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COUNTING EFFICIENCY OF BISMUTH 205 AND BRANCHING RATIO OF POLONIUM 205

Kenneth L. Hall

August 29, 1951

Berkeley, California

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ABSTRACT

The counting efficiency of Bi^{205} has been determined by relating the geiger counting rate to the alpha counting rate of its At^{209} parent. This datum was then used to determine the branching ratio of Po^{205}. By counting the geiger activity of the separated Bi^{205} daughter it is possible to calculate the electron capture disintegration rate of Po^{205}. This can then be related to the alpha activity of Po^{205} to give the branching ratio. The corresponding alpha half-life of 0.23 ± 0.05 year is then discussed in terms of a hindered decay or a deviation from the normal nuclear radius.

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I. INTRODUCTION

In the study of the correlation of alpha decay properties, abrupt changes occur in the regularities in the cases of nuclei with less than 126 neutrons.[1] In this region the decay energy, when plotted against mass number, reaches a minimum before resuming the initial trend. This irregularity is attributed to abnormally strong nuclear binding associated with 126 neutrons. It was observed that the plot of the decay energy versus half-life for the even-even nuclei define a line which agrees well with a line calculated by means of the well known Gamow formula, giving the decay constant as a function of nuclear radius, decay energy, atomic number, and mass number. Assuming that the normal nuclear radius can be expressed by $r_o = 1.48 \times 10^{-13} A^{1/3}$ cm, where A is the mass number of the daughter isotope, it has been pointed out[2] that the majority of even-even nuclei conform to this expression within 1 percent. Exceptions which appear to be real are polonium isotopes, which decay into a stable configuration of 82 protons. There appears to be a progressive shrinkage in the normal nuclear radius from Po^{218} to Po^{208}, i.e., over and above that due to a smaller number of neutrons. This is explained in terms of the effect of the 82 proton shell on the daughter nucleus. Between Po^{212}

and Po^{210} this excess shrinkage is quite large and is interpreted as the effect of the 126 neutron shell. The radius of Po^{208} apparently remains shrunken. It is of interest, therefore, to determine the behavior of the nuclear radius in the lighter polonium isotopes.

Po^{205} was chosen for this purpose, principally because the experiment is feasible. It is possible to determine the partial alpha half-life for this isotope and to compare the value so obtained with the calculated value. The difference can then be interpreted either as a deviation from the normal nuclear radius, or as a hindered decay (due to an odd type nucleus), or to a combination of both.

In order to determine the branching ratio of Po^{205} and hence the half-life, it is necessary to know the absolute disintegration rates of both the alpha and the electron capture modes of decay for this isotope. All alpha particles have a counting efficiency (number of counts per disintegration) of 0.52 when counted in an alpha chamber consisting of an argon ionization chamber connected to a scaling unit. On the other hand, the electron capture counting efficiency varies with the isotope in question and particular counting arrangement. Now it is possible to measure the counting efficiency of Bi^{205} in a separate experiment and then to use this value to calculate the disintegration rate of Po^{205} by counting its Bi^{205} daughter. In order to measure this counting efficiency the number of Bi^{205} counts per minute was related to the number of alpha disintegrations per minute of the parent At^{209}.

II. EXPERIMENTAL

A. Counting Efficiency of Bi^{205}

Astatine was prepared by bombarding bismuth metal with 70 Mev alpha particles in the 184-inch cyclotron. At this energy, only At^{209}, At^{210}, and At^{211} are produced. The target was placed in the "astatine boiler" (a stainless steel crucible in which is inserted a water cooled metal finger with a platinum disk attached to collect the astatine). The bismuth was heated just above its melting point (as measured by a thermocouple) and the astatine was collected on the disk. Samples prepared in this way were counted in the 48 channel alpha pulse analyzer to determine the ratio of At^{209} to At^{211} alpha activities. More intense samples were prepared for following the alpha decay to the 138 day alpha-emitting Po^{210} which grows from At^{210}. It is then possible to calculate the ratio of At^{209} to At^{210}. For following the electron capture activity to the 14.5 day Bi^{205} (daughter of At^{209}), extremely intense samples were prepared. The number of alpha disintegrations per minute of At^{209} was then calculated by counting the Po^{210}, and the Bi^{205} electron capture activity from the same sample was measured. Thus the counting efficiency of Bi^{205} can be calculated.

B. Branching Ratio of Po^{205}

Polonium isotopes from Po^{205} to Po^{210} were produced by bombarding Bi_2O_3 with 50 Mev protons in the 184-inch cyclotron. The polonium was separated chemically from the target material by a method based upon procedure No. 84-2 of Meinke's compilation.[3] This involved dissolving the

Bi_2O_3 target in hydrochloric acid, carrying down the astatine and polonium on tellurium, solution of the tellurium, and extraction of the polonium with a 20 percent solution of tributyl phosphate in dibutyl ether. Aliquots of this polonium solution were evaporated onto platinum disks for following the alpha decay.

After a short growth period, the Bi^{205} electron capture daughter of Po^{205} was separated chemically from aliquots of the polonium solution. The procedure used here was to wash the bismuth out of the organic phase with dilute hydrochloric acid.[4] The decay of samples made in this way was followed in the Geiger counters.

In order to determine the chemical yield, an aliquot (about 10,000 c/m) of purified RaE tracer was added to the organic phase before the bismuth was separated. Then the amount of RaE that was extracted along with the other bismuth activities was taken as a measure of the chemical yield.

This tracer bismuth was separated from its parent, RaD, by the following procedure. A few milligrams of $Pb(NO_3)_2$ were added to the RaD in hydrochloric acid solution. Lead sulfate was precipitated with a slight excess of dilute sulfuric acid, thus carrying down the RaD. Whenever tracer bismuth was needed, the lead sulfate was dissolved in a minimum amount of concentrated sulfuric acid, reprecipitated by diluting with water and the supernatant was transferred to a clean centrifuge cone. To this solution of RaE and RaF a few drops of concentrated hydrochloric acid was added to make approximately 6 N Cl^-. The RaF was then extracted away with the tributyl phosphate solution, leaving purified RaE tracer in solution.

III. CALCULATION OF RESULTS

A portion of the isotope table in the region of interest is shown below:

			At^{209} 5.5h 95%EC 5%α(5.65)	At^{210} 8.3h EC	At^{211} 7.5h 60%EC 40%α(5.89)	At^{212} 0.253s α
Po^{205} 1.5h EC α(5.21)	Po^{206} 9d EC α(5.21)	Po^{207} 5.7h EC α(5.10)	Po^{208} 2.93 y α(5.10)	Po^{209} ~200y EC α(4.86)	Po^{210} 138d α(5.30)	Po^{211} $5x10^{-3}$s α(7.43)
	Bi^{205} 14.5d EC	Bi^{206} 6.4d EC	Bi^{207} 10-100y EC	Bi^{208} long EC	Bi^{209} stable	

The notation used in the following equations is summarized:

A = counts per minute of At^{209} alpha activity

B = counts per minute of At^{211} alpha activity

C = counts per minute of Po^{205} alpha activity

D = counts per minute of Po^{210} alpha activity

E = counts per minute of Bi^{205} geiger activity

R = disintegrations per minute of At^{209} alpha activity

S = disintegrations per minute of At^{210} geiger activity

T = disintegrations per minute of Po^{205} geiger activity

r = decay constant of At^{209}

m = decay constant of At^{210}

n = decay constant of At^{211}

o = decay constant of Po^{205}

p = decay constant of Po^{210}

q = decay constant of Bi^{205}

H = ratio of alpha disintegration rate of At^{209} to disintegration rate of At^{211}

I = ratio of alpha disintegration rate of At^{209} to disintegration rate of At^{210}

J = difference between total alpha activity and that due to Po^{210}

K = total counts per minute of alpha activities

V = chemical yield

W = growth factor

X = branching ratio

Y = counting efficiency of Bi^{205}

t_o = time of separation from target

t_m = time of separation of daughter activity

t_1 = time of counting alpha activity of At^{209}

t_2 = time of counting geiger activity of Bi^{205}

t_3 = time of counting alpha activity of Po^{210}

t_4 = time of counting alpha activity of Po^{205}

A. Counting Efficiency of Bi^{205}

The counting efficiency Y (counts per disintegration) of Bi^{205} can be expressed as:

$$Y = \frac{E(t_2)}{R(t_o)} \cdot \frac{q - r}{q} \cdot \frac{1}{e^{-rt_2} - e^{-qt_2}} \cdot \qquad (1)$$

$E(t_2)$ can be measured experimentally by following the geiger decay of the extremely intense samples (about 1,000,000 geiger counts per minute) until pure Bi^{205} is obtained.

To find $R(t_o)$ it is first necessary to determine the ratio of the alpha disintegration rate of At^{209} to the disintegration rate of At^{211}, H. This is done by counting samples of approximately 1000 alpha counts per minute in the alpha pulse analyzer. The ratio of the total number of counts under the At^{209} peak (5.65 Mev) to that under the At^{211} peak (5.89 Mev) when multiplied by 2/5 gives H. (The factor of 2/5 is to take into account the 40 percent electron capture branching of At^{211} to give the 5×10^{-3} second Po^{211} alpha emitting daughter.) This is multiplied by the factor $e^{(n - r)\Delta t}$, to correct to various times.

The next step in finding $R(t_o)$ is to find I, the ratio of the alpha disintegration rate of At^{209} to the disintegration rate of At^{210}, using the ratio H just determined. For this purpose the alpha decay of the more intense samples of about 100,000 alpha counts per minute was followed until pure Po^{210} was obtained; thus, $D(t_3)$ was found. At some time, t_1, before the astatine activities had decayed away, the difference between the total alpha counting rate, $K(t_1)$, and that due to Po^{210}, $D(t_1)$ (the rate of pure Po^{210}, $D(t_3)$, corrected to time t_1) gives the sum of the alpha counts from At^{209} and At^{211}. From this difference, J, and the ratio H, the alpha activity of At^{209}, A, can be determined as

$$A(t_1) = \frac{H(t_1) \cdot J(t_1)}{1 + H(t_1)} . \qquad (2)$$

The desired ratio, I, then becomes

$$I(t_1) = \frac{A(t_1)}{0.52} \cdot \frac{1}{S(t_1)} \tag{3}$$

where $A(t_1)$ is given by equation (2), and 0.52 is the counting efficiency of these alpha particles. Furthermore $1/S(t_o)$ is given by

$$\frac{1}{S(t_o)} = \frac{P}{P - m} \cdot \frac{0.52}{D(t_1)} \cdot \left[e^{-mt_1} - e^{-pt_1}\right] \tag{4}$$

Now correcting $A(t_1)$ to time t_o by multiplying equation (2) by the factor e^{rt}, and substituting equation (4) and (2) into equation (3), the ratio $I(t_o)$ becomes

$$I(t_o) = \frac{H(t_1) \cdot J(t_1)}{1 + H(t_1)} \cdot e^{rt_1} \cdot \frac{P}{P - m} \cdot \frac{e^{-mt_1} - e^{-pt_1}}{D(t_1)} \tag{5}$$

This is multiplied by the factor $e^{(m - r)\Delta t}$ to correct to various times. Finally the disintegration rate $R(t_o)$ becomes

$$R(t_o) = I(t_o) \cdot S(t_o) = I(t_o) \cdot \frac{p - m}{0.52\ p} \cdot \frac{D(t_3)}{e^{-mt_3} - e^{-pt_3}}, \tag{6}$$

where t_o now refers to the very intense samples. The counting efficiency, Y, is then found by substituting equation (6) into equation (1):

$$Y = \frac{E(t_2)}{e^{-rt_2} - e^{-qt_2}} \cdot \frac{q - r}{q} \cdot \frac{0.52\ p}{p - m} \cdot \frac{e^{-mt_3} - e^{-pt_3}}{D(t_3) \cdot I(t_o)}. \tag{7}$$

The data are summarized in Tables I, II, and III, which give the ratio H(0), the ratio I(0), and the counting efficiency Y, respectively. The counting efficiency corrected for the geometry of the counting

arrangement is given in the last column of Table III. The Geiger tubes used were of the end window type filled with 33 ± 5 cm of argon containing less than 1 percent chlorine ("Amperex"). The thickness of the mica windows was 3.5 mg/cm^2, for tubes number A2104B and 2190B respectively. Tube number A2104B and scaler E were used in run 1, and tube number 2190B and scaler 12 were used in run 2. These tubes were mounted in the standard lucite sample holders, the whole assembly being placed in a lead housing. The platinum plates were supported on the shelves by 1 mm thick cardboard. The time t = 0 was taken as 10:35 on November 15, 1950 for run 1, and as 8:30 on July 13, 1951 for run 2.

Table I

Run	Sample	A (counts)	B (counts)	Hours after t = 0	H(0)
1	At-1	340	1625	12.40	0.1269
		330	1713	12.67	0.1180
		340	2021	13.12	0.1046
	At-5	407	2941	14.92	0.0915
		419	2816	15.17	0.0992
		413	2591	15.50	0.1074
	At-8	272	5589	51.58	0.1117
				Average	0.112 ± 0.012
2	At-9	1316	6633	6.27	0.0980
		1176	8160	16.52	0.1004
	At-11	1901	14073	15.12	0.0900
				Average(weighted)	0.095 ± 0.005

Table II

Run	Sample	$D(t_3)$ (average)	t_3 (hrs)	$K(t_1)$	t_1 (hrs)	I(0)
1	At-1	6.91	464.3	593	12.1	0.0617
	At-2	1223	102.6	65,893	15.8	0.0597
				21,995	27.2	0.0554
				10,435	36.1	0.0566
				3,152	51.8	0.0511
	At-7	1620	427.5	52,892	19.4	0.0689
				32,901	24.9	0.0659
				2,158	69.6	0.0552
				1,836	88.2	0.0624
				Average		0.0597 ± 0.0090
2	At-1	119.4	239.7	40,997	0.534	0.0753
				22,064	5.90	0.0675
				7,306	17.28	0.0651
	At-6	408.3	235.3	84,022	3.97	0.0756
				38,917	12.25	0.0764
				8,786	27.52	0.0694
				Average		0.0716 ± 0.0059

Table III

Run	Sample	Shelf	$D(t_3)$	t_3 (hrs) (average)	$E(t_2)$	t_2 (hrs)	$Y \times 10^3$	Y/ geometry
1	At-6	3	18,537	459.9	19.8 ± 0.4	292.4	2.62 ± 0.40	0.05
		2			44.2 ± 0.6	316.6	6.13 ± 0.93	0.05
2	At-2	3	25,258	481.6	17.1 ± 0.2	719.1	3.02 ± 0.25	0.06
		2			39.1 ± 0.9		6.92 ± 0.59	0.06
	At-5	3	9,978	479.9	5.2 ± 0.8	717.4	2.51 ± 0.44	0.05
		2			12.2 ± 0.5		5.93 ± 0.92	0.05

B. Branching Ratio of Po^{205}

The branching ratio of Po^{205}, X, can be expressed as the ratio of partial alpha decay constant to the partial electron capture decay constant, or as the ratio of the corresponding disintegration rates:

$$X = \frac{\sigma_\alpha}{\sigma_{EC}} = \frac{C(t_o)}{0.52} \cdot \frac{1}{T(t_o)} \tag{8}$$

Now the alpha activity of Po^{205} was found by resolving the alpha decay curve of the polonium samples. Alpha-pulse analysis showed a peak at 5.2 Mev (Po^{205} and Po^{206}) and a possible small peak at 5.1 (Po^{207}). However, it was impossible to resolve the gross alpha decay curve to show a significant amount of the 5.7 hour Po^{207} in addition to the 1.5 hour Po^{205}. So theoretical decay curves were constructed for varying percentages of the 1.5 hour Po^{205} in with the 9 day Po^{206} activity at

1.67 hours after the separation of the polonium. These were compared with the experimental points and the best fit obtained was chosen to determine the activity of Po^{205}, C. Figure I shows the theoretical curves and the experimental points for sample Po-1.

The electron capture disintegration rate $T(t_o)$, was found by following the geiger decay of the separated bismuth electron capture daughter and resolving out the 14.5 day component. Figure II shows the resolution of the decay curve of sample Bi-2 into the long lived Bi^{207}, 14.5 day Bi^{205}, and 6.4 day Bi^{206}.

Equation (8) now becomes:

$$X = \frac{C(t_o)}{0.52} \cdot \frac{q}{q - o} \cdot \frac{Y}{E(t_2)VW} \left[e^{-ot_2} - e^{qt_2}\right], \tag{9}$$

or, since $o \gg q$,

$$X = \frac{C(t_o)}{0.52} \cdot \frac{q}{o} \cdot \frac{Y}{E(t_o)VW} . \tag{10}$$

The quantity $E(t_2)VW$ in equation (9) is the corrected Bi^{205} activity taking into account the chemical yield and the growth factor. The chemical yield, V, was determined by counting the alpha activity of RaF grown from the tracer RaE introduced into the polonium sample. The ratio of this alpha activity to that from an equal aliquot of pure RaE tracer was then taken as the chemical yield. The reciprocal of the chemical yield is V. The growth factor, W, is to make allowance for the fact that not all of the Po^{205} had decayed at the time t_m, that the chemical separation was made. If all the Po^{205} had decayed, the Bi^{205} activity at the time of separation, $F(t'_m)$ would be expressed as:

$$E(t'_m) = Yq \frac{T(t_o)}{q - o} \left[-e^{-qt'_m} \right] . \tag{11}$$

Actually the activity at this time is

$$E(t_m) = Yq \frac{T(t_o)}{q - o} \left[e^{-ot_m} - e^{-qt_m} \right] e^{-q(t'_m - t_m)}. \tag{12}$$

The ratio of these two numbers is W:

$$W = \frac{E(t'_m)}{E(t_m)} = \frac{1}{1 - e^{(q - o)t_m}} . \tag{13}$$

The data are summarized in tables IV and V which give the initial alpha activity of Po^{205}, $C(t_o)$, and branching ratio, X, respectively. The activities $C(t_o)$ and $E(t_2)$ are the corrected activities including the aliquot factor shown. In all cases the same counting arrangements were used as in the counting efficiency experiments.

Table IV

Sample	Aliquot	t_4 (hrs)	$C(t_4)$	$C(t_o)$
Po-1	50/1000	1.67	1831 ± 194	79,200
Po-2	50/1000	1.67	1360 ± 185	58,800
Po-3	50/1000	1.67	2250 ± 200	92,250
			Average	76,750 ± 11,970

Table V

Sample	Aliquot	Shelf	$E(t_o)$	t_m	W	$D(t_m)$	$D(t_m)$	V	X
Bi-1	200/1000	2	3250 ± 300	4.1_5	1.17_3	218	262	1.20_2	.00073
Bi-2	200/1000	2	3000 ± 400	4.7_5	1.12_7	209	262	1.25_3	.00079
Bi-3	200/1000	2	3200 ± 350	5.8_2	1.07_3	191	262	1.37_2	.00071
					Average				.00074 ± .00016

IV. CONCLUSION

The branching ratio of Po^{205} just determined corresponds to a partial alpha half-life of 0.23 ± 0.05 years. The alpha particle energy is 5.21 ± 0.10 Mev.[5] Using the Gamow formula, values of the alpha half-life were calculated for various values of the nuclear radius and alpha energy. These were plotted in Figure III.

It can be seen from the intersection of the lines representing the best values of the alpha half-life and the alpha energy that the radius is about 2 percent smaller than the normal nuclear radius. This is assuming that the decay is not hindered. An alternative explanation is that the radius is normal and that the decay is hindered by a factor of two. However, the limits of error (shown by the dotted lines) are such that possible deviations from the normal radius are + 2 percent and - 6.5 percent. Similarly, assuming a normal radius, the decay could be superallowed by a factor of 1.8 or hindered by a factor of 11.

Now the two other even-odd polonium isotopes for which the alpha half-lives have been compared with the theory are Po^{213} and Po^{215}. The half-lives of these isotopes agree with the calculated values, indicating a normal radius and no hindrance. Thus it appears reasonable in the case of Po^{205} to assume the alpha decay is not hindered and that the radius is shrunken by about 2 percent of the normal value. This amount is in line with that of the polonium isotopes 218 through 214 and can be interpreted as the effect of the 82 proton shell. The 126 neutron shell would then be without effect on the nuclear radius.

V. ACKNOWLEDGMENTS

The advice of Dr. D. H. Templeton, who directed the work, is gratefully acknowledged. This work was performed under the auspices of the U. S. Atomic Energy Commission.

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5. D. G. Karraker, A. Ghiorso, and D. H. Templeton, Phys. Rev. 83, 390 (1951).

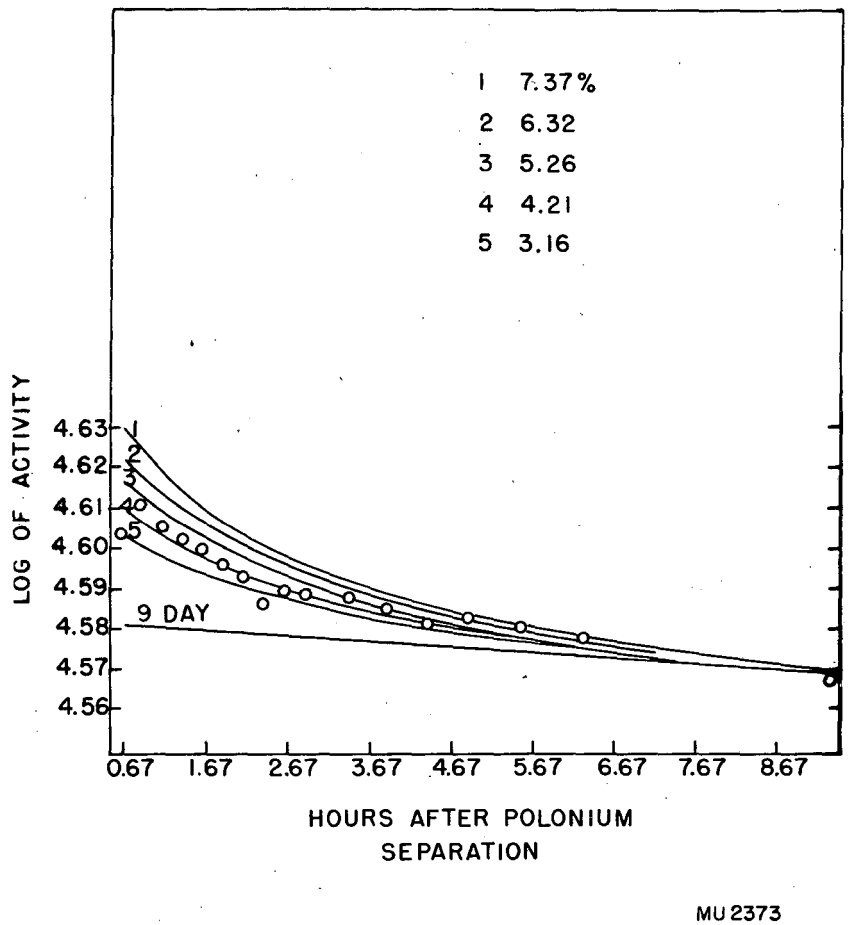


Figure I. Decay Curves of Varying Percentages of Po^{205} in with Po^{206} -- Experimental Points are for Sample Po-1.

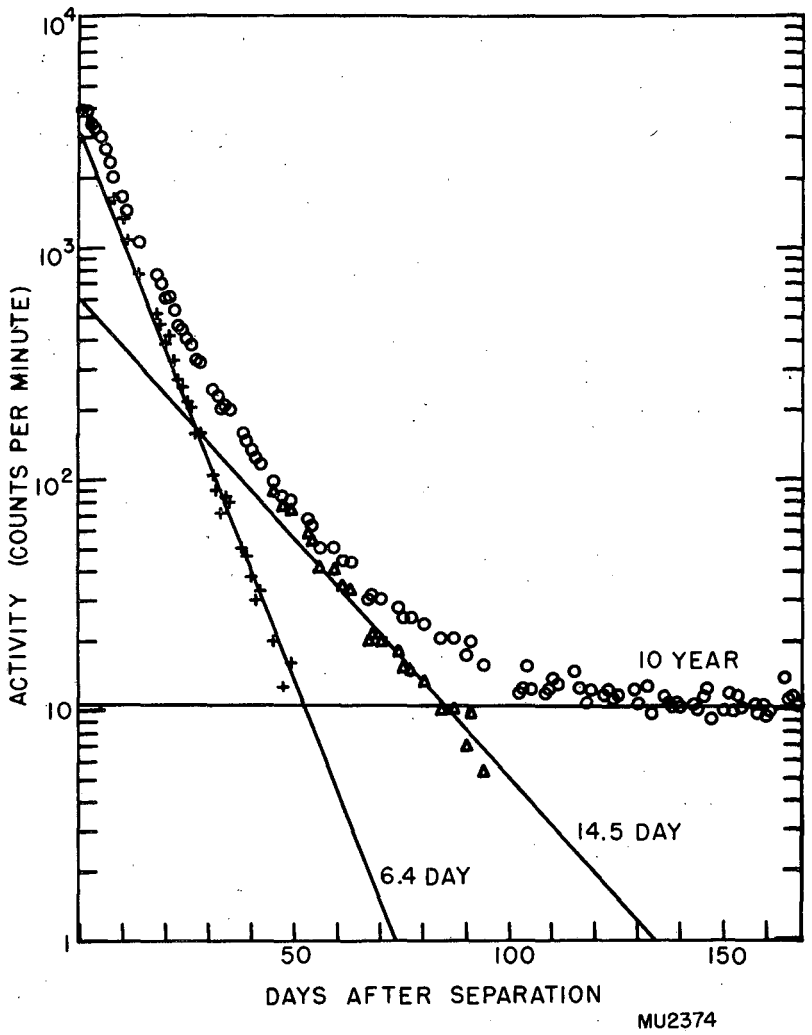
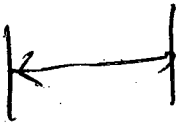
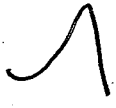
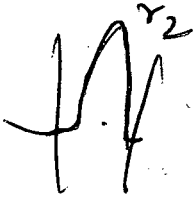


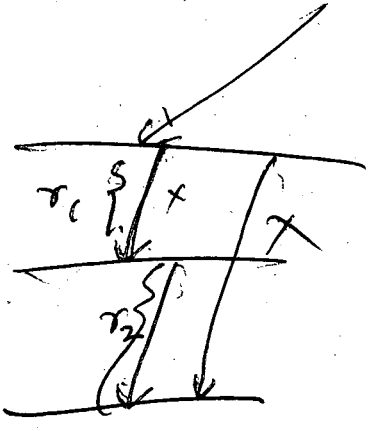
Figure II. Resolution of the Experimental Decay Curve of Sample Bi-2

r_1
r_2
x_2





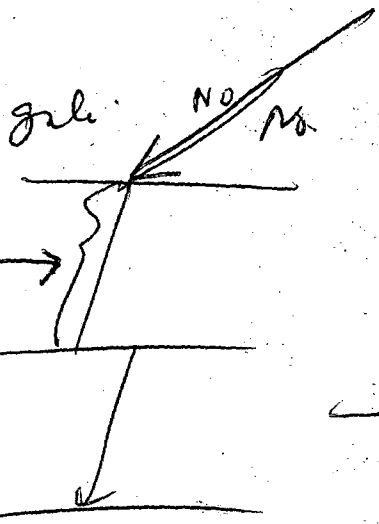

r_2

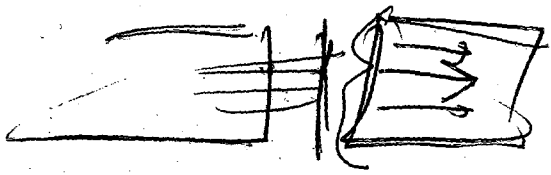

r_1
S
x
x
r_2

$$\text{Corr} = N_0 \gamma_1 \omega_1 \gamma_2 \omega_2$$

$$\frac{\text{Corrected}}{\gamma_{\text{gated}}} = N_0 \cdot \gamma_2 \omega_2$$

$$= N_0$$

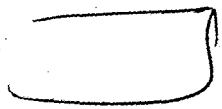




$$= N_0 \gamma_1 \omega_1 \times \gamma_2 \omega_2$$

$$\text{Cosm} = N_0 \underbrace{\gamma_1 \omega_1}_{} \times \gamma_2 \omega_2$$

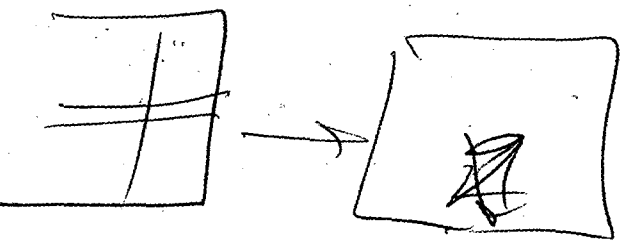
$$\text{Cos} = \gamma_{\text{single}} \times \underline{\underline{\gamma_1 \omega_1}}$$



$\gamma_1 \omega_1 \longrightarrow$ galv

$\gamma_2 \omega_2 \longrightarrow$ peak.

$\gamma - \gamma$



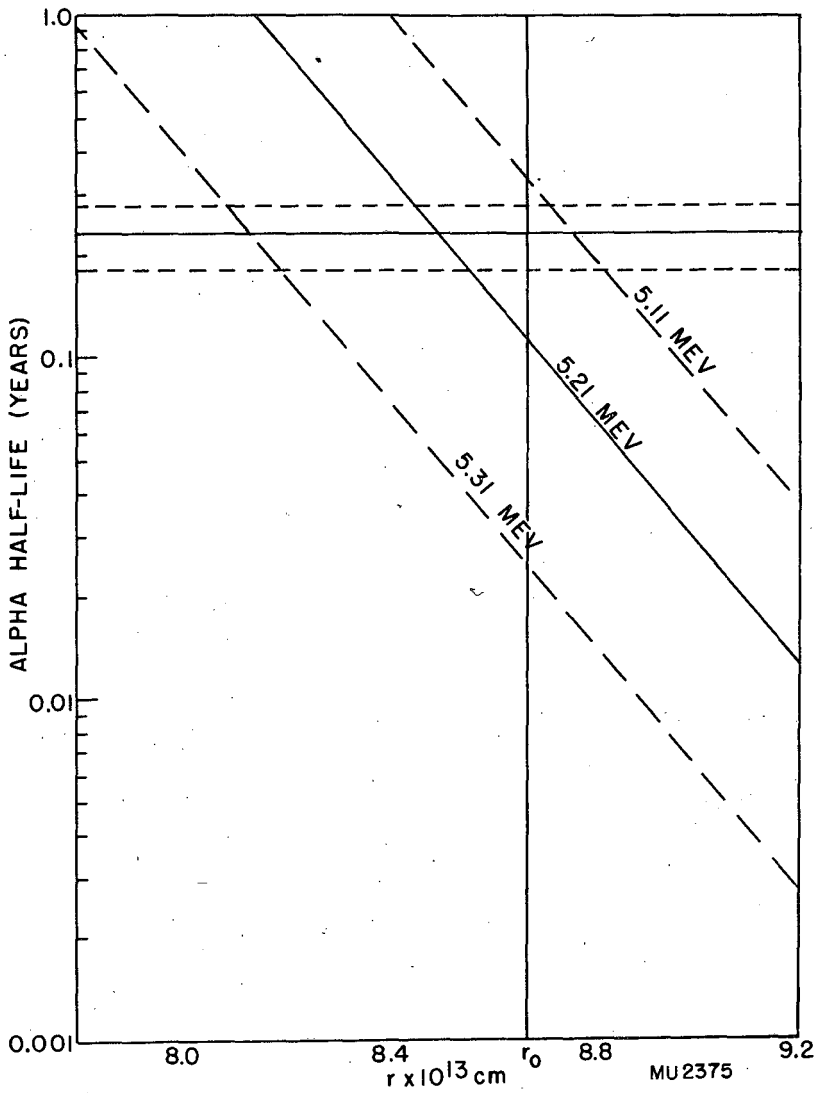


Figure III. Partial Alpha Half-life as a Function of Nuclear Radius