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Some Neutron-Deficient Strontium Isotopes

Stanley Vernon Castner

October 16, 1950

Berkeley, California

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SOME NEUTRON DEFICIENT STRONTIUM ISOTOPES

When rubidium is bombarded with protons, strontium isotopes are produced by the reactions Rb(p,xn)Sr, where x may equal 1,2,3,4, 5...., depending on the energy of the incident protons. As the only rubidium isotopes found in nature[1] are Rb^{85} and Rb^{87}, the strontium isotopes which are produced will have mass numbers less than or equal to 87. Sr^{84}, Sr^{86}, Sr^{87}, and Sr^{88} are stable,[2] although an excited state of Sr^{87} is known[3] with a half-life of 2.7 hours. In addition Sr^{85} has been reported[4,5] to decay by the following modes: an isomeric transition of 70 minutes half-life accompanied by a partially converted gamma ray of 0.170 Mev energy, which is followed by an orbital electron capture of 65 days half-life, accompanied by a gamma ray of 0.8 Mev energy. Strontium isotopes with mass number less than 84 have not been reported.

By bombarding rubidium chloride with protons at energies from 25 Mev to 100 Mev, three new strontium isotopes with mass number lower than 84 have been observed, and their half-lives and radiation characteristics have been determined. The neutron deficient strontium isotopes produced decay by positron emission and orbital electron capture, to form rubidium isotopes. Stable Rb^{85} ends the decay chain starting with Sr^{85m}. Sr^{84} is stable, and consequently no radioactive Rb^{84} will be seen from strontium decay. Sr^{81}, Sr^{82}, Sr^{83} will decay to form Rb^{81}, Rb^{82}, and Rb^{83} respectively. Karraker and Templeton[6] have reported the properties of these rubidium isotopes as follows:

ISOTOPE	HALF-LIFE	TYPE OF DECAY	ENERGY OF RADIATION IN MEV PARTICLES	GAMMA RAYS
Rb^{83}	107 days	K, γ	-----	-----
Rb^{82}	6.3 hours	β^+, γ, K	0.670	1.2, 0.7
Rb^{81}	4.7 hours	β^+, γ, K	0.990	0.95

The above rubidium isotopes were assigned to the mass numbers shown by the use of a mass spectrograph. These isotopes were used to assign the mass numbers of the strontium isotopes under discussion, by establishing parent-daughter relationships between the unknown strontium isotopes and the known rubidium isotopes.

TARGET PREPARATION

The rubidium was obtained as $RbNO_3$. Spectrographic analysis showed the cation to consist of 86% Rb, 5% Cs, 8% K, and 1% Na. The rubidium was purified by the use of a cation exchange column. The column was 11 mm in diameter and 81 cm in length. The column was filled with the hydrogen form of Dowex 50 cation resin which had been graded to remove particles smaller than about 350 mesh. The column was washed with 6N-HCl for 48 hours and then with distilled water for 48 hours. Approximately 1 cm (in length of the column) of the resin was removed and equilibrated for 45 minutes with 1.074 grams of $RbNO_3$ dissolved in a few milliliters of water. The resin was then replaced on the column. The eluting agent was 6.0 N-HCl. The flow-rate was approximately 0.2 ml per minute. The temperature was held at about 30° C. One hundred fifty samples of about 1.5 ml each were collected. An aliquot of 100 microliters was taken from

each sample and analysed spectrographically for Na, K, Rb, and Cs with the following results:

Sample #	Elements Detected	% By Weight of Total Elements Present in a Sample
1 to 33	None	---
34 to 39	Na	100
40 to 74	None	---
75	K	100
76 to 81	K Rb	3 97
82 to 100	K Rb	00.4 99.6
101 to 146	None	---
146 to 150	Cs	100

The samples numbered 82 to 100 were combined, the solution was evaporated to a small volume, and the rubidium chloride was allowed to crystallize. The RbCl was recrystallized from a 50% solution of ethanol and water and then washed with absolute ethanol and finally with dry ether. The weight of the dry RbCl was 0.605 grams or an overall yield of 68.7% on the basis of 1.074 gms of $RbNO_3$. The remaining rubidium was presumed lost in the equilibration and in the samples numbered 76 to 81. Bombardment targets were made by wrapping 25 to 50 milligrams of the purified RbCl in 0.001 inch thick aluminum foil. The targets were then bombarded with protons on the 184" cyclotron.

CHEMICAL SEPARATIONS

After bombardment the RbCl was dissolved in water. The solution was boiled to expell any inert gas isotopes formed and then cooled. A solution containing a known amount of $Sr(NO_3)_2$ was added. The strontium was precipitated with excess fuming nitric acid. The resulting suspension was chilled in an ice bath for five minutes and then centrifuged. The supernatant solution containing the RbCl was removed and the $Sr(NO_3)_2$ was dissolved in a small amount of water. A few mgs. of the impure $RbNO_3$ was added to the solution, and the strontium was re-precipitated with fuming nitric acid. After removal of the supernatant, the precipitate was redissolved and diluted to an arbitrary volume from which several accurately measured aliquots were taken. The strontium in an aliquot was again re-precipitated with fuming nitric acid. After removal of the supernatant, the precipitate was dissolved in water and the solution neutralized by bubbling NH_3 through the solution. A few ml of a saturated solution of $(NH_4)_2C_2O_4$ was added to precipitate the strontium as $SrC_2O_4 \cdot H_2O$. After removal of the supernatant the precipitate was washed once with water, once with 95% ethanol and finally slurried up in approximately one half ml of 95% ethanol and placed on a previously weighed aluminum dish of 0.002" thickness and approximately 1.0" in diameter. The ethanol was evaporated to dryness with a heat lamp. The sample was allowed to cool and was then weighed to determine the chemical yield of the strontium. The dish was mounted on a sample holder, and the activity counted with an end window geiger counter. In the initial bombardments, the purification procedures were extended to include a silver chloride precipitate scavenge, and a portion of the strontium fraction was passed through a Dowex 50 cation resin column. As no differences were noted between the decay curves of the strontium which had undergone these additional steps of purification,

and the decay curves of the strontium which had not undergone such measures, these steps were omitted in subsequent bombardments as consuming too much time.

To isolate the radioactive rubidium in an aliquot which resulted from the strontium decay, the following procedures were used: A solution containing a known amount of stable rubidium was added to the aliquot. The strontium was precipitated with excess fuming nitric acid. The resulting suspension was chilled in an ice bath for five minutes and then centrifuged. The supernatant was removed to a clean centrifuge cone and evaporated to approximately one half milliliter. A few mgs. of strontium was added and precipitated with more fuming nitric acid. Again the supernatant was removed to a clean centrifuge cone and evaporated to about one half ml. The solution was chilled and one half ml of 70% $HClO_4$ was added to precipitate the rubidium as $RbClO_4$. This precipitate was washed once with 50% ethanol, once with 95% ethanol and finally slurried up with one half ml of 95% ethanol onto a previously weighed aluminum dish. The ethanol was evaporated with a heat lamp. The sample was allowed to cool and was then weighed to determine the chemical yield of rubidium. The sample was mounted on a sample holder and counted with an end window geiger counter.

Radioactive kryptons were expelled from rubidium samples by heating the rubidium sample with a bunsen burner to dull redness. For the purposes of this separation, the rubidium was mounted on 0.020" monel metal disks.

RADIOACTIVITY MEASUREMENTS

Decay and absorption measurements were made with "Amperex," chlorine quenched, argon filled, end window, Geiger-Müller counters. The window thickness was approximately 4 mg/cm^2 of mica. Aluminum absorption measurements were made in the conventional manner. Then sufficient beryllium to absorb all particles present was interposed between the sample and the counter. An aluminum absorption of the electromagnetic radiation present was then made. Subtraction of the electromagnetic component (after correction for the effect of the beryllium) yielded complex particle absorption curves which could not be resolved readily. Soft electromagnetic components of appropriate half-thickness in aluminum were interpreted as k-x-rays. Their abundance was determined by subtraction of the harder components from the curve for aluminum absorption of electromagnetic radiation, with correction for the effect of the beryllium. Lead absorption measurements were taken at low geometry, using the counter without its conventional lead shield. Lead absorbers were placed between two beryllium absorbers of sufficient thickness to absorb all primary and secondary particles. Rubidium and Krypton K x-rays (about 13 Kev) were assumed to count with 4.5% efficiency and gamma rays over 0.5 Mev with an efficiency of 1.0% per Mev. Positrons and conversion electrons were assumed to count with only 45% efficiency. No estimate of the approximate branching ratios for the decay via positron emission and electron capture has been made due to the extreme complexity of

the radiations, i.e., at least two conversion electrons of unknown intensity and conversion coefficient with each strontium isotope, which makes it impossible to determine the true number of orbital electron capture events.

For samples of low activity or short half-life, rough measurements of the particle energies were made with a crude 180° magnetic spectrograph with about a 4 centimeter radius. This instrument was also useful to distinguish the sign of the particles in preliminary work.

The particles from Sr^{83} and Sr^{82}, which were produced in sufficient intensity were observed with the 270°, double focusing, beta-ray spectrometer described by Karraker and Templeton.[6] Samples for this instrument were mounted on tygon films of 20 to 30 micrograms per square centimeter, with about 10 to 50 micrograms of material in the sample. The backscattering from these samples was not serious in the range of energies of the particles observed.

MASS ASSIGNMENTS

A bombardment of rubidium chloride with the 184" cyclotron using protons of 25 Mev energy for a period of one hour was made. The range of energies of the protons in this bombardment was such that the maximum energy was 25 ± 1 Mev while the minimum energy was unknown but much smaller. The strontium activities formed were separated and counted within one hour of the end of the bombardment. The following strontium activities were noted: (Fig. 1) Sr^{85m} 70 minutes

half-life, Sr^{85} 65 days half-life, Sr^{87m} 2.7 hour half-life, and an unknown 38 hour half-life. Subsequent bombardments at higher energies have also shown this new activity. To assign a mass number to this isotope, the following experiment was performed. From a single aliquot of strontium activity, rubidium was separated, as described previously, several times at equal intervals of time. The yield of rubidium activities when plotted logarithmically against the time of separation will produce a straight line, the slope of which corresponds to the half-life of the parent strontium activity. In this case the time interval chosen was 24 hours and the experiment was continued for one week. The slope of the yield vs time curve was 34 hours (Fig. 2). The shortening of half-life was probably due to incompleteness of the strontium nitrate precipitation. The individual rubidium samples all showed a marked growth in activity during the first few hours (Fig. 2). This growth is due to Kr^{83m} 113 minute half-life[7] activity (Fig. 3). This part of the decay chain has not previously been reported. Decay of the individual rubidium samples was followed for a period exceeding two months, showing that each sample decayed with a half-life of approximately 100 days in good agreement with the value given by Karraker and Templeton of 107 days for this isotope (Fig. 4). In two of the samples strontium activity is to be noted giving support to the reason for the shortening of the half-life of the parent strontium.

With protons of 60 Mev energy and above, another activity in the strontium fraction was observed. The half-life of this isotope

was found to be approximately 29 minutes (Fig. 5). The experiment described above to determine the mass assignment of this activity was performed. The time interval chosen was 20 minutes, and the experiment was continued for a period of four hours. The yield of rubidium activity when plotted vs time, as before, gave 22 minutes as the half-life of the parent strontium isotope (Fig. 6). Again the shortening of the half-life was assumed due to incomplete precipitation of the strontium nitrate. In an attempt to further verify the value of the strontium activity half-life, the following experiment was carried out. Strontium was separated from several of the aforementioned aliquots at arbitrary intervals. The initial counting rates of such samples will have no "daughter" activities present. Hence if the initial counting rates are plotted vs time, the slope of the line obtained will be the true half-life of the strontium activity. This gave a value of 31 minutes fr the half-life of the strontium (Fig. 7). The decay of some of the individual rubidium samples was followed for approximately 30 hours. The half-life of the rubidium activity was 4.7 hours (Fig. 8). An absorption experiment using aluminum absorbers gave 1.1 Mev as the energy of the positrons present. (Fig. 9). The crude magnetic spectrograph showed the presence of conversion electrons as well as positrons. The conversion electrons could readily be reduced in number by flaming the sample, although the conversion electron counting rate increased very rapidly as soon as the heating ceased. The energy of the conversion electrons was

not measured precisely but from the distribution given by the crude spectrograph, the energy was estimated as 0.2 Mev. These conversion electrons belong to 13 sec Kr^{81}. This rubidium activity is therefore the 4.7 hour Rb^{81}. Thus the 29 minute half-life belongs to Sr^{81}.

With protons of 40 Mev energy and above, a 25.5 day half-life strontium activity was observed (Fig. 10). The experiment to determine the mass assignment of the strontium activity was performed but no rubidium activity of half-life longer than a few minutes or shorter than a year was observed. This 25.5 day half-life activity has been tentatively assigned to Sr^{82} from the energy protons needed to produce it. The 6.3 hour half-life of Rb^{82} has not been observed. If this 25.5 day half-life activity is Sr^{82}, there must exist isomerism for Rb^{82}. Sr activity when passed through a cation exchange column showed no difference, in decay curves or in the ratio of 25.5 day half-life activity to 38 hr. half-life activity, from another aliquot of Sr activity which was not passed through the cation exchange column. In addition, no other isotopes have been reported which have approximately this half-life with the correct radiation characteristics. It is not surprising that Rb^{82} exhibits isomerism, since many other odd-odd nuclei in this immediate region are known to do so.

The mass relations of these isotopes are shown in Table I. Radiation characteristics of each isotope will be discussed separately.

Table I

Isotope Chart[a]

Mass No. / Element	79	80	81	82	83	84	85	86	87	88
Sr			29 m e^-,K β^+,γ	25.5d e^-,K β^+,γ	38 h e^-,K β^+,γ	Stable	70 m e^-,γ 65 d K,γ	Stable	e^-,I.T., γ, 2.7 h Stable	Stable
Rb			4.7 h K,β^+,γ	6.3 h K,β^+,γ	107 d K,e^-,γ	34 d K,β^+,γ	Stable	19.5 d β^-,γ	10^{10} y e^-,γ,β^-	17.5 m β^-
Kr	34 h K,β^+,γ	Stable	13 s IT? e^-,γ	Stable	113 m IT e^-,γ Stable	Stable	4.5 h β^-,γ	Stable	74 m β^-	3 h β^-
Br	Stable	4.4 h IT,e^-,γ 18m β^-,γ,β^+	Stable	34 h β^-,γ	2.4 h β^-	30m β^-,γ	3.00m β^-			

a G. T. Seaborg and I. Perlman, Rev. Mod. Physics 20, 585 (1948).

RADIATION CHARACTERISTICS

38-hour Sr^{83}

Measurements on the 270° beta-ray spectrometer have shown that Sr^{83} has positrons with 1.15 ± 0.05 Mev energy limit as determined by a Kurie plot[8] of the spectrum (Fig. 11). Measurement of the conversion electrons with the 270° beta-ray spectrometer showed the presence of five pairs of lines corresponding to K and L conversion of gamma rays of 0.040, 0.074, 0.101, 0.151, and 0.165 Mev energy.

The presence of K x-rays has been established by following the decay of strontium activity through sufficient beryllium to absorb all particles emitted and then through the same amount of beryllium plus sufficient silver to reduce the intensity of the K x-rays by a factor of 0.6 (60.41 mg Ag/cm^2). The resulting curves show identical half-lives, and the percentage difference between the two curves remains the same throughout the experiment. (Fig. 12). The presence of gamma rays of 38-hr. half-life was noted in lead absorption measurements, but it was not possible to determine the energy of these gamma rays because of the presence of 25.5 day Sr^{82} activity.

25.5-Day Sr^{82}

Measurements on the 270° beta-ray spectrometer have shown that Sr^{82} has positrons with 3.15 ± 0.03 Mev energy limit as determined by a Kurie plot of the spectrum (Fig. 13). Conversion electrons of Sr^{82} were observed on the crude magnetic spectrograph, but the intensity of these electrons was too low to observe on the 270° beta-ray spectrometer. The distribution given by the crude magnetic spectrograph indicates at least two lines of conversion electrons corresponding to

gamma-ray energies of 0.15 and 0.40 Mev. K x-rays have been identified by absorption measurements in aluminum (Fig. 14). A gamma-ray of 0.95 Mev energy and one gamma-ray of 0.15 Mev energy have been observed by an absorption measurement in lead (Fig. 15). The measurements on this isotope were made approximately five weeks after bombardment and was consequently free of all other activities.

As no rubidium daughter activity was observed in this fraction, the mass assignment is still in some doubt. If we assume, as is probable, that the long range positron belongs to the rubidium daughter, the half-life of this daughter, if an unforbidden decay, will be of the order of ten seconds and would not have been observed by the separation techniques used.

28-minute Sr^{81}

Since the half-life of this isotope is so short, and the activities due to Sr^{82} and Sr^{83} are always present whenever investigations of Sr^{81} are being made, the use of the 270° beta-ray spectrometer was not possible. Use of the crude spectrometer shows the presence of both positrons and conversion electrons, the energies of which have not been determined. The presence of K x-rays has not been definitely established although there is some evidence for their presence. The half-life of this isotope is so short that time did not permit a lead absorption measurement for detection of gamma-rays.

107-day Rb^{83}

Approximately 4×10^4 counts per minute of this activity have been isolated from the 38-hour Sr^{83}. Aluminum absorption measurements

have shown the presence of K x-rays (Fig. 16). A gamma ray of 0.8 Mev energy has been observed in an absorption in lead (Fig. 17). The activity was too little to use the 270° beta-ray spectrometer, but the crude spectrograph has shown the presence of two lines of conversion electrons corresponding to gamma rays of approximately 0.15 and 0.45 Mev energy. No positrons were observed in the sample but the sensitivity for detection is poor. Assuming no positrons present, the ratio, conversion electrons: K x-rays: gamma-rays is 3×10^{-2}:1:1.

(1)	A. O. Nier	Physical Review	50,	1041 (1936)
(2)	A. O. Nier	" "	54,	275 (1938)
(3)	L. A. Dubridge and J. Marshall	" "	66,	706 (1939)
(4)	" "	" "	57,	348 (1940)
(5)	" "	" "	58,	7 (1940)
(6)	D. G. Karraker and D. H. Templeton	" "	In press	
(7)	A. Langsdoorf Jr. and E. Segrè	" "	57,	105 (1940)
(8)	R. Hayward	U.C.R.L.---582 (Unclassified)		

Fig. 1

Decay of Strontium Activities Produced at

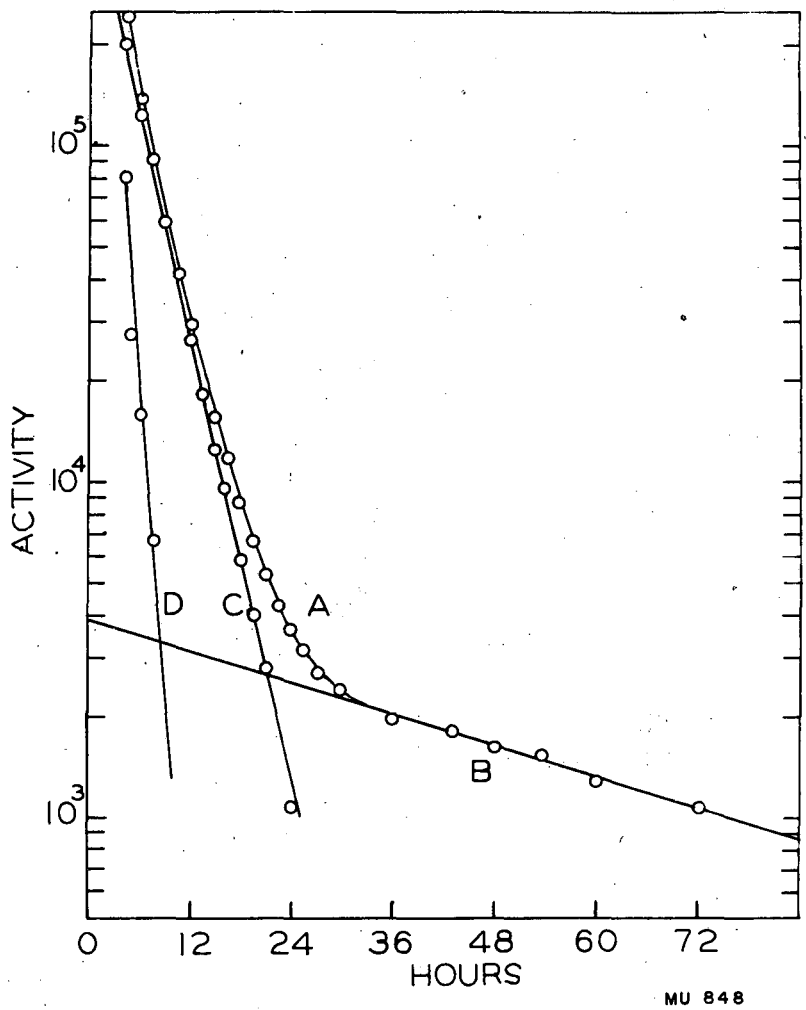
25 Mev Energy

A. Gross decay with 350 counts/min of 65-day Sr^{85} subtracted

B. 38-hour Sr^{83}

C. 2.7-hour Sr^{87m}

D. 70-minute Sr^{85m}



167241

Fig. 2

Yield of Rb^{83} Vs. Time of Separation From Sr^{83}

A, B, C, D, E, F. Rubidium samples showing initial growth and little decay during the week of the experiment. All samples corrected for chemical yield. Samples A & B corrected for presence of strontium contamination.

G. Yield vs. time curve, slope equals 34-hour half-life.

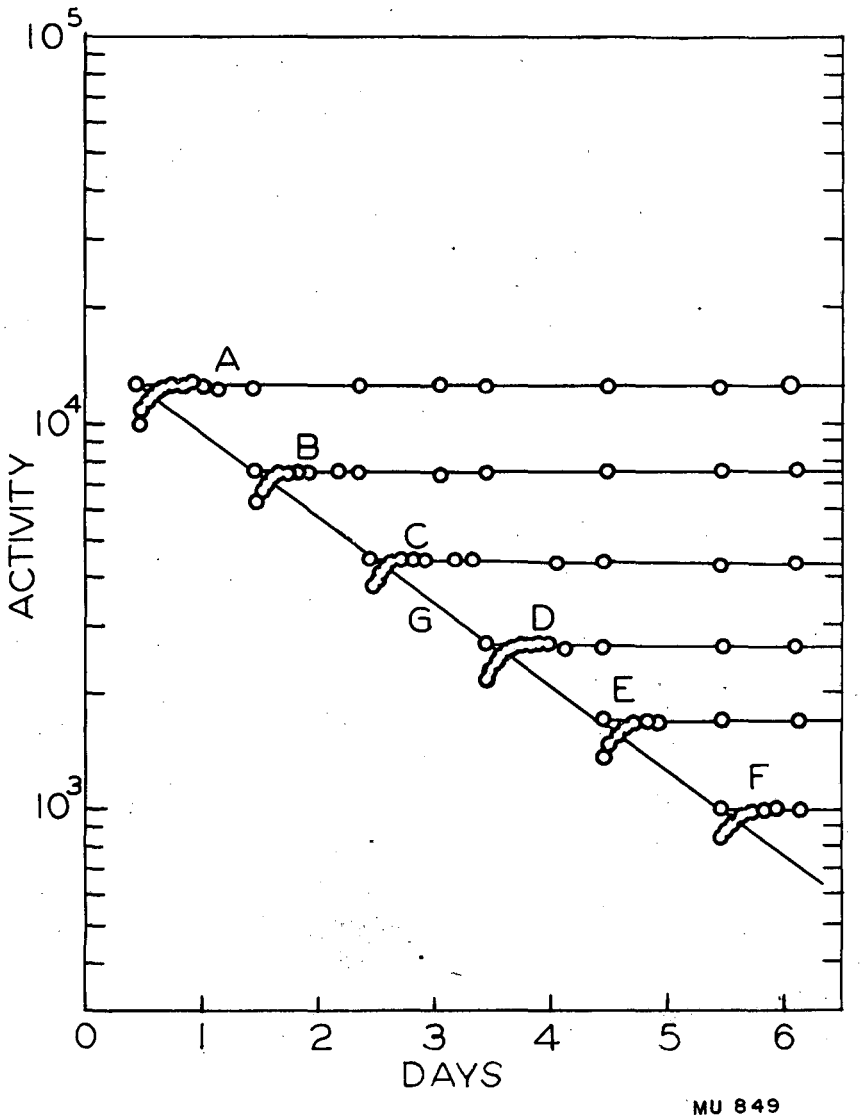
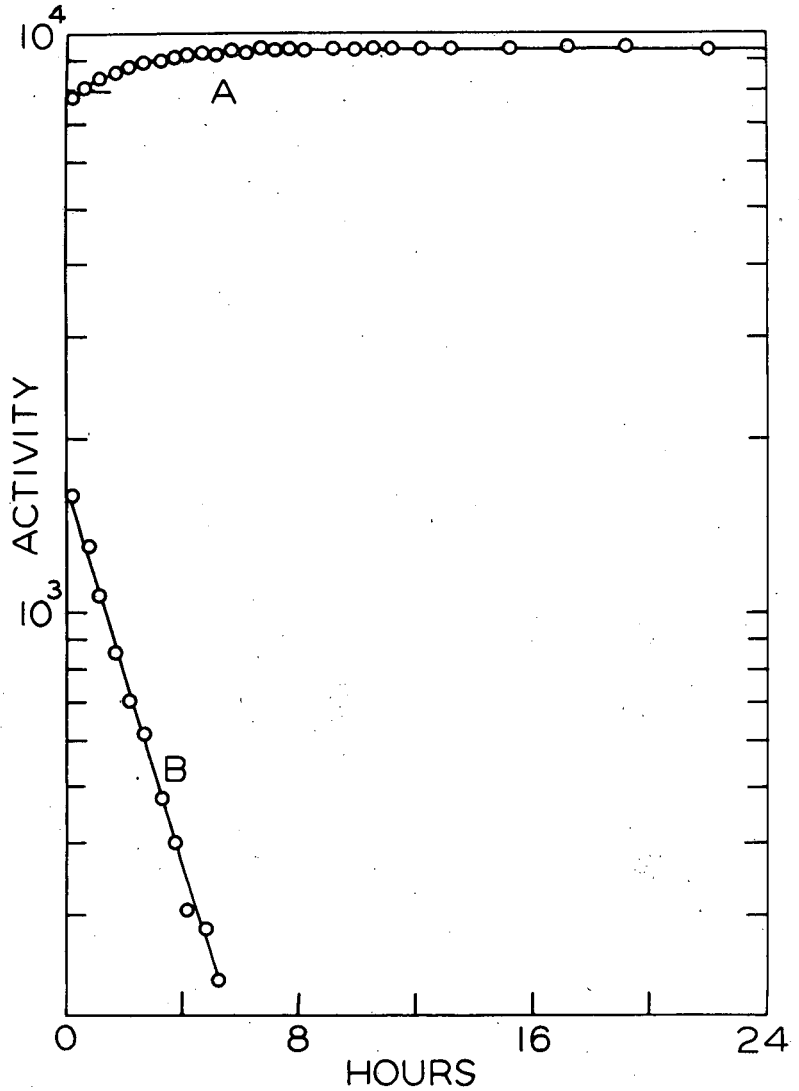

10^5
10^4
10^3
ACTIVITY
A
B
C
G
D
E
F
0
1
2
3
4
5
6
DAYS
MU 849

Fig. 3

Rb^{83} Sample Showing Growth of Kr^{83}

A. Gross decay showing growth

B. Negative of Kr^{83} growth curve. Slope shows 111-minute half-life for Kr^{83}.



MU 850

Fig. 4

Gross Decay of Rb^{83} After Initial Growth Period Is Over

Uncorrected for Chemical Yield.

A. and B. Samples contaminated by Sr^{82} and Sr^{83}. Slope equals 80 days.

C, D, E, and F. Samples uncontaminated. Slopes equal 95 to 105 days.

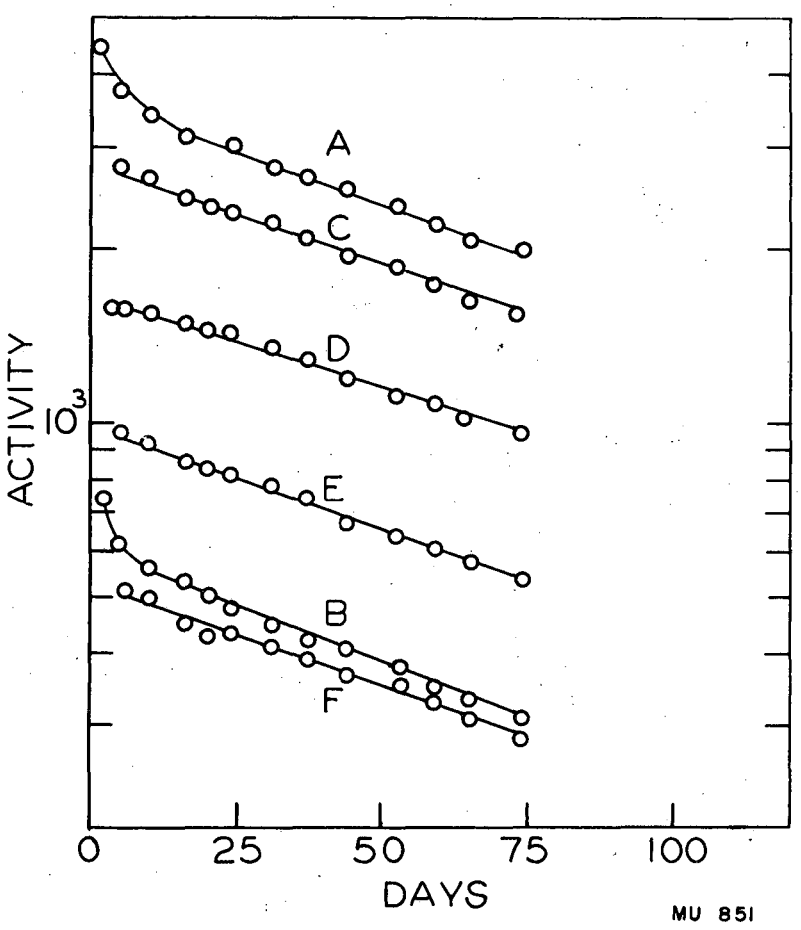

A
C
D
E
B
F
ACTIVITY
10^3
0
25
50
75
100
DAYS
MU 851

Fig. 5

Decay of Strontium Activities Produced at or Above 60 Mev Energy

A. Extrapolation of 25.5-day Sr^{82} to beginning of experiment.

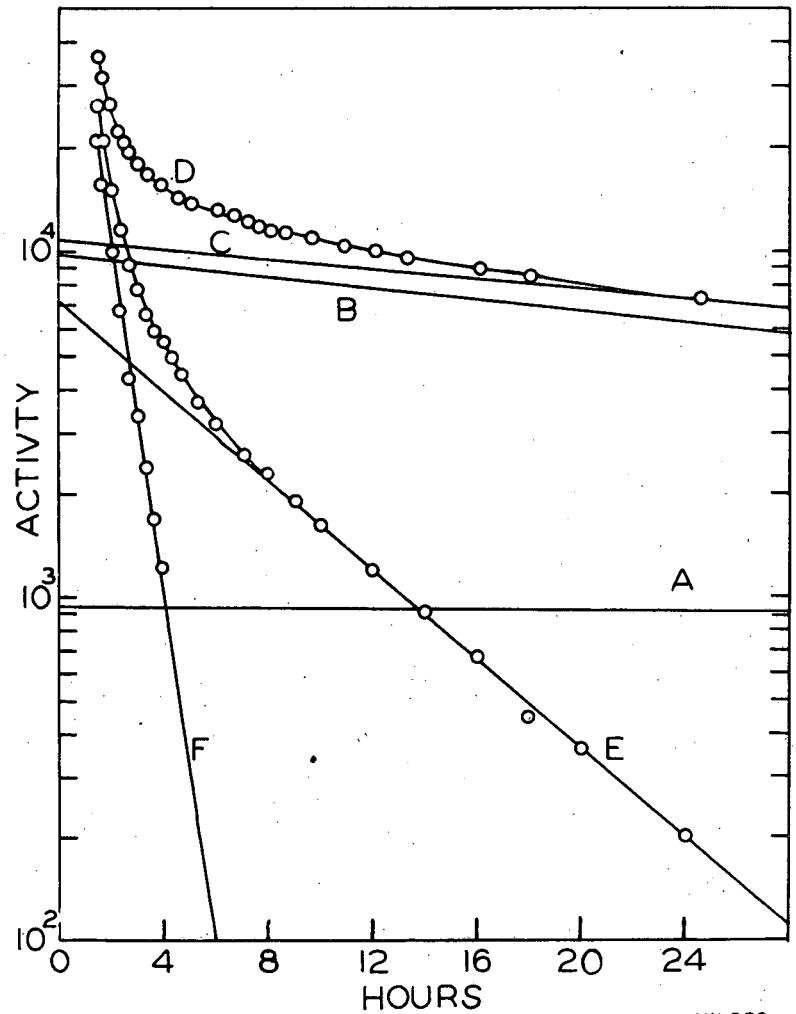
B. Extrapolation of 38-hour Sr^{83} to beginning of experiment.

C. Combination of curve B and A.

D. Gross decay curve.

E. 4.8-hour slope due to Rb^{81}. Curve D minus Curve C.

F. 29-minute slope due to Sr^{81}.



MU 852

167281

Fig. 6

Yield of Rubidium Activity Vs. Time Showing Slope of 22-minutes.

Corrected for Chemical Yield and Presence of Small Amount of Rb^{83}

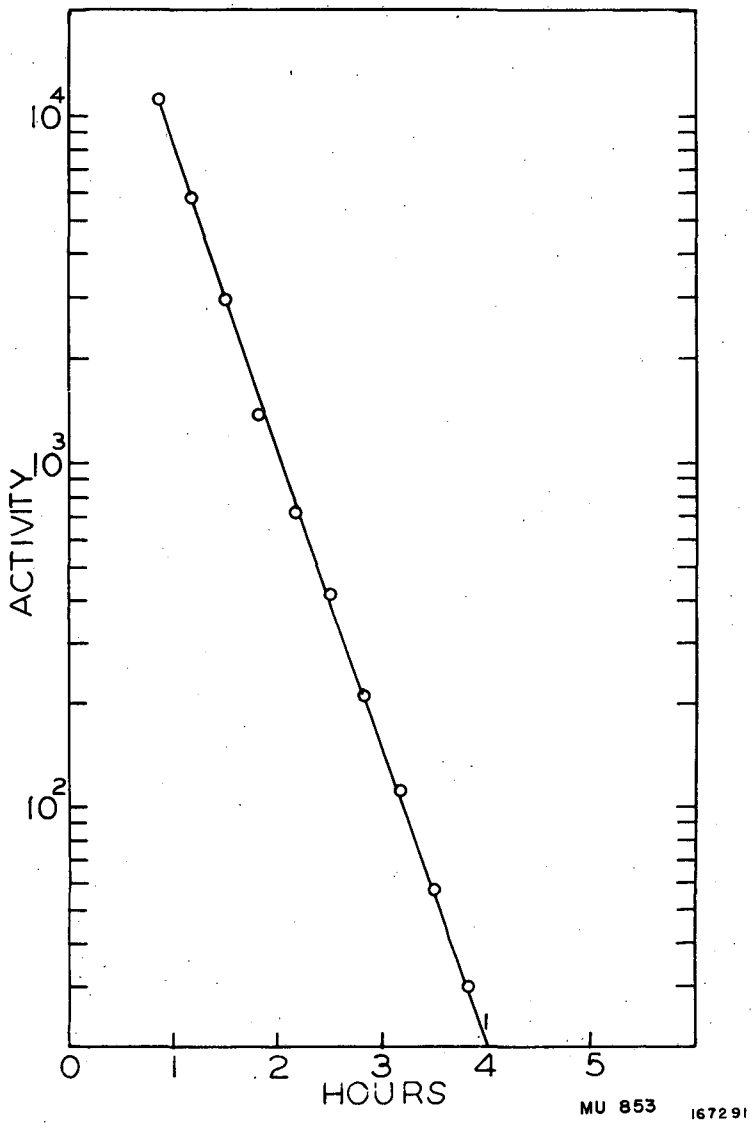

ACTIVITY
10^4
10^3
10^2
0
1
2
3
4
5
HOURS
MU 853
167291

Fig. 7

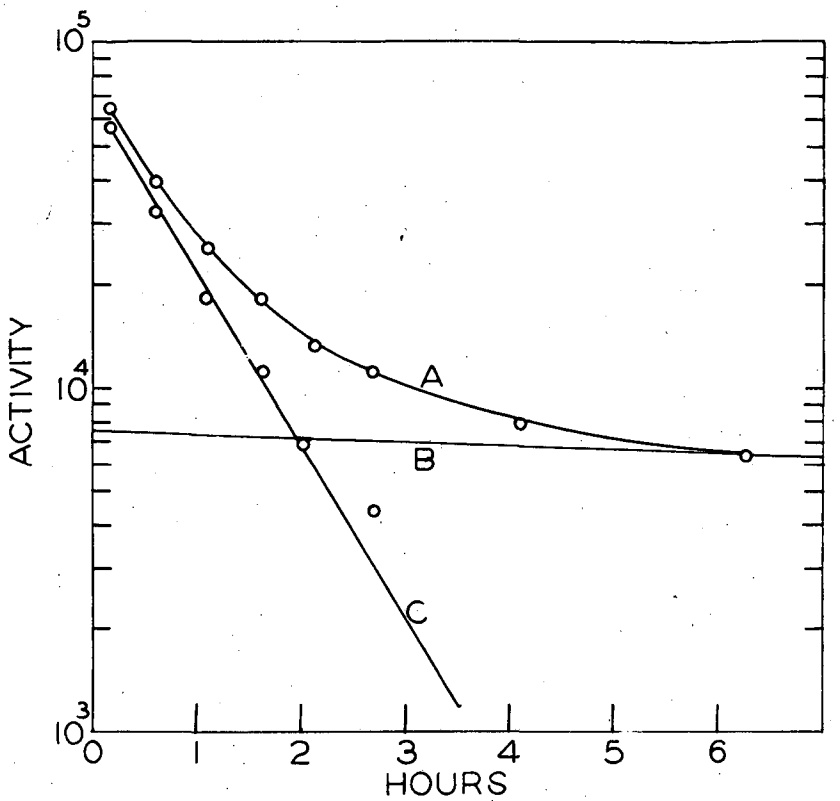
Initial Counting Rate Vs. Time for Strontium Samples.

Corrected for Chemical Yield.

A. Gross decay curve.

B. 38-hour Sr^{83}

C. 31-minute Sr^{81}

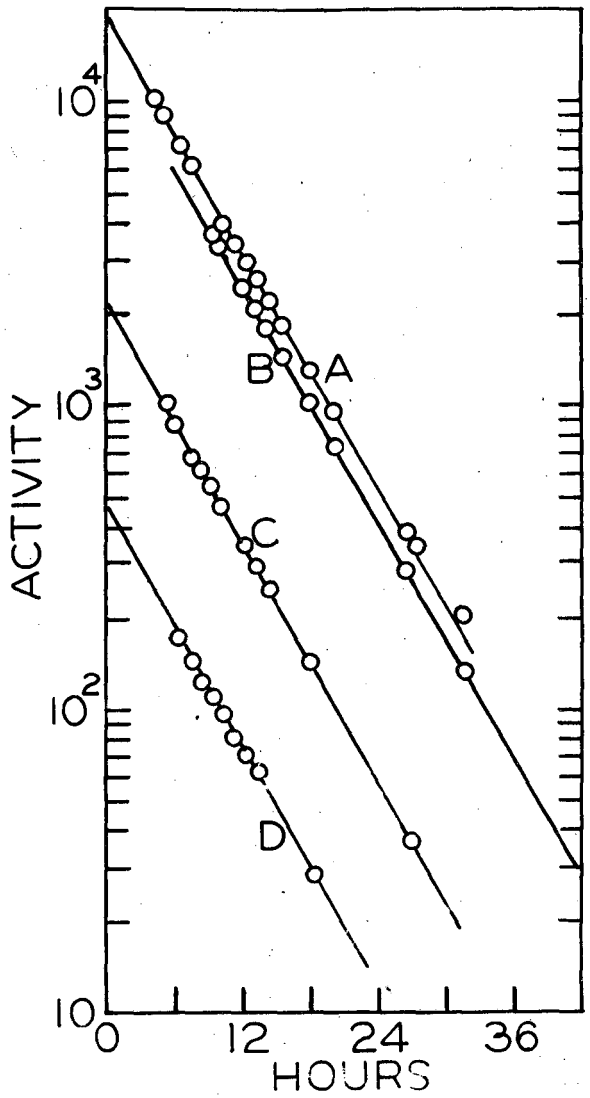


MU 854

167301

Fig. 8

Rubidium Samples Separated From Sr^{81}. Slope Shows 4.5 to 4.9 Hours.



MU 855

Fig. 9

Aluminum and Aluminum Plus Beryllium Absorption Measurements on Rb^{81}

A. Aluminum absorption (no beryllium present)

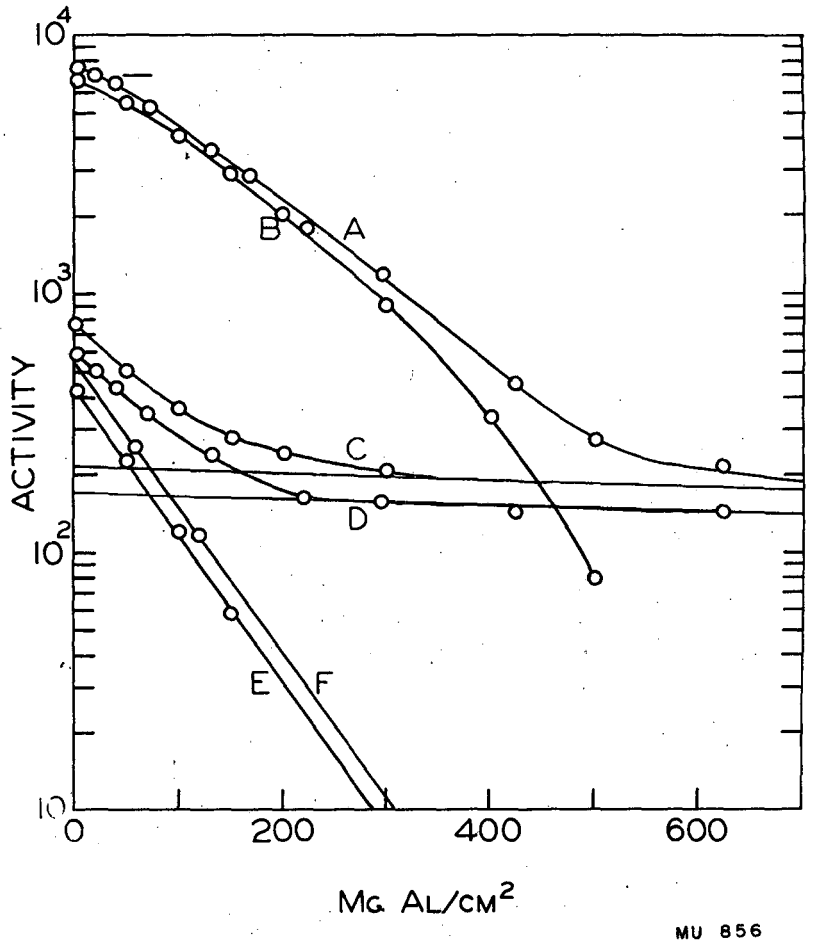
B. Absorption of particles showing range to be equivalent to approximately 1.1 Mev energy.

C. Electromagnetic Component Corrected for Beryllium.

D. Aluminum absorption with 800 mg Be/cm^2.

E. Krypton K x-rays half thickness equals 70 mg Al/cm^2

F. Krypton K x-rays corrected for effect of beryllium.



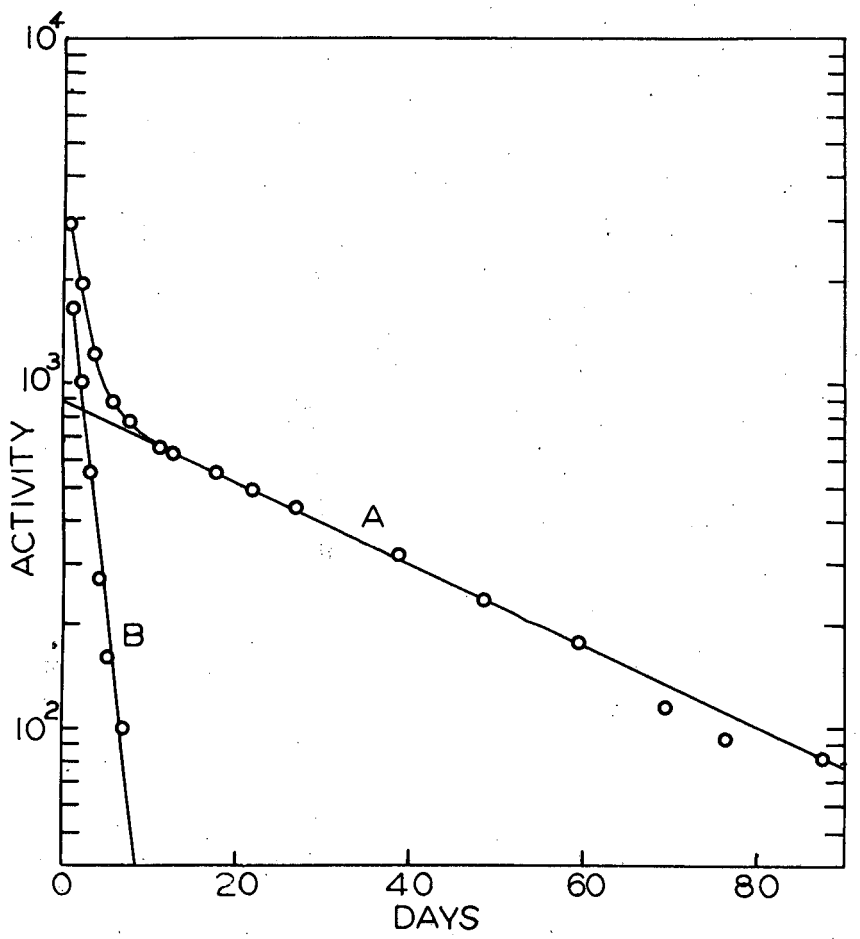
167321

Fig. 10

Decay of Strontium Activities Produced at or Above 40 Mev Energy

A. 25.5-day Sr^{82}

B. 38-hour Sr^{83}



MU 857

Fig. 11

Kurie Plot of Positron Spectra of Sr^{82} and Sr^{83}

A. Kurie plot of both spectra combined.

B. Extrapolation of data pertaining to Sr^{82} only.

C. Curve C equals the square root of the difference of the squares of curves A and B. The end point energy is 1.15 ± 0.05 Mev. Curve C corrected for 38 hour decay as 12 hrs. were required to take all data.

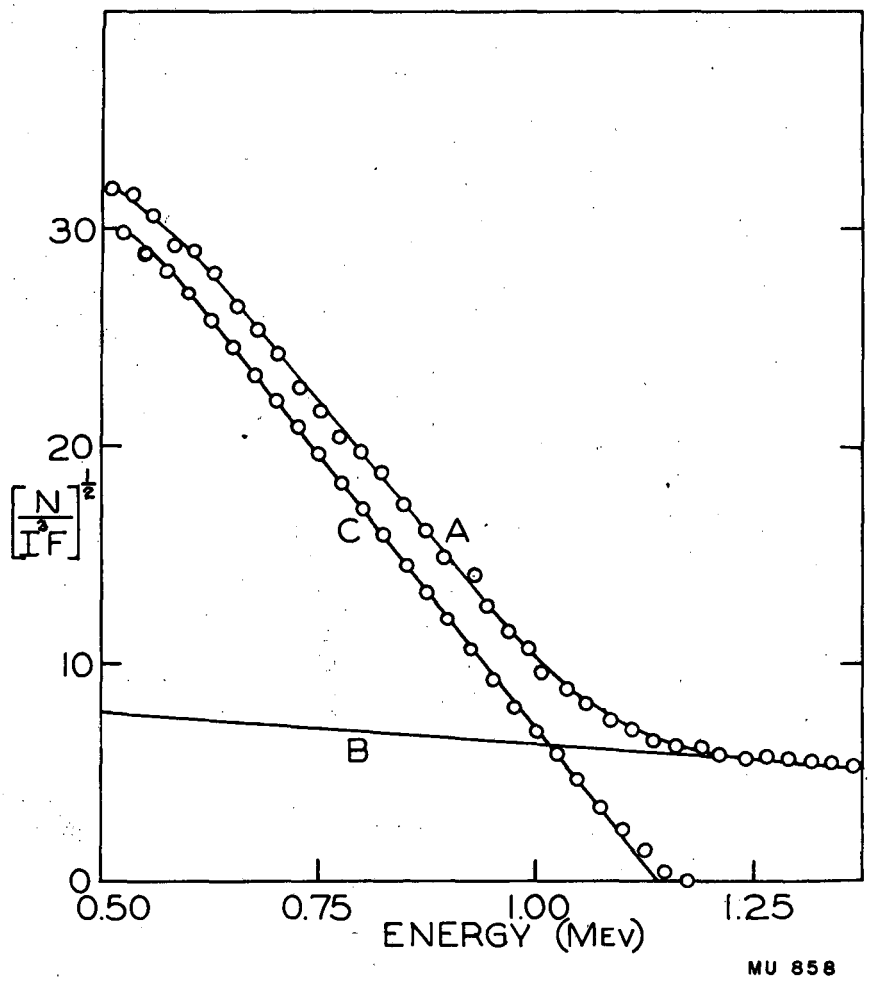

30
20
10
0
$\left[\frac{N}{I^2F}\right]^{\frac{1}{2}}$
A
B
C
0.50
0.75
1.00
1.25
ENERGY (MEV)
MU 858

Fig. 12

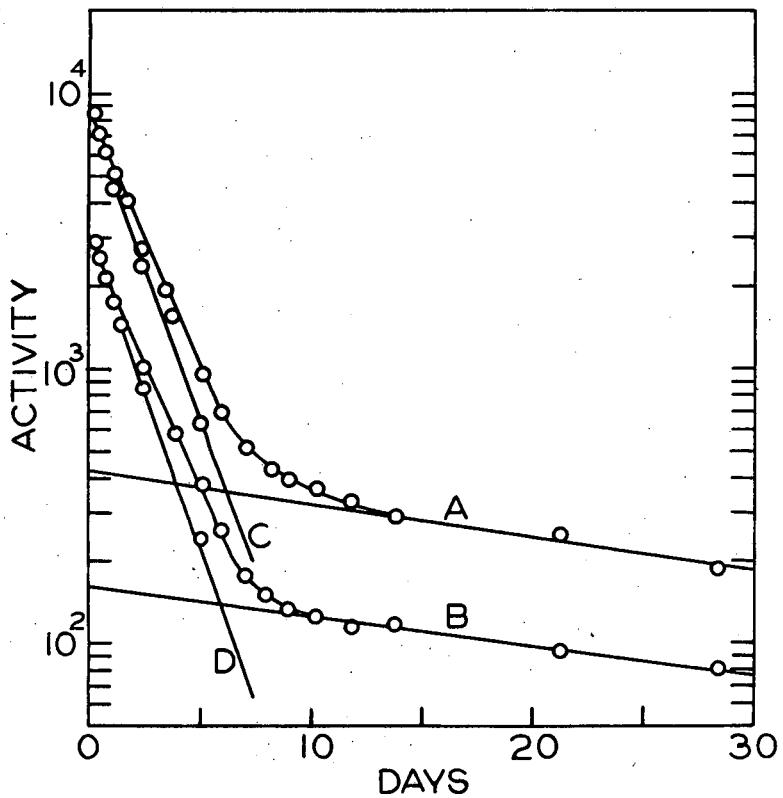
Decay of Strontium Activities Through

1876 mg Be/cm^2 and 1876 mg Be/cm^2 plus 60.41 mg Ag/cm^2.

Note percentage difference remains the same throughout the experiment.

A. Decay through beryllium only. Curve resolves into 25.5 day Sr^{82} and 38-hour Sr^{83} curve C. Activity counted is rubidium K x-rays and gamma-rays.

B. Decay through beryllium and silver. Curve also resolves into 25.5-day Sr^{82} and 38-hour Sr^{83} curve D. Activity counted is rubidium K x-rays(in reduced intensity) plus gamma rays.



MU 859

167351

Fig. 13

Kurie Plot of Sr^{82}. End Point is 3.15 $\pm$ 0.03 Mev energy. Initial part of curve represents activity due to Sr^{83}. This activity decays with 38 hr. half-life, leaving the 3.15 Mev positron which decays with 25.5 day half-life.

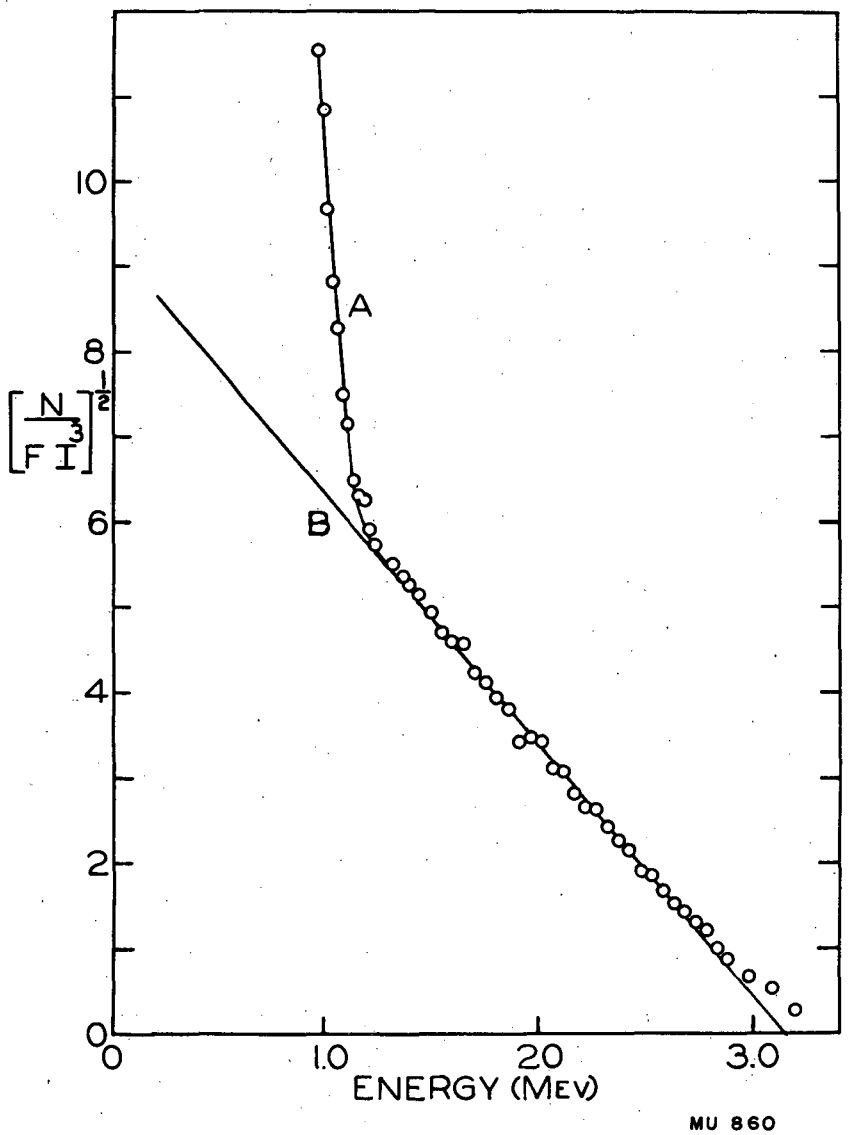
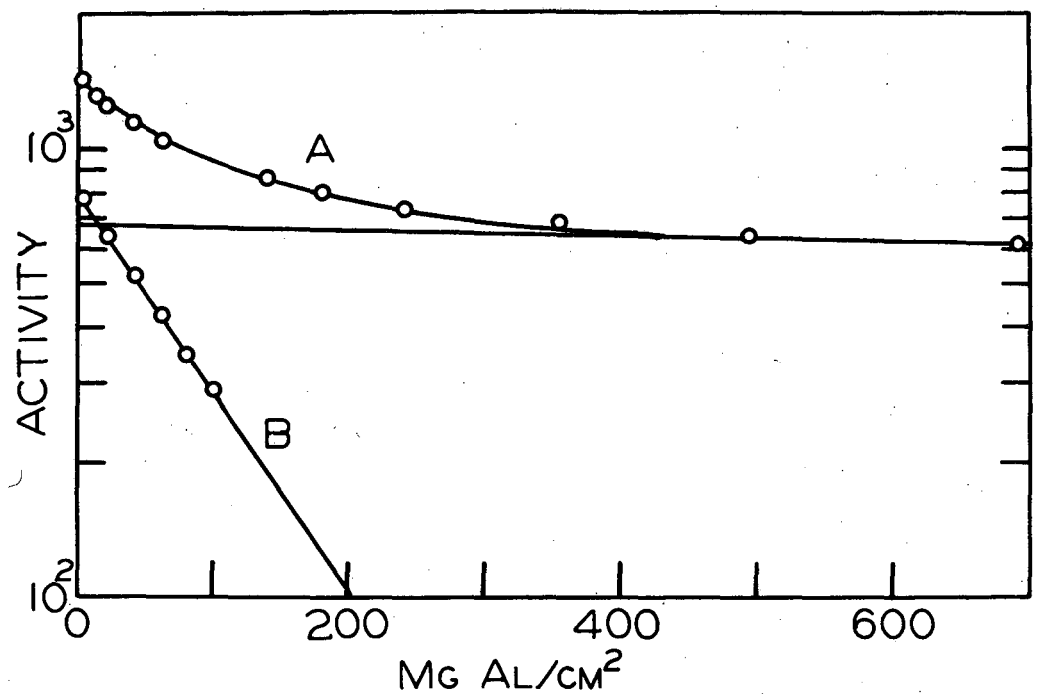

10
8
6
4
2
0
$\left[\frac{N}{F I^3}\right]^{\frac{1}{2}}$
A
B
0
1.0
2.0
3.0
ENERGY (MEV)
MU 860

Fig. 14

Absorption of Sr^{82} Activity Through Aluminum Plus 1870 mg Be/cm^2

A. Total electromagnetic component of Sr^{82}

B. Rb K x-rays half-thickness equals about 70 mg Al/cm^2.



MU 861

Fig. 15

Absorption of Sr^{82} Activity in Lead

A. 0.15 Mev gamma-ray (half-thickness is 208 mg Pb/cm^2)

B. 0.95 Mev gamma-ray (half-thickness is 7.3 gm Pb/cm^2).

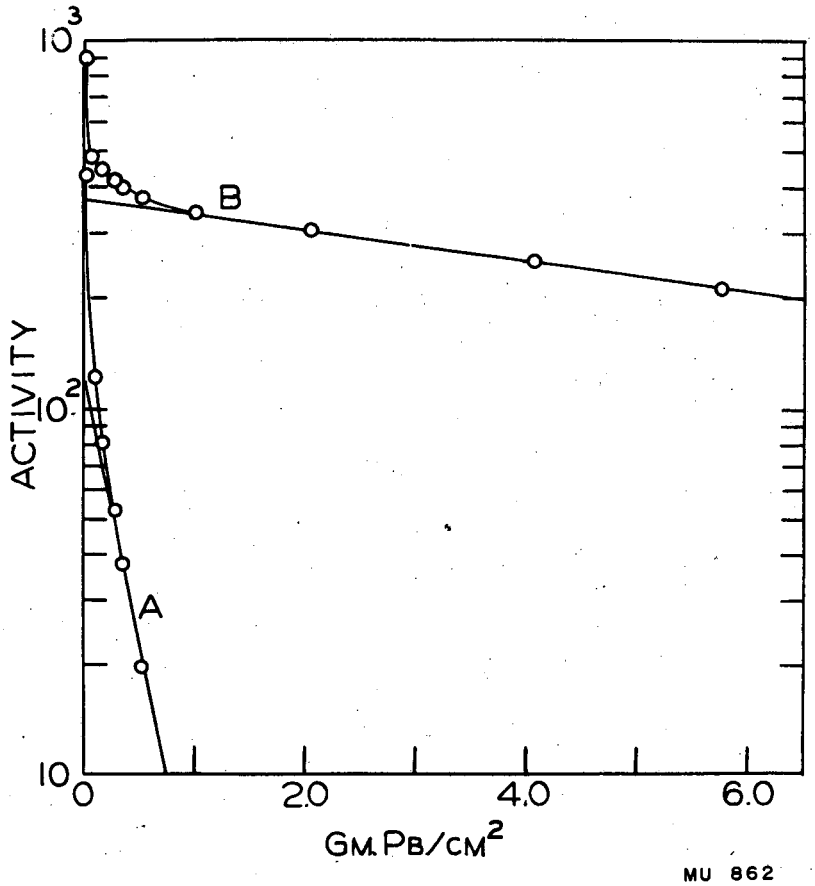

10^3
10^2
10
ACTIVITY
B
A
0
2.0
4.0
6.0
GM. PB/CM^2
MU 862

Fig. 16

Aluminum and Aluminum Plus Beryllium Absorption Measurements On Rb^{83}

A. Conversion electrons of 0.15 and 0.45 Mev energy.

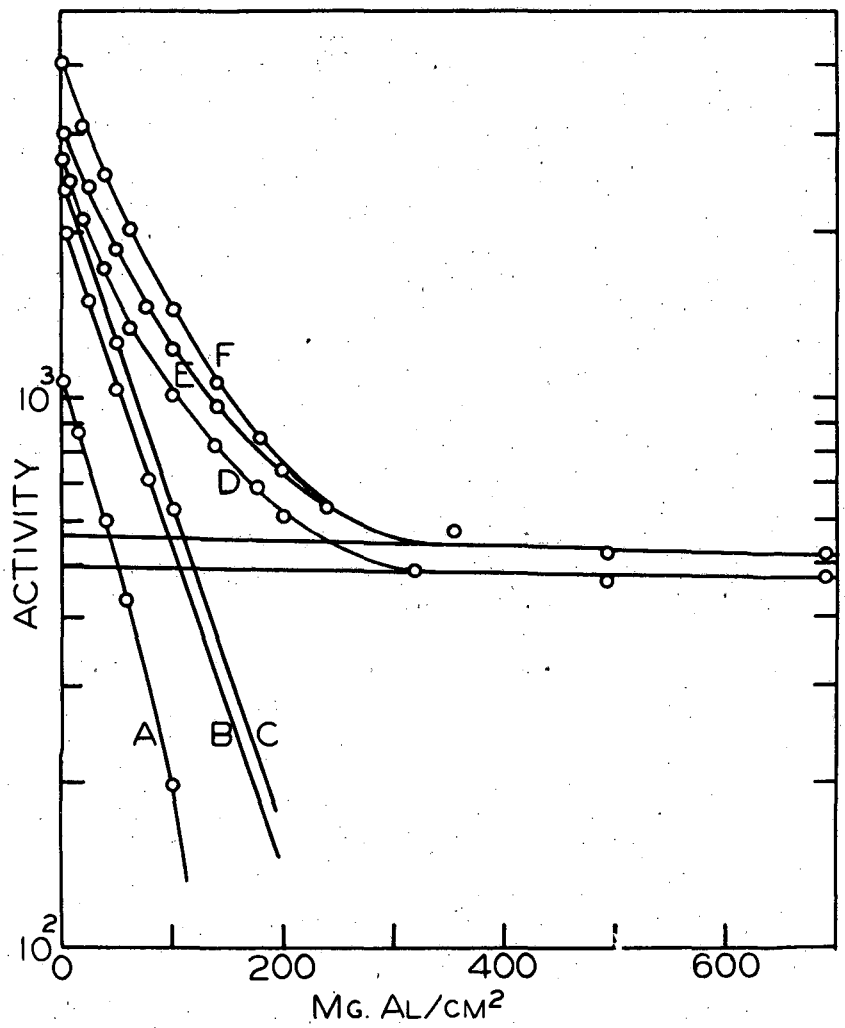
B. Krypton K x-rays

C. Krypton K x-rays corrected for the effect of the beryllium.

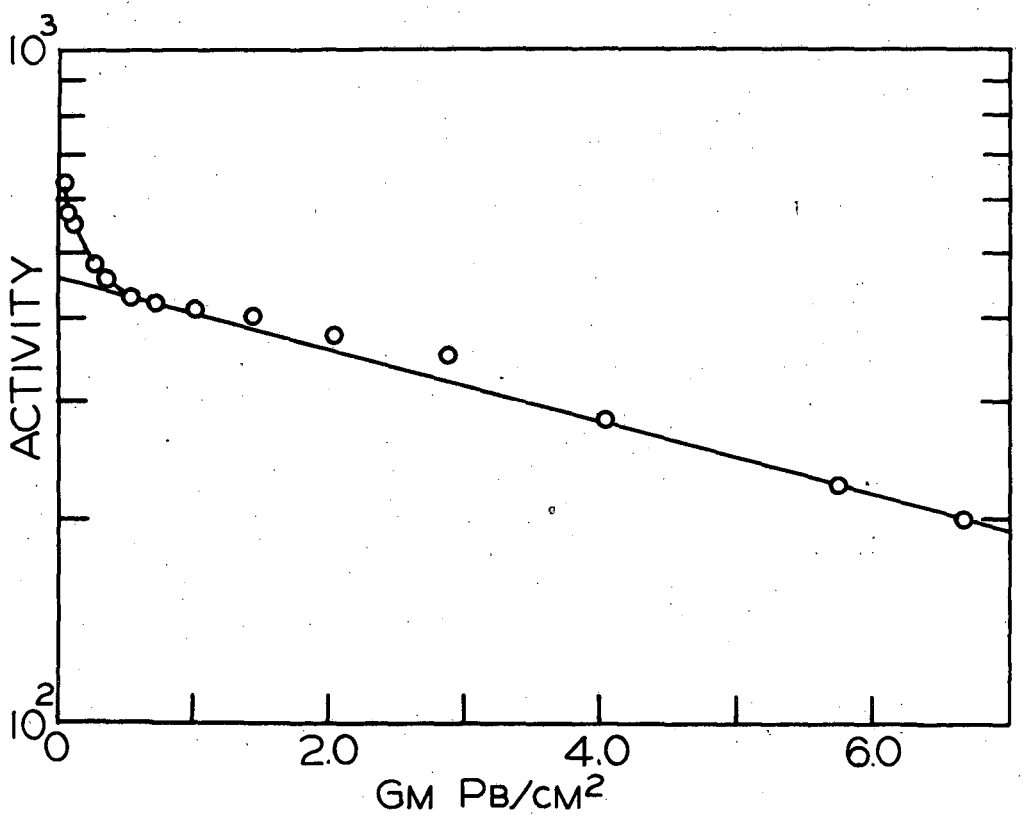
D. Absorption in aluminum plus 188 mg Be/cm^2. Electromagnetic component of Rb^{83}.

E. Electromagnetic component corrected for the effect of the beryllium.

F. Absorption in aluminum only.



MU 863



MU 864

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