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ABSTRACT

The new isotopes E^{249}, E^{250}, E^{251} and E^{252} were produced by bombardment of Bk^{249} with helium ions. A recoil method was used to separate the bombardment products from the target material, so that the target could be used repeatedly.

Approximate excitation functions for the formation of the einsteinium isotopes were measured as well as their half lives, α-particle energies, and electron-capture branching ratios.

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INTRODUCTION

The bombardment of heavy-element nuclei with charged particles leads mainly to fission.[1] The cross sections of reactions such as (α, xn) are therefore much lower for target nuclei of $Z > 90$ than they are for lighter elements. The excitation functions of several reactions of the (α, xn) and (α, xpyn) types have been studied for target nuclei up to Cm^{244}.[1, 2] It is of some interest to investigate similar reactions with even heavier targets to see whether the same pattern of competition between fission and spallation is maintained.

In the work described herein, the 280-day β^--emitting Bk^{249} was bombarded with helium ions of from 20 to 40 Mev, and reactions of the type (α, xn) were studied radiochemically. Such reactions can produce four previously unobserved isotopes of einsteinium (symbol E, atomic number 99) with mass numbers from 249 to 252. E^{248} could presumably be produced by (α, 5n) reaction at the highest energies, but in fact it was not observed.

The production of Bk^{249} by multiple neutron captures and β^- decays, starting with Pu^{239}, has already been described.[3] The amount of Bk^{249} available was only 1.2×10^{-8} g, or about 3×10^{13} atoms. Hence it was necessary to use all of it in each bombardment, and to use the highest available helium-ion beam density in the Crocker Laboratory 60-inch cyclotron.

These requirements were met by using a deflector-channel target assembly previously developed for the production of the new element mendelevium (atomic number 101).[4] The target material was deposited as a very thin layer over a

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

rectangular area 0.6 x 6 mm on a 0.005-cm-thick gold foil. (This small target area was chosen to match the area of the most intense helium-ion beam.) The target area was exposed to a collimated helium-ion beam whose intensity varied from about 20 to 75 μamp /cm^2, depending on how well the cyclotron was adjusted. The beam passed first through the gold foil and then through the Bk^{249} target, so that nuclear reaction products were ejected from the target in the forward direction. They were caught on a 0.00025-cm-thick gold "catcher" foil placed about 5.5 mm from the target foil. By this method it was possible to bombard the Bk^{249} target a large number of times without the necessity of dissolving it. Some 45 bombardments, many of them for as long as 9 hours, were made with no loss of target material.

The energy of the helium ions was varied by inserting aluminum degrading foils into the beam ahead of the target. The helium-ion beam intensity was measured with a Faraday cup placed behind the thin gold catcher foil. The Bk^{249} target also contained a known amount of Cm^{244}, from which Cf^{246} was produced by the (α, 2n) reaction. From the known cross section for this reaction[5] and the measured amount of Cf^{246} finally separated from the catcher foil, the over-all yield was calculated for each bombardment. The yield value obtained in this way included both the recoil collection efficiency and the chemical handling loss. In almost every case, yields between 60 and 90% were obtained.

EXPERIMENTAL

1. Target Preparation

The recoil of At^{211} atoms produced by bombardment of Bi^{209} with helium ions was investigated by Chetham-Strode.[4] For helium ion energies of 20 to 40 Mev, he found that high ejection yields of nuclear reaction products should be obtained if the density of the target deposit was less than about 30 μg /cm^2. Since the target area was only 0.036 cm^2, the total weight of target and impurities could not be allowed to exceed 10^{-6} g. Preliminary experiments with targets of Cm^{244} showed that it was possible in practice to obtain ejection yields very close to 100% for the (α, 2n) reaction. The Cf^{246} produced in this case was found to be projected forward into a rather narrow cone.

Radiochemically pure Bk^{249} was isolated by precipitation and ion-exchange methods, which have been described earlier.[6] At this point, a known amount of Cm^{244} was added. Final chemical purity was obtained by elution of the berkelium and curium with pure 6M hydrochloric acid through a bed, 4 cm by 0.3 cm in diameter, of purified Dowex-50 cation-exchange resin (4% cross-linked). By use of a very low flow rate of eluant, the berkelium and curium were eluted as a very sharp band in a volume of only about 0.1 ml. The solution was then evaporated to dryness.

The deposition of the mixture of Bk^{249} and Cm^{244} onto the gold target foil was made electrolytically. The Bk and Cm were dissolved in 6M ammonium chloride of high purity at a pH of about 2, and transferred to a cell in which the annealed gold target foil was the cathode. The bottom of the cell (Fig. 1) defined the area over which the target materials would deposit. A carefully cleaned platinum wire anode was used. A current density of about 1 amp/cm^2 of cathode surface, maintained for about 40 min, brought about the deposition of 80 to 90% of the target material onto the gold foil. After the ammonium chloride was rinsed off, the foil was washed with acetone and heated to a dull red to expel volatile substances. Finally it was fastened into a stainless steel holder (v. infra) with Epon* resin.

The amount of Bk^{249} in the target was estimated by counting aliquots of the cell solution before and after the deposition. A windowless proportional counter was assumed to give a counting efficiency of 90% for the β^- particles from Bk^{249} mounted on thick platinum plates.

2. Cyclotron Probe

An expanded schematic view of the deflector-channel probe is shown in Fig. 2. The helium-ion beam was first collimated by means of the water-cooled copper and graphite collimator A. The second collimator B reduced the beam to the required size. Stainless steel holder C supported to 0.04-cm-thick duraluminum foil which sealed the rest of the probe assembly from the cyclotron vacuum. Aluminum absorber strips could be mounted behind the duraluminum foil.

* An epoxy resin supplied by Shell Development Company, Emeryville, California

Holder D supported the target. The foils C and D were cooled by means of rapid circulation of helium between them. The gold "catcher" foil E was mounted on the end of rod F with Scotch tape. The rod F terminated in a water-cooled aluminum block which served as the Faraday cup. The space between the target and the catcher foil was evacuated. The several cooling and vacuum systems were interlocked so that the cyclotron could not be operated until proper conditions were obtained.

The rod F could be very rapidly withdrawn after completion of a bombardment, so that no time was lost in examining the products. Holders C and D sealed into the assembly with O-rings so that they too could be readily withdrawn to permit a change of target or absorber.

The target and collimator B assembly could be moved vertically by remote control so that the most intense beam could be located. Cooling of the foils was adequate to permit a beam as large as 200 μamp /cm^2 to be used over prolonged periods of time, but such intense beams were not commonly available.

3. Chemical Separations

At the end of a bombardment, the Faraday cup rod was removed and the catcher foil torn off. The foil was dissolved in a few drops of 8M hydrochloric acid containing a little nitric acid. The solution was then forced rapidly through a 5-by-0.3-cm bed of Dowex-1 anion-exchange resin. The resin bed was heated to 87°C by means of an outer jacket through which trichloroethylene vapor was circulated. Under these conditions, gold was very completely retained by the resin, while the tripositive actinide elements passed through. The resin bed was washed with a few drops of 8M hydrochloric acid.

The solution thus obtained was evaporated to dryness. The several actinide elements were then separated from one another by means of elution through a bed of Dowex-50 cation-exchange resin, with a solution of ammonium α-hydroxy isobutyrate as eluant. This, and the other chemical procedures, have been fully described elsewhere.[6] The α-radioactivity of the separated actinide element fractions was then examined with a gridded ionization chamber and pulse-height analyzer.

In experiments where the electron-capture activities were to be examined, further decontamination from radioactive impurities was required. The actinide element fraction from the Dowex-1 anion-exchange bed was purified by coprecipitation of the actinide elements with a carrier of lanthanum fluoride. The precipitate was redissolved in a saturated solution of boric acid. The actinide elements were further purified (and separated from the lanthanum carrier) by elution through a bed of Dowex-50 cation-exchange resin with saturated hydrochloric acid containing 20% ethyl alcohol.[6] The final separation of the actinide elements from one another was then made as described above. The full chemical procedure required about 2 to 2.5 hours.

NUCLEAR PROPERTIES OF EINSTEINIUM ISOTOPES

Four previously unobserved isotopes of einsteinium were produced in this work. Their mass assignments were made initially from the systematics of decay[7] and confirmed by means of excitation functions. Their properties are summarized in Table I.

Table I. Nuclear Properties of Einsteinium Isotopes

Isotope	Type of decay	Half life	α-Particle energy (Mev)	Ratio of electrons capture to branching
E^{249}	EC, α	2 hr	6.76	760
E^{250}	EC, α?	8 hr	Not observed	α Not observed
E^{251}	EC, α	1.5 days	6.48	190
E^{252}	α, EC?	~140 days	6.64	EC Not observed
		$t_{1/2\ \beta^-} > 20$ yr		

The electron-capture decays were observed in a windowless proportional counter, for which a counting efficiency of 80% was assumed for thin samples

mounted on platinum plates. It was not possible to measure the counting efficiency for any of the nuclides involved, but the assumed counting efficiency is based on the general experience of this laboratory.

The observed α-particle energies (not necessarily ground-state transitions) of the known isotopes of californium and einsteinium are plotted against their mass numbers in Fig. 3. The einsteinium isotopes show a break associated with a neutron number of 152, which was first observed for the californium isotopes.[8] A similar break has also been observed for the isotopes of fermium.[9] (In Fig. 3, data are included for the recently discovered nuclides E^{248} and Fm^{251}.[10, 11])

The half-life measurements were most accurately made by resolution of proportional-counter decay curves such as that shown in Fig. 4. The decay of E^{252}, however, was observed only by means of α-pulse analysis, which yielded the decay curve shown in Fig. 5. The decay of the other nuclides was also studied by α-pulse analysis to check the half-life values obtained in the proportional counter.

REACTION CROSS SECTIONS

The yields of the four new isotopes of einsteinium were measured in some experiments with a windowless proportional counter and in others by α-pulse analysis. The accuracy of the cross-section measurements was limited to about ± 30% by (1) uncertainty in the amount of Bk^{249} in the target (the very soft β^- activity of Bk^{249} is not easy to count accurately, since the counting yield is very sensitive to the thickness of the sample), and (2) counting statistics in those experiments in which yields were measured by α-pulse analysis. For example, less than 20 events of the 6.76-Mev α-particle of E^{249} were usually observed.

The β^- decay of the Bk^{249} produced Cf^{249}, so that bombardment of the target yielded progressively larger amounts of Fm isotopes whose electron-capture decay produced further amounts of E^{250}, E^{251}, and possibly E^{249}. Reactions such as Cf^{249} (α, p2n) E^{250} presumably also occurred.

The Bk^{249} (a, n) and (a, 2n) reactions were studied at a time when the amount of Cf^{249} in the target was negligible. The E^{250} was produced by Bk^{249} (a, 3n), by Cf^{249} (a, p2n), and by Cf^{249} (a, 3n) $Fm^{250} \xrightarrow{E.C.} E^{250}$. Although the atom ratio of Bk^{249} to Cf^{249} in the target was always larger than 2:1, a large fraction of the E^{250} was probably produced by the Cf^{249} (a, p2n) and (a, t) reactions, since reactions of this type appear to have unexpectedly large cross sections.[12] It was not possible to distinguish between the various modes of production of E^{250}, so that the excitation curve shown in Fig. 6 is a sum of the cross sections and serves mainly to confirm the mass assignment of E^{250}. The cross sections given for the (a, n) curve are based on the assumption that E^{252} is β-stable. This is almost certainly not true. Hence the cross sections for Bk^{249} (a, n) E^{252} represent lower limits, but probably fairly close ones.

The (a, n), (a, 2n), and (a, 4n) curves in Fig. 6 are very similar in shape and magnitude to those obtained for other somewhat lighter nuclides such as Pu^{238} or Pu^{239}. The cross sections are comparable in magnitude, showing that fission is not rapidly superseding spallation as the Z and A of the target are increased. (For a more detailed discussion, see Ref. 1.) The flatness of both the (a, n) and (a, 2n) curves is not readily to be explained by the usual compound-nucleus mechanism. To avoid fission or further particle evaporation, the residual nucleus must be left with less than about 6 Mev of excitation. This implies that much more energy is carried off by the neutrons in the (a, n) and (a, 2n) reactions than is required by an evaporation mechanism involving commonly accepted values for nuclear temperatures. The small peak in the (a, 2n) curve might then be attributed to a compound-nucleus-evaporation mechanism, while the long "tail" at higher energies must be due to a different mechanism- - perhaps a direct interaction of the helium ion with one or two neutrons.

Such a non-compound-nucleus mechanism implies the transfer of less momentum to the reaction product, and hence that the recoil collection efficiency might be less than expected. However, the Bk^{249} target deposit was sufficiently thin that recoils with much less than the full momentum would

escape. (This was shown by measuring in several bombardments the small amount of Bk^{249} ejected from the target by elastic and inelastic scattering processes, which gives a very sensitive measure of the thickness of the target.[13]) In addition, the measurement of the yield of Cf^{246} from the Cm^{244} added to the target automatically compensated for any such effects insofar as they occurred equally for nuclear reactions of Bk^{249} and for the (α, 2n) reaction of Cm^{244}.

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FIGURE LEGENDS

Fig. 1. Cell for electrodeposition of target.

Fig. 2. Cyclotron target assembly (schematic).

Fig. 3. α-particle energy vs neutron number for Z = 99 to 100.

Fig. 4. Windowless proportional counter decay curve for sample containing E^{249}, E^{250}, and E^{251}.

Fig. 5. Decay curve of 6.64-Mev α activity of E^{252}.

Fig. 6. Excitation functions for formation of einsteinium isotopes.

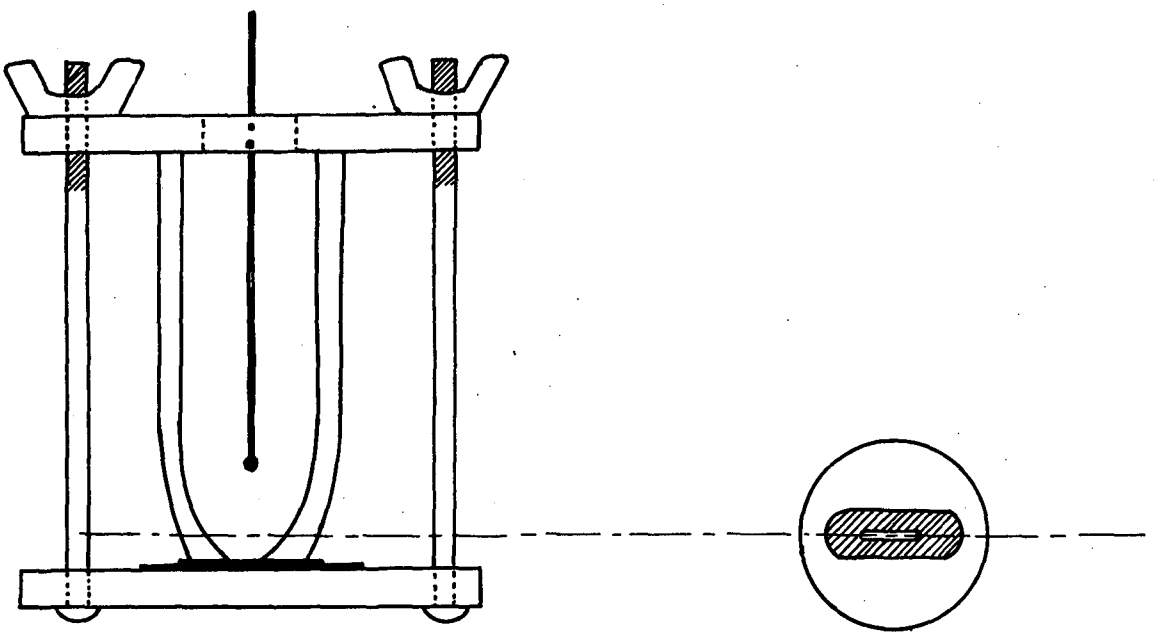
key

▼ Bk^{249} (α, n) E^{252}

● Bk^{249} (α, 2n) E^{251}

■ Bk^{249} (α, 3n) E^{250} + Cf^{249} (α, 3n) Fm^{250} → E^{250} + Cf^{249} (α, p2n) E^{250} + Cf^{249} (α, t) E^{250}

▲ Bk^{249} (α, 4n) E^{249} + Cf^{249} (α, p3n) E^{249}.



MU-11742

Figure 1

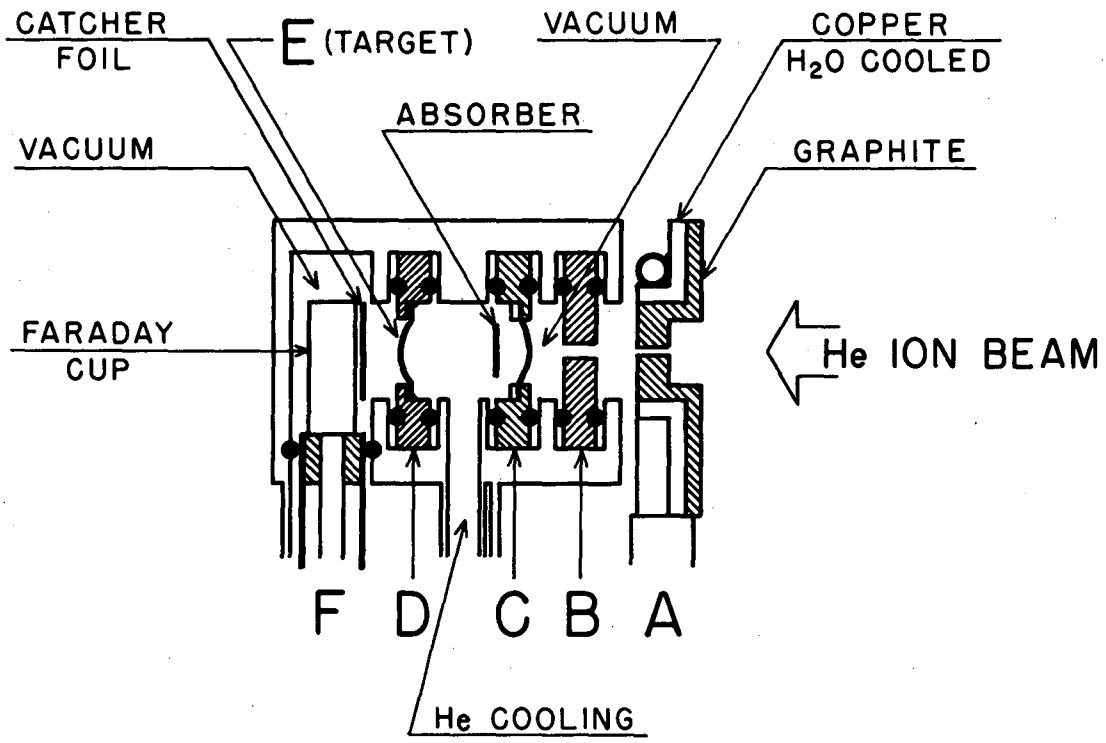

CATCHER FOIL
E (TARGET)
VACUUM
COPPER
H_2O COOLED
ABSORBER
VACUUM
GRAPHITE
FARADAY CUP
He ION BEAM
F D C B A
He COOLING
MU-11743

Figure 2

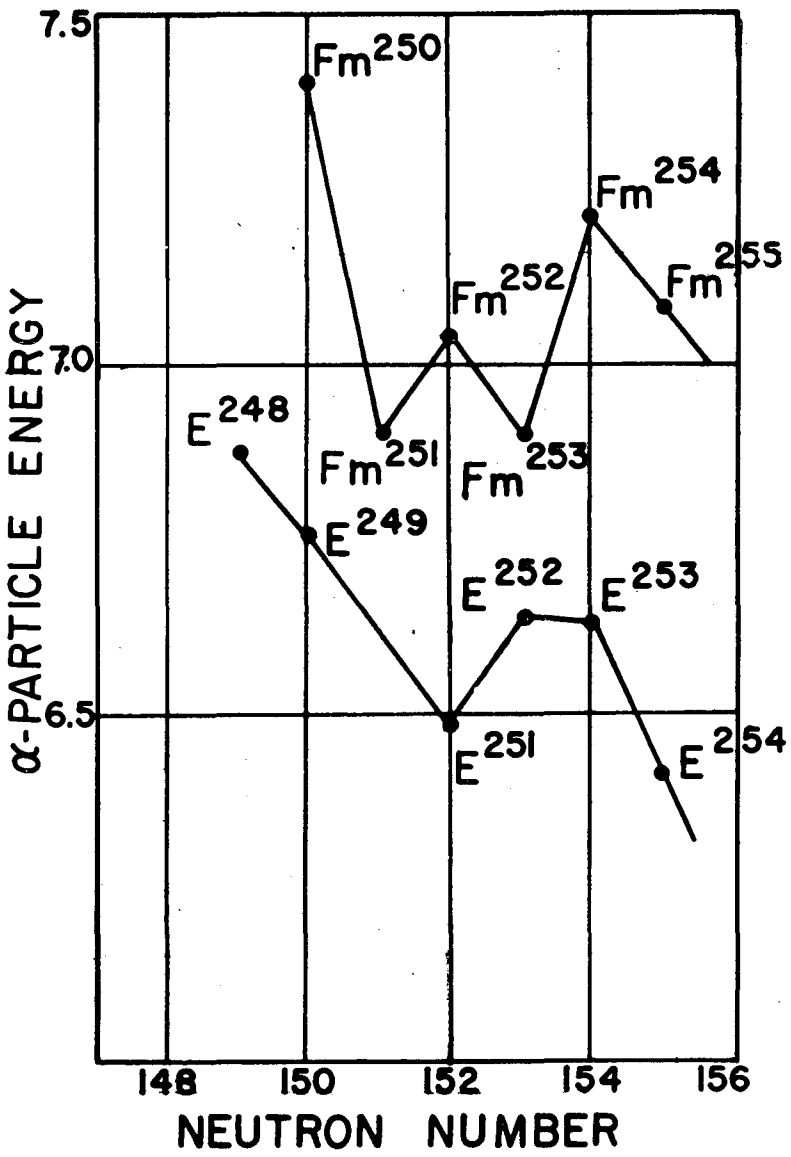

7.5
7.0
6.5
α-PARTICLE ENERGY
Fm250
Fm254
Fm255
Fm252
Fm251
Fm253
E248
E249
E252
E253
E251
E254
148
150
152
154
156
NEUTRON NUMBER
MU-11744

Figure 3

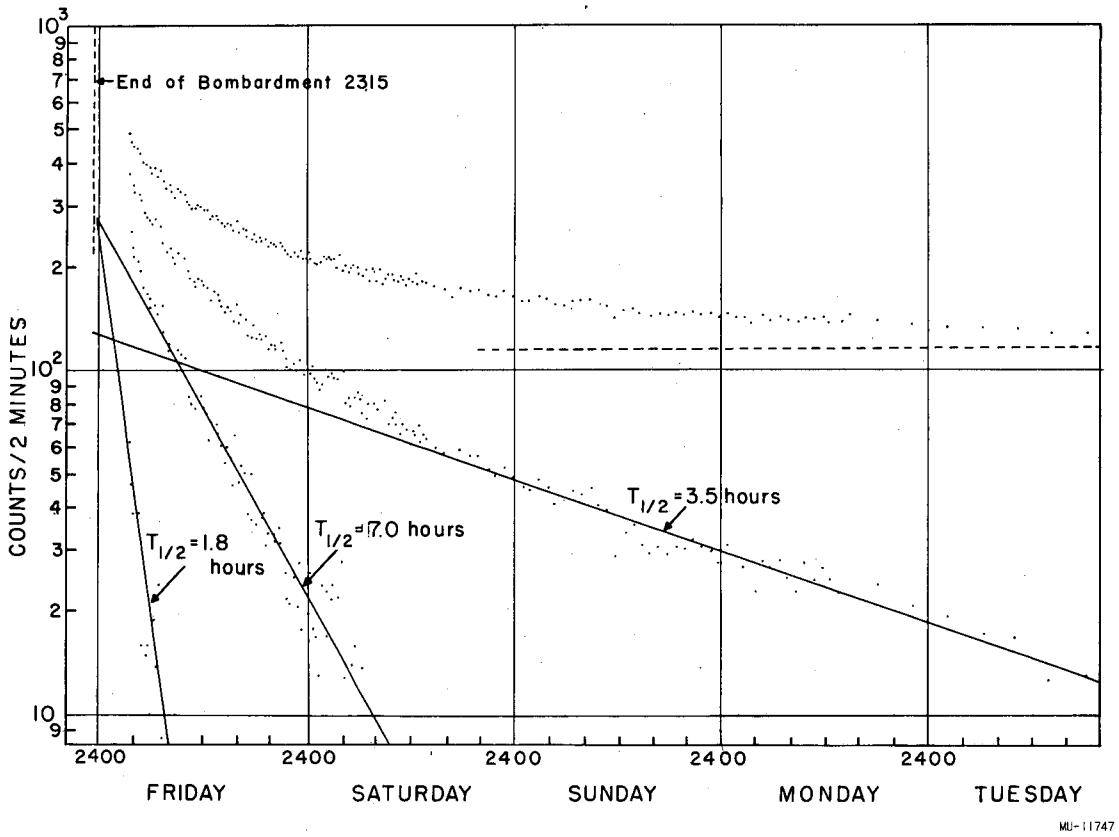

End of Bombardment 2315
COUNTS/2 MINUTES
$T_{1/2}$ = 1.8 hours
$T_{1/2}$ = 17.0 hours
$T_{1/2}$ = 3.5 hours
2400
FRIDAY
SATURDAY
SUNDAY
MONDAY
TUESDAY
MU-11747

Figure 4

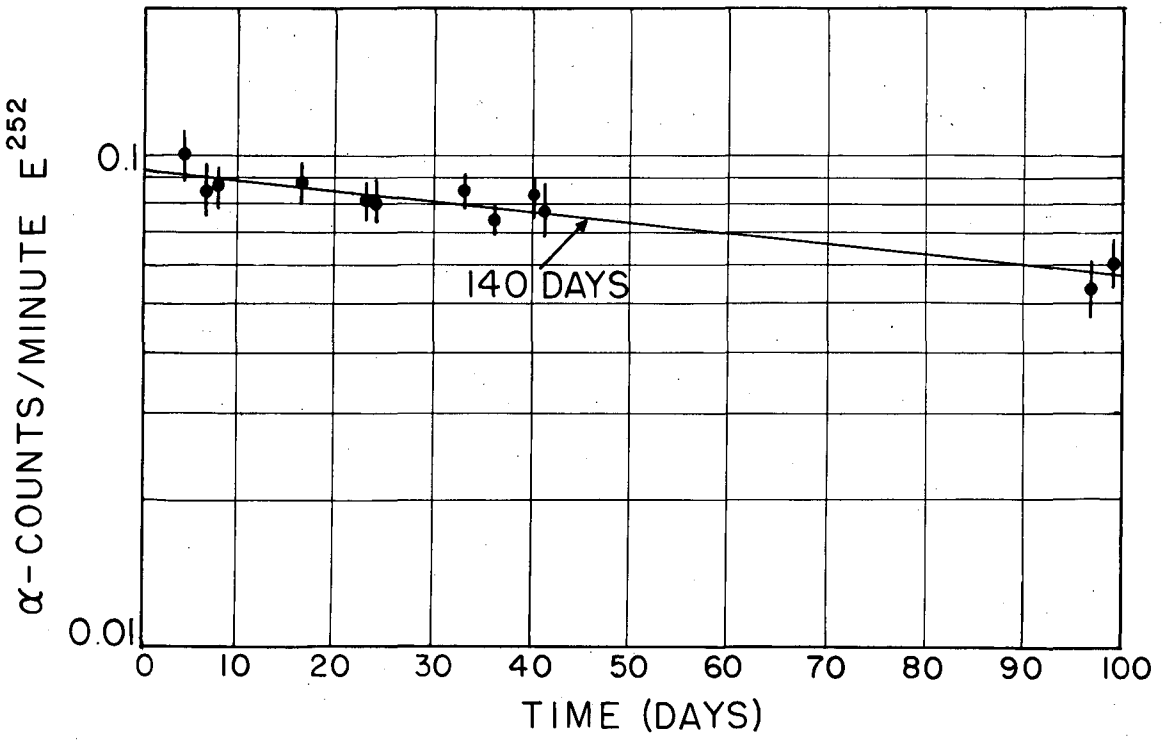

α-COUNTS/MINUTE E252
0.1
0.01
140 DAYS
0
10
20
30
40
50
60
70
80
90
100
TIME (DAYS)
MU-11745

Figure 5

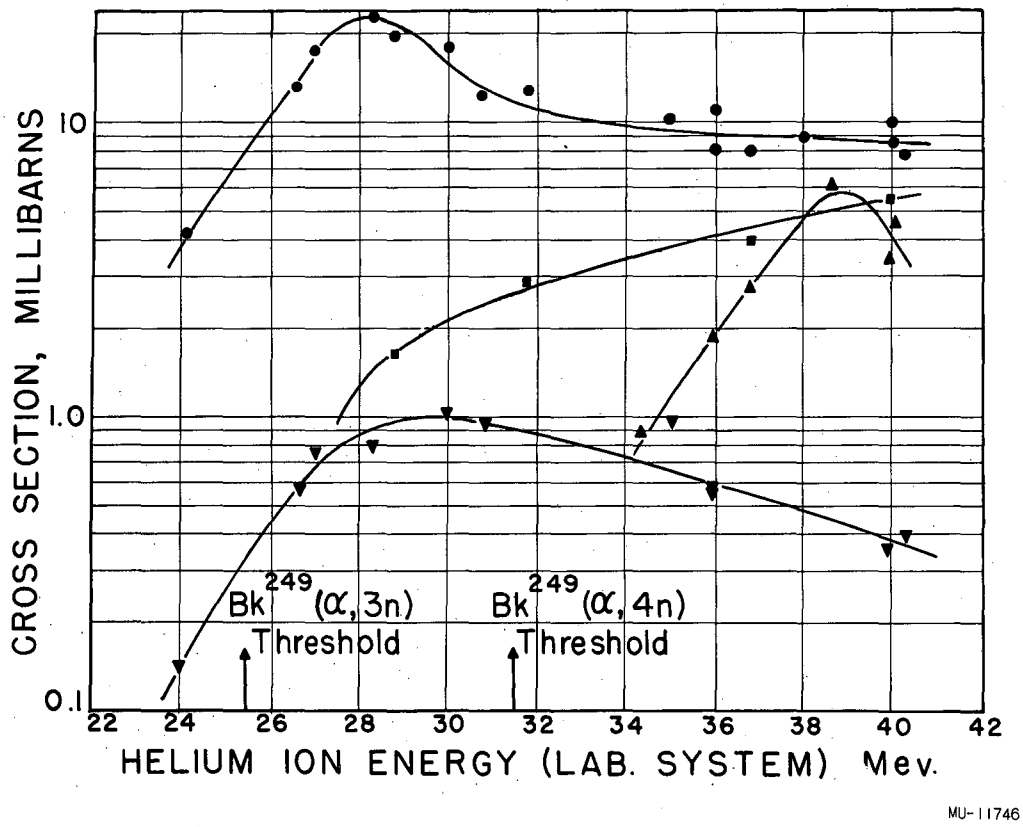

CROSS SECTION, MILLIBARNS
10
1.0
0.1
22
24
26
28
30
32
34
36
38
40
42
HELIUM ION ENERGY (LAB. SYSTEM) Mev.
Bk249(α, 3n) Threshold
Bk249(α, 4n) Threshold
MU-11746

Figure 6