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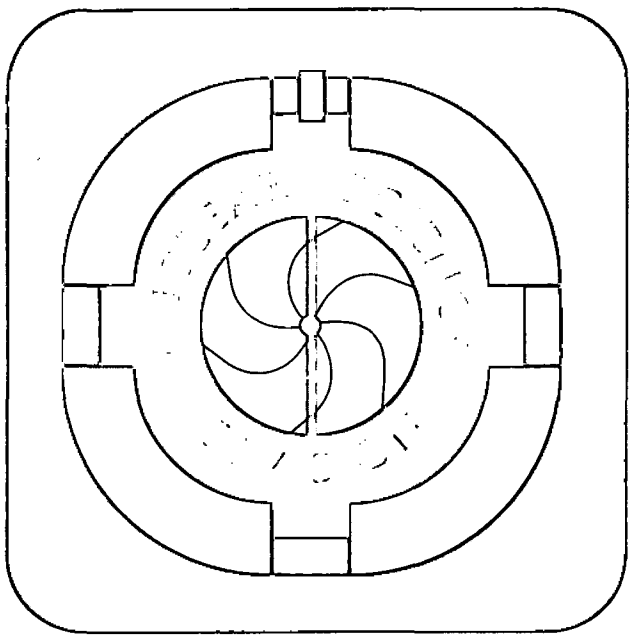

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DISCOVERY OF ^{253}Md

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Abstract

We have measured the half-life and production cross section of the new isotope ^{253}Md, produced via the ^{243}Am(^{13}C,3n) reaction. Isolation of Md from other activities was accomplished using elution with ammonium α-hydroxyisobutyrate from a cation exchange resin column. Experiments were performed with different irradiation time intervals, but the chemical separation always began and ended at exactly the same length of time after the end of irradiation. All separations with the same irradiation lengths were combined and analyzed for growth and decay of the 3.0-d ^{253}Fm daughter and 20.47-d ^{253}Es, granddaughter of ^{253}Md. The amount of ^{253}Es in each fraction depends on the length of each irradiation and the ^{253}Md half-life. An increase in the length of irradiation will cause a corresponding increase in the amount of the new isotope ^{253}Md and hence, in the amount of ^{253}Es produced, provided the length of irradiations are not very long compared to the half-life of ^{253}Md. In this way, the Md half-life was estimated to be about 6 minutes with a production cross section of the order of 50 nb.

1. INTRODUCTION

Element 101 was discovered in 1955 by A. Ghiorso et al. [1] at the Lawrence Berkeley Laboratory (LBL). In this experiment an ^{253}Es target was bombarded at the 60-Inch cyclotron with 40-MeV helium ions to produce ^{256}Md via the (α,n) reaction. It was found that ^{256}Md decays with a half-life measured at that time to be a little under an hour (now known to be 76 minutes), by electron capture (EC) to ^{256}Fm, which decays primarily by spontaneous fission with a half-life of 2.63 hours. Since the discovery of Md, all of the isotopes from ^{247}Md through ^{260}Md [2], with the exception of ^{253}Md, have been identified via excitation-function measurements and studies of genetic links to previously known isotopes of Fm and Es [3,4,5]. ^{253}Md has 152 neutrons, believed to be a deformed neutron subshell [6]. Based on the predicted Q values of 1.89 MeV and 7.70 MeV for the electron-capture and alpha-decay modes, respectively, this isotope is expected to have a half-life and decay modes similar to those for neighboring isotopes of Md. The half-life should be on the order of 10 minutes with electron capture being the dominant decay mode [7]. The alpha-decay branch should be less than a few percent. Presumably,the reason this isotope has not been identified is because the isotopes of Md from mass number 249 to 256 have similar electron-capture Q values and, therefore, similar half-lives, assuming the same type of transitions occur. In the past, the detection of ^{253}Md has probably been obscured by these other activities. The observation of the decay modes of this nuclide is of interest because it will add to the information available on the N=152 deformed subshell which may be important in the understanding of a similar, but stronger, deformed shell which has been predicted [8] to exist around N=162 to 164 for atomic numbers near 109. The immediate goal of this experiment was to discover this isotope and measure its half-life and production cross section.

2. PRODUCTION

^{253}Md was produced at the LBL 88-Inch Cyclotron by the ^{243}Am(^{13}C, 3n) reaction. Three ^{243}Am targets (containing 0.32 mg/cm^2, 0.34 mg/cm^2, and 0.37 mg/cm^2, 72% isotopic purity, 28% ^{241}Am), deposited by the molecular plating method [9] in 0.7-cm diameter spots on 2.59 mg/cm^2 molybdenum foil were used. The ^{243}Am targets were mounted in the Light Ion Multiple (**LIM**) target system [10], (Fig.1), with spacing of approximately two cm between the targets. A 25-μm beryllium foil served as the volume limiting foil, and another 25-μm beryllium foil served as the vacuum window for the system. The beam energy was chosen so as to result in ^{13}C^{4+} energies ranging from 66 to 74 MeV (laboratory system) in the targets, near

the maximum of 69 MeV for the 3n excitation function. A production cross section of 2.3 μb was calculated using the **SPIT** [11] program. A schematic diagram of the target system is shown in Figure 2. The 86-MeV $^{13}C^{4+}$ beam from the cyclotron was degraded to 74 MeV by passing through the 25 μm beryllium vacuum window, 0.17 mg/cm^2 of He and the 2.59 mg/cm^2 Mo target backing foil before passing through the first ^{243}Am target material. The beam then passed through 0.36 mg/cm^2 of He (1.2 atm), the second Mo target backing foil, and into the second ^{243}Am target material, at a energy of 70 MeV. After passing through the third 0.36 mg/cm^2 of He, the third Mo target backing foil, and the third ^{243}Am target material, at a energy of 66 MeV, it entered a fourth zone of 0.36 mg/cm^2 of He, before passing through a Be volume limiting foil. The beam intensity was typically on the order of 0.5-0.75 pμA. The reaction products were transported via the He/KCl aerosol jet transport system. The KCl aerosols for the He/KCl gas jet system were generated by heating crystalline KCl to 690 ^{o}C inside a quartz tube. Helium gas was used to sweep the aerosols out of the tube and into a 'presorter' capillary, where the larger aerosol particles were allowed to settle out of the He flow. The presorter is a 5 meter length of 2-mm i.d. polypropylene capillary tube which has been wound into several coils of about 15 cm diameter. The 'presorted' aerosols were transported directly into the target chamber via an 0.95-cm i.d. tygon tube as shown in figure 2. The recoiling reaction products were collected on the aerosols in helium, which swept out the volume behind each target continuously. The activity-laden aerosols were transported by the helium through 1.2-mm i.d. polyvinylchloride capillary tube to a collection site in a chemistry laboratory 6 m away.

3. CHEMICAL SEPARATION

A chemical separation procedure was designed for the isolation of Md from other recoiling activities. The separation was carried out using an ammonium α-hydroxyisobutyrate (α-HIB) elution from a cation exchange resin column [12]. This elution separates actinides and lanthanides according to their ionic radii. By adjusting the concentration and the pH of the α-HIB, a good separation between the Md and the Fm fractions was obtained. In order to locate the elution position of Md, ^{166}Ho^{m}, which has nearly the same radius as Md [13], was used. All separations were carried out using the Automated Chromatographic Chemical Element Separator System, **ACCESS**, which assures a rapid and reproducible separation [14]. Experiments were carried out with the same separation times but three different collection lengths, 5 min, 10 min and 20 min. These collection periods were alternated cyclically so that any variations in the beam current would average out in the final analysis. Beam currents were

also recorded for each experiment. The recoiling activities on KCl aerosols were collected on a Teflon surface, picked up with 0.035 ml of 0.05 M α-HIB solution with pH=2.0, and transferred to the top of a 2.0-mm i.d. by 70-mm long quartz column containing Hamilton, D-50, X-12 cation exchange resin, 10-15 μm particle size, kept at 80 ^{o}C. The separation began exactly one minute after the end of each irradiation period. The Md fraction was eluted with a solution of 0.50 M ammonium α-HIB, pH=3.4, with the separation ending exactly 10 min after the end of bombardment. The column was then rinsed with a 0.50 M α-HIB solution with pH=4.0 which washed all remaining activity off the column. The column was regenerated with 1 M ammonium chloride solution followed by a water rinse before the next experiment.

All fractions with the same irradiation times (34, 38, and 34 individual separations for 5, 10, and 20 min irradiation times, respectively) were combined, resulting in three ≈40 ml α-HIB solutions containing the three different Md fractions. A known amount of ^{241}Am tracer and 1.70 ml of concentrated hydrochloric acid (HCl), to protonate the α-HIB, were added to each of the three solutions. Each combined fraction was then passed through a 3.0-mm diameter by 30-mm long column of Dowex-50, X-4, 100-200 mesh cation exchange resin. The Md and daughter activities were adsorbed on the resin while the α-HIB passed through. The column was rinsed with 3.0 ml of 1.0 M HCl to remove mass and extraneous activity, and the activity was eluted from the column with 2.0 ml of 6.0 M HCl. This eluant was evaporated on a tantalum foil and analyzed for alpha activity with 20% efficiency using a Si(Au) surface barrier detector, fission/alpha spectrometer system. The chemical yields were estimated to be 91%, 99%, and 88% , for the 5, 10, and 20 min fractions respectively. A chemistry flow chart is presented in Figure 3.

4. DETERMINATION OF HALF-LIFE

The experiments were carried out so that the separations were always completed at the same time after the end of irradiations but cycles of different irradiation lengths were conducted, as stated above. The standard equation for the number of ^{253}Md atoms present at end of separation is:

$$N = (R/\lambda)(1-e^{-\lambda t})e^{-\lambda s} \quad (1)$$

where,

N= Number of ^{253}Md atoms at s

R= Production rate

λ= Decay constant for ^{253}Md

t= Irradiation time interval, and

s= Time from end of irradiation to Md separation

After the ^{253}Md decays by electron capture, the 12% alpha branch of the 3.0-d ^{253}Fm daughter at 6.943 MeV can be detected by alpha-pulse-height analysis. The amount of ^{253}Fm daughter in each fraction is dependent on the length of irradiation, and the Md half-life. ^{253}Fm also decays by electron capture (88%) to ^{253}Es ($t_{1/2}$=20.47 d) whose 90.6%, 6.663 MeV alpha branch can be measured (Fig.4).

5. RESULTS AND DISCUSSION

The direct observation of ^{253}Fm was not possible because ^{252}Fm ($t_{1/2}$=25.4 h, α=7.04 MeV), the daughter of ^{252}Md, produced by a 4n reaction, masks this peak. A comparison of Figures 5 and 6 clearly shows the decay of ^{252}Fm and ^{253}Fm. The 6.633 MeV ^{253}Es peak is also apparent in these spectra. After 20 half-day, 6 one-day, and 6 three-day counts for a total of 34 days of continuous counting, the ^{253}Es decay curves were analyzed using a maximum likelihood method computer program [15]. The decay of the ^{253}Es region (6.4-6.6 MeV) of each spectrum was fit with a parent-daughter plus one component. The half-life of the parent ^{253}Fm was fixed at 3.0 d, and the ^{253}Es half-life and activity at the end of bombardment were fixed at 20.47 d and zero, respectively. The half-life of the third component was fixed at 1.06 d, as it was probably from tailing from the ^{252}Fm peak. The number of ^{253}Fm atoms was obtained from the observed ^{253}Es activity for each different collection time. A growth curve

was constructed by use of these numbers and an error-weighted least squares fitting program based on equation (1) (Fig.7), was used to give the best values for λ and R. A half-life of 6.4 minutes (+11.6 min, -3.6 min) and a production cross section of 46 nb (+243 nb, -31 nb) has been calculated for this isotope. The error limits calculated are the approximate 68% confidence limits. Production cross section values calculated by **SPIT** [11] and **JORPLE** [16] are 2.3 μb and 1.6 μb, respectively. Now that we have found the half-life of ^{253}Md to be about 6 min, a future experiment in which several irradiation intervals both shorter and longer than the half-life should be performed in order to arrive at a better value.

We have also determined the half-life (Fig.8) and production cross section of ^{252}Md from these same samples by measuring the 7.04-MeV alpha group of ^{252}Fm, the EC daughter of ^{252}Md. A half-life of 4.8 minutes (+0.8 min, -0.5 min) and production cross section of 1.4 μb (+0.6 μb, -0.4 μb) was measured for this isotope. Our production cross section is consistent with the value of about 1.0μb estimated from the paper by Eskola [3]. However, these experimental values are much lower than the values of 13 μb and 20 μb calculated by **SPIT** [11] and **JORPLE** [16], respectively. The irradiation intervals of 5, 10, and 20 minutes used in this experiment were not optimal for measuring this half-life. Another experiment with irradiation intervals between 2 min and 10 min should be performed. Our measured half-life for this isotope is between the 2.3±0.8 min half-life observed by Eskola [3] in 1972 and the half-life of 8 min reported for the ^{238}U(^{19}F,5n)^{252}Md reaction by Donets, Schegolev, and Ermakov [17] in 1965.

6. ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

1. Illustration of the multiple target system used for this study (taken from ref. 9).
2. Horizontal cross-section view of the LIM target system. The KCl aerosols in helium enter the target chamber as shown. The recoiling reaction products are collected on the aerosols in the helium, which sweeps out the volume behind each target continuously. The activity-laden aerosols are then transported by the helium through 1.2-mm i.d. polyvinylchloride capillary tube to a collection site 6m away.
3. Flowchart of the chemical separations from the end of irradiation to the end of separation.
4. Decay scheme of ^{252}Md and ^{253}Md to their daughter and granddaughter and relevant branching ratios and alpha decay energies.
5. Alpha spectrum from half a day to seven days after bombardment, an integral 6.5-day count. The 7.040 MeV and 6.943 MeV ^{252}Md and ^{253}Md peaks, respectively, are apparent in the spectrum.
6. Alpha spectrum from seven days to 34 days after bombardment, an integral 27-day count. A comparison of this spectrum with figure 5 clearly shows the decay of ^{252}Fm and ^{253}Fm.
7. Growth curve constructed from the measured number of ^{253}Fm daughter atoms and an error-weighted least squares fitting program based on equation (1). This curve was used to give the best values for λ and R.
8. Growth curve constructed from the measured number of ^{252}Fm daughter atoms and an error-weighted least squares fitting program based on equation (1). This curve was used to give the best values for λ and R.

Light Ion Multiple (LIM) target system

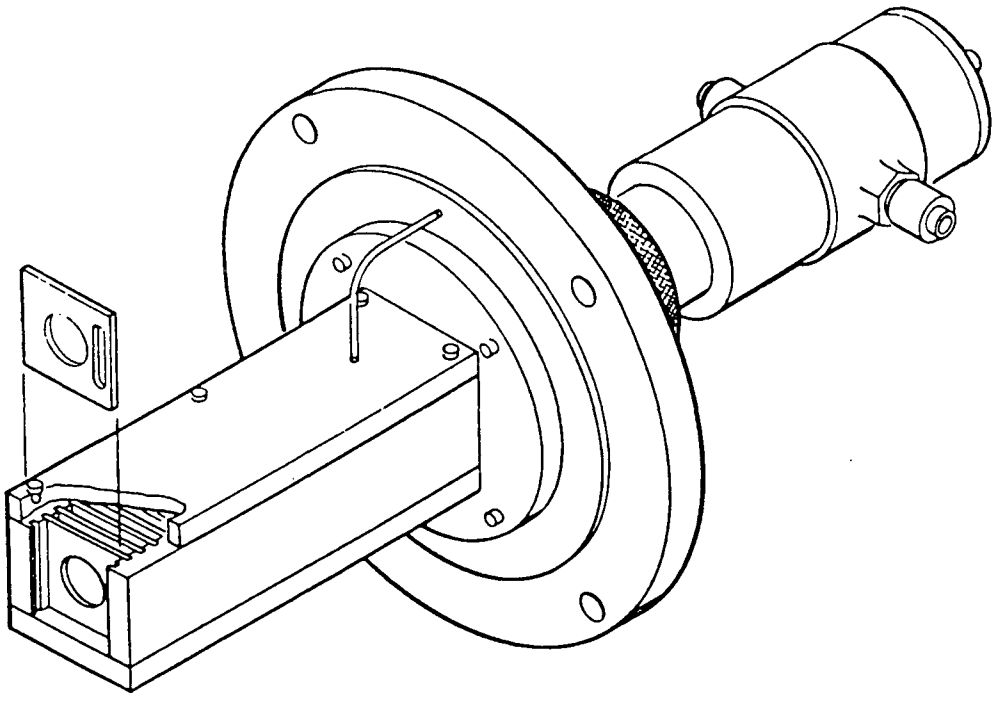

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Fig.1

Light Ion Multiple (LIM) target system

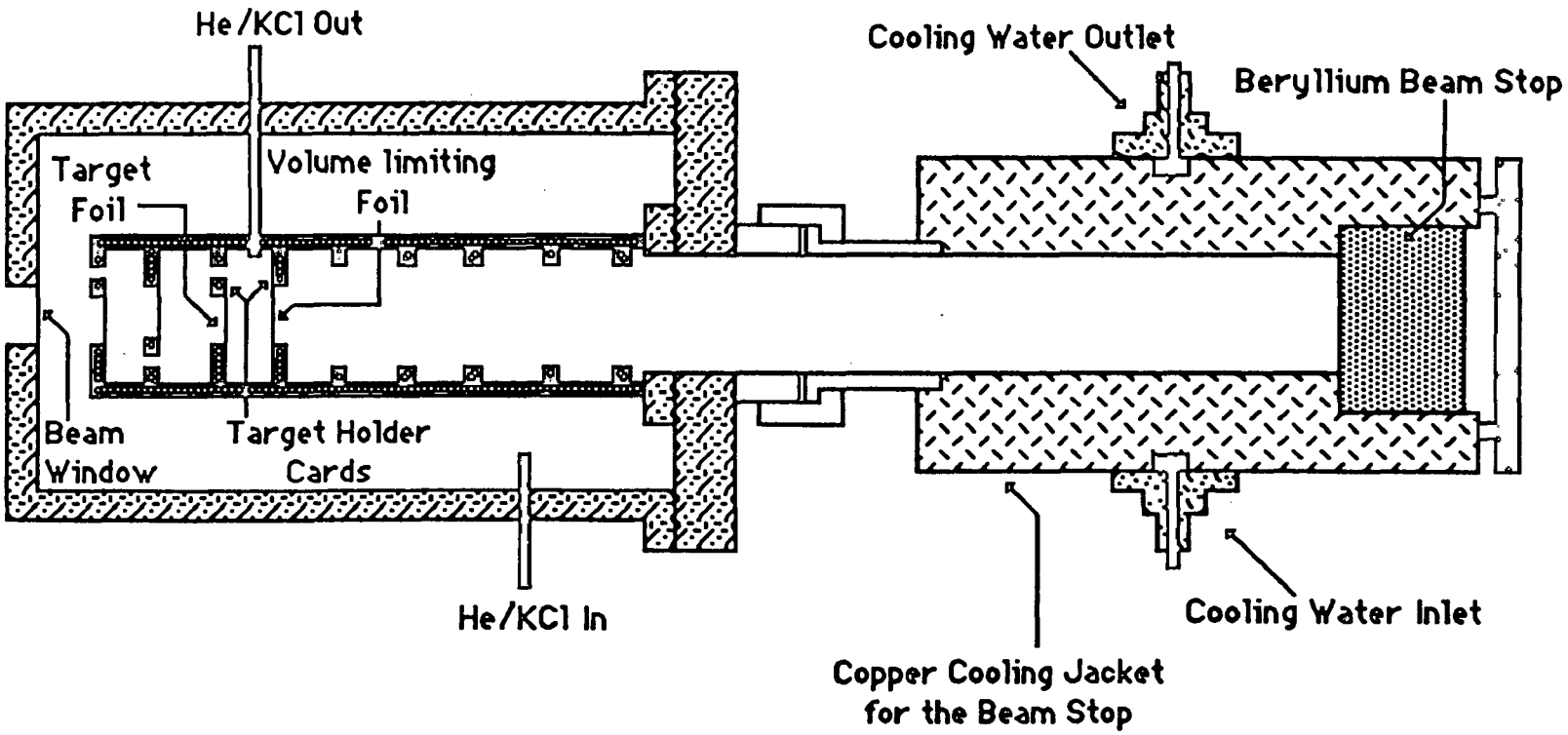

Fig.2

FLOW CHART OF THE CHEMICAL SEPARATION

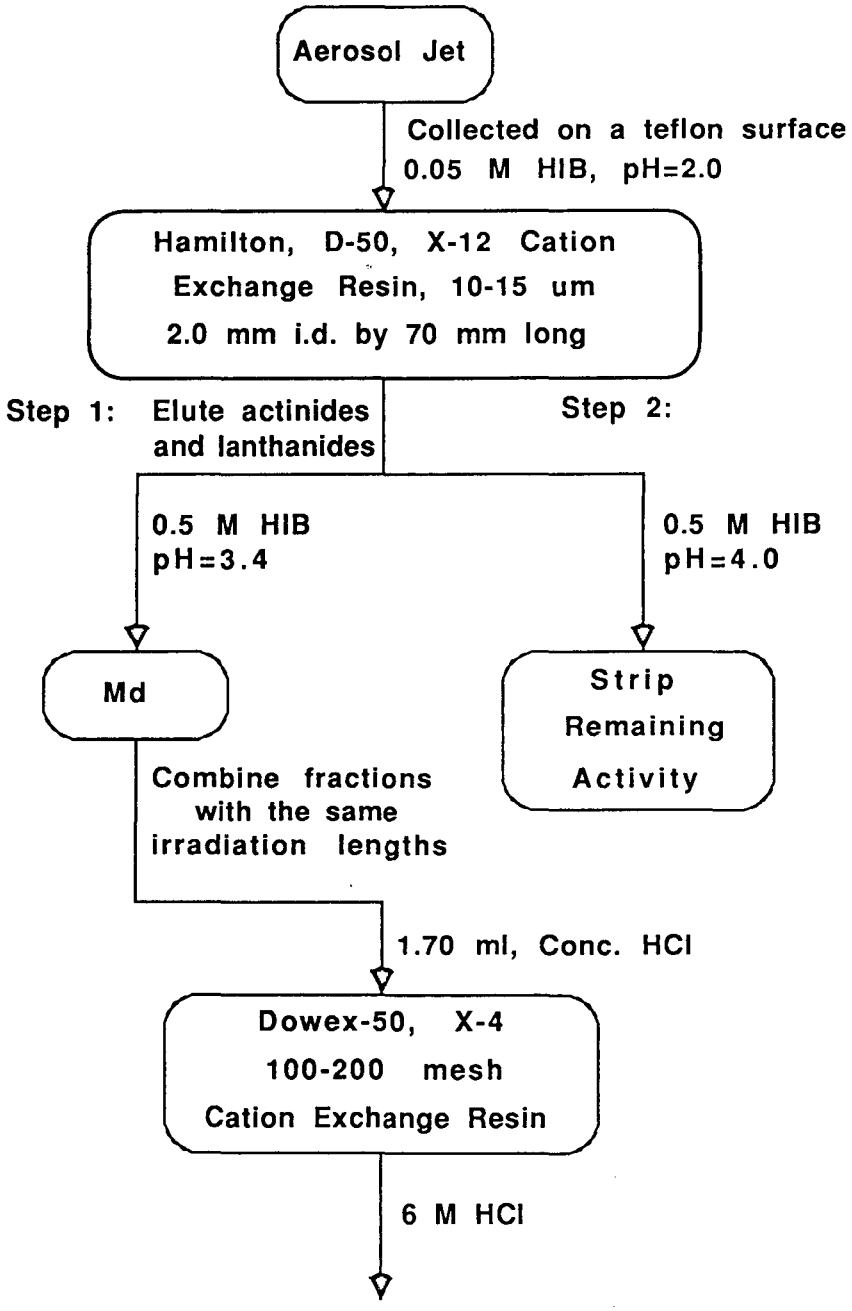

Fig. 3

Decay of ^{252}Md and ^{253}Md

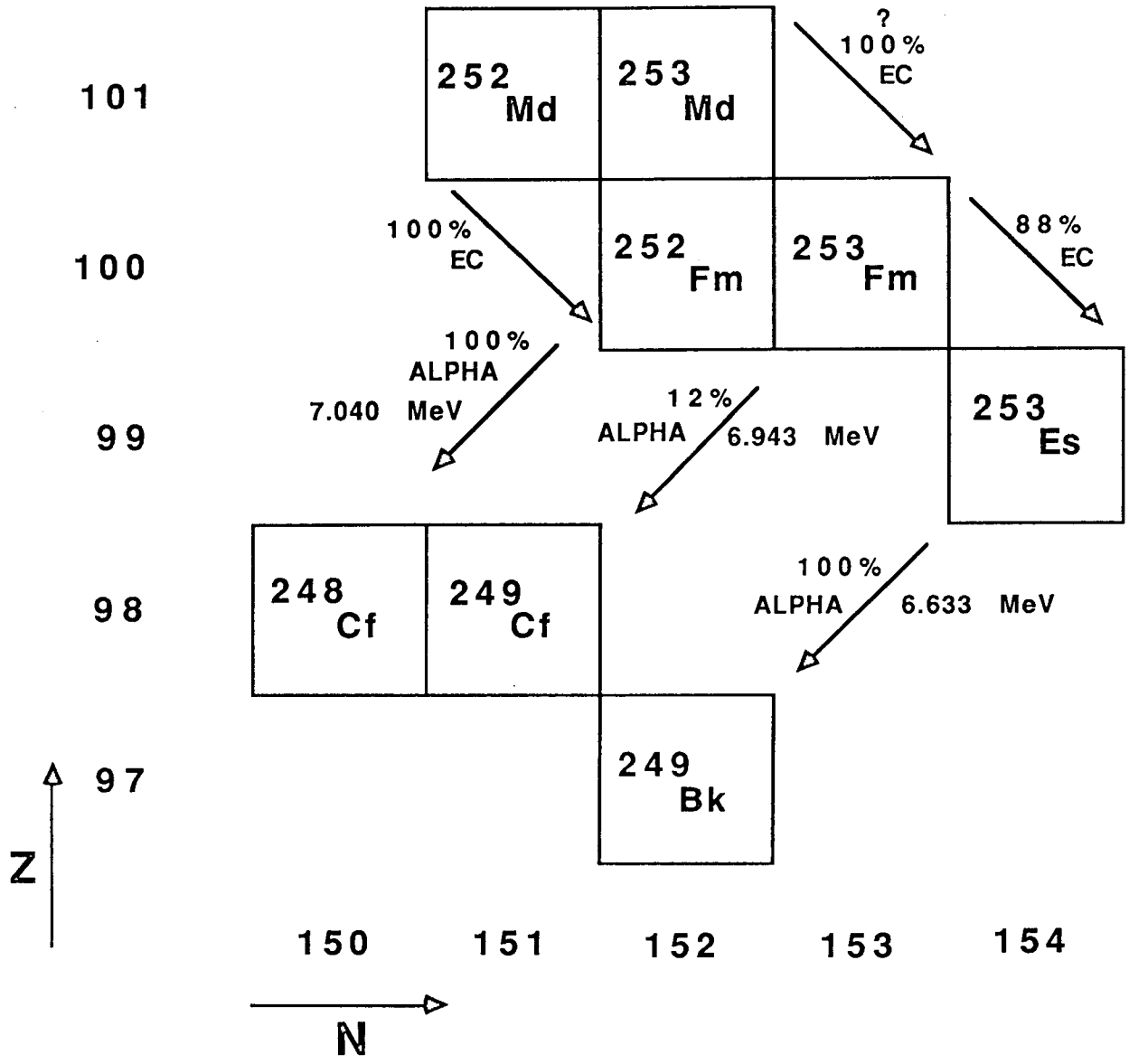

Fig. 4

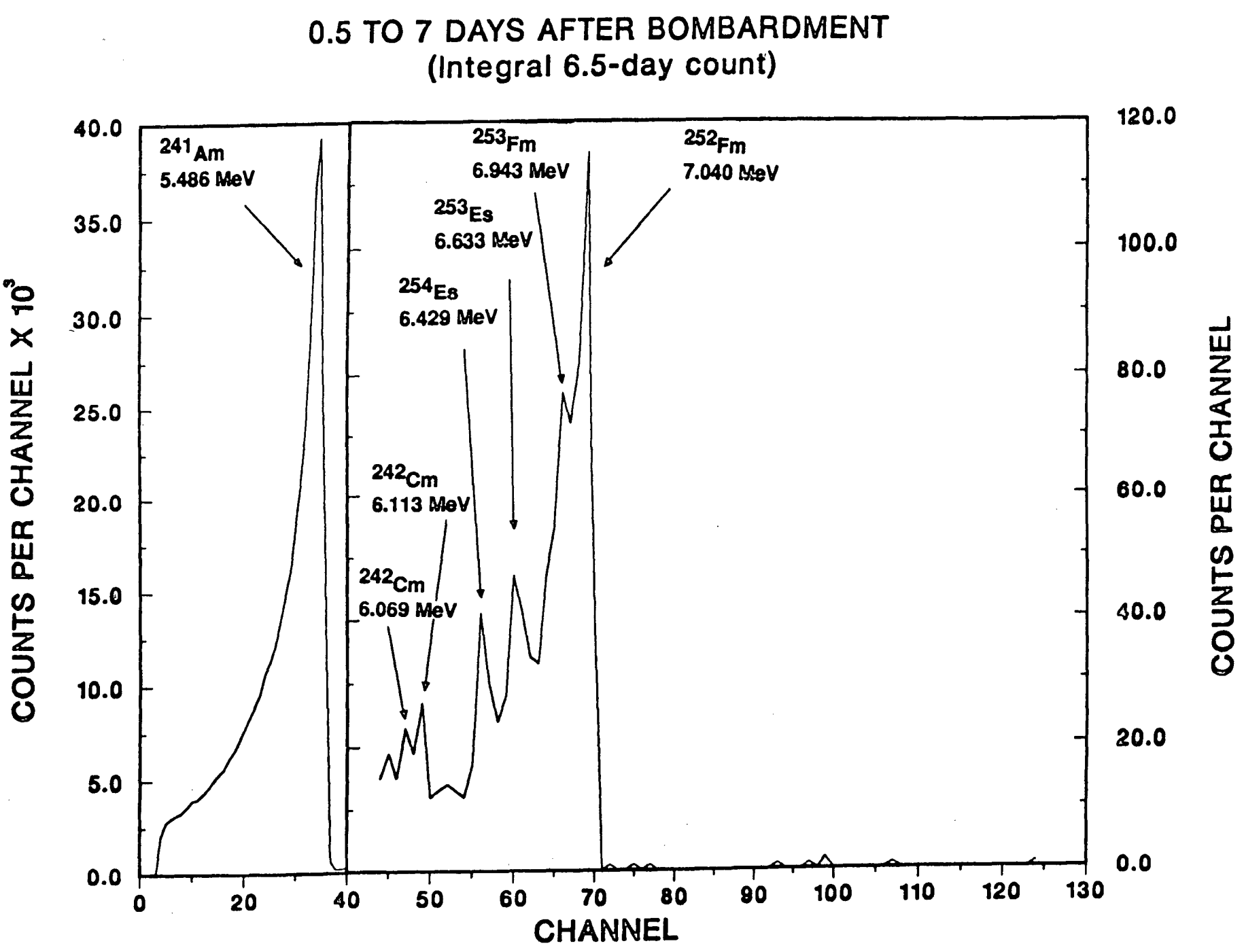
0.5 TO 7 DAYS AFTER BOMBARDMENT
(Integral 6.5-day count)
241Am
5.486 MeV
253Fm
6.943 MeV
252Fm
7.040 MeV
253Es
6.633 MeV
254Es
6.429 MeV
242Cm
6.113 MeV
242Cm
6.069 MeV
COUNTS PER CHANNEL X 10³
COUNTS PER CHANNEL
CHANNEL
0.0
5.0
10.0
15.0
20.0
25.0
30.0
35.0
40.0
0.0
20.0
40.0
60.0
80.0
100.0
120.0
0
20
40
50
60
70
80
90
100
110
120
130

Fig.5

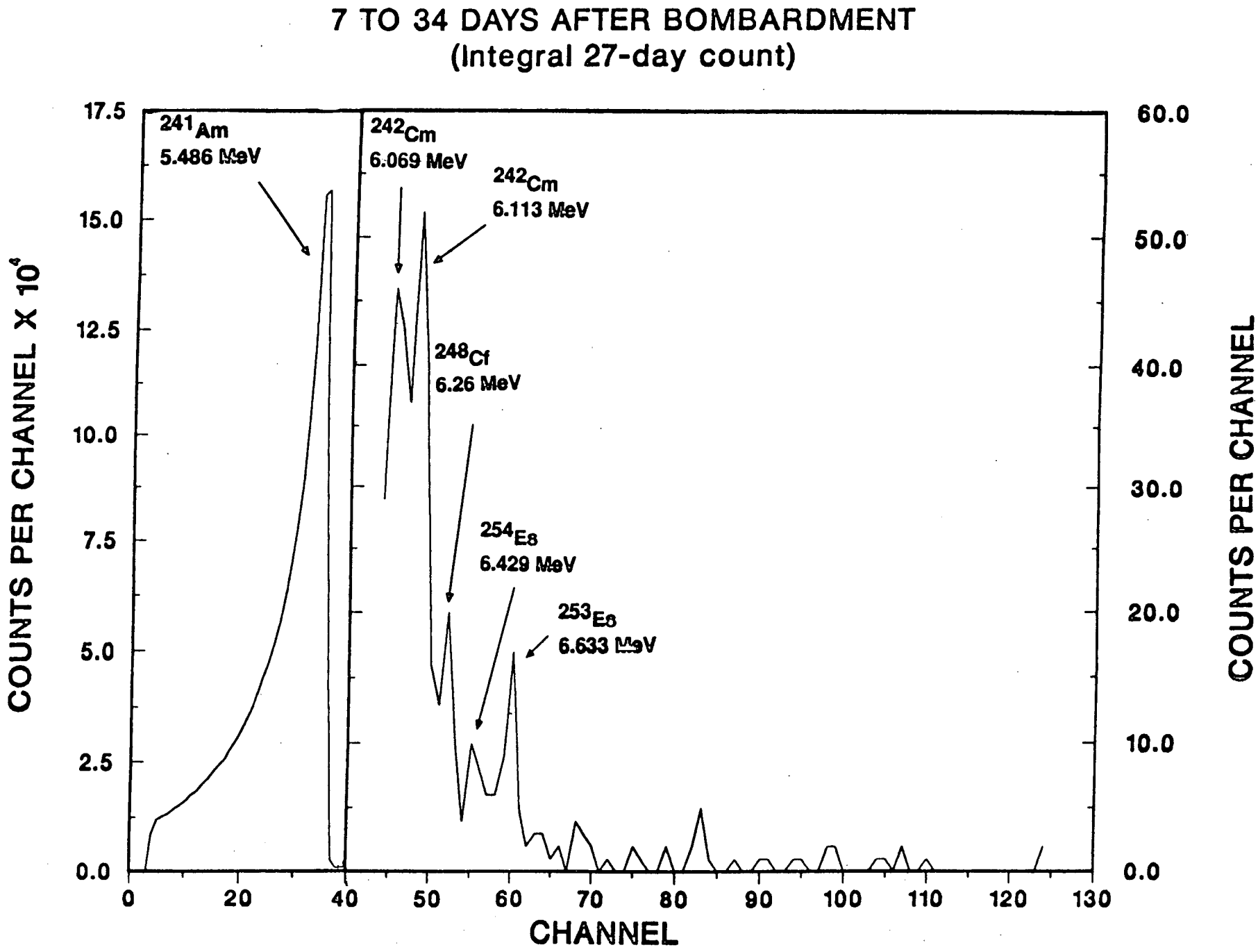
7 TO 34 DAYS AFTER BOMBARDMENT
(Integral 27-day count)
241Am
5.486 MeV
242Cm
6.069 MeV
242Cm
6.113 MeV
248Cf
6.26 MeV
254Es
6.429 MeV
253Es
6.633 MeV
COUNTS PER CHANNEL X 10⁴
COUNTS PER CHANNEL
CHANNEL
17.5
15.0
12.5
10.0
7.5
5.0
2.5
0.0
60.0
50.0
40.0
30.0
20.0
10.0
0.0
0
20
40
50
60
70
80
90
100
110
120
130

Fig.6

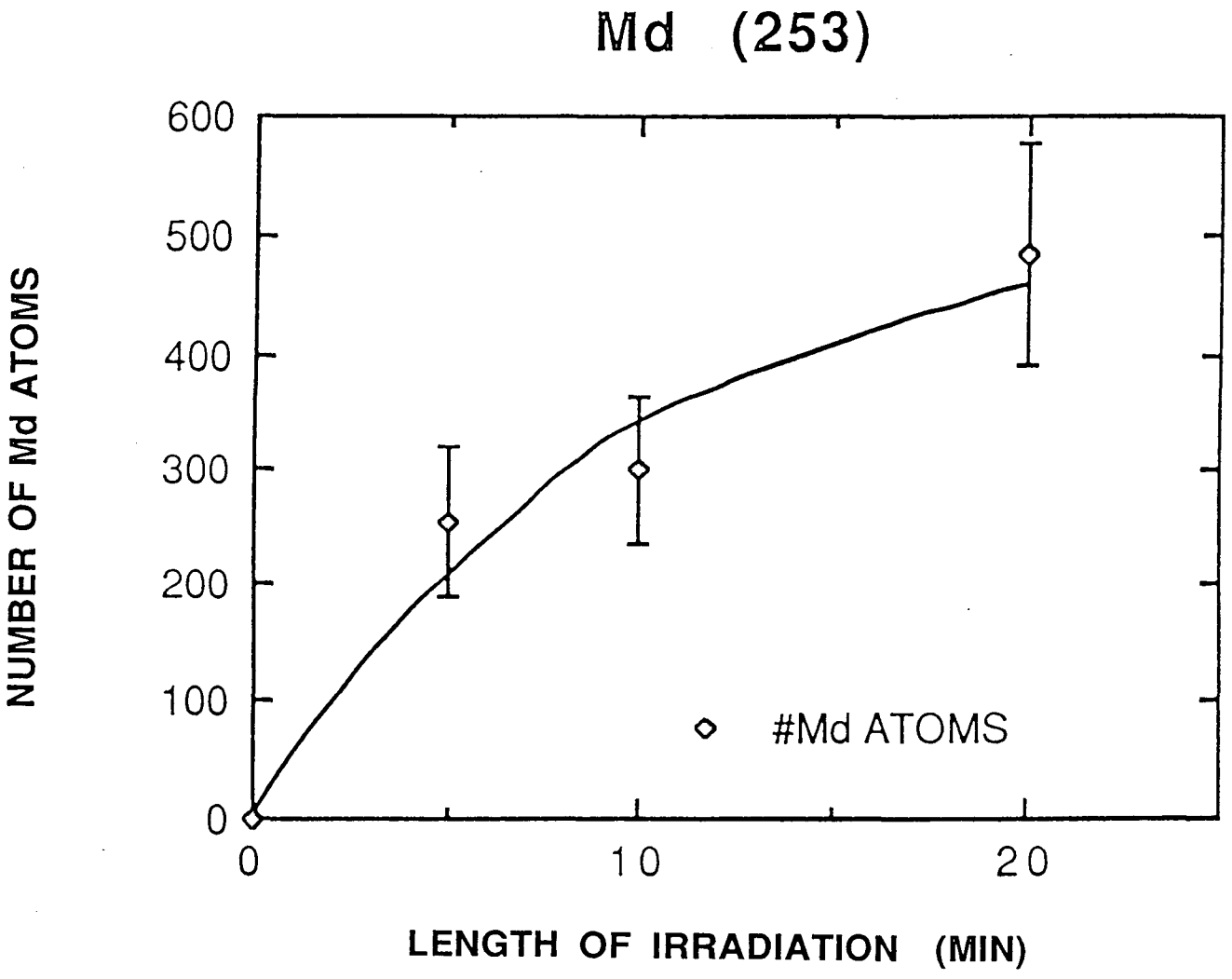
Md (253)
NUMBER OF Md ATOMS
600
500
400
300
200
100
0
0
10
20
#Md ATOMS
LENGTH OF IRRADIATION (MIN)

Fig. 7

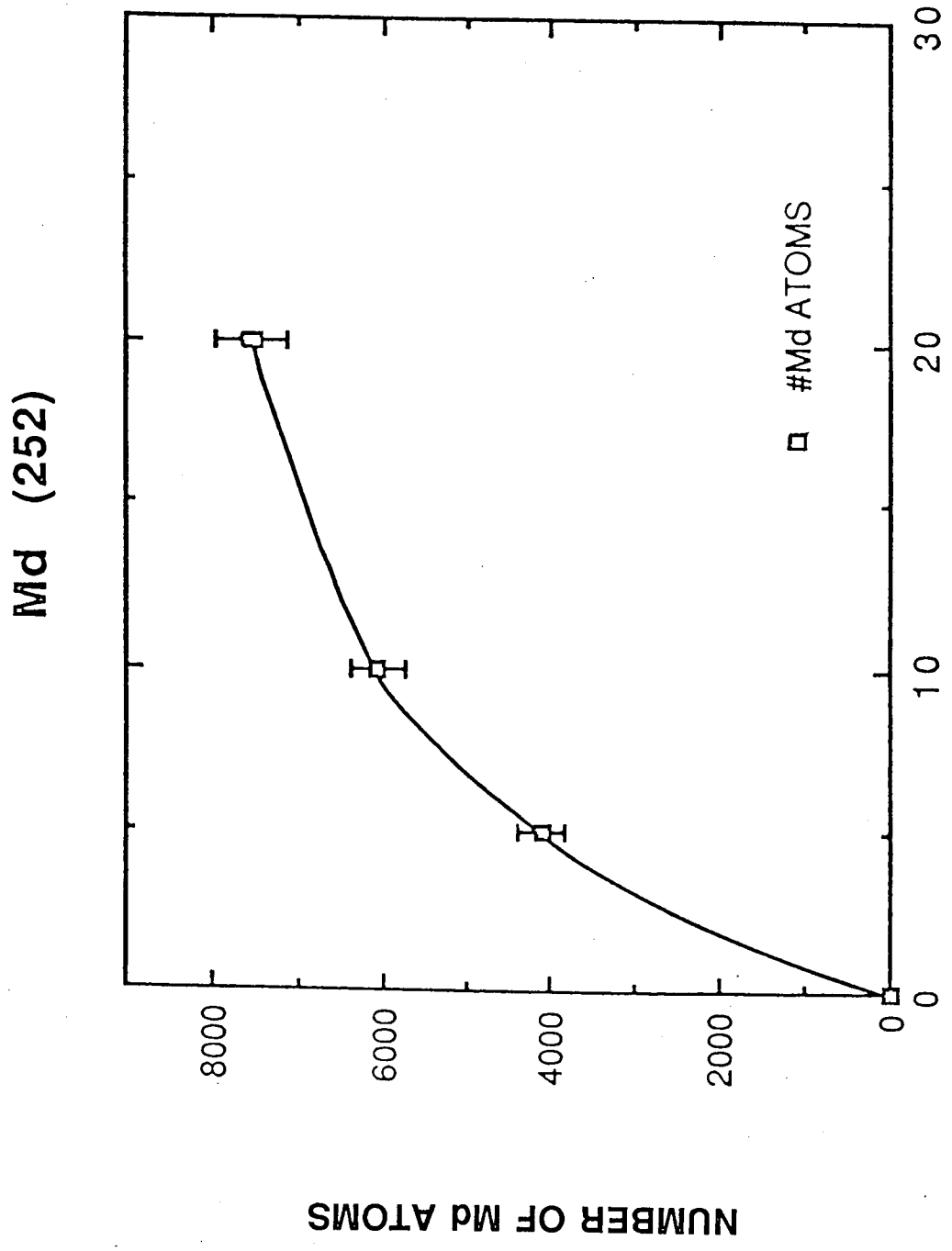
Md (252)
NUMBER OF Md ATOMS
8000
6000
4000
2000
0
0
10
20
30
LENGTH OF IRRADIATION (MIN)
#Md ATOMS

Fig. 8

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