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LOW MASS FRANCIUM AND EMANATION ISOTOPES OF HIGH ALPHA STABILITY

E. K. Hyde, A. Ghiorso and G. T. Seaborg

October 10, 1949

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LOW MASS FRANCIUM AND EMANATION ISOTOPES OF HIGH ALPHA STABILITY

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ABSTRACT

Isotopes of francium with 126 or fewer neutrons have been looked for in bombardments of Th^{232} with 350 Mev protons from the 184-inch cyclotron. Fr^{212} with an apparent half-life of 19.3 minutes for branching decay by alpha emission (44%) to At^{208} and by orbital electron capture (56%) to Em^{212} has been found. Em^{212} is shown to be a 23-minute alpha-emitter. At^{208} decays primarily (99.5%) by orbital electron capture to Po^{208}, but shows 0.5% alpha-branching. The francium and emanation isotopes have alpha half-lives completely out of line with the predictions based on the previously known isotopes of these elements. Their high alpha stability is believed to be due to a closed shell of 126 neutrons in analogy to the behavior of elements 83-85. The non-existence of long-lived francium in nature is discussed in the terms of this and other recent work on francium isotopes.

* On leave of absence from Argonne National Laboratory, Chicago, Illinois.

Recent work with the 184-inch cyclotron has increased greatly the number of alpha emitting isotopes and stimulated the re-examination of alpha decay systematics in the heavy region.[1,2] The present paper describes experimental work carried

(1) I. Perlman, A. Ghiorso and G. T. Seaborg, Phys. Rev. 74, 1730 (1948).
(2) I. Perlman, A. Ghiorso and G. T. Seaborg, Phys. Rev. 75, 1096 (1949).

out to get more information concerning the abnormalities in alpha stability for nuclei in the region of 126 neutrons.

A plot of alpha energy versus mass number for nuclei of elements above atomic number 85 produces a series of nearly parallel curves (see Fig. 1, ref. 1). The curves for bismuth, polonium, and astatine (elements 83, 84 and 85) differ markedly in showing sharp maxima and minima as shown in Figure 1 below. It was of interest to know whether those for emanation (element 86) and francium (element 87) show the same behavior. If the emanation and francium curves of Figure 1 are simply extrapolated back to lower mass numbers, the alpha energies predicted for all low mass isotopes correspond to half-lives of fractions of microseconds. But if the behavior of the astatine, polonium, and bismuth curves is imitated, one might expect that Fr^{213}, Em^{212}, and isotopes of lower mass number would exhibit alpha half-lives long enough to permit their chemical isolation from targets bombarded in the 184-inch cyclotron. Should it be possible to do this, it would constitute further evidence for a region of stability at 126 neutrons.[3]

(3) Maria G. Mayer, Phys. Rev. 74, 235 (1949).

In addition to the above considerations, it is of interest to learn more about element 87. No isotopes of this element were known until Perey's work on the

21-minute β-emitting AcK.[4,5] Since then the five shorter-lived isotopes listed

(4) M. Perey, Compt. rend. 208, 97 (1939).
(5) M. Perey, J. Phys. et le Rad. 10, 435 (1939).

in Table I have been discovered. Any new information concerning the isotopes or the chemical properties of this little known element should be welcome.

Table I

KNOWN ISOTOPES OF FRANCIUM

Isotope	Half-Life	Mode of Decay	Energy of Particle(Mev)	Ref.
Fr^{218}	very short	α	7.85	(6)
Fr^{219}	0.02 sec	α	7.30	(7)
Fr^{220}	27.5 sec	α	6.69	(7)
Fr^{221}	4.8 min	α	6.30	(8,9)
Fr^{222}	14 min	β^-		(10)
Fr^{223} (AcK)	21 min	β^-,γ	1.20	(4,5)

(6) A. Ghiorso, W. W. Meinke and G. T. Seaborg, Phys. Rev. 74, 695 (1948).

(7) W. W. Meinke, A. Ghiorso and G. T. Seaborg, Phys. Rev. 75, 314 (1949).

(8) F. Hagemann, L. I. Katzin, M. H. Studier, A. Ghiorso and G. T. Seaborg, Phys. Rev. 72, 252 (1947); more completely given in U.S.A.E.C. declassified document MDDC 1186 (Dec. 10, 1947).

(9) A. C. English, T. E. Cranshaw, P. Demers, J. A. Harvey, E. P. Hincks, J. V. Jelley and A. N. May, Phys. Rev. 72, 253 (1947).

(10) E. K. Hyde, A. Ghiorso and G. T. Seaborg, unpublished work (June, 1949).

Therefore we undertook to prepare new francium isotopes by cyclotron bombardment. Without the aid of the high energy beam of the 184-inch cyclotron it would have been impossible as the closest suitable target material is Th^{232}. However, the

use of a high energy proton beam (maximum 350 Mev) on thorium easily effected the preparation of isotopes in the region of interest by spallation reactions. We were able to isolate francium and establish the new family of isotopes shown in Figure 2. The experimental evidence for this scheme is given below.

In addition evidence was found for Fr^{211} decaying principally by successive orbital-electron captures to At^{211} with a half-life of 8 minutes. This particular work is not complete.

It is quite evident then that the emanation and francium curves in Figure 1 show the same sharp break in the neighborhood of 126 neutrons as do the curves for elements 83 - 85. One may safely predict that higher elements will show the same effect, but the possibility of observing it by the method used here for francium and emanation grows slim, because the half-lives for alpha-decay become very short and the region of greater beta instability is being penetrated. These matters are discussed in much more detail in a forthcoming paper by Perlman, Ghiorso and Seaborg.(11)

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(11) I. Perlman, A. Ghiorso, and G. T. Seaborg, "Systematics of Alpha-Radioactivity", Phys. Rev. 77, Jan. 1, 1950.

Many writers of the past decade have pointed out the improbability of the natural occurrence of any long-lived isotopes of element 87. It may be well to reiterate this view in the light of the recent increase of the number of known francium isotopes from one to seven. Known isotopes of mass less than 222 are quite short-lived with respect to alpha-emission. Even the stabilization which occurs around 126 neutrons, although it enormously lengthens the expected half-life, is not enough to raise it to as much as an hour. On the other hand, isotopes 222 and 223 are short-lived with respect to beta-emission and those of higher mass are expected to be even more unstable. At this time we can be virtually certain that no very long-lived francium will ever be found. One cannot completely eliminate

the possibility of the existence of a long-lived isomer corresponding to a highly forbidden transition.

EXPERIMENTAL SECTION

(1) Bombardment Thorium foils 0.005 inch in thickness and 0.5 inch wide were bombarded on edge in the 184-inch cyclotron by full energy protons (max. 350 Mev) for periods ranging from 15 to 50 minutes. Numerous bombardments were made to obtain all the data because of the short half-lives involved.

(2) Chemical Isolation of Francium and Proof of Element Assignment It was necessary to separate francium out of an exceedingly complex mixture of activities produced by (p,xn), (p,pxn), and other spallation and fission reactions.

This was done with cesium carrier, using a procedure based on the fission product assay method for cesium reported by Nelson and Glendenin.[12]

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(12) C. M. Nelson and L. E. Glendenin, U. S. A. E. C. declassified document AECD 2556C (March 24, 1949).

The thorium foil was dissolved in hot 6N HCl containing 0.1N $(NH_4)_2SiF_6$. After dissolution, 2 mg of cesium carrier was precipitated as cesium silicotungstate from 6N HCl, washed in HCl, dissolved in NaOH, scavenged of co-precipitated impurities by means of by-product ferric hydroxide precipitation and finally precipitated as cesium perchlorate from anhydrous alcoholic perchloric acid solution.

The activity listed in Figure 2 as Fr^{212} was identified as the alkali element, francium, by the facts of its co-precipitation on cesium silicotungstate and cesium perchlorate, and its failure to co-precipitate on ferric hydroxide, thorium fluoride, or barium carbonate. It was possible to partially separate it from cesium by passing a 1N HNO_3 solution through a short resin column containing colloidal Dowex 50, the

francium coming off last as the results of Cohn and Kohn[13] for the other alkali

(13) W. E. Cohn and H. W. Kohn, J. Am. Chem. Soc. 70, 1986 (1948).

metals would indicate.

In addition, it showed the volatility behavior reported for Fr^{221} (8) and the daughters showed the specific behavior of astatine and emanation. It might also be mentioned that the Fr^{223}(AcK) produced in the bombardment followed the behavior of the 19-min. alpha-emitter identically.

(3) Determination of Half-Life of Fr^{212} The gross alpha activity of francium samples did not show a straight line decay. (See Figure 8.) This curve is a composite of the growth and decay of the alpha-activity of Em^{212} superimposed on the direct alpha decay of Fr^{212} with small contributions from At^{208} and from the alpha-emitting daughters (AcX chain) of Fr^{223}. Ordinarily such a situation can be easily resolved by frequent differential pulse analyses of the alpha spectrum by which method the peak due to any isotope can be followed independently of the rest. In our alpha differential pulse analyzer[14] alpha pulses from an ionization chamber are passed

(14) A. Ghiorso, A. H. Jaffey, H. P. Robinson and B. Weissbourd, National Nuclear Energy Series Vol. 14B, No. 16.8 (1949); shorter description given in U.S. A.E.C. declassified document MDDC-23 (June 14, 1946).

through a series of 48 electronic channels. Each channel records only the pulses falling within a narrow energy band. By plotting alpha counts per channel versus the channel number, a curve is obtained with peaks which indicate the energy and abundance of each alpha group.

This powerful method failed in this case because the near identity of the Fr^{212} and Em^{212} alpha energies made it impossible to resolve them accurately.

In the method used, aliquots of a francium solution were taken at intervals of 15-30 minutes and volatilized from one platinum plate heated briefly to red heat to a second cooled plate 2 mm above. This volatilization is believed to be due to the disproportionation of the francium oxide to volatile metal and peroxide coupled with the thermal decomposition of the peroxide in analogy to the behavior of cesium oxide. Thin francium plates could be 99% volatilized.

The francium aliquots thus freed of Em^{212} and the Ra^{223} daughter of Fr^{223}, which was also present, were immediately counted before any appreciable re-growth of these had occurred. Corrections were made for an appreciable contribution by At^{208} activity in the later aliquots.

A half-life value of 19.3 ± 0.5 minutes is obtained from the data plotted in Figure 3.

(4) Identification of Astatine Daughter Alpha-pulse analyses of francium samples after three hours decay clearly showed a new alpha activity of $5.65 \pm .02$ Mev energy. This activity could be volatilized from one plate to another at temperatures much lower than the francium could be moved.

High volatility is characteristic of astatine,[15] and this 5.65 Mev activity

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(15) G. L. Johnson, R. F. Leininger, and E. Segre, J. Chem. Phys. 17, 1 (1949). Also published as U.S.A.E.C. declassified document AECD-1952 (May 10, 1948).

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was judged to be the At^{208} daughter of Fr^{212}. Chemical proof involved carrying the activity on tellurium metal precipitated from 2M HCl with SO_2 and extraction into benzene from a 2N H_2SO_4 solution saturated with SO_2.

Benzene-extracted astatine samples were observed to have a straight line alpha decay with a half-life of 1.7 hours. These samples emitted x-rays which also decayed with a 1.7-hour half-life. Representative curves are shown in Figure 4.

By following decay through 2 grams of beryllium absorber with the sample at 10% geometry beneath a xenon-alcohol (10 cm pressure) filled end-window Geiger tube,

we observed one Geiger count per two alpha disintegrations. This Geiger activity is principally K and L x-rays, but includes some γ-rays. A value of 1.1 Mev was determined by lead absorption for the most energetic γ-ray.

A value of the At^{208} K/α branching ratio was obtained by comparing the initial alpha counting rate of a pure sample of At^{208} to the alpha counting rate of the Po^{208} daughter from electron-capture observed on complete decay. From 7830 alpha c/m At, we obtained 88 c/m of 3-year Po^{208} alpha activity.

From this we obtain 178/1 as the K/α branching ratio for At^{208}. Duplicate samples gave scattered results, the average value being 180 $\pm$ 20. This would make the partial alpha half-life of At^{208} about 13 days.

It should be pointed out that Barton has found evidence for an isomer of At^{208} in astatine fractions of bismuth targets bombarded with high energy helium ions.(16)

(16) G. W. Barton, Jr., A. Ghiorso and I. Perlman, unpublished work (1949).

This isomer decays by orbital electron-capture to Po^{208} with a half-life of 7 hours.

(5) Identification of Polonium Daughter The alpha activity found after complete decay of the 1.7 hour astatine activity in the samples mentioned in section (4) was shown by pulse analysis to be of 5.15 $\pm$ 0.05 Mev energy. It showed no decay in 90 days. The only known polonium isotope which fits this description is the 3-year 5.14 Mev Po^{208} reported by Templeton et.al.(17) Large samples of initially

(17) D. H. Templeton, J. J. Howland and I. Perlman, Phys. Rev. 72, 758 (1947).

pure Fr^{212} (10^5 - 10^6 α c/m) were found weeks later to have the amounts of 5.14 Mev alpha activity consistent with the production of 3-year Po^{208} by the branching scheme of Figure 2. This evidence plus the isolation of the same polonium activity from an initially pure sample of the 23 minute emanation is the evidence on which the

mass assignments of Figure 2 rest.

(6) The Emanation Isotope The apparatus shown in Figure 5 was used in the isolation and identification of Em^{212}.

A platinum plate with a sample of francium on it was placed in the apparatus. After evacuating the entire system, the part between valves A and B was filled with an atmosphere of pure argon. The platinum plate was brought to red heat with the induction heater to volatilize the francium and deposit it as a readily emanating film on the walls of the vessel. By opening valves B, C and E with D closed and letting argon flow in from A until the pressure reached atmospheric, the greater part of the emanation could be passed through the dry ice-cooled trap into the counting chamber. The trap removed astatine, water vapor and any low volatility impurities. By inserting this chamber into a special breech and setting the pre-amplifier unit of an alpha counter into the top of the breech, the central wire of the special chamber made contact with the high voltage lead to the counter (Figure 5(b)). The unit then functioned as an argon-filled ionization chamber for the collection of alpha pulses. This comparatively crude arrangement was quite adequate for relative measurements and for half-life determinations.

Using this method an alpha-emitting emanation activity of 23-minutes half-life was discovered. (See Figure 6.) This activity as well as a second alpha-emitting low mass emanation of 2.3-hours half-life, had been previously found in this laboratory.(18) The present work determined its mass assignment and alpha energy.

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(18) A. Ghiorso, W. W. Meinke and G. T. Seaborg, Phys. Rev. 76, Nov. 1, 1949.

To establish that the 23-minute emanation was the daughter of the 19.3-minute francium, the following "milking" experiment was carried out. After flashing an active francium sample into an atmosphere of argon as above, the system was evacuated. Fresh argon was let in to the section of the apparatus between valves A and B

and the emanation permitted to grow for exactly 20 minutes. At the end of this time the gas was transferred to the counting chamber and immediately counted. Then the chamber was again placed in the apparatus and pumped out. The emanation which had grown in during a second 20-minute period was transferred to the counting chamber and counted. This procedure was repeated 8 times. Results are shown in Figure 7. The decay line represents the decay of the francium parent. Considering the experimental error the half-life value is consistent with the assignment of the 19.3-minute francium as the parent of the emanation.

The alpha energy of Em^{212} is established as nearly identical to that of Fr^{212} from the fact that one peak in pulse analysis curves of the francium fraction accounts for the major part of the activity even though the gross alpha decay is definitely non-linear. More careful work with the energy scale of the pulse analyzer spread out revealed a difference in the two alpha-energies. A difference of about 80 Kev was found by pulse analyzing a francium sample freshly freed of Em^{212} by volatilization and continuing to pulse analyze it as the daughter re-grew. Our best energy values are $6.25 \pm .03$ Mev for Fr^{212} and $6.17 \pm .03$ Mev for Em^{212}. These values depend on calibration of the instrument with the Ra^{226} series.

The polonium daughter of alpha-emitting Em^{212} was shown to be identical to the orbital-electron capture daughter of At^{208} by pumping an active sample of emanation through two dry ice traps and letting it decay in an isolated glass tube. When the acid used to wash out this glass tube several hours later was evaporated on a platinum plate and pulse analyzed, alpha activity of $5.15 \pm .05$ Mev energy was found. This activity apparently due to Po^{208}, did not decay in a period of 60 days.

(7) Determination of Fr^{212} K/α Branching Ratio In principle, several methods might be used for making this important measurement, but all involve experimental difficulties.

Counting the francium x-rays directly, is rendered impossible by the presence of a high background of cesium fission product activity.

Quantitatively isolating and counting the Em^{212} from a measured sample of Fr^{212} would require developing better counting equipment for the gaseous activity.

One could quantitatively determine the amount of Po^{208} produced in an initially pure alpha-counted Fr^{212} sample. The excess of Po^{208} activity over the amount that would be produced if Fr^{212} decayed only by alpha emission would determine the amount of K-branching. This is insensitive because of the long half-life of Po^{208} and the low K/α branching of Fr^{212}.

The method selected was to determine carefully the shape of the alpha growth and decay curve for an initially pure francium sample and compare this curve with theoretical curves drawn for various values of the K/α branching. A sample of francium containing 10^5 alpha c/m was freed of emanation by volatilizing it from one platinum plate to another, quickly sprayed with a zapon solution to insure the complete retention of Em^{212}, and immediately placed in an alpha counter. This sample was counted continuously for several hours. The experimentally observed decay points were corrected for a background of Ra^{223} (from the decay of AcK present in the initial sample) amounting to 0.2% of the initial count. A small contribution to the counting rate due to the growth and decay of the At^{208} daughter was also subtracted. The resulting points were normalized to make the counting rate equal 100 at the time of volatilization and plotted as shown in Figure 8. The curves shown were derived from the theoretical equation below using a Fr^{212} half-life of 19.3 minutes and an Em^{212} half-life of 23.0 minutes.

$$\text{TOTAL ALPHA COUNTING RATE} = -\lambda_1 N_0 e^{-\lambda_1 t} - \frac{\gamma \lambda_2 \lambda_1 N_0 (e^{-\lambda_1 t} - e^{-\lambda_2 t})}{(\lambda_2 - \lambda_1)}$$

λ_1 and λ_2 are the disintegration constants of Fr and Em. t is the time. γ is the K/α branching ratio of Fr^{212}. N_0 is the initial amount of Fr^{212}. $\lambda_1 N_0$ is arbitrarily set equal to 100. From Figure 8 we estimate the K/α branching to be 1.3 ± 0.1.

This would place the partial alpha half-life of Fr^{212} at 43 minutes, the partial orbital electron-capture half-life at 34 minutes.

Acknowledgements The authors are indebted to Mr. James Vale and the operating crew of the 184" cyclotron for their assistance in the numerous bombardments required for this work. This research was performed under the auspices of the U. S. Atomic Energy Commission.

October 10, 1949

Figure 1 Plot of alpha particle energy versus mass number for isotopes of elements 83-89. Lines join all isotopes of a single element.

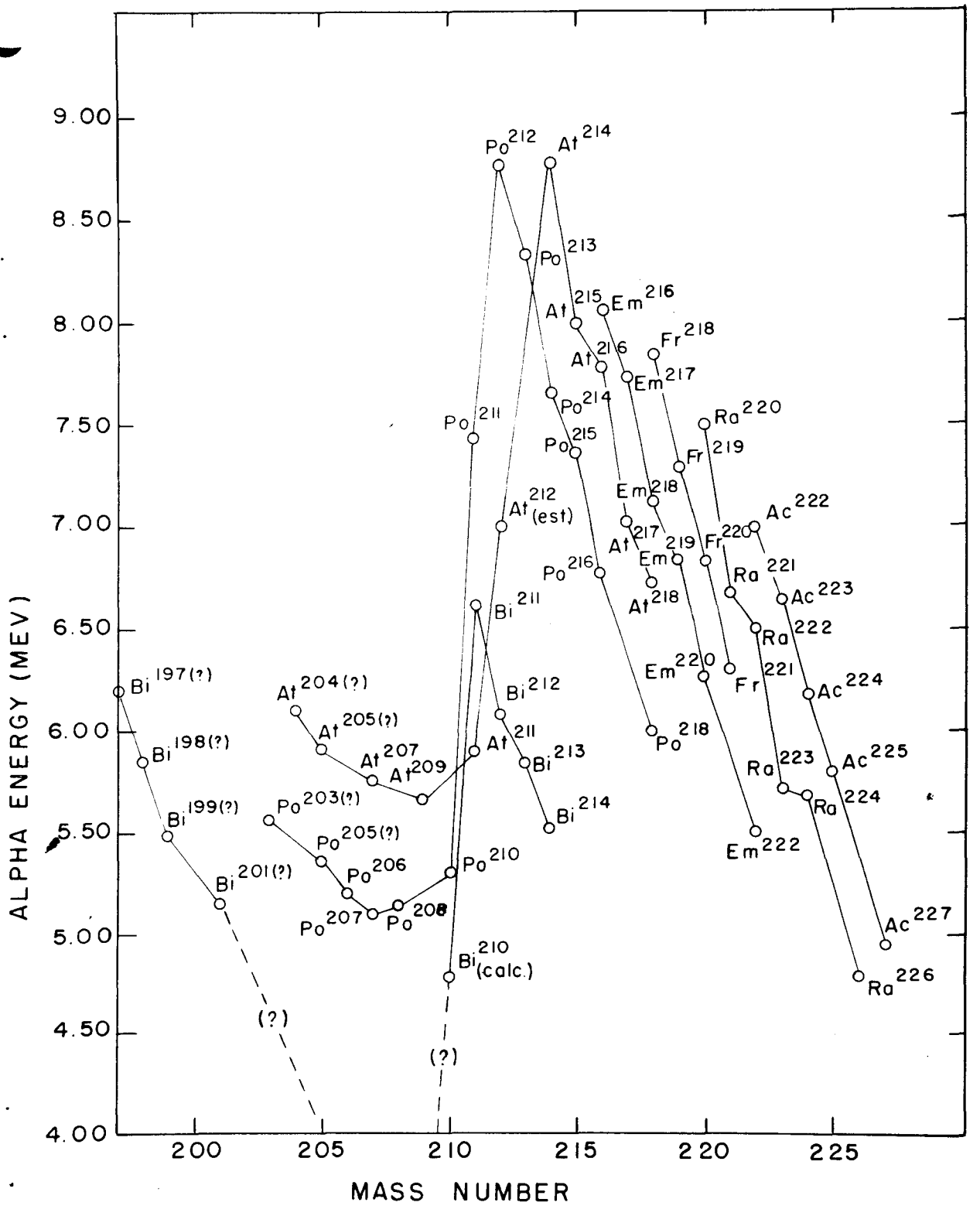
ALPHA ENERGY (MEV)
MASS NUMBER
9.00
8.50
8.00
7.50
7.00
6.50
6.00
5.50
5.00
4.50
4.00
200
205
210
215
220
225
Po212
At214
Po213
At215
Em216
Fr218
At216
Em217
Po214
Ra220
Po211
Po215
Fr219
Em218
At212 (est)
Ac222
At217
Fr220
Em219
Ra221
Ac223
Po216
Bi211
At218
Ra222
Em220
Fr221
Ac224
Bi212
Po218
At204(?)
At205(?)
At211
Bi213
At207
At209
Ac225
Ra223
Po203(?)
Bi214
Ra224
Po205(?)
Em222
Po206
Po210
Bi197(?)
Bi198(?)
Bi199(?)
Po207
Po208
Ac227
Bi201(?)
Bi210 (calc.)
Ra226
(?)
(?)

FIG. 1

Figure 2 Disintegration scheme of Fr^{212}.

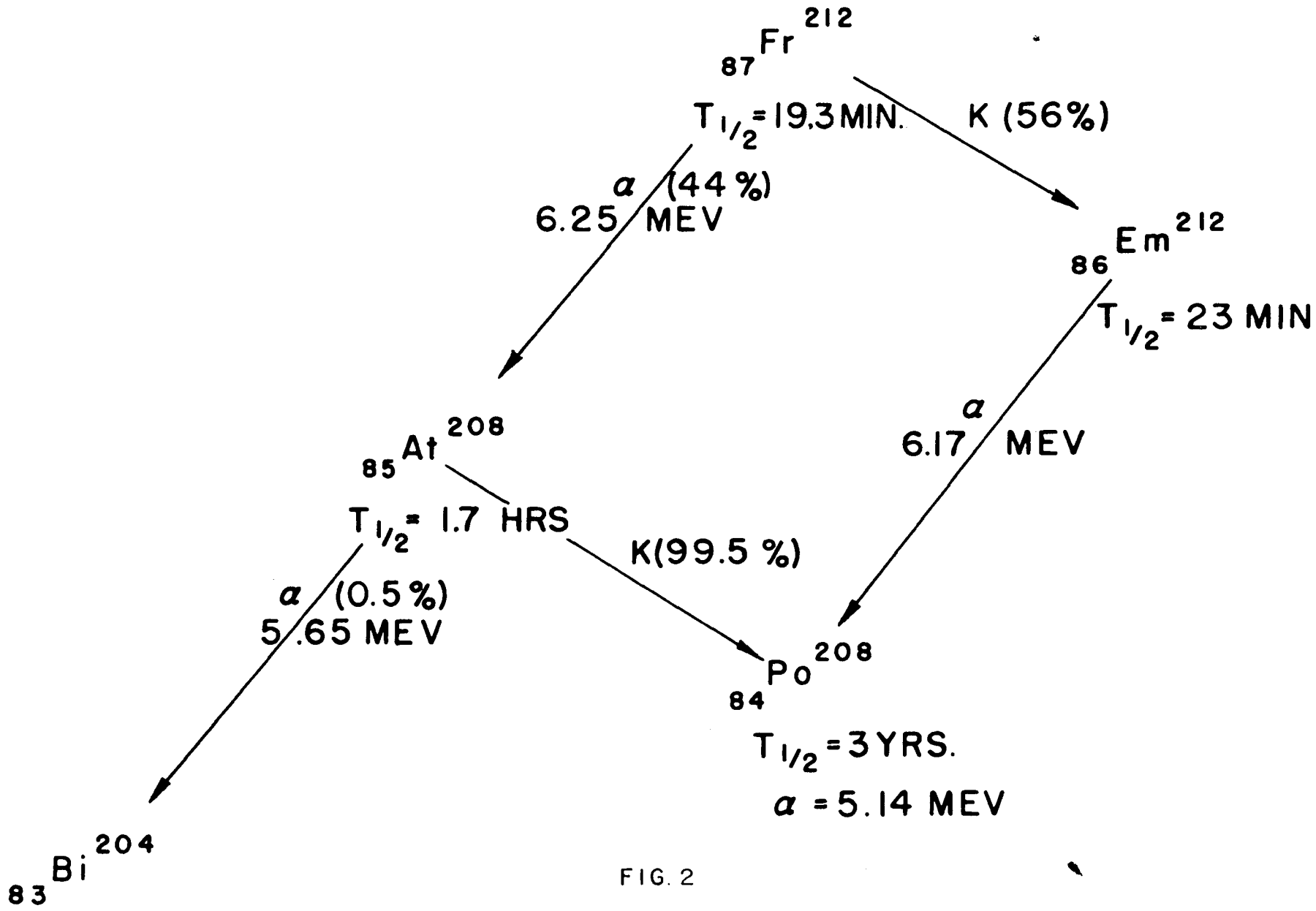
$_{87}Fr^{212}$
$T_{1/2}$ = 19.3 MIN.
K (56%)
α (44%)
6.25 MEV
$_{86}Em^{212}$
$T_{1/2}$ = 23 MIN
α
6.17 MEV
$_{85}At^{208}$
$T_{1/2}$ = 1.7 HRS
K(99.5 %)
α (0.5%)
5.65 MEV
$_{84}Po^{208}$
$T_{1/2}$ = 3 YRS.
α = 5.14 MEV
$_{83}Bi^{204}$

FIG. 2

Figure 3 Half-life of Fr^{212}. Points represent counting rates of equal sized aliquots of a francium solution counted immediately after purification by volatilization.

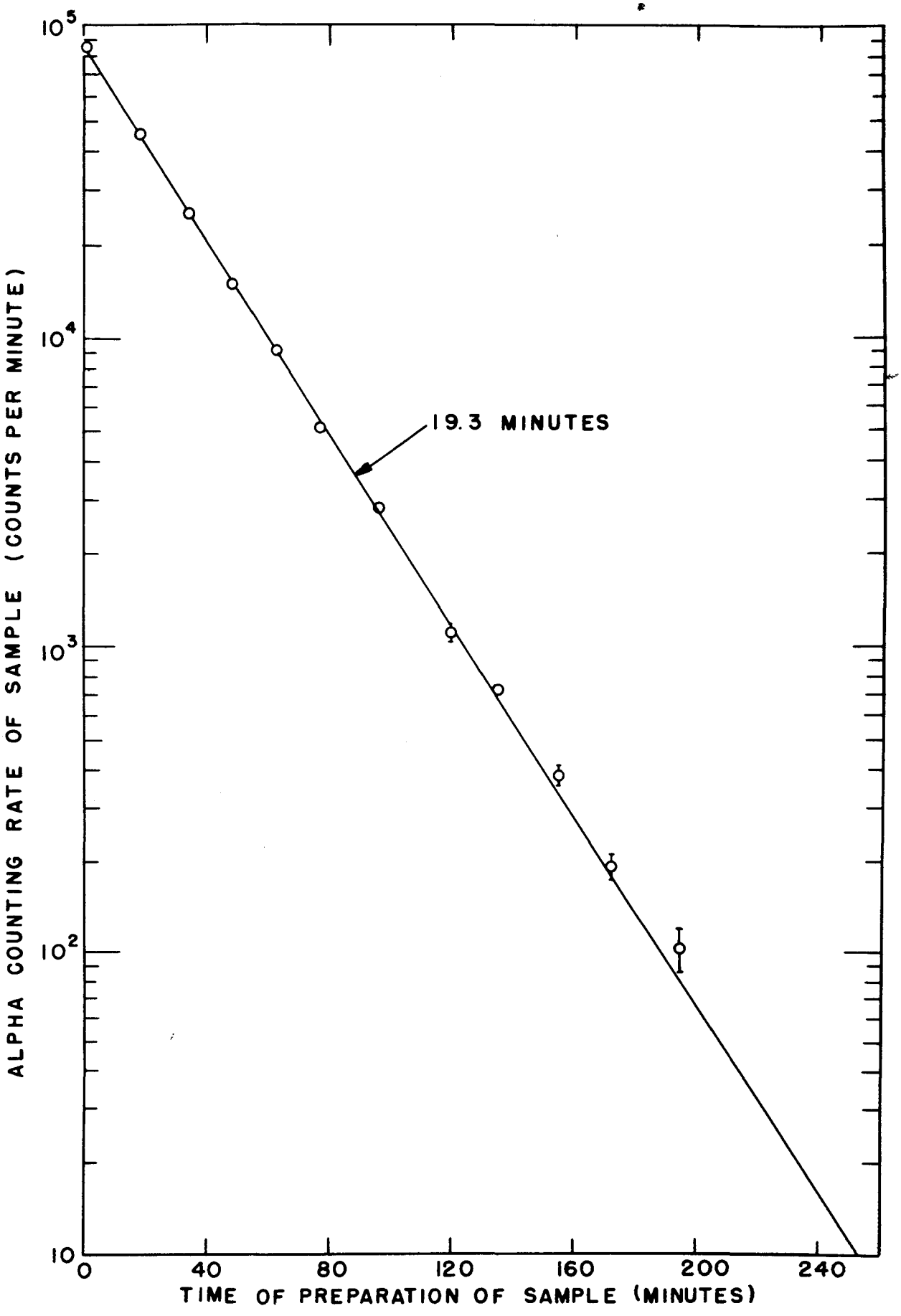
ALPHA COUNTING RATE OF SAMPLE (COUNTS PER MINUTE)
10^5
10^4
10^3
10^2
10
19.3 MINUTES
0
40
80
120
160
200
240
TIME OF PREPARATION OF SAMPLE (MINUTES)

FIG. 3

Figure 4 Half-life of At^{208}. Upper curve shows decay of x-ray and γ-activity through 1.87 grams/cm^2 beryllium absorber. Lower curve shows alpha decay.

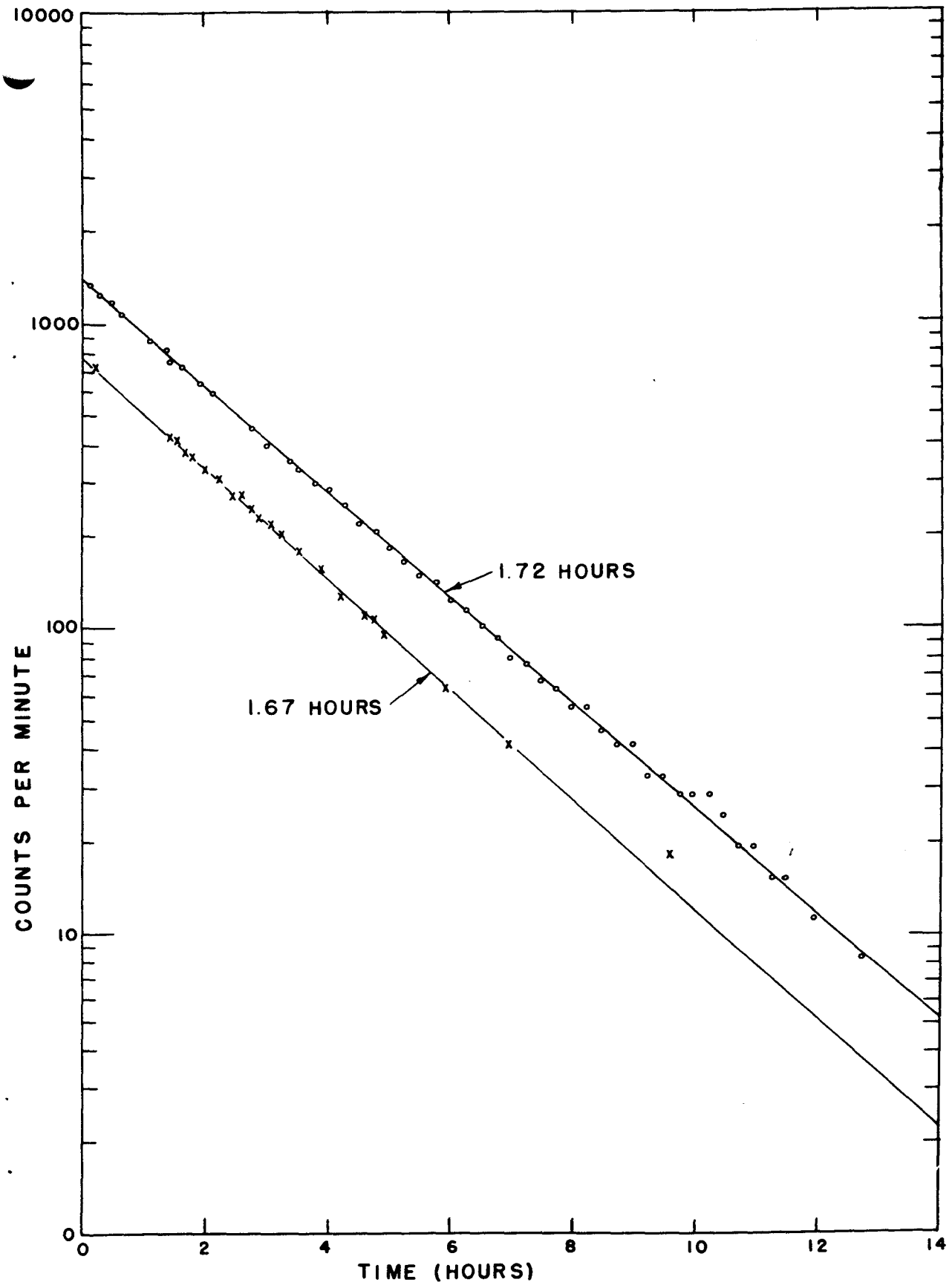
10000
1000
100
10
0
COUNTS PER MINUTE
1.72 HOURS
1.67 HOURS
0
2
4
6
8
10
12
14
TIME (HOURS)

FIG. 4

Figure 5 Apparatus used in the isolation (a) and alpha counting (b) of Em^{212}.

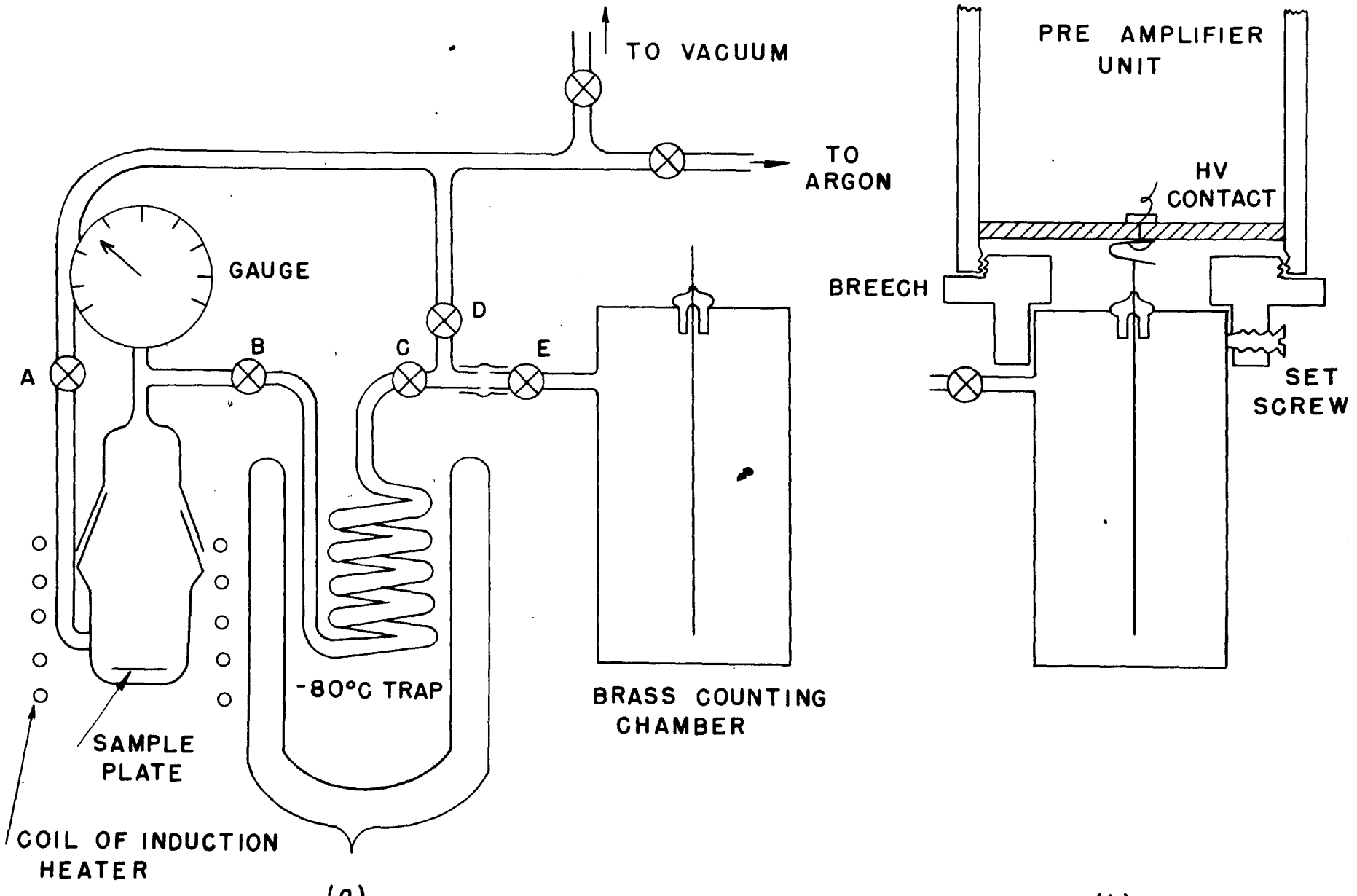
TO VACUUM
TO ARGON
GAUGE
A
B
C
D
E
-80°C TRAP
BRASS COUNTING CHAMBER
SAMPLE PLATE
COIL OF INDUCTION HEATER
(a)
PRE AMPLIFIER UNIT
HV CONTACT
BREECH
SET SCREW
(b)

FIG. 5

Figure 6 Half-life of Em^{212}.

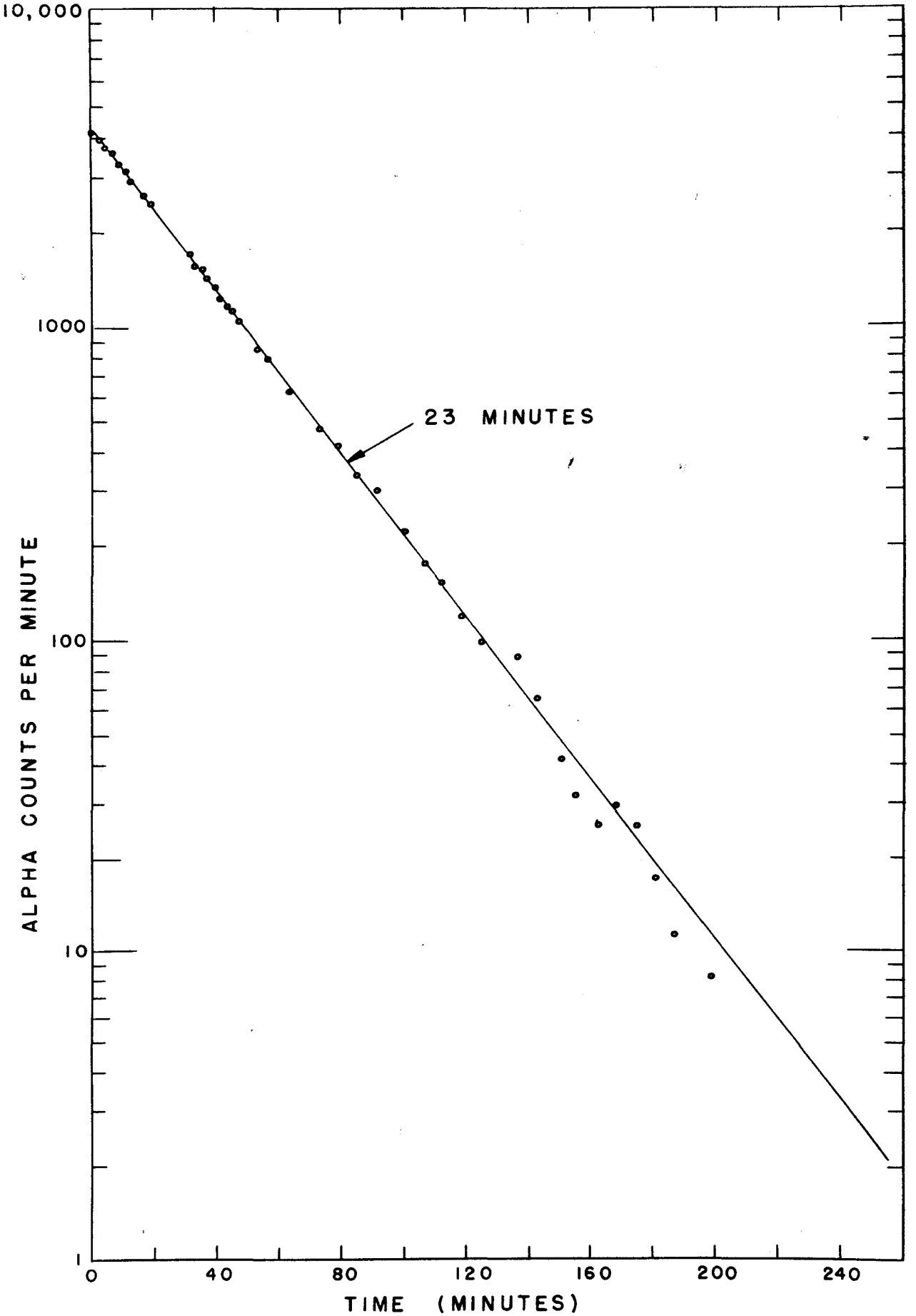
10,000
1000
100
10
1
ALPHA COUNTS PER MINUTE
23 MINUTES
0
40
80
120
160
200
240
TIME (MINUTES)

FIG. 6

Figure 7 Half-life of parent of Em^{212}. Points represent counting rate of Em^{212} grown into a sample of Fr^{212} during 20-minute period immediately preceding count.

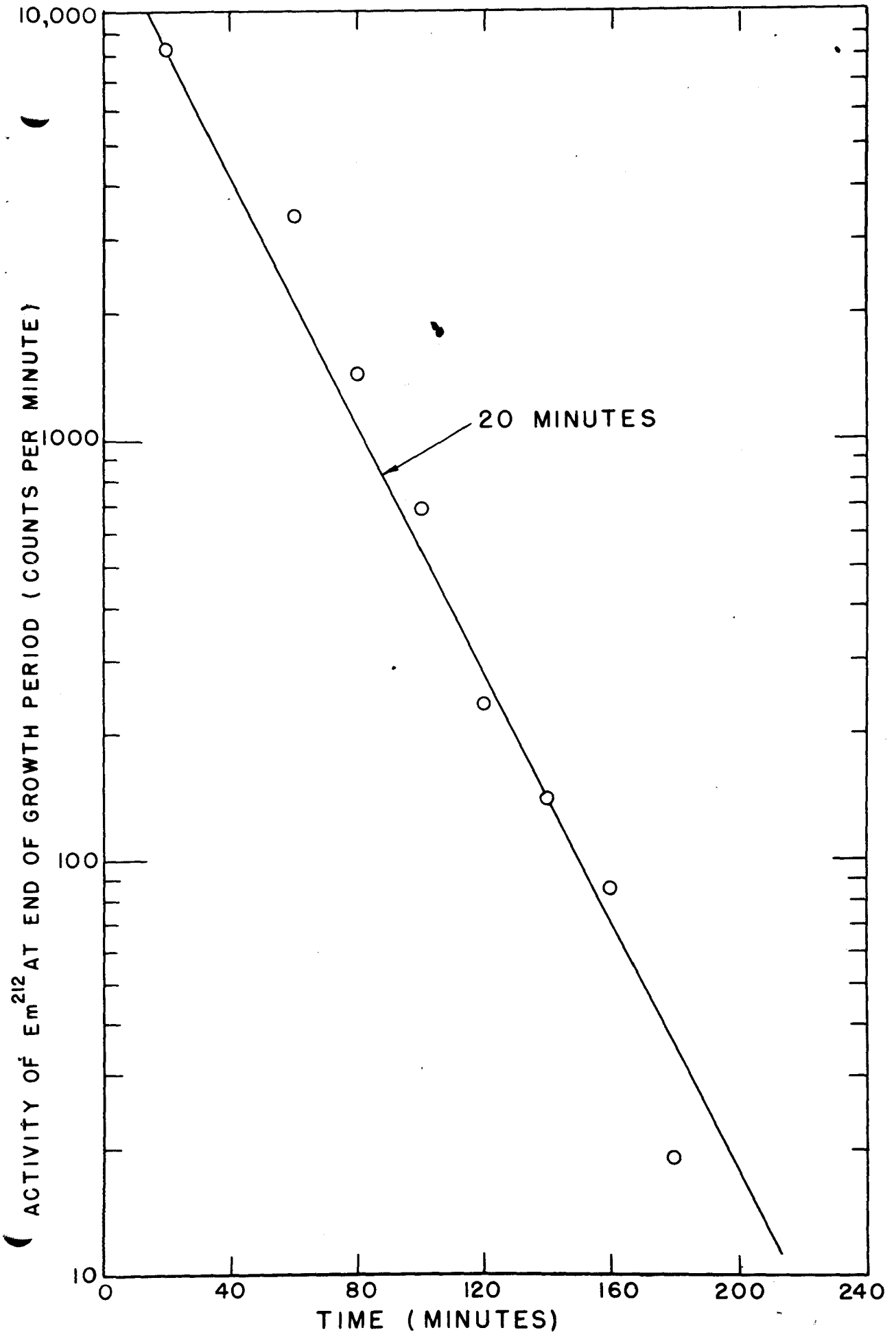
10,000
1000
100
10
ACTIVITY OF Em^{212} AT END OF GROWTH PERIOD (COUNTS PER MINUTE)
20 MINUTES
0
40
80
120
160
200
240
TIME (MINUTES)

FIG. 7

OZ 701

Figure 8 Determination of the K/α branching ratio of Fr^{212}. X points are experimental values corrected for small contributions from At^{208} and Ra^{223} and normalized to an initial count of 100. Lines are theoretical curves for the total alpha counting rate of a 19.3-minute alpha-emitting parent producing a 23-minute alpha-emitting daughter by K capture with the branching ratio indicated.

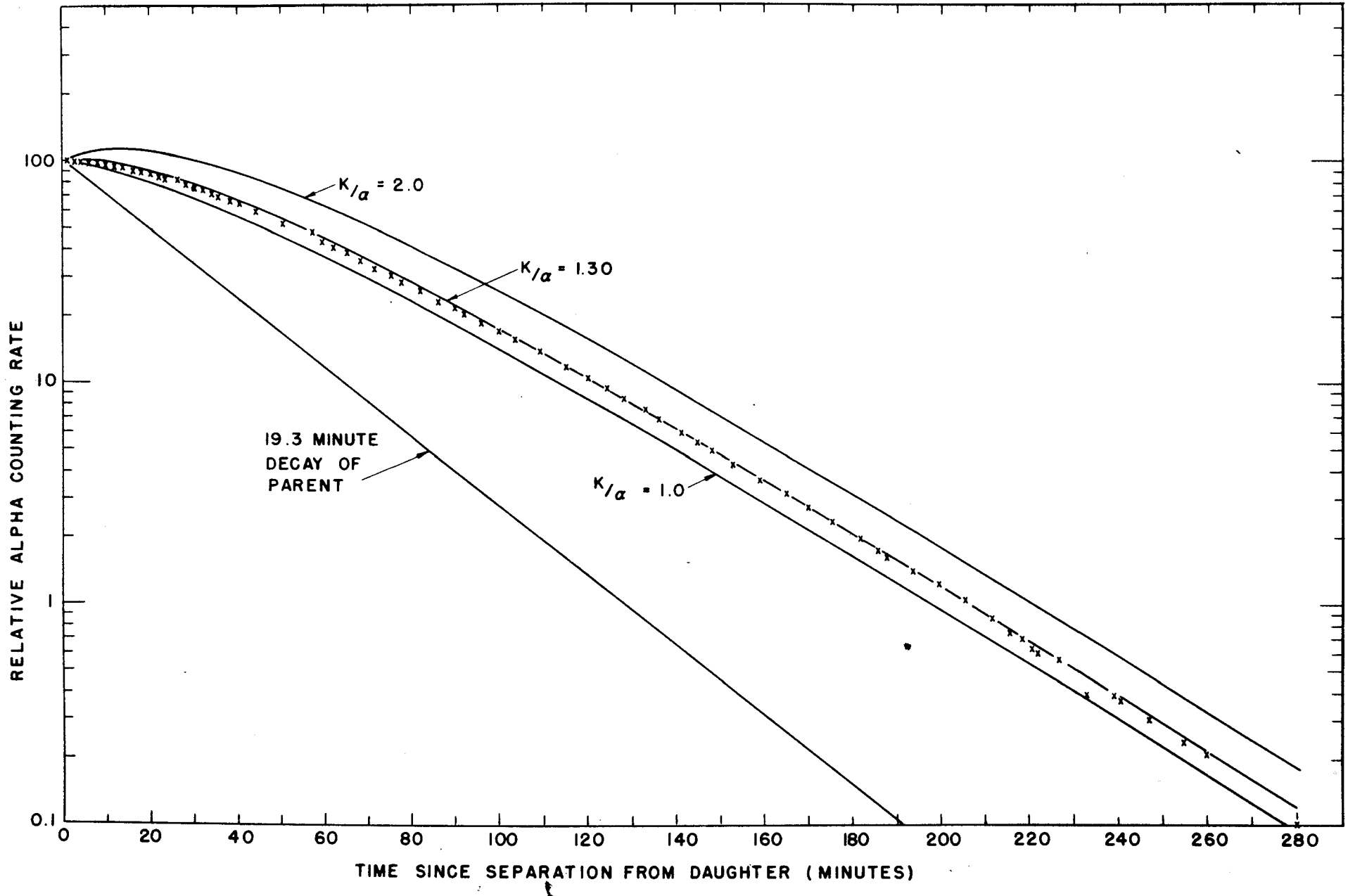

FIG. 8