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DECAY PROPERTIES OF THE NUCLIDES FERMIUM-256, 257, AND 258 AND MENDELEVIUM-255, 256, AND 257

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DECAY PROPERTIES OF THE NUCLIDES FERMIUM-256, 257, AND 258 AND MENDELEVIUM-255, 256, AND 257

T. Sikkeland, A. Ghiorso, R. Latimer, and A. E. Larsh

April 1965

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SUMMARY

The decay properties of fermium and mendelevium nuclides produced by B^{11}, C^{12}, and C^{13} ion bombardments of californium have been studied. Some new characteristics have been observed for known nuclides and some new nuclides have been elucidated.

I. INTRODUCTION

Heavy ions from the Berkeley HILAC have been used in the production and identification of several new nuclides. In most of these cases the nuclides were formed in "neutron out" reactions. These reactions are characterized by a complete fusion of ion and target nuclei followed by evaporation. The excitation function for a particular isotope exhibits a distinct maximum, the position of which can be used to determine the number of neutrons evaporated and hence the mass number of the isotope. These reactions will yield neutron-deficient isotopes.

In addition to these reactions, incomplete fusion also takes place in which one or more nucleons are exchanged between ion and target nuclei. Here a variety of nuclei is formed and an excitation function can no longer

be used for mass assignment. Instead, this now has to rest on chemical methods and decay characteristics.

The advantage of using the latter type of reactions is that neutron-rich nuclides can be produced. This is particularly important in the region of the heaviest elements. Such isotopes may also be produced by neutron irradiation by the use of very high fluxes.

The present work was undertaken in an attempt to produce relative heavy nuclides of transcalifornium elements by the use of the heavy ion stripping method. This report does not give a complete account of the isotopes that are produced. Rather it must be regarded as a progress report from a continuing program. The data given have been collected over a period of about four years.

As target nucleus Cf^{252} was used because this is the heaviest isotope available in reasonable quantities. The target was bombarded with B^{11}, C^{12}, and C^{13} in order to determine the effect of Z and N of the ion on the yield of a particular nuclide. Efforts were concentrated on production and identification of unknown nuclides and of isotopes whose decay characteristics are not well known.

II. EXPERIMENTAL

The target was made by electroplating a carefully purified Cf sample onto a 2.1 mg/cm^2 thick Ni foil. The target was approximately 0.3 cm in diameter and consisted of 35 μg Cf^{252} and 7 μg Cf^{250}. The ions from the accelerator have an energy of 10.4 MeV/nucleon and their energy was reduced known amounts by the use of weighed Al or Be foils.

Ions with energies of few MeV above the Coulomb barrier for the ion and target nuclei were used. This corresponds to laboratory energies of about 75 and 65 MeV for the carbon and boron ions respectively. In general the yield of the spallation products from multinucleon transfer in the heavy element region is at a maximum at these energies. The average intensity of the beams was usually 0.5 μAmp.

Typically the bombardments lasted three hours. Reaction recoils were caught in 1 mg/cm^2 thick Pd foils. The transcalifornium elements were separated from Pd by the use of an anion exchange column and from each other by the use of a "But" column (Dowex-50 ion exchange resin and ammonium α-hydroxy-isobutyrate eluant).[1]

The drops from this column were individually analyzed for α and spontaneous fission (SF) activities in a twenty-sample solid state detection device. This analyzer also gave a rough indication of the α energies involved. The important fractions were analyzed by the use of two Frisch-grid ionization chambers each connected with 200 channel pulse height analyzers. As calibration standards the 5.80 MeV, 6.40 MeV, and 6.63 MeV alphas respectively from Cm^{244}, Es^{254}, and Es^{253} were used.

The Fm fraction was immediately recognized by its high α activity. Around 4,000 α/min and 300 SF/min were typically recorded at the end of separation. The α activity was due to Fm^{253}, Fm^{254}, and Fm^{255} and that of SF was due to Fm^{256}. The Fm fractions from different runs were, after the decay of the lighter Fm isotopes, combined, repurified, and analyzed for α and SF radiations from Fm isotopes heavier than 256.

The Md fraction was easily identified by the growth in of SF activity from the isotope Fm^{256} via Md^{256}. Around 3000 SF were recorded per experiment in this fraction. In addition Y and Tm tracers were used to further internally calibrate the But-column.

No α or SF activities were observed in the 102 or 103 fractions. The time between end of bombardment and separation, Δt, was as much as 20 minutes. Separate experiments were performed in which recoils were analyzed directly without chemical separation. Here Δt was reduced to 1/2 minute. Again no radiation was detected that could be definitely attributed to 102 or 103 isotopes.

Most of the efforts in the short bombardments were concentrated on the Md fraction. Only drops from the elution peak and its leading edge were analyzed to prevent interference from Fm activities. To further prove to our satisfaction that the observed activities were indeed due to isotopes of Md or their daughters, additional experiments were performed in which the Md fraction was repurified by the use of a second But-column. In general, only one such column was used.

III. RESULTS AND DISCUSSION

A. Mendelevium Fraction

In approximately twenty experiments that were undertaken, similar alpha pulse height spectra of the Md fraction were obtained. In Fig. 1, one is shown what is a composite of four runs. There are five distinct alpha groups of 6.85, 7.07, 7.17, 7.24, and 7.34 MeV. The energies are believed to be accurate to better than 30 KeV.

It is evident that especially the group at 7.07 MeV is a composite of at least two groups. At the end of the decay of this group, its energy is closer to 7.04 MeV; however, we will in the following label this group as 7.07 MeV.

Frequently observed were higher energy pulses around 7.5 and 7.6 MeV that decayed relatively quickly. These probably belong to ground state transition alphas for some of the Md isotopes (see Section IV). Not enough data were available to analyze them and they will therefore be ignored in the discussion.

The decay curves of the different alpha groups and of the SF activity are shown in Fig. 2. In Table 1 are listed the nuclides to which we have assigned these activities together with their decay characteristics as deduced from our data. In the same table are also given results from other experiments. The mass assignments were based on the following arguments.

1. Md^{255}

The experiments with two But-columns excluded the possibility that the groups at 7.17, 7.24, and 7.34 MeV and the 3-hour component of the 7.07 MeV group were due to daughters of Md isotopes. The long-lived component of the latter group was assigned to Fm^{255} and is thus the electron capture (EC) daughter of Md^{255}. By comparing the relative intensity of Fm^{255} after the first and the second column, it was concluded that Md^{255} has a half-life of about 1/2 hour. Consequently we assigned the 1/2 hour, 7.34 MeV alpha group to Md^{255}. The EC branching was estimated from the yield of Fm^{255}. The decay properties agree well with earlier work.[2]

2. Md^{256} and Fm^{256}

The growth and decay of the SF activity is identical to that observed[2,3] in the Md fraction from helium bombardment of Es^{253} and is known to be due to Fm^{256}, the EC daughter of Md^{256}. The half-life of the latter isotope is 1.5 hours as deduced from the SF decay curve. The α group at 7.17 MeV decays with a similar half-life and was therefore assigned to Md^{256}.

The growth and decay of the 6.85 MeV alphas suggests they are due to a daughter of Md with a half-life of around three hours and thus are assigned to Fm^{256}. However, the curve is not identical to that of the SF from Fm^{256}. In particular, the value for the ratio SF/(6.85 MeV α) is increasing with time. A possible explanation for this discrepancy is the presence in the 6.85 MeV peak of alphas from other relatively short lived Md isotopes such as Md^{258} or lighter. For that reason the ratio SF/α listed in Table I was taken at the end of the decay curves and consequently the value is not very accurate due to poorer statistics at this time.

3. Md^{257}

The 3-hour component of the 7.07 MeV and 7.24 MeV alpha groups was assigned to the previously unobserved isotope Md^{257}. The assignment of the latter group is questionable. Its energy and half-life are characteristic also for those of Fm^{254}. This isotope[4] can be present both as the result of contamination and as a consequence of production of the hypothetical EC isotope Md^{254}. On account of the few events observed we feel these possibilities are not entirely ruled out. It was not possible to observe the growth of its α and EC daughters, Es^{253} and Fm^{257}, due to their relatively long half-lives. The mass assignment can be argued as follows. Results from experiments with $Es^{253} + He^{4}$ exclude the possibility that this emitter is Md^{256} or lighter.[2] From decay systematics, that are discussed later, it is suggested that for $A \geqslant 258$ the α half-lives are too long and the yields too low to account for the intensity of the 7.07 MeV peak. These arguments make Md^{257} the most reasonable choice.

The value for the branching ratio (EC + SF)/α given in Table I was not based on a direct measurement but was deduced from the estimated yield of Md^{257}. In Table II the cross section of the 7.07 MeV 3-hour alpha emitter is compared to those of the Md and Fm isotopes. The yield of a particular

isotope was within a factor of two independent of the ion used. For these values no corrections were made for self-adsorption of recoils in the target since it was assumed that the Md isotopes are made by mechanisms that lead to similar ranges of the products. The cross sections given therefore have a value on a relative basis. What is of importance here is that the cross sections for Md^{255} and Md^{256} are nearly equal whereas that for the production of the 7.07 MeV alphas is an order of magnitude lower. The cross section for Md^{257} was taken as the average value of those for the two other Md isotopes for which the (EC + SF) branching was computed.

An upper limit of 10% can be set on contribution to the SF curve from a 3-hour SF emitting Md isotope and so the partial SF half-life for Md^{257} is at least 30 hours. This further means that this species is decaying predominately by EC to Fm^{257}. The long half-life of the latter prevents it from being detected in a 3-hour bombardment. A prolonged bombardment is not of any help in determining the EC branching of Md^{257} since the Fm^{257} is also produced directly.

The decay systematics of these isotopes as discussed in Section IV furnish additional evidence for the correctness of the mass assignments.

B. Fermium Fraction

1. Fm^{257}

Evidence for a long lived alpha emitting Fm isotope, tentatively assigned as Fm^{257}, was obtained in these experiments in 1964.* The energy

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*This information was reported by one of us (A.G.) at the Los Alamos meeting of the Ad Hoc Transplutonium Advisory Committee in February, 1964.

of the alphas was slightly less than 6.6 MeV, close to that of Es^{253} alphas. In our experiments Es^{253} grows in, both from Fm^{253} and Fm^{257}, via Cf^{253}. The production cross section for Fm^{253} is around 200 μbarns and that of Fm^{257} (directly and via Md^{257}) approximately 15 μbarn. For this reason the final But-column for the combined Fm fractions was run after about two months had elapsed since the last bombardment. By that time the three day[5] Fm^{253} was reduced by a factor of 10^6.

The fermium fraction was electroplated and Cm^{244} added as a tracer to check yield and stability of the analyzer. Following the separation no alphas from Fm^{253} or Es^{253} were recorded. Instead a complex α spectrum was observed consisting of three groups at 6.525 MeV ± 5 KeV, 6.57 ± 10 KeV, and 6.60 MeV ± 10 KeV. The combined activity of these groups was ca. 0.5 c/m (1.5 dis/m) and the ratios between them 2:2:1. There was considerable tailing of the peaks towards higher energies indicating the presence of electrons in coincidence with the alphas and thus that these alphas are not ground state transitions.

The peak at 6.60 MeV and partly that at 6.56 MeV in time became obscured due to the growth in of an alpha group at 6.63 MeV coming from Es^{253} and hence the identification of these groups is less certain. They may in fact consist of 6.525 MeV alphas and coincident conversion electrons. Typical spectra from early and later parts of the analysis are given in Fig. 3.

The growth in of Es^{253} can only be explained by the presence of Fm^{257}:

$$Fm^{257} \xrightarrow{\alpha} Cf^{253} \xrightarrow[18\ d]{\beta-} Es^{253} \xrightarrow[20\ d]{\alpha}$$

The decay of the peak at 6.525 MeV (since this is not effected by alphas from Es^{253}) and growth in of the 6.633 MeV alphas from Es^{253} were

analyzed. The former had a half-life of 80 days and the latter corresponded to a grandmother with similar half-life.

The analysis was terminated after about 50 days during which some 3×10^4 alphas had been recorded. The sample then was bombarded with neutrons as will be discussed later. At this time the intensities of the Es^{253} peak and that of the three others were about equal. This shows that the bulk of the alphas observed after separation was indeed due to Fm^{257}.

Hulet et al[6] have reported Fm^{257} to decay mainly by alpha emission and with a half-life of 79 ± 8 days. This isotope was produced in prolonged bombardment of Cm with neutrons in a reactor. They also observed a complex alpha group around 6.6 MeV with an intensity of 0.4 dis/m that they were not able to resolve. This intensity was shown to increase by 20% over a period of 60 days after which it decayed. The sample was analyzed for about 100 days and the conclusion was drawn that this increase was due to the growth in of Es^{253} and thus that the alphas in the fresh sample were due to Fm^{257}. The results from these two sets of experiments are thus in good agreement.

2. Fm^{258}

The isotope Fm^{258} is expected to emit alpha particles with an energy of about 6.6 MeV with an alpha half-life of some 300 days.[7] Spontaneous fission half-life guesses have ranged from seconds to days. The alpha daughter would be Cf^{254} which decays by SF with a half-life of 56 days.[8]

The ratio of SF to the alphas assigned to Fm^{257} was observed to be 1:500 and 1:600 in the experiments by Hulet et al[6] and by us respectively. No evidence could be presented for this SF activity to be due to Fm^{258} or Cf^{254} and hence for Fm^{258} to have a half-life longer than one month.

Gatti et al detected an SF emitter with a half-life of about 11 days in the fermium fraction after neutron irradiation of curium.[9] They suggested this emitter to be either Fm^{257} or Fm^{258}. The evidence presented above eliminates the former possibility.

To look specifically for this emitter, our Fm^{257} sample was subjected to neutron flux in the Idaho Materials Testing Reactor (MTR). Here it received 1.2×10^{20} n/cm^2 over a period of eight days. The fermium fraction was chemically separated after bombardment so that the effective number of Fm^{257} atoms irradiated could be established and to eliminate Cf^{254} interference in the sample. The latter isotope would be produced in the pile from the capture of neutrons by Cf^{253}, the alpha daughter of Fm^{257}.

The analysis of the fermium fraction was started one day after the end of bombardment. Again the alphas from Fm^{257} were observed. Only four drops of the fermium peak were analyzed and were found to contain 2.5×10^4 atoms of Fm^{257}.

Only 41 fission events were recorded of which 7 were background events over a period of 50 days. Within statistical errors (one standard deviation this SF activity could be due to Fm^{257}. An upper limit of 2 barns can be set for the production of a 10-day SF emitter in neutron irradiation of Fm^{257}. This is a very low neutron capture cross section for an even-odd nucleus. However it would not be unique in this region. The nuclide Cf^{253} has a cross section of only 2 barns.[9]

The nuclide Fm^{258} should have been produced in our heavy ion bombardments. Here no direct evidence was found for the existence of a 10-day SF emitter. If this species has an SF half-life shorter than days its presence will be masked by the intense SF activity from Fm^{256}.

IV. DECAY SYSTEMATICS

In Table III are given predicted values, taken from Ref. 7 for Q_α and Q_{EC}, of the energy releases in ground state α and EC transitions respectively. We have further given the experimental values Q_α^{exp} for the alpha groups observed and the calculated values[7] for the partial alpha half-life t_α in an unhindered decay of energy Q_α^{exp}. The values for t_α are to be compared to those for the experimental partial α half-life t_α^{exp}. The partial half-lives t_{SF} for spontaneous fission are also listed.

For the even-even isotope Fm^{256} we should have $Q_\alpha = Q_\alpha^{exp}$. The agreement is good. The estimated partial α half-life for this isotope is longer than the experimental one. However in the alpha group, in addition to alphas to the ground state transition, are included alphas to the 2+ rotational level. Generally the energy of the latter alphas will be around 45 KeV lower in energy than the former and their abundance around 15%. Corrections for this effect will bring the half-lives in reasonable agreement with each other. This suggests two things: (1) the α decay is favored, and (2) the isotope decays predominantly by α emission. In addition assuming the Q_α value to be correct, the alpha emitter is indeed Fm^{256}.

For odd A and odd odd isotopes we should in general have $Q_\alpha > Q_\alpha^{exp}$ since here the dominant alpha group normally does not correspond to the ground state transition. This appears to be the case also for such isotopes listed in Table III.

The energy level diagram of Nilsson has been successful in predicting the particle levels of odd A nuclei.[11] According to his scheme the ground state of the nuclides Md^{255} and Md^{257} and their daughters Es^{251} and Es^{253} are [514] 7/2- and [613] 7/2+ respectively and hence the transition between these two states will be unfavored. However an excited proton level,

[514] 7/2-, in the daughters is expected to which a favored decay will take place. As seen from Table III the dominant groups of Md^{255} and Md^{257} do in fact decay with half-lives that are close to the estimated ones. These transitions are therefore favored. From the values for Q_α we deduce the [514] 7/2- proton state in Es^{251} and Es^{253} to be around 350 KeV above the ground state. Nilsson's diagram indicates a relatively large gap between the [633] 7/2+ and the [514] 7/2- levels although it does not pretend to predict their energies quantitatively.[11] This gap will be responsible for relatively long α half-lives for odd A Md isotopes. However the energy difference between these levels does not necessarily have to be independent of A (Cfr. decay schemes of Am^{239}, Am^{241}, and Am^{243} given in Ref. 12 where it increases slightly with increasing A.)

The alpha group at 7.24 MeV for Md^{257} has a hindrance factor around 50 and belongs perhaps to a transition to the rotational band based on the [521] 3/2- level in Nilsson's particle diagram.[11]

The ground state configuration of Fm^{257} cannot readily be deduced from Nilsson's diagram[11] since there are many close-lying levels. Apparently the 6.525 MeV alphas belong to a favored transition between the same neutron states in Fm^{257} and Cf^{253}.

For odd odd nuclides no clear systematics exists. We see that Md^{256} α decays very nearly unhindered to a level in Es^{252} that is around 350 KeV above the ground state.

V. CONCLUSIONS

In the course of these experiments we have discovered the nuclides Md^{257} and Fm^{257} and for the first time observed alphas from Md^{256} and Fm^{256}. We have further confirmed previously reported decay properties for Md^{255}, Md^{256}, and Fm^{256}. We have not been able to verify the existence of a 10-day SF emitter of fermium.

As to the possibility of producing still heavier Fm and Md isotopes by the use of heavy ions, one can only speculate. The extrapolation of the cross section to Md^{257}, as was done above, is probably valid. This isotope is produced by the net transfer of (3p + 2n) of Cf^{252} and (3p + 4n) to Cf^{250}. According to results from reactions with lighter targets there is a maximum in the yield when the number of transferred neutrons and protons is equal.[14] Hence the cross sections for Md^{258} and Md^{259} should be comparable to those of the lighter ones. For still heavier isotopes the yield will decrease with increasing mass number. Higher yields would be expected when neutron rich ions such as B^{11} and C^{13} are used.

From the variation of EC half-lives with Q_{EC} for the Md isotopes it is apparent that Md^{258} might have an EC half-life of the order of hours. Its alpha half-life should be several days and consequently alphas from this isotope should be very difficult to detect.

The isotope Md^{259} is listed by Swiatecki and Myers[13] as having positive Q_{EC}; however, Foreman and Seaborg[7] predict this isotope to be β stable. In any case its alpha half-life should be too long to be observed in the present experiments.

The cross sections for the Fm isotopes show an interesting pattern. The values for isotopes of mass 255 and lighter are at least one order of magnitude higher than that for Fm^{256} and Fm^{257}. The former are probably produced

in an alpha transfer followed by neutron evaporation. Such a transfer has a high probability and is presumably connected with an alpha structure of the ion. The isotope Fm^{256} corresponds to the net transfer of (2p + 2n) to Cf^{252} or of (2p + 4n) to Cf^{250}. Hence, according to arguments used above for Md isotope production, there will be a decrease in cross section with increasing A past A = 256. Again a neutron rich ion might be more advantageous to use. It appears to be possible to produce Fm^{258} and Fm^{259} with a reasonable cross section by this method. The latter isotope will have an alpha half-life of the order of years and hence its alphas could not be observed by us. It is either β^- unstable[7] or β stable.[13]

VI. ACKNOWLEDGMENTS

We are indebted to Alfred Wydler for the design and building of the solid state detection device, to Mrs. Roberta Garrett for help in the chemical separations, to Charles Corum for the design of the target ensemble, and to the HILAC crew for their patient cooperation during the many months of bombardment. The initial separation of californium from the neutron irradiated curium was undertaken by Sherman Fried and his group for which we express our gratitude. Miss Judy Hartwell showed much patience in typing this manuscript.

Table I. Nuclear properties of fermium and mendelevium isotopes studied.

Nuclide	Mode of Decay	Half-Life	Alpha-Particle Energy (MeV)	Branching Ratio
Md^{255}	α,EC	30 min[a] 35 ± 10 min	7.34[a] 7.34	EC/α = 9± 2
Md^{256}	α,EC	1.5 hr[a] 1.5 ± 0.2 hr	7.17	EC/α = 30 ± 5
Md^{257}	α,EC,SF?	3 ± 0.5 hr	7.07, 7.27?	(SF + EC)/α = 12 ± 5 SF/α < 1
Fm^{256}	A,SF	2.7 hr[a] 2.7 ± 0.1 hr	6.85	SF/α = 35 ± 10
Fm^{257}	α,SF	79 d[b] 80 ± 5 d	6.6 complex[b] 6.525, 6.57? 6.60?	SF/α = 2 x 10^{-3}[b] SF/α = 1.4 x 10^{-3}
Fm^{258}	SF?	11 d[c]	-----	-------

[a]Taken from Ref. 2

[b]Taken from Ref. 6

[c]Taken from Ref. 9

Table II. Maximum cross section[a] in μbarn (10^{-30} cm) for production of fermium and mendelevium isotopes with B^{11}, C^{12}, and C^{13} incident on $Cf^{250,252}$.

Ion	Fm^{253}	Fm^{254}	Fm^{255}	Fm^{256}	Fm^{257}	Md^{255}	Md^{256}	7.07 MeV - 3 hr
B^{11}	300	150	150	10		4	9	.5
C^{12}	---	350	350	9	7.5[b]	6	7	.5
C^{13}	---	300	150	20		3	4	.3

[a]Not corrected for loss of recoil atoms in the target.
[b]Average value for the three ions used.

Table III. Decay systematics of fermium and mendelevium isotopes studied.

	Md^{255}	Md^{256}	Md^{257}		Fm^{256}	Fm^{257}			Fm^{258}
Q_α(MeV)[a]	7.83	7.76	7.50		6.95	6.78			6.62
Q_{EC}(MeV)[a]	1.1	2.1	0.5		Stable	Stable			Stable
Q_α^{exp}(MeV)	7.46	7.28	7.18	7.38	6.96	6.63	6.67?	6.70?	-------
t_α	3.9 hr	20 hr	51 hr	8 hr	150 hr	175 d	110 d	70 d	300 d
t_α^{exp}	6.0 hr	47 hr	39 hr	400 hr	100 hr	200 d	200 d	400 d	-------
t_{SF}	-----	------	>30 hr		2.7 hr	3.7×10^4 d[b], 4.8×10^4 d (±30%)			------

[a]Taken from Ref. 7

[b]Taken from Ref. 6

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CAPTIONS

Fig. 1

Alpha spectrum of the Md fraction from four 3-hour bombardments of 40/μg of $Cf^{250,252}$ with B^{11}, C^{12}, and C^{13}. Average beam intensity was about 7 μAmp/cm^2.

Fig. 2.

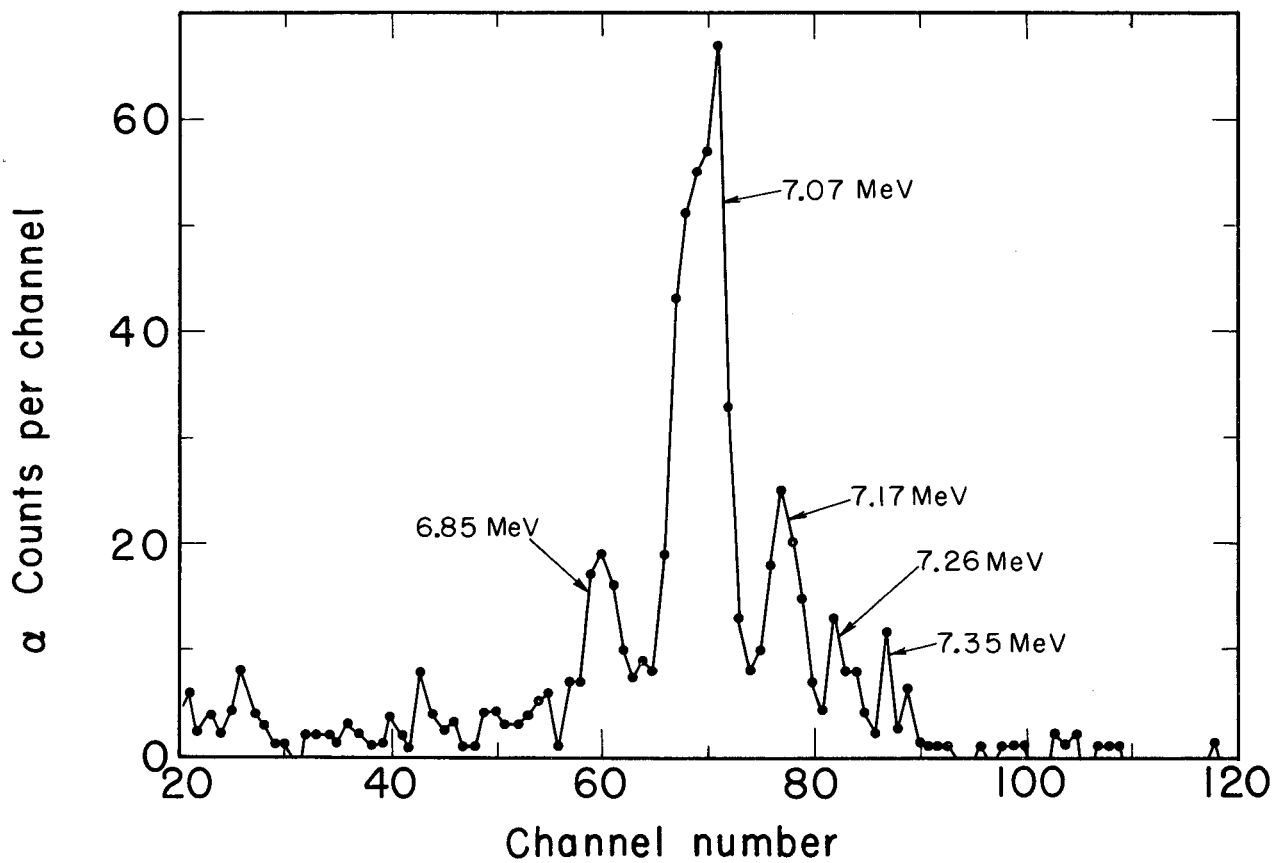
Decay curves of the alpha groups of Md shown in Fig. 1 and SF in the Md fractions.

Fig. 3.

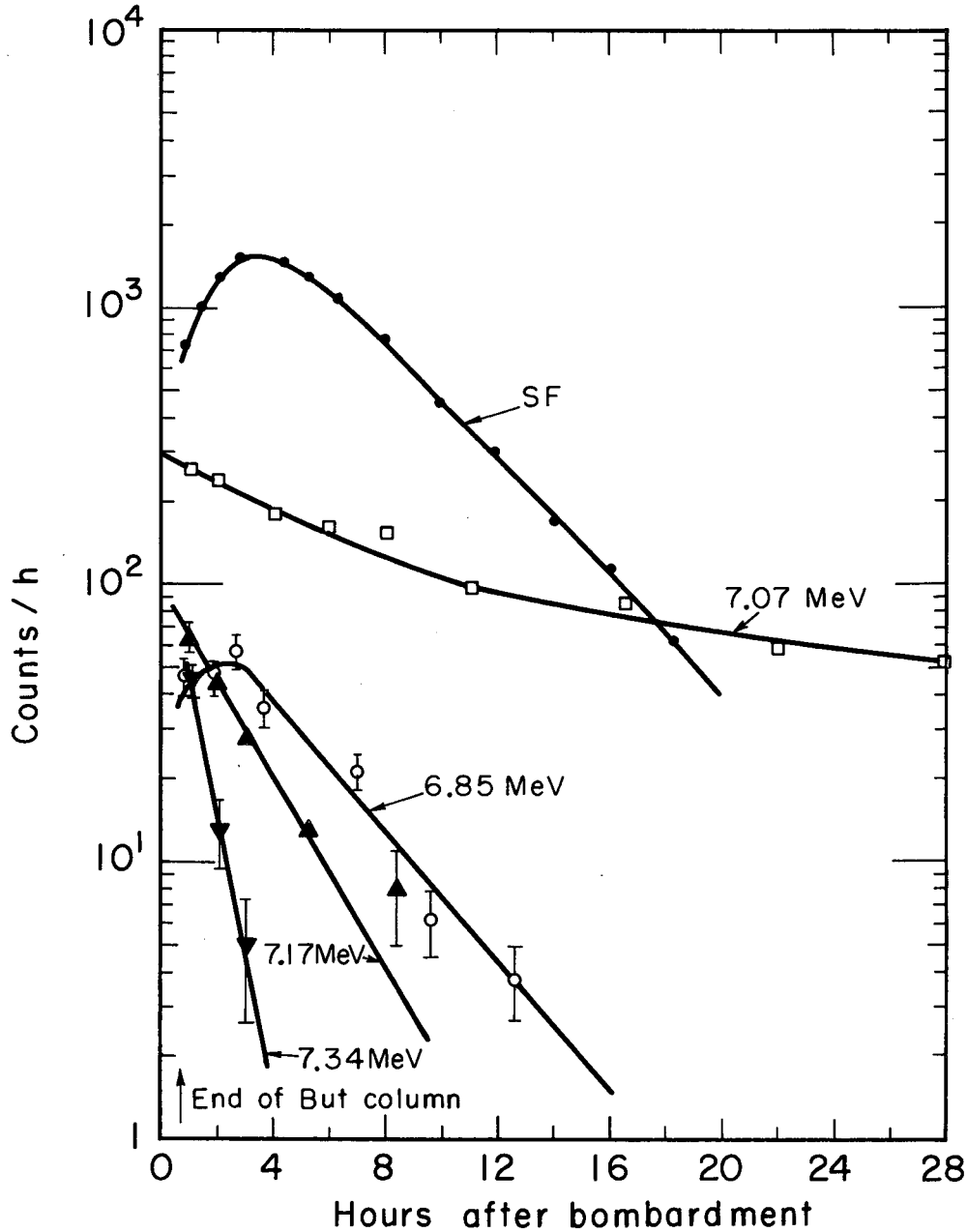
Alpha spectra of the Fm fraction. Last But column was run 14 July 1964.

Legend - o - accumulated counts from 24-31 July 1964,

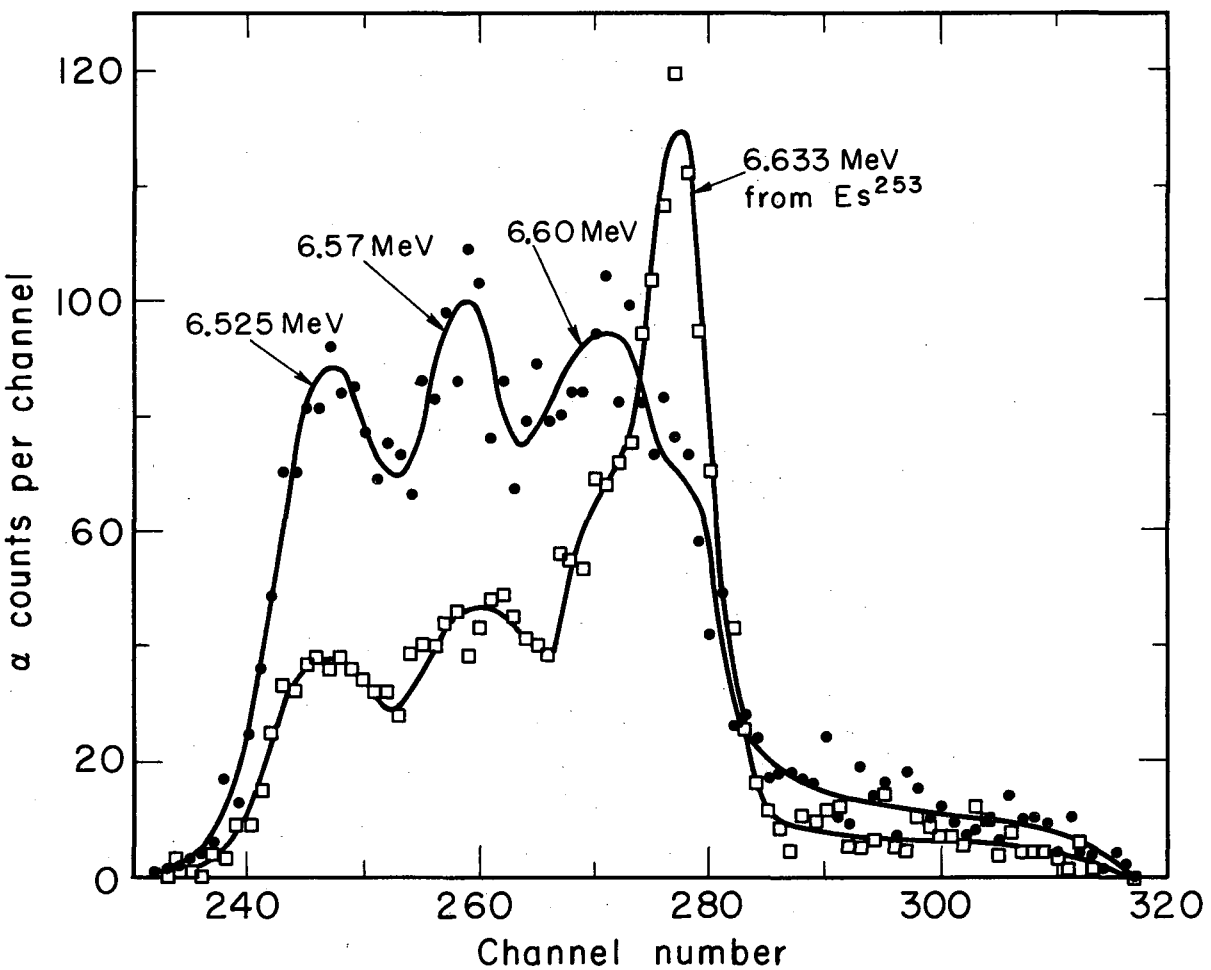
Legend - x - accumulated counts from 20 August - 3 September 1964.


60
40
20
0
α Counts per channel
7.07 MeV
7.17 MeV
6.85 MeV
7.26 MeV
7.35 MeV
20
40
60
80
100
120
Channel number

MUB-6294


10^4
10^3
10^2
10^1
1
Counts / h
SF
7.07 MeV
6.85 MeV
7.17MeV
7.34MeV
End of But column
0
4
8
12
16
20
24
28
Hours after bombardment

MUB-6295


120
100
60
20
0
α counts per channel
240
260
280
300
320
Channel number
6.525 MeV
6.57 MeV
6.60 MeV
6.633 MeV
from Es253

MUB-6296

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