation analysis
of sodium in silicate minerals is described. Counting of
the 2.754 m.e.v. γ of Na^{24} with a $NaI(Tl)$ crystal and an
integral discriminator provides a method in which the
precision is 2% and the interference by trace elements
less than 1%. Na values obtained for the standard granite
G-1 are slightly lower than the mean of previous chemical
analyses, while values for the standard diabase W-1 are
slightly higher.

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I

INTRODUCTION

The analysis for Na in silicate minerals presents a problem to the investigator. The chemistry of dissolving the original mineral and ultimately separating it into its constituent compounds is complicated (1). Each step introduces a new possibility of contamination, or loss during precipitation. In the conventional procedure of rock analysis, the total alkalies are removed in a group filtrate and Na and K are precipitated from this filtrate as chlorides. The K is then determined and the Na value calculated from the difference. The value determined for sodium, as Na_2O, will be in error if either the total alkalies value, or the K_2O value is invalid (2). An independent method of analysis is highly desirable.

A study of the precision of chemical analysis of silicate rocks by Fairbairn (3) has revealed the poor reproducibility achieved by conventional techniques. In the analysis of a standard granite, G-1, and of a standard diabase, W-1, by 35 laboratories, the standard deviation for sodium was 5.2% of the determined value in G-1 and 9.3% in W-1. Chayes (4) stated well the seriousness of this lack of reproducibility: "Normative variation is so

extreme that in the absence of direct evidence to that effect there would be little reason to suppose all these analyses were of the same two rocks." Although the potassium determination depends primarily on the complete extraction of the alkalies, and is essentially independent of other constituents, the standard deviation of determined potassium in the G-1 analyses was 6.8% and in W-1 23%! In viewing this considerable range, one must remember the dependence which the sodium value has on the determined potassium. Winchester (5) successfully employed neutron activation analysis as a highly precise method of determining the potassium composition of silicate and other minerals. The analysis required no chemical treatment before irradiation. In adopting this tool as a means of determining sodium composition of silicate minerals, some difficulty was encountered in adopting the correct electrical circuit to handle the high flux of low energy γ 's. The final experiment, however, yielded encouraging results.

For neutron activation analysis to be useful for the analysis of a particular element in a mineral, the element under investigation must possess a radionuclide producible by irradiation with thermal neutrons, which has a form of activity unique among all other radionuclides produced during the irradiation.

The primary constituents of W-1 and G-1 are Si, Al, Fe, Mg, Ca, Na, K, and O (6). A study of the GE chart of the nuclides (7) shows that a short irradiation period (several minutes or less) followed by a "cooling off" period of 30 hours will leave activity which is essentially all 15 hour Na^{24} and 12.5 hour K^{42}. Na^{24} decays by emitting 1.394 m.e.v. β, 1.368 m.e.v. γ and 2.754 m.e.v. γ for 100% of the decays (8). K^{42} decays by emitting 3.55 m.e.v. β in 82% of the decays, and 1.99 m.e.v. β and 1.53 m.e.v. γ in 18% of the decays (8).

Using the proper electronic circuitry, it is possible to count only gammas with energies greater than 2.75 m.e.v. This ensures recording solely the activity of Na^{24}. The apparatus employed consists of a thallium activated, NaI scintillation crystal monitored by a photo-multiplier tube. The output of the tube is amplified and then sent through an electronic integral discriminator which is biased so as to pass only those pulses corresponding to γ's with energies of 2.75 m.e.v. or higher. From the dis-criminator the signal is fed to a scaler on which the decay rate is recorded.

The experimental procedure is as follows: 10 mg Na_2CO_3 standards, and 50 mg. mineral samples (both finely ground) are weighed and heat sealed in polyethylene packets.

Standards and samples in their individual packets are ir-
radiated as a unit for 10 seconds in the nuclear reactor
and "cooled" for 30 hours. The activity of each is then
measured with the NaI crystal and the counting rate per
unit weight of standard is compared with that of the
mineral. The per cent Na is computed. At the high bias
of the discriminator the background activity is low.

II

PROCEDURE FOR ANALYSIS

1) Grind the mineral sample and the Na_2CO_3 standard (note 1) to a fine powder in an agate mortar. Dry at 110° C (note 2).

2) Weigh 10 mg. portions of standard and 50 mg. portions of sample. Place each portion on a cleaned 0.001" polyethylene sheet (note 3) and heat seal a second polyethylene sheet over the sample (note 4). Prepare several blank polyethylene packets (note 5).

3) Number each packet (note 6).

4) Place samples and standards alternately in a sealed polyethylene container (note 7). Irradiate as a unit at a thermal nuetron flux $(n\nu)$ 5 x 10^{12} n/cm^2- sec for 10 seconds.

5) Allow specimens to cool for 30 hours (note 8).

6) Mount the packets on aluminum planchets (note 9).

7) Measure the 2.75 m.e.v. γ activity (notes 10 and 11).

8)   Compute the per cent Na in the sample (note 12).

Notes

1)   Runs number 1, 2, and 4 utilized Na_2CO_3 as a standard. Because there was the possibility that the Na_2CO_3 was adsorbing significant water vapor, NaF was used as the standard for Run #3.

2)   Drying temperature should be no higher than 110° C; at elevated temperatures Na_2CO_3 decomposes with the evolution of CO_2. This will upset the stoichiometry of the compound.

3)   The polyethylene sheet may be cleaned by wiping with a dry cotton cloth to remove any solids which are contaminating the surface.

4)   To carry out the sealing operation place the specimen in the center of a polyethylene square. Lay a second square over the sample and transfer the unit to the horizontal face of a heat sealing iron. With a bottle cap of 3/8" diameter press the top sheet firmly down onto the undersheet so as to enclose the specimen. Hold for several

seconds and then remove the newly formed plastic packet
from the iron. Check the seal carefully for leaks. It
is difficult to effect a satisfactory seal if the speci-
men powder has previously been scattered on to the part
of the plastic which is to be sealed. The operator should
wear gloves and handle the packet with tongs in order to
avoid contamination. The sodium content of the body is
high.

5) A blank packet consists of two clean polyethylene
sheets of a size similar to those used on the actual
specimens heat sealed together.

6) A convenient method for identifying the packets is
to write the packet number on a piece of paper and then
tape this paper to the edge of one of the polyethylene
sheets. Before counting remove the tape and paper since
the tape, and ink, may have become radioactive during the
irradiation.

7) By placing the samples and standards alternately one
is ensured that both are subjected to the same neutron
flux.

8) This period allows the short-lived radionuclides to

to decay to very low levels of activity, while the 15 hour
Na^{24} passes through only two half-lives and has 25% of its
activity remaining.

9)   After cutting off the tape and number tag, attach the
packet to the flat side of a planchet using fresh tape.
Taping, and handling, the packet after irradiation does
not contaminate the specimen since neither the tape, nor
one's body, contains any appreciable radioactivity.
Record the sample number on the reverse side of the plan-
chet.

10)   The thallium activated NaI crystal used as the scin-
tillating material provides a high light pulse per unit
energy of impinging particle, and has a low (2×10^{-7}
seconds) resolving time (9).

11)   It is possible to count the specimens manually or
automatically. To count manually, each specimen is
individually placed on a rack at a fixed distance from
the NaI crystal. The radiation is counted for a suffi-
ciently long time so that the statistical variation of
the count is less than 2%. To count automatically, the
samples and standards are placed alternately in the maga-
zine of a Baird-Atomic automatic sample changer which feeds

and removes the specimens after a predetermined number of counts is reached. The time taken to reach this number is printed on a tape next to the specimen number. In both manual and automatic counting, the activity of background and blank polyethylene packets is measured and this activity is subtracted from the gross number of counts to give the net γ activity.

12) After determining the net counts per mg. for the sample and standard, and knowing that Na_2CO_3 is 43.4% Na, it is possible to compute the per cent Na in the sample by the following:

$$\frac{\text{net counts/mg sample}}{\text{net counts/mg standard}} \times 43.4\% \ .$$

III

ANALYTICAL RESULTS

Four separate groups of analyses were made during
the course of this investigation. Runs 1 through 3 were
carried out using a differential discriminator as the
means of counting only 2.75 m.e.v. γ's. This apparatus
allows the selection of a maximum and minimum γ pulse
height. All γ's with energies which lie within these
limits will be counted. For run #4 an integral discrimin-
ator was used. With this, a minimum base line setting
is chosen and all γ's with energies greater than this
base line are counted.

Tables 1-3 give a summary of the data obtained
for the first three runs. Specimens for run #1 were ir-
radiated by thermal neutrons for one minute (neutron
flux 5 x 10^{12} n/cm^2-sec.) and were "cooled" for 24 hours
before counting, while specimens for runs #2 and 3 were
irradiated for 6 minutes and had cooling periods of 35 and
32 hours, respectively.

In column 4 of tables 1-3 is tabulated the
corrected net counting rate of all observed γ's having
energies between 2.675 m.e.v. and 2.925 m.e.v., the mini-

Counting Data for Run #1

SA No.	Type	SA Wt. (mg.).	Corrected Net c/s	Corrected Net c/s-mg	% Na
1	Na_2CO_3	12.9	265	20.6	
2	"	11.0	244	22.2	
3	"	11.1	259	23.4	
Average				22.1 ± .8	
4	G-1	106.4	142	1.33	2.62
5	"	103.1	138	1.34	2.63
6	"	105.2	140	1.33	2.62
Average					2.62 ± .01
Trace element correction					.02
Net					2.60 ± .01
7	W-1	101.6	88	0.866	1.45
8	"	103.2	90.5	0.876	1.46
9	"	108.0	88.5	0.819	1.37
Average					1.43 ± .03
Trace element correction					0.01
Net					1.42 ± .03

TABLE 1

Counting Data for Run #2

SA No.	Type	Wt. (mg.)	Corrected net c/s	Corrected net c/s-mg.	% Na
1	Na_2CO_3	10.07	1060	105.2	
2	"	8.90	918	103.2	
3	"	10.32	1150	111.3	
4	"	10.51	1170	111.2	
Average				107.8 ± 2.6	
5	G-1	29.23	172	5.88	2.37
6	"	30.71	193	6.28	2.53
7	"	31.03	182	5.87	2.37
8	"	29.50	180	6.11	2.46
9	"	29.59	184	6.22	2.51
Average					2.45 ± .03
Tr. El. Cor.					.02
Net					2.43 ± .03
10	W-1	29.85	117	4.26	1.72
11	"	29.81	122	4.08	1.65
12	"	29.52	123	4.17	1.68
13	"	30.03	128	4.26	1.72
Average					1.69 ± .02
Tr. El. Cor.					.01
Net					1.68 ± .02

TABLE 2

Counting Data for Run #3

SA No.	Type	Sa Wt (mg)	Corrected net c/s	Corrected net c/s-mg	% Na
1	NaF	10.70	2270	212	
2		10.15	2160	213	
3		10.10	2150	213	
4		11.40	2530	222	
Average				215 ± 2.3	
5	G-1	31.31	244	7.78	1.98
6		30.70	236	7.66	1.95
7		31.17	231	7.40	1.88
9		29.82	222	7.42	1.88
Average					1.92 ± .02
Tr. El. Cor.					.02
Net					1.90 ± .02
10	W-1	31.15	138	4.41	1.12
11		31.42	146	4.62	1.17
12		32.16	149	4.61	1.17
13		31.64	143	4.50	1.14
Average					1.15 ± .01
Tr. El. Cor.					.01
Net					1.14 ± .01

TABLE 3

mum and maximum pulse height settings of the differential
discriminator. The correction refers to the failure of the
differential analyser to show a linear response in the
observed counting rates when linear changes in the activity
level of the specimen were made. A chart comparing the
actual disintegration rate with the observed counting rate
was made by Winchester (see fig. 2, Discussion of Errors).
From this figure it is seen that for all activities yield-
ing an observed counting rate of greater than 40 c/s some
correction in the observed counts must be made; the
higher the level of observed activity, the greater the
correction. Because the level of activity of the Na
standard was consistently higher than either the G-1
or W-1 samples, the correction required for it was greater.
The chart apparently did not compensate for the entire
error, since even with the use of it, the values of per
cent Na in G-1 and W-1 determined in the first three
runs are consistent neither among themselves nor with
the literature values.

To obtain the net counting rate, the activity
of the blank polyethylene packets was measured and sub-
tracted from the gross counting rate of the specimens.
In all cases this correction was less than 0.5% of the
lowest sample counting rate for the particular run. The

statistical counting errors were never greater than 1%.
The errors indicated for the average activity of the stand-
ards and for the average per cent Na are the standard
deviations of the mean.

Trace element correction was determined by
theoretically predicting the activity induced in elements
present in G-1 and W-1 which have γ radiations similar
to Na and comparing this activity level to the measured
counting rate of the Na (see Discussion of Errors). The
correction is well under 1% of the determined Na value.

Table 4 summarizes the data obtained from
Run #4. Counting was carried out using an integral dis-
criminator, and all γ's with energies greater than 2.70
m.e.v. were counted. Preliminary trials on this apparatus
indicated that a considerably higher counting rate than
the permissible 40 c/s of the differential discriminator
could be handled without introducing a dead time error.
The irradiation time of the specimens was reduced to 10
seconds.

The fifth column lists the decay corrected, net
c/M - mg. This decay correction was necessary because the
use of the automatic sample changer in this run increased
the time taken to complete one counting cycle of the samples

Counting Data for Run #4

SA No.	Type	Sa Wt (mg)	Net c/M	Net c/M-mg	Decay corrected net c/M-mg	% Na
1	Na_2CO_3	9.53	1392	146.3	140.8	
3	"	11.25	1486	132.0	135.2	
4		9.31	1226	131.7	139.2	
Average					138.8 ± 1.8	
5	G-1	44.97	366	8.14	7.86	2.46
6		49.70	392	7.86	7.86	2.46
7		52.21	396	7.58	7.76	2.43
8		50.31	347	6.89	7.31	2.29
Average						2.41 ± .04
Tr. El. Cor.						.02
Net						2.39 ± .04%
9	W-1	47.74	268	5.59	5.43	1.71
10		49.94	254	5.12	5.12	1.61
11		48.97	239	4.88	5.04	1.58
12		50.37	231	4.59	4.90	1.54
Average						1.61 ± .04
Trace El. Cor.						.01
Net						1.60 ± .04%

Two additional runs of the above cycle yielded the following results:

Net % Na in G-1	2.38 ± .04%
	1.60 ± .03%
Net % Na in G-1	2.41 ± .04%
in W-1	1.62 ± .03%

TABLE 4

and standards. In decay correcting, the zero time point
was chosen at a time approximating the half-way mark of
the cycle. This kept the average correction as low as
possible. The results of three cycles run over a period
of two days are listed. All of the determined per cent
Na values lie within the standard deviation of the mean
of the most extreme value.

Table 5 is a summary of Na analyses including
literature values for the per cent Na in G-1 and W-1 as
well as the results of Run #4. New chemical analyses of
the G-1 indicate a decrease in the Na found as compared
with past work, while in W-1 the new chemical analyses
indicate an increase in the observed Na composition.
The results of the determinations by neutron activation
analysis follow this trend of increasing Na in W-1 and
decreasing Na in G-1.

Summary of Sodium Analyses of G-1 and W-1

		% Na_2O	% Na
G-1 Granite			
This work Run #4			2.40 ± 0.04
Ref. 1, P. 38	median of preferred new values	3.28	2.43
	median of all preferred values	3.30	2.45
W-1 Diabase			
This work Run #4			1.61 ± 0.03
Ref. 1, P. 39	median of preferred new values	2.10	1.56
	median of all preferred values	2.09	1.55

TABLE 5

IV

DISCUSSION OF ERRORS

For the Na analysis of a mineral to be adaptable
to neutron activation analysis there must be no appreciable
amount of elements present in the mineral under investiga-
tion which will contribute conflicting radiations. One
must be certain that the γ's counted are those which
originate from the decay of Na^{24}.

The first section of table 6 lists the prominent
constituents of G-1 and W-1 and their radionuclides pro-
duced during irradiation by thermal neutrons. The
second half of the table lists other elements which, upon
irradiation, give rise to radionuclides which have similar
decay characteristics to Na^{24}. The second column lists
the radionuclide and its half-life; the third column,
the energy, or energy range of the γ's given off; and
the fourth column, the activity in disintegrations per
second obtained when 1 gm of the pure parent element is
irradiated for 10 sec. under a thermal neutron flux of
10^{13} n/cm^2 sec. and then allowed to cool for 30 hours. Of
the prominent elements present in G-1 and W-1 it is seen
that only Mn has a γ sufficiently energetic to interfere
with the Na determination. Of the other elements listed,

Prominent elements and their activities
Prominent elements of G-1 and W-1

Parent element	Radionuclide		E_γ (Mev)(7),(8)	Activity of 1 gm of parent after t_{ir} = 10 sec t_d = 30 hr (d/s)
Na	15h	Na^{24}	2.75	4.46×10^6
Si	2.62h	Si^{31}	1.26	1.95×10^2
Al	2.30m	Al^{28}	1.78	--
Fe	4.5d	Fe^{59}	0.19-1.29	5.60×10^2
Mg	9.5m	Mg^{27}	0.18-1.02	--
Ca	4.7d	Ca^{47}	0.5-1.30	2.06×10^1
K	12.5h	K^{42}	0.32-1.5	3.36×10^5
Ti	5.80m	Ti^{51}	0.32-0.93	--
P	14.5d	P^{32}	no	2.05×10^5
Mn	2.58h	Mn^{56}	0.85-3 (50% with E_γ 2.65)	3.48×10^5

other elements

Parent element	Radionuclide		E_γ (Mev)(7),(8)	Activity of 1 gm of parent after t_{ir} = 10 sec t_d = 30 hr (d/s)
Ni	2.56h	Ni^{65}	0.37-1.5	4.63×10^2
Cu	12.8h	Cu^{64}	1.34	8.76×10^6
Zn	3.0h	Zn^{71}	0.4-0.6	3.26×10^1
Ga	14.1h	Ga^{72}	0.6-3.35 (83% with E_γ 2.508)	4.11×10^6
Ge	26.7h	As^{76}	0.55-2.1	1.14×10^7
Sn	10d	Sn^{125}	0.33-2.0	9.18×10^1
La	40.2h	La^{140}	0.11-2.9 (48% with E_γ 2.6)	1.10×10^7
Ir	19h	Ir^{194}	0.29-2.0	7.86×10^7

TABLE 6

Ga, La, Mn, and possibly Ge may interfere. For equal weights of parent element, La will yield approximately 2.5 times the activity of Na after 30 hours of decay. Ahrens and Fleischer (10) have compiled the known trace element composition of W-1 and G-1. In table 7, the second and fourth columns, this data is listed for Ga, Ge, La, and Mn. The third and fifth columns list the apparent per cent Na which these elements will account for when counting the 2.75 m.e.v. Na^{24} γ. This apparent per cent Na is arrived at by determining what fraction of the counting rate which is observed as Na^{24} decay is, in reality, originating from these four elements. The correction is found to be small in both G-1 and W-1.

The foregoing gives a theoretical verification of the adaptability of the determination of Na in G-1 and W-1 by neutron activation analysis. A delicate practical test is the comparison of the γ spectra of G-1, and W-1, with the γ spectrum of pure Na as obtained from a Na_2CO_3 standard. A rigorous check on the similarity of the spectra is to take point-by-point ratios of the counting rates over a wide range of energy levels and to plot these ratios as a function of the energy level. The result should be a horizontal straight line. Figure 1 is such a plot. The upper line is the ratio of G-1 to Na_2CO_3 and

| | G-1 | | W-1 | |
Element	Content (PPM)(10)	Apparent % Na	Content (PPM)(10)	Apparent % Na
Ga	18	0.00138	16	0.00123
Ge	1	0.000000847	1.6	0.00000136
La	150	0.0178	30	0.00356
Mn	210	0.000819	1300	0.00508
Total apparent % Na		0.0202		0.0099

γ Interferences in G-1 and W-1

TABLE 7

the lower is W-1 to Na_2CO_3. The specimens used to obtain
the data for this figure were prepared in the same manner
as ones which were later used in the experiments. Thus a
test of possible contaminants in the polyethylene container,
as well as the samples, is made. While there is consider-
able scatter there does not appear to be any trend other
than horizontal. An upward slope, if it did occur, would
occur most likely at the low energy range, since it is in
this region that most of the residual contaminating activ-
ity would be. This activity would augment the Na activ-
ity of the specimen and increase the value of the ratio.

In light of this apparent lack of conflicting
activity, and the very low background and polyethylene
packet activity, there was some question as to the cause
of the erratic data obtained in the first three runs.
Plotting the decay curves of the samples over several
days revealed half-lives of the standards in the order of
20 hours, half-lives of the G-1 samples in the order of
16 hours, and half-lives of the W-1 samples close to the
normal 15 hours. From tables 1-3, column four, it is seen
that this increasing of the half-life, from W-1 to the
standard follows the same trend of the increasing level of
activity from W-1 to the standard. This gives the possible
implication that the erratic data may be connected with

figure 1

Ratio of G-1 and W-1 γ spectra to Na₂CO₃ γ spectrum
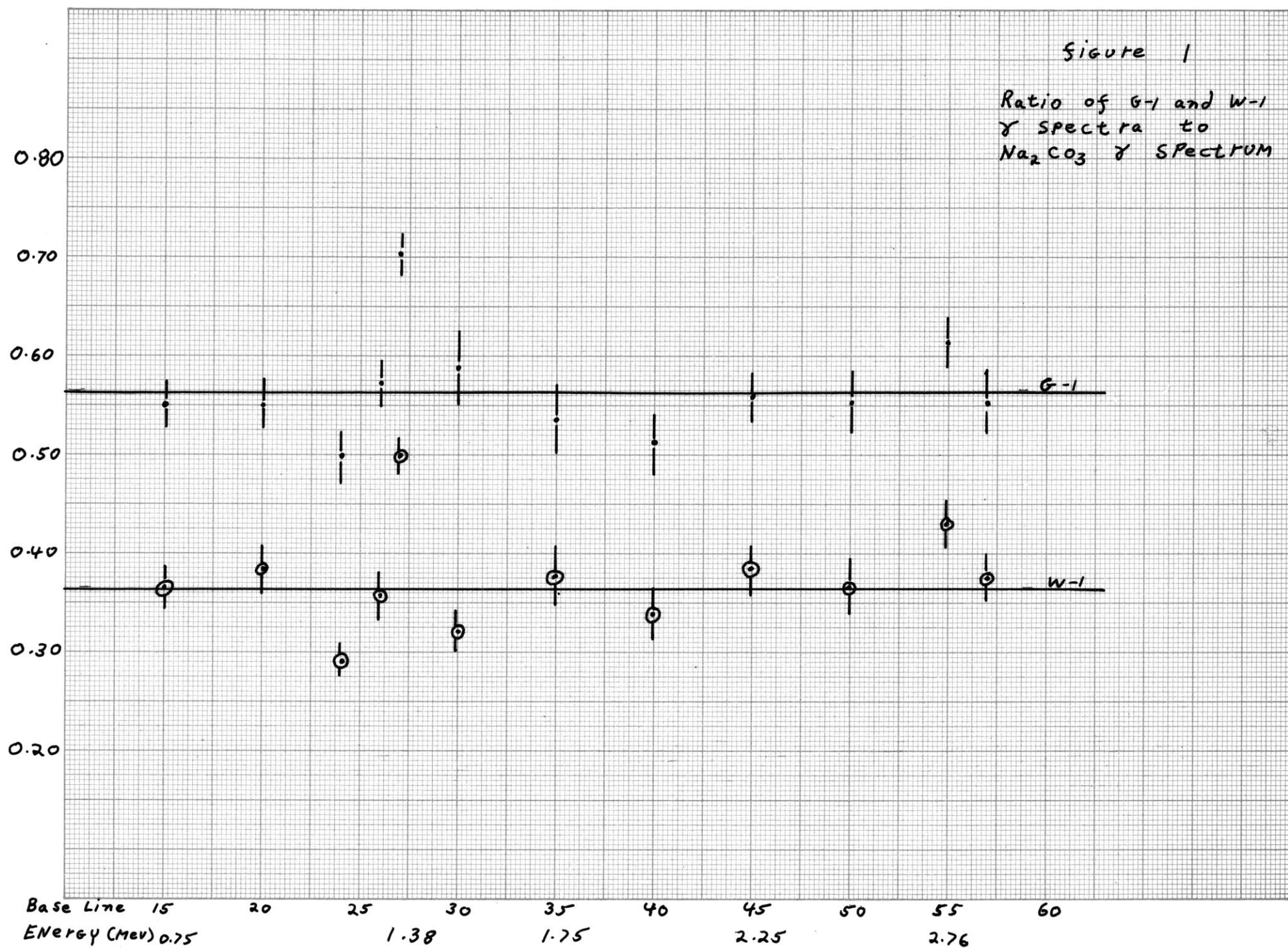
electronic counting equipment and not with the feasibility
of the experiment. Because the observed counting rate is
relatively low as compared with the theoretical capacity
of the apparatus, the difficulty may lie with the high
flux of not recorded low energy gammas and their effect
on the dead time of the apparatus. The higher the level
of observed activity the proportionately higher the level
of low energy activity and hence the greater the effect on
observed counting rate, and measured half-life. Figure
2, prepared by Winchester, is the counting correction
chart for the first 3 runs. It was obtained as follows:
A group of Na standards with a wide range of activity
levels known by theoretical predictions was prepared. The
activity of these standards was counted using the differen-
tial discriminator (which was employed during the first
3 runs) as the pulse height analyser and the observed
counting rate versus the relative disintegration rates
of the standards was plotted on logarithmic paper. The
line should be a straight line following a 45° angle;
that is, increasing the relative disintegration rate
(or specimen strength) by a factor of ten should also
increase the observed counting rate by a factor of ten.
Up to 40 c/s the observed counting rate followed the 45°
line; however, above this the variation continually became
more pronounced until at an observed counting rate of 1400

figure 2

Winchester's correction curve for differential analyser

Counts/sec

65.2%

Theoretical counting rate →

Measured counting rate
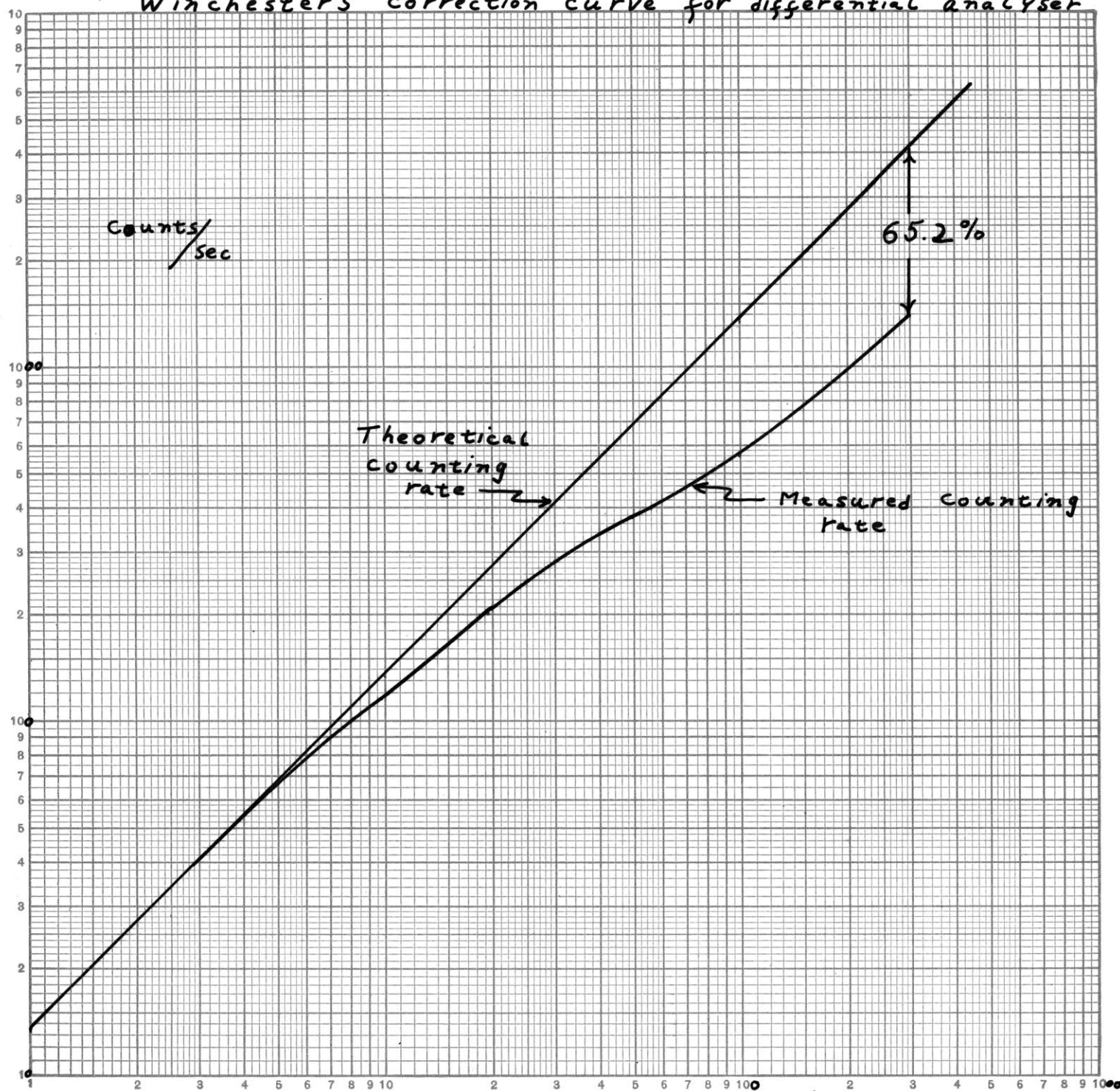
Relative Disintegration Rate

c/s one is actually counting 65% below the theoretical counting rate.

A similar test was performed using the integral pulse height analyser which was used for Run #4. The observed counting rate was found to follow the 45° line to above 100 c/s. This was satisfactory for the experiment, and as a safety factor, the irradiation time was reduced to 10 sec. so that the maximum counting rate was below 60 c/s at all times. Above 100 c/s the trend of the measured counting rate was to be greater than the 45° theoretical ine. This may result from coincidence adding of several low energy γ 's to appear as a single high energy .

As a sensitive check on the reproducibility of the counting data obtained in Run #4 the decay of the specimens was measured over a 3-1/2 day period and the ratios of the G-1 and W-1 counting rates to the Na_2CO_3 counting rate was plotted (fig. 3) While there is considerable scatter, all the points for G-1, and most for W-1, lie within their statistical counting error of the lines drawn. The horizontal lines found for G-1 and W-1 indicate that there is no trend for either of the samples to have half-lives other than 15 hours, and therefore the observed counting is assumed to be correct, and resulting all from the 15 hour Na^{24}.

Figure 3

Ratio of G-1 and W-1 decay curves to Na_2CO_3 decay curve
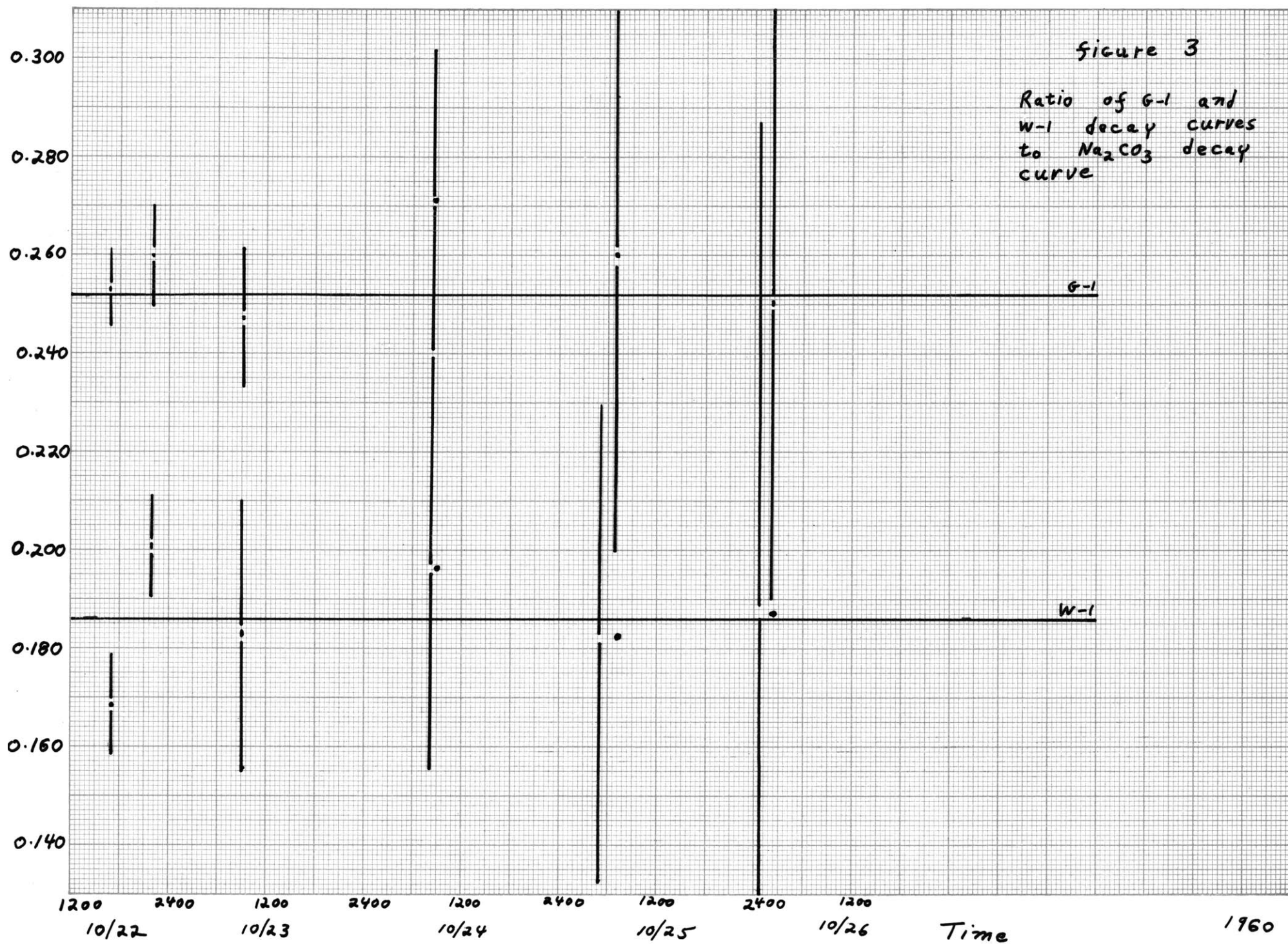
V

CONCLUSIONS

Barring difficulties encountered in the electrical phase of the counting, determining the Na composition of G-1 and W-1 by neutron activation analysis appears practical. The calculated trace element interference in both G-1 and W-1 is less than the standard deviation of the mean. The purity of the Na^{24} γ is verified by the 15 hour half-life measured in the decay of the minerals. Care must be taken to keep the observed level of activity low enough so that the low energy γ's do not cause dead time complications with the counting apparatus. In any case, the characteristics of the counting apparatus should be studied before attempting an experiment.

It is possible to determine theoretically whether the minerals are adaptable to this form of analysis, provided previous chemical analyses are available. Using the data of table 6, an estimation of the interference other elements present in a mineral can be made.

The method of heat-sealing the specimens in polyethylene provides an efficient and contamination-free means of handling.

VI

ACKNOWLEDGEMENTS

VII

APPENDIX

A. Description of Apparatus

Primary detection of the radiation was effected
by a 2" x 2" cylindrical, thallium activated NaI crystal.
At a height of 2 cm. above the face of the crystal a
fixed aluminum rack was located. This held the specimens
during their counting and ensured identical counting
geometry in all instances. The NaI($T\ell$) crystal was
monitored by a 10 stage photomultiplier tube which relayed
the scintillation information to a Baird-Atomic amplifier
and pulse height analyser, and ultimately to a scaler.
During the first three runs of the experiment a differ-
ential analyser was used. For the last run an integral
analyser was chosen. Coupled to the integral pulse
height analyser and its scaler was an automatic sample
changer. This enabled the investigator to preselect
the total number of counts desired for each sample and
to   have a register print out next to the sample number,
the time taken for the scaler to record this number of
counts.

Weighing of the specimens was performed on a

Mettler balance, with a maximum sensitivity of 0.001 mgs.
For fine determinations in weighing an excess of specimen
should be placed on the balance pan and weighed. From
this the desired amount of specimen is removed and a
second reading is taken. The weight of the specimen
taken is simply the difference of the two readings.

B. Irradiation Facilities

All irradiations were carried out in the M.I.T.
Reactor. Sealed samples are placed in a cylindrical
"rabbit" 2.5 cm. in diameter and 7 cm. long. This rabbit
is taken into and removed from the pile by a pneumatic
tube. As a test for uniformity of the neutron flux over
the length of the rabbit, accurately weighed Na standards
were placed at intervals along the rabbit and the rabbit
was fed into the pile and irradiated. The absolute
activity of each sample was then determined and found to
be the same within statistical limits. Thus it is assumed
that the neutron flux is the same over the entire rabbit.
It is good practice, even so, to alternate samples and
standards when packing the rabbit with several small

specimens.

Thermal neutron flux $(n \nu)$ equals
5×10^{12} n/cm^2-sec.

C. Statistics of Counting[1]

Starting with the binomial distribution law
and applying the general characteristics found in most
counting circumstances it is possible to arrive at a
convenient equation describing the probability of obtain-
ing a particular number of counts in a given time.

The binomial distribution law is

$$W(r) = \frac{n!}{(n-r)! \, r!} \, P^r (1- P)^{n-r}, \qquad (1)$$

where, in a very large set of objects, P is the proba-
bility of the occurance of a particular object W. n
objects are removed. $W(r)$ is the probability that r
of the objects are of the kind W.

To find the probability, $W(m)$, that m disinte-
grations will occur in time t from N_0 original active

[1] Friedlander & Kennedy, Nuclear and Radiochemistry,
John Wiley & Sons, Inc., N. Y., 1960. Pp. 255-263.

nuclides, let N_0 equal n, and m equal r. After time t, the number of atoms left N from the original N_0 is just $N_0 e^{-\lambda t}$. Thus the probability of an atom surviving t seconds is $\frac{N}{N_0} = e^{-\lambda t}$. This equals 1- P. Therefore $P = 1 - e^{-\lambda t}$.

Substituting in the original equation yields

$$W(m) = \frac{N_0!}{(N_0-m)! m!}(1-e^{-\lambda t})^m (e^{-\lambda t})^{N-m} . \quad (2)$$

Now if the number of active atoms is large, the observation time t short compared with half life, then $\lambda t << 1$, $N_0 >> 1$ and $m << N_0$. Approximating as follows:

$$e^{\lambda t} = 1 + \lambda t$$

$$X! = \sqrt{2 \pi X} \, X^X e^{-X}$$

$$(1 - \frac{m}{N_0})^{N_0} = \lim_{N_0 \to \infty} (1 - \frac{m}{N_0})^{N_0} = e^{-m}$$

and applying this to equation (2) we have

$$W(m) = \frac{M^m e^{-M}}{m!} \quad \text{where M is the average to}$$

be expected. This is Poisson's Law. It is accurate for N_0 as low as 100 and λt as large as .01.

Studying this equation shows that $W(M) = W(M-1)$. For large numbers of counts (M) the distribution is nearly symmetrical about M. From this the

fact that $\sigma = \sqrt{m}$ may be obtained. That is, the standard deviation in counting is just equal to the square root of the total number of counts.

It is extremely useful to be able to carry out simple arithmetic functions on numbers containing standard deviations. The sum of two Poisson distributions is also a Poisson distribution. The variance σ_s^2 is found by summing the individual variances. Thus $\sigma_s = \sqrt{\sigma_1^2 + \sigma_2^2 + \cdots}$ The difference of two Poisson distributions, however, is not a Poisson distribution. The variance σ_d^2 is found by summing the individual variances. Again $\sigma_d = \sqrt{\sigma_1^2 + \sigma_2^2 + \cdots}$

To find the standard deviation of the quotient of two counting rates, $\sigma_q = Q \sqrt{\left(\dfrac{\sigma_1}{R_1}\right)^2 + \left(\dfrac{\sigma_2}{R_2}\right)^2}$

where Q is the quotient.

The standard deviation of a product is found in a similar manner. $\sigma_p = P \sqrt{\left(\dfrac{\sigma_1}{R_1}\right)^2 + \left(\dfrac{\sigma_2}{R_2}\right)^2}$

D.   Interaction of Gammas With Matter

Unlike α and β rays, the γ has zero charge
and zero rest mass. Its velocity in a given medium is
constant and equal to velocity of light (3.0×10^{10} cm/sec.
in vacuum) in that medium. Since it adheres to the wave-
particle theories concerning matter, the energy of the
ray is determined by its frequency. The range of a
beam of γ's is truly infinite and falls off exponentially
according to the equation

$$I = I_0 \, e^{-\mu x}$$

where   I = intensity of the beam
         I_0= original intensity
         X = distance traversed
         μ = absorption coefficient

         = σ abs . n atoms/cc.

Thus a plot of the log c/s versus gm/cm^2 of absorber will
yield a straight line with a negative slope proportional
to μ .

The γ interacts with matter in the three follow-
ing ways, considered here in the order of increasing energy
requirements. In Compton scattering the γ transfers a
small fraction of its energy to an atomic electron in
a nearly elastic collision. The electron flies off and
the γ changes direction accordingly, to conserve momentum.

If the direction and velocity of one of the participating particles is known, the direction and velocity of the other may be calculated by classical methods.

In the photoelectric effect the γ transfers all of its energy to a single atomic electron which is ejected with a kinetic energy equal to the energy of incoming γ minus the ionization potential of the electron. After this photoelectric emission of the electron, characteristic X-rays are given off as the energy levels which the electron has vacated are refilled. The reaction cross-section for the photoelectric effect is proportional to Z^4 and inversely proportional to $(KE_\gamma)^3$.

For a true understanding of pair production, one must first have a knowledge of the theory of what positrons are.

In the normal state of nature, electrons at rest have an energy of $M_o C^2$ relative to a state of zero energy. When given motion the energy of the electron increases by the value of the kinetic energy. Electrons may also have a negative rest energy equal to $-M_o C^2$ and negative kinetic energies. Normally these negative levels are all filled. To create an electron pair one must impart sufficient energy to an electron in the neg-

ative state to move it into the positive state. The
minimum energy requirement would be to move a stationary
electron in the negative state (at $-M_oc^2$) to a stationary
position in the positive state (at $+M_oc^2$). Thus a
total of $2\,M_oc^2$ is needed. This is equal to 1.02 Mev for
an M_o of an electron. What one sees as a positron is merely
a space or a hole in the otherwise completely filled nega-
tive state. The destruction of a positron is always accom-
panied by the destruction of an electron. This is
because what actually actually happens is that the electron
falls back into the space in the negative state. Thus the
electron is gone, and so is the space.

In pair production, a γ , having an energy
equal to, or greater than 1.02 Mev strikes an electron
which is in the negative state, and moves it into the
positive. Thus a new electron is seen in the positive
state, and a positron (actually a hole in a negative
field, thus appearing as a positive charge) is found to
accompany it. The two quanta conserve the momentum of the
original γ . When the positron encounters an electron,
the two disappear and a γ , which conserves the momentum
of the two disintegrated quanta, is formed.

E.   Scintillation Counters

The basic mechanism in the operation of the
scintillation counter is the interaction of a quantum,
usually an alpha or a gamma, with a phosphor. The result-
ing pulse of light is observed by a photomultiplier,
amplified and recorded. By selecting a large enough
crystal, usually thallium-activated sodium iodide, gamma
counting efficiencies, which are very low in proportion-
al counters, approach 100%. The pulse height of the
interaction is found to be proportional to the energy
of the incident particle. This enables the construction
of multi-channel counters which when coupled with continu-
ous recorders are able to plot pulse height, and hence
incident particle energy, versus the number of pulses at
that particular pulse height. A sample placed in the count-
ing chamber can have its entire spectrum of gammas anal-
ysed in a single operation.

The construction of the counter may be accom-

plished by placing a thin phosphor directly on the surface
of a photomultiplier tube, or, more generally, by coating
a transparent light pipe such as lucite with the phosphor
and have this lucite conduct the light to the photomulti-
plier. Recent developments have brought forth fluorescent
plastics which combine the action of the phosphor and the
lightconductor. In coating a lightconductor one must
arrive at an optimum thickness for the phosphor which
will maximize the interaction with the incident quanta
and minimize the self-absorption of the light pulse pro-
duced.

Since the fluorescent material must be
shielded from all external light certain difficulties
arise if the sample is located outside of the counter
housing. A thin aluminum cover may be placed over the
phosphor; however, if alphas are to be counted their short
range dictates that this cover be of absolute minimun
thickness. Ideally an alpha source should be placed
directly in the housing. This is most convenient when
dealing with gases.

A marked advantage of the scintillation counter
is its extremely low dead, or resolving, time. The
thallium-activated sodium iodide crystal has a resolving
time in the order of 2×10^{-7} second, thus enabling the

metering of an extremely high alpha or gamma flux.

F. Chi Squared Test

It is often useful to be able to perform a rapid
test on a counter and scaler to determine whether or not
the unit is functioning properly. The chi squared test
performs just such a service. If a counter is register-
ing incorrectly, the scattering of the data obtained will
follow a pattern such that when the probability of the
next set of data falling within the standard deviation
of the previous data is calculated the probability will
either be too high (>0.98) or too low (<0.02). To
determine the probability take a set of data, find the
average value ($\bar{x}$), and then x^2

$$x^2 = \frac{(x_1 - \bar{x})}{\bar{x}}$$

on page   in Evan's The Atomic Nucleus (11)
a chart relates x^2 to the number of degrees of freedom
F which is equal to the number of sets of data taken minus
one. An example of the test allows using background

noise as a source. This is a convenient source to use, since its rate is low and the low numbers allow for rapid calculations.

G. Analytical Data for Sodium Standards
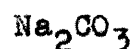
Na_2CO_3

     Produced by: J. T. Baker Chemical Co., N.J.

     Grade: Reagent

     Per cent Na_2CO_3: 99.8

NaF

     Produced by: Merck Chemical Co., N. J.

     Grade: Reagent

     Per cent NaF: 99.4

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