Title
A NEW ISOTOPE, SULFUR-38

Authors
Nethaway, F.R.
Caretto, A.A.

Publication Date
1957-08-01

UCRL 3881

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

BERKELEY, CALIFORNIA

DISCLAIMER

UCRL-3881

UNIVERSITY OF CALIFORNIA

Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

A NEW ISOTOPE, SULFUR-38

D. R. Nethaway and A. A. Caretto, Jr.

August 1957

Printed for the U. S. Atomic Energy Commission

A NEW ISOTOPE, SULFUR-38

D. R. Nethaway and A. A. Caretto, Jr.

Radiation Laboratory
University of California
Berkeley, California

August 1957

ABSTRACT

A new isotope of sulfur, S^{38}, has been produced by the (α, 3p) reaction on Cl^{37}. It was found to have a half-life of 172 ± 1 minutes, and to decay by the emission of two beta groups with end-point energies of 1.1 and 3.0 Mev. The 1.1-Mev beta was found to be in coincidence with a 1.88-Mev gamma ray. No other gamma rays were observed. The 3.0-Mev beta occurs in 5% of the disintegrations, and leads to the Cl^{38} ground state. The log (ft) values of the 1.1- and 3.0-Mev beta groups are 5.0 and 8.2, respectively. A comparison is made of the (α, 3p) reactions on Al^{27}, Cl^{37}, and Cu^{65}.

A NEW ISOTOPE, SULFUR-38*

D. R. Nethaway† and A. A. Caretto, Jr.††

Radiation Laboratory
University of California
Berkeley, California

August 1957

INTRODUCTION

Until now the only available radioactive isotope of sulfur that was suitable for tracer studies was S^{35}. The low decay energy of this isotope (167-kev β^-) leads to inaccuracy in absolute counting, so that its usefulness in many applications is greatly reduced. A new isotope of sulfur, S^{38}, has now been produced by alpha-particle bombardment of chlorine, and in some ways this nuclide is better suited for tracer studies than S^{35}.

An earlier search for S^{38} was made by J. W. Jones,[1] who tried to produce it by high-energy proton bombardment of scandium, but was unable to find direct evidence for its existence. He was able, however, to set exclusion limits for the half-life as less than 3 hours, or greater than 50 years. Jean-Claude Roy and T. P. Kohman[2] had also obtained evidence that the half-life was about 6 minutes. The trend in the half-lives of the even-even isodiaspheres of the series including S^{38} indicated that its half-life might be long enough to be useful as a tracer. Table I

† Present address - Dept. of Chemistry, Washington University, St. Louis, Mo.

†† Present address - Dept. of Chemistry, Carnegie Institute of Technology, Pittsburgh, Pa.

* This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] J. W. Jones, Spallation Yields from Chlorine with 45-430-Mev Protons; A Search for Unknown Medium-Light Even-Even Nuclides; and Beta Lifetime Statistics, (Ph. D. Thesis), Carnegie Institute of Technology, NYO-6627, May 1956.

[2] Jean-Claude Roy and T. P. Kohman, private communication.

gives the even-even isodiaspheric series with N-Z = 6.

Table I. Even-even isodiaspheric series with N-Z = 6. (Isotopic abundances given)

Cr^{54}	Ti^{50}	Ca^{46}	A^{42}	S^{38}
Stable (2%)	Stable (5%)	Stable (0.003%)	> 3.5 years	-----

EXPERIMENTAL PROCEDURE

The S^{38} was produced by alpha-particle bombardment of NaCl, the reaction of interest being Cl^{37} (α, 3p) S^{38}. A similar reaction, Al^{27} (α, 3p) Mg^{28} was known to produce Mg^{28} in good yield.[3] The target material was reagent-grade (99.9+%) NaCl crystals which had been ground into a powder. The powder was placed in an aluminum holder and covered with a piece of 0.001-inch aluminum foil. The target was exposed to the 48-Mev alpha-particle beam of the 60-inch cyclotron of the University of California for a period of 30 minutes. The beam intensity was about 4 to 6 microamperes. After the bombardment the NaCl was removed from the target assembly and the sulfur produced was chemically purified from interfering radioisotopes. The chemical separation used is described in Appendix I.

The radiations emitted from the sulfur sample were measured in a methane-flow beta proportional counter. The radioactivity found could be resolved into two groups: the first with a half-life of about 172 minutes, and the second with a much longer half-life, presumably due to the 87-day S^{35}. The beta radiation of the latter group was stopped completely by a 40-mg/cm^2 aluminum absorber, verifying that it was due to the low-energy (167-kev) radiation from S^{35}.

A preliminary experiment showed that the beta activity, when measured through the aluminum absorber, had a growth characteristic of the well-known 37.3-minute Cl^{38}, indicating that the 172-minute period was due to the parent, S^{38}. For further proof of the genetic relationship, a "milking" experiment was performed. A sulfur sample was purified of

[3] J. Hudis, J. Inorg. Nucl. Chem. 4, 237 (1957).

Cl^{37}, Cl^{38}, and other interfering radioisotopes. It was then allowed to stand for a period of 90 minutes, at which time the chlorine activity was separated. This sulfur-chlorine separation was then repeated at 90-minute intervals. The chemical separations used are described in Appendix II.

The beta activity of the chlorine fractions was measured in a proportional counter. All the chlorine samples were observed to decay with a single 37-minute half-life. The observed counting rates were extrapolated to the time of the sulfur-chlorine separation and then corrected for the chemical yield of the chlorine carrier added and for the loss of sulfur in each separation step. In Fig. 1 these results are presented in a plot of corrected chlorine activity versus the time of separation. The slope of the line indicates that the parent of Cl^{38} has a half-life of about 172 minutes, and the repeated chemical isolation of Cl^{38} from the sulfur fraction verifies the assignment as S^{38}.

The gamma-ray spectrum of S^{38} was investigated by use of a 100-channel pulse-height analyzer together with a thallium-activated sodium iodide crystal. The 1.65- and 2.15-Mev gamma rays from the decay of Cl^{38} were observed to grow in and then decay with a 172-minute half-life. In addition, one other gamma ray, of 1.88 Mev, was found, which decayed with a pure 172-minute period. No other gamma rays were observed in the energy range up to 3 Mev; in particular, the 660-kev isomeric transition from the Cl^{38m} isomer was not seen.

By integration of the areas under the photopeaks and application of corrections for counting efficiency,[4] it was possible to obtain an approximate value of 70% for the abundance of the 1.88-Mev gamma ray. This value was found by comparison with the 47% abundant 2.15-Mev gamma ray in an equilibrium mixture of S^{38}-Cl^{38}, and is uncertain owing to errors in the integration and counting efficiencies.

Preliminary measurements on the beta spectrum were made by taking a set of aluminum absorption curves on an initially pure S^{38} source over a period of time as the Cl^{38} grew into equilibrium. Each set of measurements with a particular absorber was extrapolated to the time of sulfur-chlorine separation. That part due to the Cl^{38} was subtracted in

---

[4] M. I. Kalkstein and J. M. Hollander, UCRL Report No. 2764, October 1954.

order to get a set of points for pure S^{38}. These points were then plotted as an absorption curve, and are shown in Fig. 2. A straight line could be drawn through the points, giving a half thickness in aluminum of 56 mg/cm^2. This corresponds to an end-point energy of about 1.1 Mev.

Beta-spectrum measurements were made by use of an anthracene crystal in connection with a 100-channel pulse-height analyzer. The beta spectrum is complicated by the fact that the Cl^{38} daughter has beta groups with end-point energies of 1.11 Mev (31%), 2.76 Mev (16%), and 4.81 Mev (53%). Hence it was necessary to make a series of measurements of the beta spectrum starting with a freshly separated S^{38} source and continuing until the Cl^{38} had grown into equilibrium.

Fermi plots[5] were made on each set of measurements. Each plot indicated the presence of three beta groups, at 1.1, 2.9, and 4.8 Mev. Each beta group was subtracted in turn and the counting rates due to each particular group were calculated. A plot of the counting rate versus time was then drawn for each of the three energy groups. From the amount of growth occurring, it could be seen that the 1.1- and 2.9-Mev beta transitions occurred in the decay of both S^{38} and Cl^{38}, while the 4.8-Mev beta was from Cl^{38} decay alone. The end-point energies of the two beta transitions occurring in the decay of S^{38} were taken from the Fermi plot of the first measurement when the least Cl^{38} was present, and are 1.1 ± 0.1 Mev and 3.0 ± 0.2 Mev. In order to determine the relative abundances of the two beta groups in S^{38} decay, synthetic growth plots for the 1.1-, 2.9-, and 4.8-Mev betas were constructed. This was done by taking varying percentages for the unknown abundances in the parent, and combining with the known amounts growing in from the daughter decay. The synthetic plots were then drawn showing the growth as a function of time.

Figures 3, 4, and 5 show the synthetic plots for the three energy groups. The experimental points are superimposed on each plot. By comparison it was determined that the 3.0-Mev beta occurs in 5 ± 3% of the disintegrations. The data taken for the 1.1-Mev beta are less definite, and the abundance was found to be between 70 and 100%. A value of 95%

---

[5] See: Tables for the Analysis of Beta Spectra, National Bureau of Standards, AMS-13, June 2, 1952.

was chosen so as to be in agreement with the other abundance (5%), since no other transitions were found.

A beta-gamma coincidence counter, with anthracene and NaI(Tl) crystals, was used to determine if the 1.1-Mev beta was in coincidence with the 1.88-Mev gamma ray. This measurement was complicated by the fact that the Compton distribution from the 2.15-Mev gamma is in coincidence with the 1.11-Mev beta in the decay of Cl^{38}. If the coincidences are gated with the 1.88-Mev gamma ray, a differentiation must be made between that photopeak and the Compton distribution. The coincident beta spectrum was measured with the gamma-ray gate set at 1.81, 1.88, and 2.04 Mev, corresponding to the peak and valleys of the 1.88-Mev photopeak. At each setting the coincident beta spectrum had an end point at about 1.15 Mev. The measured coincidence rate of the 1.1-Mev beta at a gamma gate of 1.88 Mev was 2.2 times that at 1.81 Mev, and 2.5 times that at 2.04 Mev. It was assumed therefore that the 1.1-Mev beta transition was followed by the 1.88-Mev gamma ray and that the 3.0-Mev beta was the transition to the ground state of Cl^{38}.

The half-life of S^{38} was determined by following the decay of five separate samples for periods up to seven half-lives and treating the data by means of least-squares analyses. The measurements were made by using a 40-mg/cm^2 aluminum absorber to block out the activity from the S^{35} present. No trace of impurity was found in any sample. The results found are: 174.6, 170.0, 173.2, 170.0, and 172.7 minutes. The average value is 172 ± 1 minutes.

The threshold energy for the reaction Cl^{37} (α, 3_p) S^{38} is E^o = 39.4 Mev. Owing to the unusual nature of this reaction, and to the small excitation energy available, the measured cross section was rather low. A much higher yield should be obtained with higher-energy alpha particles. For a thick NaCl target, an average of 2×10^6 atoms of S^{38} per microampere-hour was formed; when an aluminum covering foil 0.002 inch thick was used, the yield dropped to 1.3×10^6. The cross sections for the similar reactions, Al^{27} (α, 3p) Mg^{28}, E^o = 35.3 Mev, and Cu^{65} (α, 3p) Ni^{66}, E^o = 40.2 Mev, were measured at various energies by using stacked aluminum and copper foils. The results of these measurements are shown in Fig. 6. The thin-target cross sections are plotted (in microbarns) as

a function of the bombarding energy above the threshold energy ($E_\alpha - E^o$). The indicated cross sections for S^{38} production are based on approximate calculations to convert from thick-target measurements. Figure 6 shows the Z dependence of the cross sections for the (α, 3p) reaction. The decrease in cross section for the (α, 3p) reaction with increasing Z would be expected if the reaction involves the evaporation of protons from a compound nucleus. The disintegration rate of Mg^{28} was measured by means of both the 1.78-Mev gamma and the 2.87-Mev beta which occur in the decay of the Al^{28} daughter. The disintegration rate of Ni^{66} was measured by beta counting, with the assumption that the effective counting efficiency was 2.

The decay of S^{38} has been shown to proceed by the emission of two negatron groups, 1.1 ± 0.1 and 3.0 ± 0.2 Mev. The 1.1-Mev beta is followed by a 1.88-Mev gamma transition to the Cl^{38} ground state. The abundances of the 1.1- and 3.0-Mev beta groups were found to be ~95% and 5 ± 3%, respectively.

S^{38}, being an even-even isotope, would be expected to have a spin of 0 and even parity. The spin and parity of Cl^{38} have been measured as I = 2(-). The log (ft) value for the 3.0-Mev beta transition is calculated as 8.2,[6] corresponding to a first forbidden (unique) decay, in agreement with the known spin and parity changes. The log (ft) value for the 1.1-Mev beta transition is 5.0, with an allowed (normal) classification; hence it must lead to a level of Cl^{38} at 1.88 Mev with I = 0 or 1, (+). Owing to the sample thickness and the low counting rates, no conclusion could be drawn from the Fermi plots regarding the shape of the beta spectrum. The 1.88-Mev gamma occurs in a transition with ΔI = 1 or 2 (yes), and hence can be assigned as an E-1 or M-2. The most accurate measurements on the level scheme of Cl^{38} were made by Paris, Buechner, and Endt,[7] but their work was not extended to this high-energy range. The known lower-lying levels of Cl^{38} are probably not populated by S^{38} decay, owing to the large spin changes. The complete decay scheme of S^{38} is shown in Fig. 7.

---

[6] S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

[7] C. Paris, W. Buechner, and P. Endt, Phys. Rev. 100, 1317 (1955).

The total decay energy of S^{38} is 3.0 Mev. Most of the published tables of isotopic masses or decay energies either do not extend into this mass region or are of insufficient accuracy there. However, the predicted decay energy given by A. G. W. Cameron is 3.3 Mev,[8] in good agreement with the measured value.

* * * * * * * * * * *

ACKNOWLEDGMENTS

The authors would like to express their appreciation to W. B. Jones and the crew of the University of California 60-inch cyclotron for their aid with the bombardments, to J. R. Grover for his many helpful discussions, and to Dr. Frank Stephens for his help with the coincidence measurements.

---

[8] A. G. W. Cameron, A Revised Semi-Empirical Atomic Mass Formula, Chalk River Report 690, March 1957.

APPENDIX I

Standard sulfur purification procedure: The NaCl was poured from the target assembly into an iron crucible. About 30 mg of K_2SO_4 carrier, Na_2CO_3, K_2CO_3, powdered iron, and charcoal were added, and the mixture heated at 850° C for 20 minutes. The sulfide was then distilled from 6 N HCl into a chilled plumbate solution. The PbS was washed and then oxidized by an HCl-bromine mixture. $BaSO_4$ was precipitated and then metathesized with Na_2CO_3 and NaOH. The $BaCO_3$ was removed and the supernatant liquid boiled with HCl and HNO_3; $BaSO_4$ was then precipitated and mounted for experimental purposes.

APPENDIX II

Sulfur-chlorine "milking" procedure: The NaCl was dissolved in NaOH, H_2O_2, 20 mg sulfate carrier, and 1 mg HF. $PbSO_4$ was then precipitated from an acidified solution. The $PbSO_4$ was dissolved in HNO_3 and the solution fumed with HCl and HNO_3. A $Fe(OH)_3$ scavenging precipitation was done with NaOH. $PbSO_4$ was again precipitated from an acidified solution. The time was noted here as the beginning of the Cl^{38} growth. The $PbSO_4$ was dissolved in NaOH. A mixed NaCl-$NaClO_3$ carrier solution was added. A small aliquot was removed for assay to determine the sulfur yield. After the growth time was over, $PbSO_4$ was precipitated with addition of $Pb(NO_3)_2$ solution and $HClO_4$. The time was noted as the end of the Cl^{38} growth period and the start of the next growth. $FeSO_4$ was added to the supernatant solution from the final $PbSO_4$ precipitation to reduce the chlorate carrier. The resulting $PbSO_4$ precipitate was separated and AgCl precipitated from the supernatant solution. This was dissolved in NH_4OH and a final AgCl precipitation was made from an acidified solution. The AgCl was mounted for measurement of the Cl^{38} activity.

FIGURE LEGENDS

Fig. 1. Plot of Cl^{38} activity separated from S^{38} versus time of separation.

Fig. 2. Aluminum absorption curve of S^{38}.

Fig. 3. Synthetic growth curves for 1.1-Mev beta. Cl^{38} = 31%. S^{38} values indicated.

Fig. 4. Synthetic growth curves for 3.0-Mev beta. Cl^{38} = 16%. S^{38} values indicated.

Fig. 5. Synthetic growth curve for 4.8-Mev beta. Cl^{38} = 53%. S^{38} = 0%.

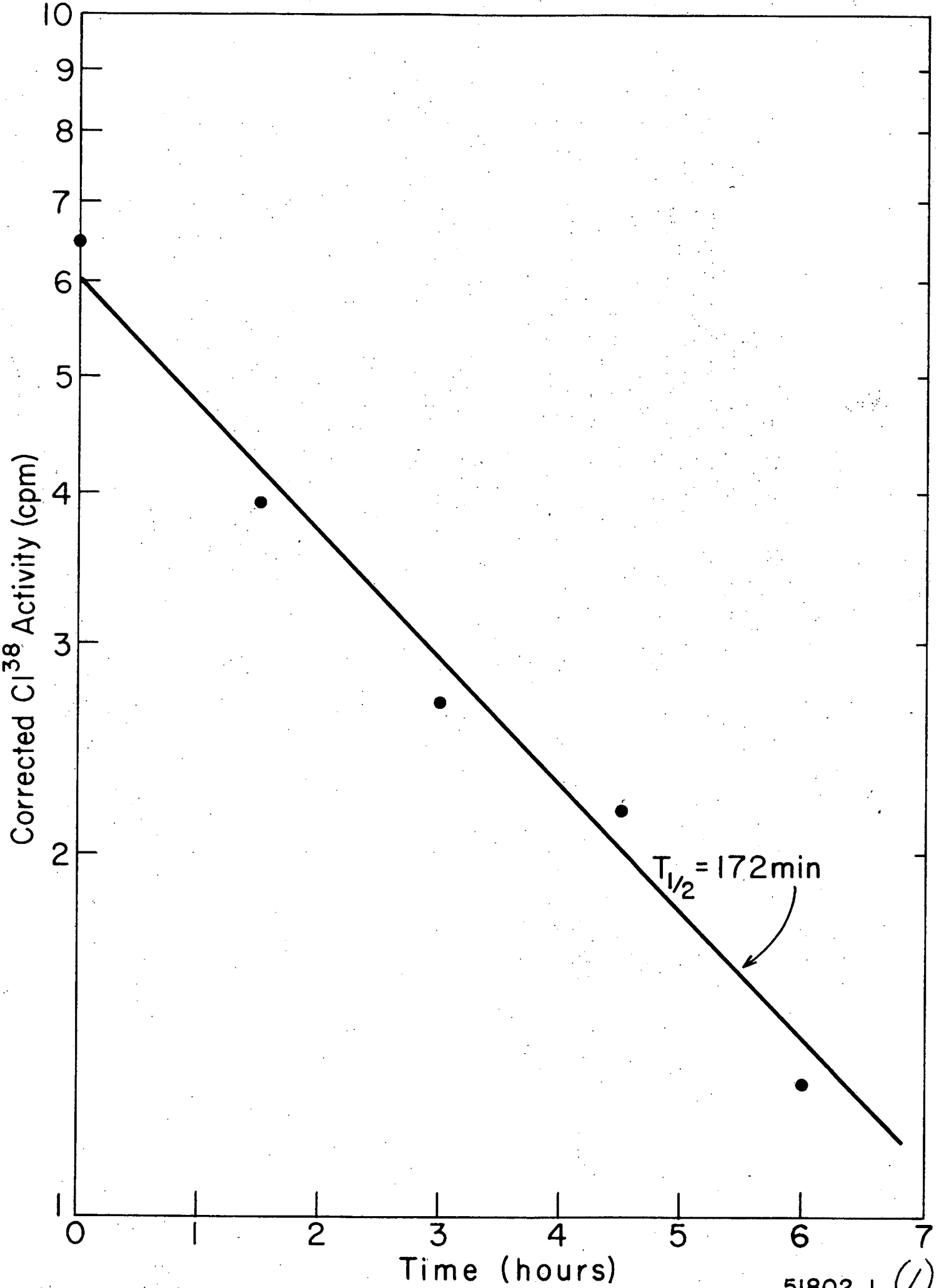
Fig. 6. Excitation functions for (a, 3p) reactions.

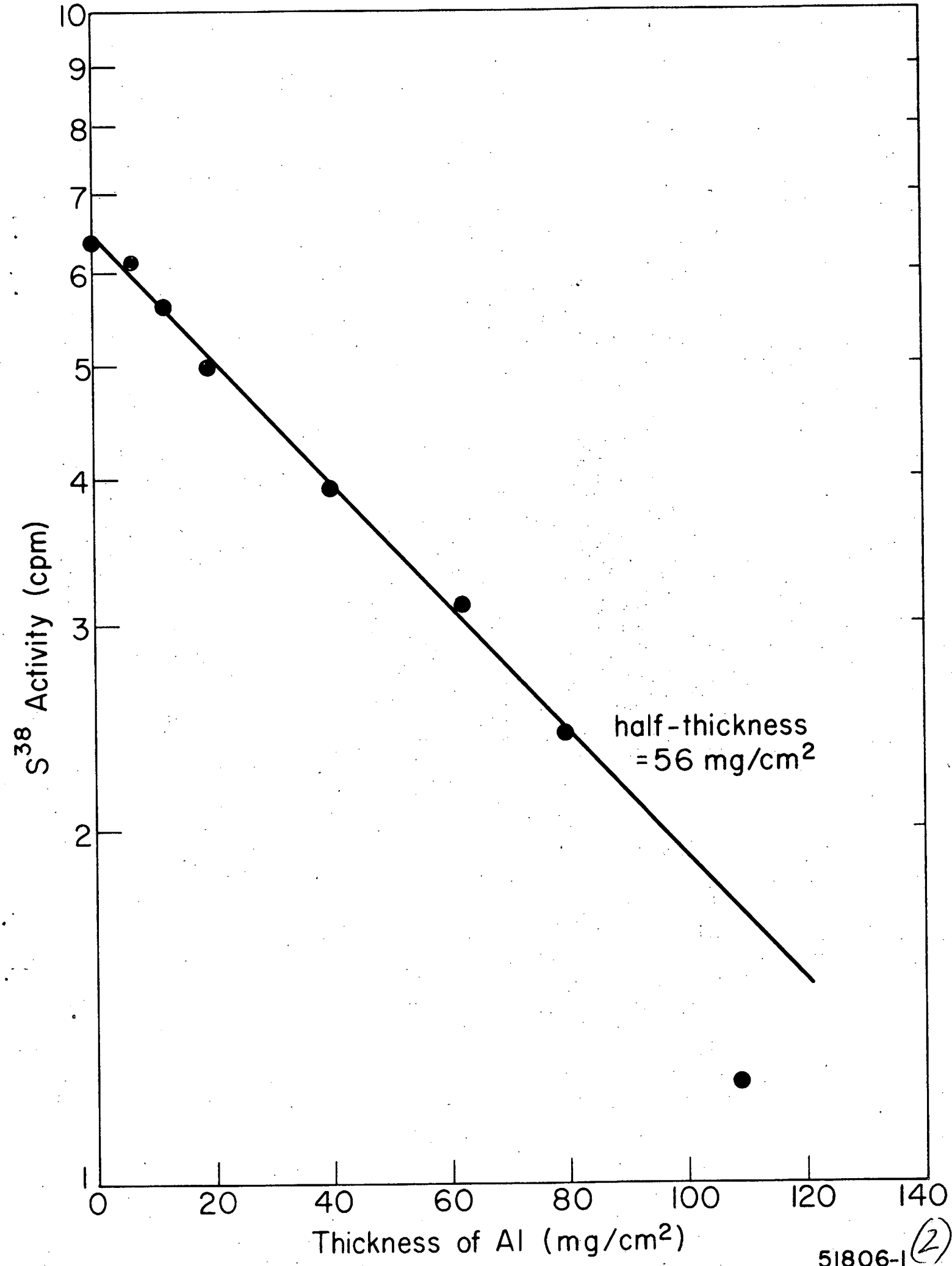
Al^{27} (a, 3p) Mg^{28} ●

Cl^{37} (a, 3p) S^{38} ×

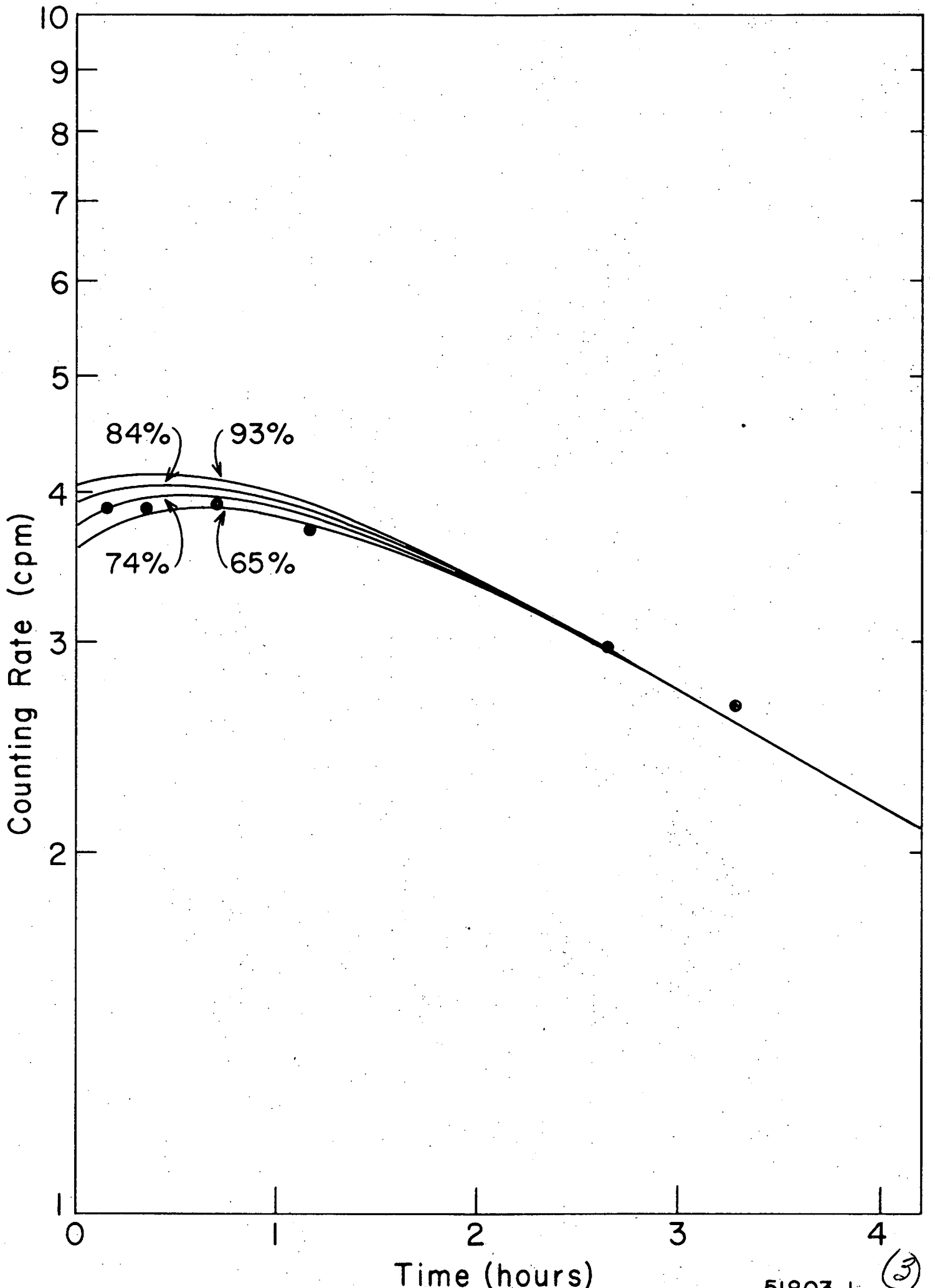
Cu^{65} (a, 3p) Ni^{66} ■

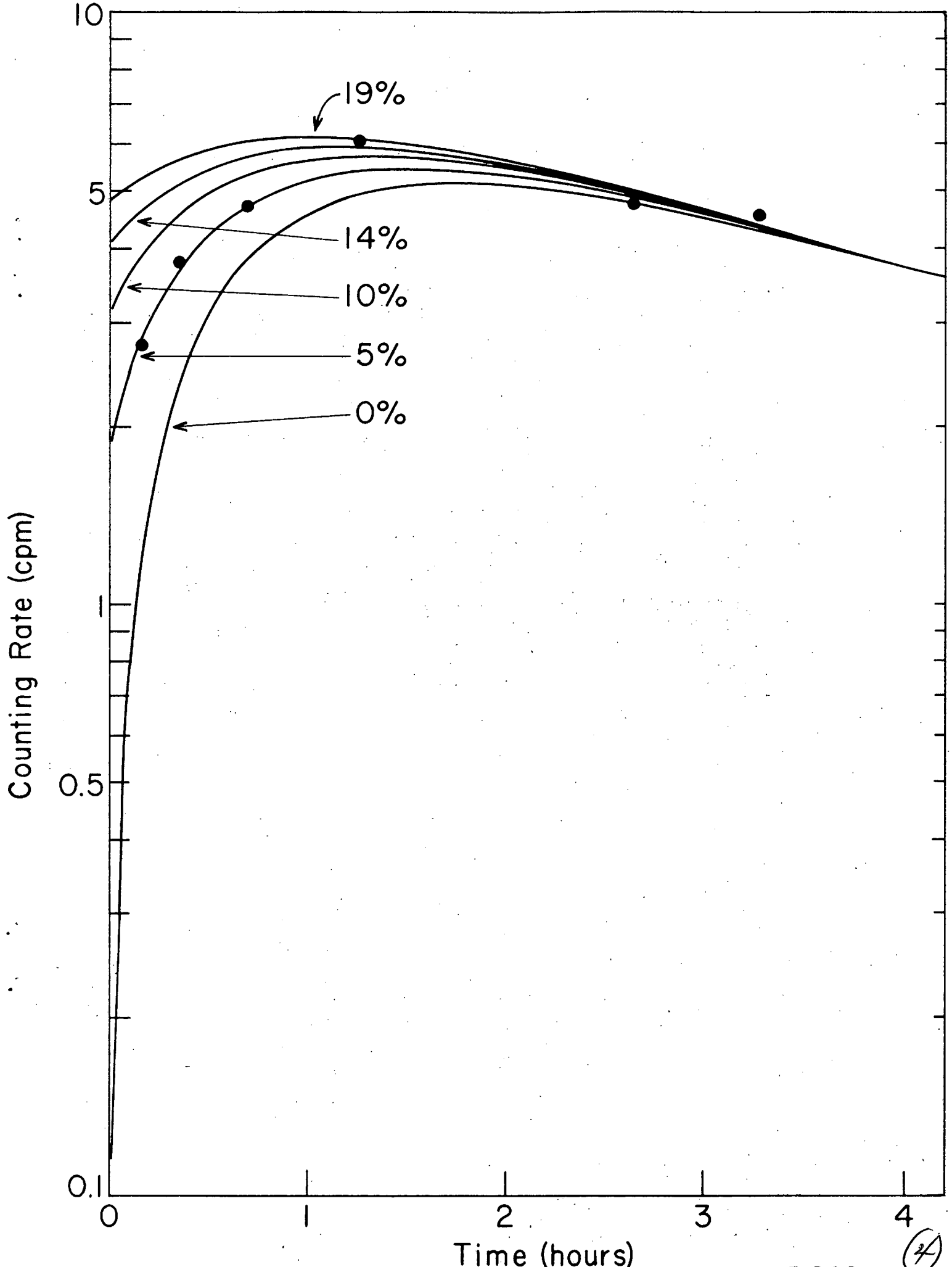
Fig. 7. Decay scheme of the Mass-38 chain.

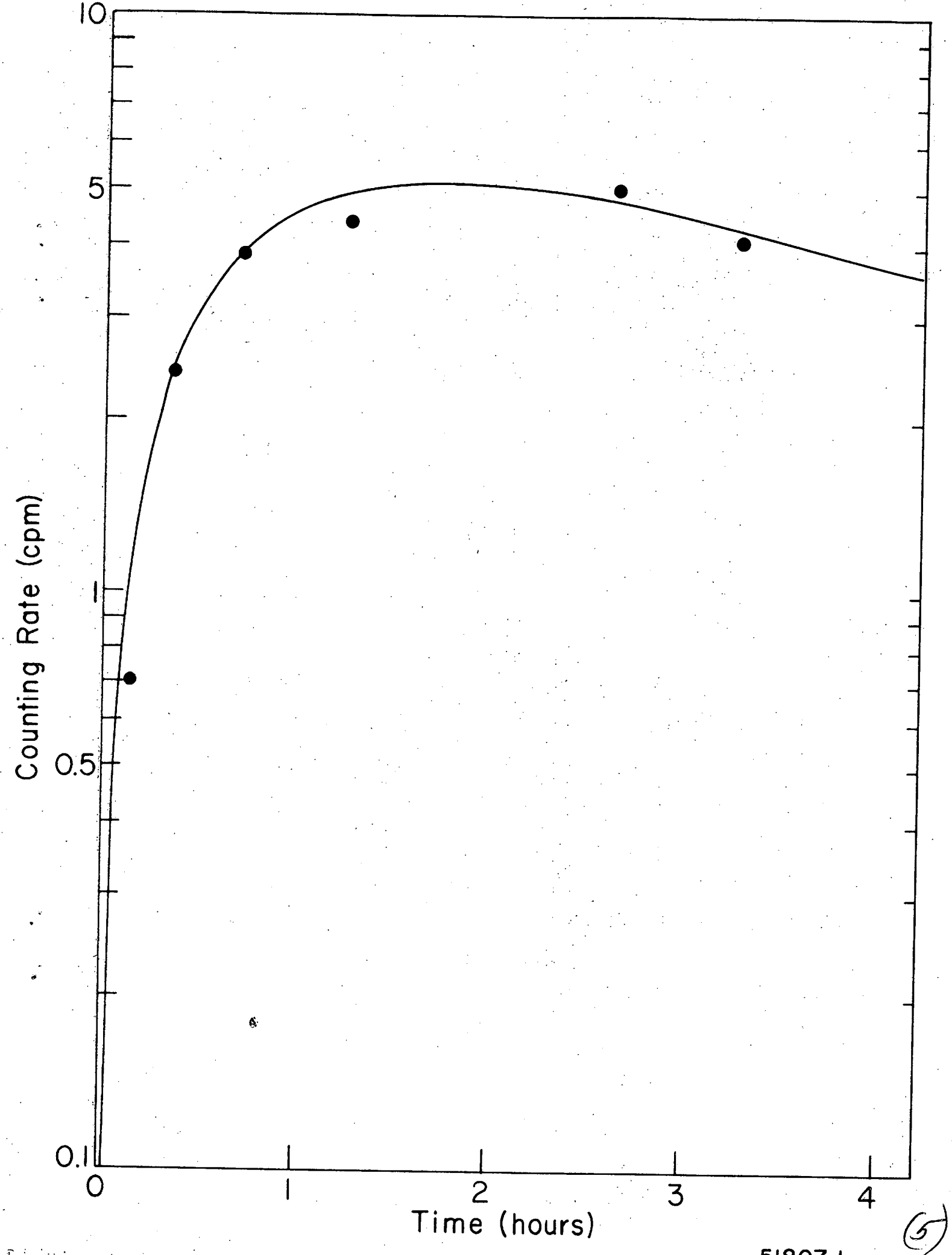

Corrected Cl^{38} Activity (cpm)
10
9
8
7
6
5
4
3
2
1
0
1
2
3
4
5
6
7
Time (hours)
$T_{1/2}$ = 172 min
51802-1


10
9
8
7
6
5
4
3
2
1
S^{38} Activity (cpm)
half-thickness
=56 mg/cm^2
0
20
40
60
80
100
120
140
Thickness of Al (mg/cm^2)

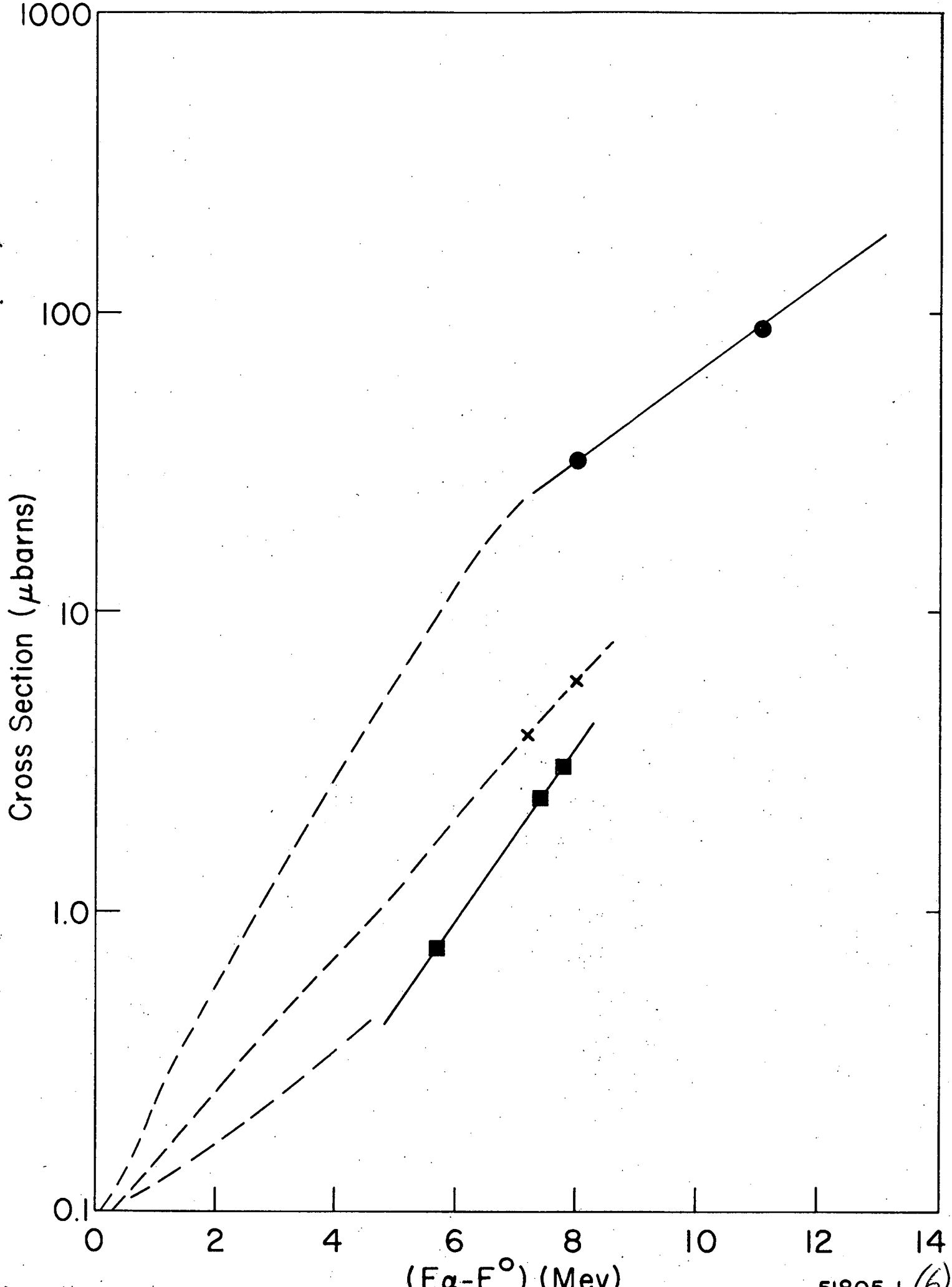
51806-1 (2)


10
9
8
7
6
5
4
3
2
1
Counting Rate (cpm)
84%
93%
74%
65%
0
1
2
3
4
Time (hours)
51803-1
(3)

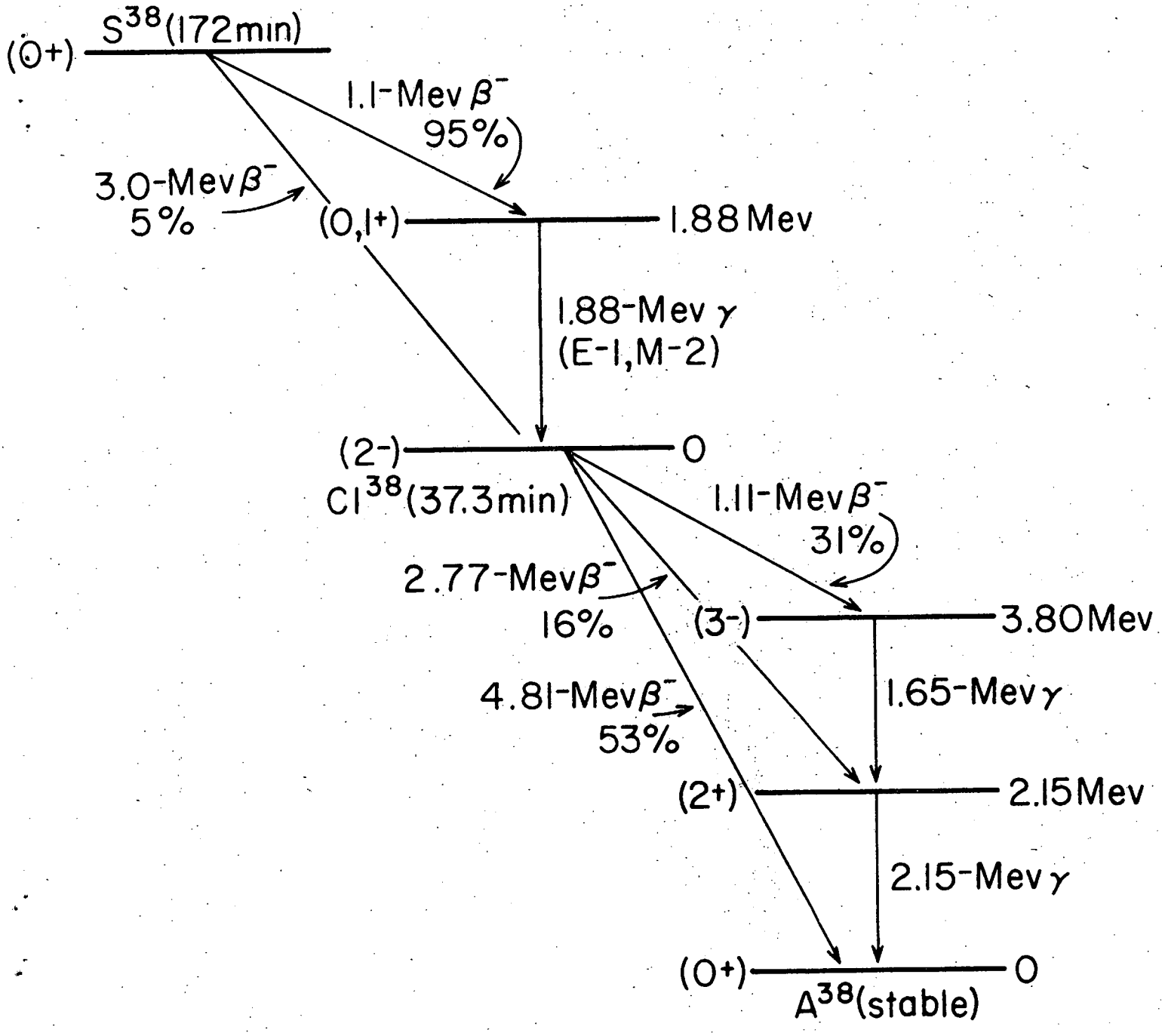

10
5
1
0.5
0.1
19%
14%
10%
5%
0%
Counting Rate (cpm)
0
1
2
3
4
Time (hours)
51808-1



51807-I



51805-1 (6)


(0+) S^{38}(172min)
1.1-Mev β⁻
95%
3.0-Mev β⁻
5%
(0,1+)
1.88 Mev
1.88-Mev γ
(E-1,M-2)
(2-)
0
Cl^{38}(37.3min)
1.11-Mev β⁻
31%
2.77-Mev β⁻
16%
(3-)
3.80Mev
4.81-Mev β⁻
53%
1.65-Mev γ
(2+)
2.15Mev
2.15-Mev γ
(0+)
0
A^{38}(stable)

(7)

51804-1