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THE HEW CHAIN BARIUM 126-CESIUM 126

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THE NEW CHAIN BARIUM 126 - CESIUM 126

M. I. Kalkstein and J. M. Hollander

May 19, 1954

Berkeley, California

THE NEW CHAIN BARIUM 126 - CESIUM 126

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ABSTRACT

The neutron deficient chain  $\text{Ba}^{126} - \text{Cs}^{126}$  has been produced from nitrogen-ion bombardments of indium in the 60-inch cyclotron, by the reaction  $\text{In}^{115}(\text{N}, 3\text{n})$ . Studies have been made of this new chain with a 50-channel scintillation spectrometer, a scintillation coincidence spectrometer, and a time-of-flight mass spectrograph. Element assignments and genetic relations have been verified chemically, and the mass number assigned with the isotope separator. Barium<sup>126</sup> decays principally by orbital electron capture with a half-life of  $96.5 \pm 2.0$  minutes, and its daughter  $\text{Cs}^{126}$  is a positron emitter of  $1.6 \pm 0.2$  minute half-life and electron capture branching of  $15 \pm 4$  percent. The decay of  $\text{Cs}^{126}$  proceeds by allowed transitions,  $\sim 63$  percent to the ground state of  $\text{Xe}^{126}$  and  $\sim 37$  percent to the first excited state at 385 kev. The positron spectrum has a maximum energy of  $3.8 \pm 0.4$  Mev. On the basis of its decay properties,  $\text{Cs}^{126}$  appears to have a ground state configuration of (1+).

## THE NEW CHAIN BARIUM 126 - CESIUM 126

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### INTRODUCTION

A program has been underway in this laboratory to study the nuclear reactions of cyclotron accelerated heavy ions and to utilize these ions in the synthesis of new isotopes of the heaviest elements.<sup>1-4</sup> In principle at least, heavy ion bombardment should also find application to the study of neutron deficient isotopes of medium and low Z elements, because it provides an almost unique way to prepare these isotopes free of heavier ones. To date, the major limitation on this application has been the low beam intensities available, which has severely restricted the feasibility of spectroscopic investigations. Nonetheless, it was felt that the possible advantages of using heavy ions for the production of neutron deficient nuclei should be examined.

In the work reported here, a new neutron-deficient chain,  ${}_{56}\text{Ba}^{126} - {}_{55}\text{Cs}^{126}$ , has been produced by the irradiation of indium (Z = 49) targets in the nitrogen ion beam of the Crocker 60-inch cyclotron. The genetic relationship between these nuclides has been verified by means of chemical "milking" experiments, and their radiations have been studied with NaI and anthracene scintillation detectors coupled to a 50-channel differential analyzer. Some coincidence studies have also been made by using the scintillation coincidence spectrometer. Assignment to the mass 126 chain has been accomplished by means of a time-of-flight mass spectrometer.

Barium 126 is found to have a half-life of 96.5 minutes, and to decay by orbital electron capture. Its daughter,  ${}_{55}\text{Cs}^{126}$ , decays with a 1.6 minute half-life, chiefly by the emission of positrons of maximum energy about 3.8 Mev.

A 12 minute barium activity, which may be  $\text{Ba}^{127}$  or possibly  $\text{Ba}^{124}$ , has also been observed in some short nitrogen ion bombardments of indium. This will be reported only briefly, since no detailed study of this nuclide has been undertaken.

#### BOMBARDMENT CONDITIONS

In most of the bombardments, targets of indium oxide ( $\text{In}_2\text{O}_3$ )<sup>5</sup> weighing ~100 mg were mounted in 0.25 or 0.5 mil platinum envelopes for use with the internal heavy ion probe. A few runs were also made using targets of indium metal melted to the copper backing plate of a watercooled probe. Irradiations were made at a dee radius of 23.5 inches. The energy of the nitrogen ion beam has a continuous distribution with a maximum of ~140 Mev.<sup>3</sup> Because of the thick targets used, it was not possible to monitor the beam during bombardments, but nitrogen ion currents measured separately have averaged around 0.1 microampere. Bombardment times were either short (two to five minutes) or of the order of an hour, depending upon the half-lives of the barium activities being studied.

#### CHEMICAL PURIFICATIONS

The chemical procedures used both for the initial separation of a pure barium fraction and for the subsequent cesium-barium "milkings" are described in Appendix I.

## EXPERIMENTAL RESULTS

Half-lives. -- The barium chemical fractions from the nitrogen ion bombardments of indium exhibited, after the decay of an initial  $\sim 12$  minute component, a single activity which followed a period of  $96.5 \pm 2.0$  minutes through as many as ten half-lives without deviation. One of the G-M counter decay curves is shown in Fig. 1. When the chemical purification was performed more than an hour after the end of bombardment, only the 97 minute activity appeared in the barium fraction, proving its chemical identity as an isotope of barium.

By making a series of rapid chemical separations of cesium away from a solution containing the 97 minute barium activity, a cesium daughter has been isolated whose half-life was measured from five milkings to be  $1.5 \pm 0.3$  minutes.

Mass assignment. -- With the aid of Dr. M. C. Michel, the mass assignment of the new Ba-Cs chain has been made on a medium-resolution, high transmission ( $\sim 20$  percent) isotope separator employing the time-of-flight principle.<sup>6</sup>

The sample, containing  $\sim 10^5$  dis/min of the 97 minute barium in equilibrium with its cesium daughter, was deposited onto a tungsten filament in the form of  $\sim 50 \mu\text{g BaSO}_4$ . Because of the weakness of this source, it did not seem feasible to attempt collection of barium ions, consequently advantage was taken of the much greater ionization efficiency of the short-lived cesium daughter. By "flashing" the ion source at a temperature sufficient to ionize cesium but not vaporize barium it was possible to collect



cesium ions alone, accomplishing simultaneously both chemical and mass separation. By so retaining barium on the filament, the experiment could be repeated several times with essentially no loss in yield of cesium at the collector end.

After flashing the source for ~10 seconds, the vacuum system was quickly let down to air, and the collector plate removed for counting. This procedure consumed about two minutes. Activity was found to collect only at the mass 126 position, and this decayed with a half-life of  $1.6 \pm 0.2$  minutes, as shown in Fig. 2. The new chain is thus identified as  $Ba^{126}-Cs^{126}$ , produced by the reaction  $In^{115}(N, 3n)Ba^{126}$ .

Gamma spectra. -- Nitrogen ion bombardments of around an hour's duration yielded less than  $10^6$  disintegrations per minute of the  $Ba^{126}-Cs^{126}$  chain, so it was not possible to utilize either of the laboratory's high resolution magnetic spectrometers in an examination of the beta and gamma spectra. Consequently, we have made use principally of a NaI (Tl) scintillation detector coupled to a 50-channel differential analyzer designed by A. Ghiorso and A. E. Larsh.<sup>7</sup>

In these experiments, the gamma ray spectrum of the barium-cesium equilibrium mixture was first studied, and it was verified that all peaks decayed with a 97 minute half-life. Then a pure sample of the 1.5 minute cesium daughter was prepared by the "milking" procedure described in Appendix I, and its gamma spectrum was examined. The spectrum due to the 97 minute barium parent was then obtained by making the appropriate subtraction of the cesium spectrum from that of the equilibrium mixture. Figures 3, 4, and 5

show respectively the spectra between 0 and 850 keV of the equilibrium mixture, the pure  $\text{Cs}^{126}$ , and the subtracted spectrum obtained for the  $\text{Ba}^{126}$  parent. It is noted that the x-rays (at ~30 keV) belong mainly to the  $\text{Ba}^{126}$  while the annihilation radiation accompanies the decay of  $\text{Cs}^{126}$ .

The gamma spectrum of  $\text{Cs}^{126}$  is quite simple; other than annihilation radiation and ~30 keV x-rays only a gamma ray of  $385 \pm 5$  keV is in evidence. The peak (in Fig. 4) at ~170 keV is due to  $180^\circ$  backscattered annihilation radiation, and that at ~75 keV is the lead fluorescent x-ray generated in the counter shield. From the observed<sup>8</sup> photopeak abundances, it is calculated that  $\text{Cs}^{126}$  decays principally by positron emission, with an electron capture branching of  $15 \pm 4$  percent. The 385 keV gamma ray appears in  $37 \pm 7$  percent of the disintegrations. No gamma ray of energy greater than 600 keV is observed with an abundance higher than 5 percent of the disintegrations.

The gamma spectrum of  $\text{Ba}^{126}$ , obtained by subtraction, is more complex. Gamma rays of  $225 \pm 10$  keV, and  $700 \pm 30$  keV are definitely observed, with relative intensities 2.5:1. Weak gamma rays at  $370 \pm 20$  keV and ~900 keV are probable. The peak at 115 keV appears to be largely backscattered 225 keV radiation, since its relative intensity varies with position of the source in the counter shield. In the equilibrium ( $\text{Ba}^{126} - \text{Cs}^{126}$ ) spectrum, the 510 keV peak intensity is slightly too high to be attributed entirely to the annihilation gamma ray of  $\text{Cs}^{126}$ , so it is concluded either that  $\text{Ba}^{126}$  has a small amount (~10 percent) of positron branching, or that it has a nuclear gamma ray of about 500 keV. X-rays are the

predominant feature of the decay of  $\text{Ba}^{126}$ , but because of the uncertainties accompanying the subtraction of the gamma spectra and because of the unknown contribution of x-rays from internal conversion of the gamma rays, a ratio of electron capture to positron emission in  $\text{Ba}^{126}$  cannot be given with confidence.

The direct decay of the gamma ray lines from the 1.5 minute  $\text{Cs}^{126}$  was observed by performing the fast cesium "milking" from the parent, placing the separated cesium fraction into the scintillation spectrometer, and photographing the dials of the spectrometer with a Leica camera at half-minute intervals following each "milking". By subtraction of the dial readings of one negative from those of the next negative, the spectrum during each half-minute period could be examined. In this way, decay curves have been constructed for the photopeaks of the annihilation gamma ray, the 385 kev gamma ray, and for the K x-rays. These exhibit a half-life of  $1.6 \pm 0.2$  minutes, as shown in Fig. 6.

Beta spectrum. -- The end point of the positron spectrum was measured in a sample of the Ba-Cs equilibrium mixture by means of an anthracene scintillation crystal coupled to the 50-channel analyzer, and also by absorption in beryllium. Both methods gave a value of  $3.8 \pm 0.4$  Mev. As noted above, these positrons are associated with the decay of  $\text{Cs}^{126}$ .

Coincidence measurements. -- Several experiments were done in which the  $\text{Ba}^{126}$ - $\text{Cs}^{126}$  chain was examined with a coincidence scintillation spectrometer designed by Professor J. O. Rasmussen. The apparatus consists of a NaI or anthracene gate detector coupled to a single channel analyzer, and a NaI coincidence detector coupled

to the 50-channel analyzer. In these experiments, the resolving time was  $\approx 8 \times 10^{-8}$  seconds. The anthracene detector in the gate circuit was employed when positron-gamma coincidences were being examined; for gamma-gamma coincidences NaI crystals were used in both circuits. The two crystals were shielded from each other by a 2-inch lead brick and beryllium absorbers were placed in front of the gamma ray detector to absorb electrons. Figure 7 is a schematic representation of the coincidence equipment.

Although the short half-life of the activity under study prevented the accumulation of sufficient events per experiment to give good statistics, several qualitative results were obtained by sweeping the gate channel over the gamma spectrum in increments of  $\sim 35$  keV and recording the coincident gamma spectrum at  $90^\circ$  on the 50-channel analyzer. Figure 8 is a plot of the coincidence rate at  $\sim 385$  keV on the 50-channel analyzer as a function of gate setting, showing a maximum rate at a gate setting of  $\sim 510$  keV. Figure 9 similarly indicates the coincidence rate at  $\sim 510$  keV, showing a maximum at a gate setting of  $\sim 385$  keV. These experiments verified that the 385 keV gamma ray follows positron decay in  $\text{Cs}^{126}$ . They also serve to point out that there is no gamma ray of  $\sim 500$  keV following positron decay of  $\text{Cs}^{126}$ , since few 500-500 gamma-gamma coincidences are observed. An upper limit of 4 percent per disintegration can be set for a possible gamma ray at  $\sim 500$  keV. This fact could not be deduced from the gross gamma spectrum alone, because of interference from the prominent 510 keV annihilation peak. Even those few 510 $\gamma$ -510 $\gamma$  coincidences that are observed at  $90^\circ$  may be

explained partially as being due to annihilation-annihilation coincidences, since an appreciable fraction of positrons annihilate in flight, giving annihilation quanta at angles other than  $180^\circ$  with respect to each other.

In scanning high energies on the gate, a maximum of gamma-gamma coincidences in the region of 225 kev was obtained when the gate was set at 700 kev, offering some evidence that the 225 kev gamma ray in  $\text{Ba}^{126}$  may be in coincidence with the 700 kev gamma ray.

Using the anthracene crystal in the gate circuit, positron-gamma coincidences were run at  $180^\circ$ . With a gate setting  $\ll 3$  Mev, coincidences were observed at  $\sim 390$  kev and  $\sim 510$  kev. At a gate setting of  $\sim 3.5$  Mev, the  $\sim 390$  kev peak was not observed, but the positron-annihilation coincidences remained. Since no gamma ray other than that at 385 kev is observed in the decay of  $\text{Cs}^{126}$ , it is inferred from the above that a ground state positron transition takes place between  $\text{Cs}^{126}$  and  $\text{Xe}^{126}$ .

#### CONCLUSIONS

From the observed positron and gamma ray abundances in  $\text{Cs}^{126}$ , some conclusions can be drawn about its decay scheme. Assuming that the observation of only a single gamma ray in the  $\text{Cs}^{126}$  is correct,  $\log ft$  values of 4.7 and 4.8 are calculated<sup>9</sup> for the positron decay to the ground and first excited states of  $\text{Xe}^{126}$ , respectively. These are both within the range of values expected for "allowed" transitions, and suggest that the configuration of the ground state of  $\text{Cs}^{126}$  is  $(1+)$ , since  $(0+)$  and  $(2+)$  doubtless represent the configurations of the first two states in  $\text{Xe}^{126}$ . The energy of the

first excited state of  $\text{Xe}^{126}$  found in this work,  $385 \pm 5$  keV, is in agreement with that reported from studies of the beta decay of  $\text{I}^{126}$  by Perlman and Friedlander<sup>10</sup> ( $386 \pm 2$  keV) and by Mitchell et al.<sup>11</sup> ( $395 \pm 5$  keV).

In a further study of the radiations of  $\text{I}^{126}$ , Perlman and Welker<sup>12</sup> have reported a second excited state in  $\text{Xe}^{126}$  at  $\sim 870$  keV, with a configuration also (2+). It is of interest to note that in the present study this state was not observed in the decay of  $\text{Cs}^{126}$ . A limit of 4 percent for the population of this state was set by the absence of coincidences between the 5.10 keV gamma ray and a gamma of 485 keV which would be expected to de-excite an 870 keV state if the spin sequence were (2+)→(2+)→(0+) as reported by Perlman and Welker. If the same selection rules are operative for decay of  $\text{Cs}^{126}$  to all these states, one would expect an 870 keV (2+) state in  $\text{Xe}^{126}$  to be populated from  $\text{Cs}^{126}$  to the extent of perhaps 20 percent, which is substantially above our experimental upper limit. A crossover gamma ray of 870 keV was also not observed in the gamma spectrum of  $\text{Cs}^{126}$ , but its expected abundance might be as low as the upper limit of 5 percent set from the gross gamma spectrum.

Because only a rough measurement of the positron energy of  $\text{Cs}^{126}$  could be obtained in these experiments, the disintegration energy cannot be specified precisely. However, the value of  $4.8 \pm 0.4$  MeV follows the general trend of the energy surface shown by the "beta-systematics" curves of Way and Wood.<sup>13</sup>

The electron capture branching of  $\text{Cs}^{126}$ ,  $15 \pm 4$  percent, compares with a figure of  $\sim 16$  percent predicted from the theoretical curves of Feenberg and Trigg<sup>14</sup> for an allowed transition with  $Z = 54$  and positron abundances and energies as reported here.

A decay scheme consistent with these data is presented in Figure 10.

#### THE 12-MINUTE BARIUM ACTIVITY

In several short nitrogen-ion bombardments of indium, an approximately 12-minute activity appeared in the barium fractions in lower yield than the 97-minute  $\text{Ba}^{126}$ . This activity may be  $\text{Ba}^{127}$ , produced by the  $\text{In}^{115}(\text{N}, 2\text{n})$  reaction. Barium 127 has been observed by Lindner and Osborne,<sup>15</sup> who have shown that it decays with a 12-minute half-life into 6.3-hour  $\text{Cs}^{127}$ . Although no 6-hour "tail" has been seen in the Geiger counter decay curves of our barium fractions, the low  $\beta^+/\text{EC}$  branching ratio (<10 percent) of  $\text{Cs}^{127}$  reported by Mathur and Hyde<sup>16</sup> would indicate that this isotope could have a counting efficiency low enough to have escaped detection in our gross decay curves.

On the other hand, it is possible that the half-life of the unknown isotope  $\text{Ba}^{124}$  could be of the order of 12 minutes, if it decays by an allowed transition. Such a half-life would require a disintegration energy of  $\sim 3$  Mev, which is not an unreasonable value.<sup>13</sup> Barium 124 would be produced in these experiments by  $\text{In}^{113}(\text{N}, 3\text{n})$  and  $\text{In}^{115}(\text{N}, 5\text{n})$  reactions.

### ACKNOWLEDGMENTS

We should like to thank A. Ghiorso, Professor J. G. Hamilton, W. B. Jones, and G. B. Rossi, and the crew of the 60-inch cyclotron for their constant efforts to develop and improve the heavy ion beams, and for their assistance to us in making these bombardments. It is also a pleasure to acknowledge helpful discussions with Professor J. O. Rasmussen and Dr. E. K. Hyde, and to thank Dr. M. C. Michel and Dr. F. A. Asaro for their help with several of the experiments.



## APPENDIX I

### Chemical Procedure

A typical procedure for the initial preparation of a pure barium fraction was the following:

1. The  $\text{In}_2\text{O}_3$  was dissolved in a minimum of concentrated HCl (or  $\text{HNO}_3$ , if indium metal targets were used), and carriers of Sn, Sb, Te, I, Cs, and Ba added.
2.  $\text{Ba}(\text{NO}_3)_2$  was precipitated by the addition of ~5 cc fuming  $\text{HNO}_3$ , washed, then dissolved in  $\text{H}_2\text{O}$ . This step was repeated.
3.  $\text{BaCl}_2$  was precipitated by the addition of 5-10 cc of ether-HCl reagent (5/1 concentrated HCl/diethyl ether), washed, then dissolved in  $\text{H}_2\text{O}$ . This step was repeated.

The following procedure was used for the rapid separation of cesium from a solution containing the  $\text{Ba}^{126}$ - $\text{Cs}^{126}$  equilibrium mixture. Total elapsed time from the initial precipitation to the beginning of count was about three minutes.

1. To a water solution of  $\text{BaCl}_2$  containing the 96 minute  $\text{Ba}^{126}$ , excess sodium cobaltinitrite reagent was added. This solution was chilled in an ice bath.
2. Several milligrams cesium carrier were added, precipitating yellow cesium cobaltinitrite.
3. The precipitate was quickly separated from the supernatant solution by pouring through a sintered glass filter funnel under low vacuum. It was washed in the same manner.
4. The  $\text{Cs}_3\text{CO}(\text{NO}_2)_6$  precipitate was dissolved from the sintered glass filter by pouring through several cc's of fuming  $\text{HNO}_3$ .

5. Several milligrams  $Ba^{++}$  carrier were added to the fuming  $HNO_3$  solution, precipitating  $Ba(NO_3)_2$ .

6. The supernate, containing pure cesium, was separated from the  $Ba(NO_3)_2$  precipitate by pouring through another sintered glass funnel. This solution was placed in the counting chamber.

## APPENDIX II

### Calibration of the NaI(Tl) Spectrometer

A 1 1/2 inch diameter by 1 inch thick thallium activated sodium iodide crystal was used in conjunction with the 50-channel analyzer. A diffuse reflector of MgO surrounded the crystal, and a thin beryllium window separated it from the sample. The crystal was bonded to a thin quartz disk which in turn was optically coupled to a Dumont 6292 photomultiplier tube by a layer of mineral oil. An aluminum-lined lead shield housed the detector assembly, and samples were mounted in a standard G-M tube five position shelf holder. In these experiments, most measurements were made at a distance of  $\approx 1.3$  inches from the crystal, which represented a geometry of 4.9 percent (as determined with the 60 kev gamma ray of a standardized  $Am^{241}$  sample).

Energy calibration of the spectrometer was made during each experiment, with the following gamma ray standards:  $Am^{241}$  (60 kev),  $U^{235}$  (143, 184 kev),  $Au^{198}$  (412 kev),  $Cs^{137}$  (32, 661 kev), and  $Na^{22}$  (510 kev, 1.28 Mev). In addition, the annihilation radiation of  $Cs^{126}$  served as an internal standard, so that any shift in the energy scale could be observed and corrected.

The following counting efficiencies were used to calculate the relative intensities of the various gamma rays:

X-rays (Cs, Xe)	100%
225 kev	66%
385 kev	23%
510 kev	12.3%
700 kev	9.3%

With the exception of the value for 510 kev, these efficiencies are as reported by Kahn and Lyon<sup>17</sup>; we have made similar absolute determinations at 412 kev (Au<sup>198</sup>), and 661 kev (Cs<sup>137</sup>) and find close agreement with their results. The 510 kev efficiency is the result of our own measurement of the relative abundance of the 510 kev and 1.28 Mev photopeaks in Na<sup>22</sup>, using Kahn and Lyon's value of 3.6 percent for the 1.28 Mev efficiency. (In absence of an absolute determination of Na<sup>22</sup> during these experiments, we have chosen to use Kahn and Lyon's value at 1.28 Mev rather than their value at 510 kev (13.7 percent), because of better agreement at 1.28 Mev with other investigators).

The observed x-ray intensities were corrected for fluorescence yield and absorption by comparison with the x-rays from a Cs<sup>137</sup> standard examined under identical experimental conditions.

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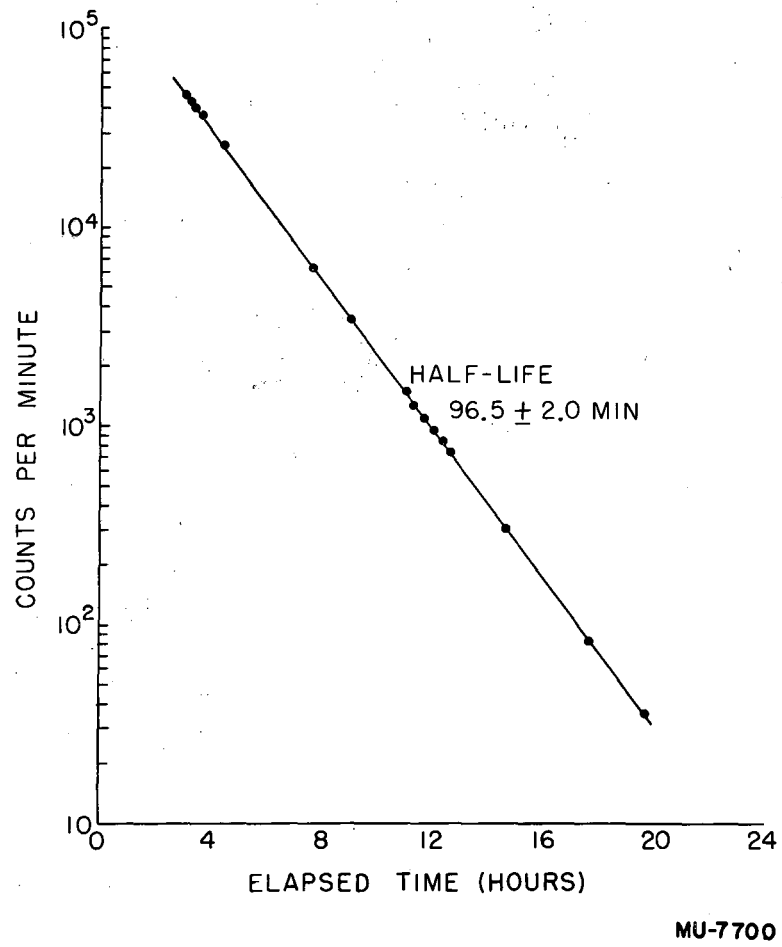


Fig. 1. Geiger-counter decay curve of Ba126.

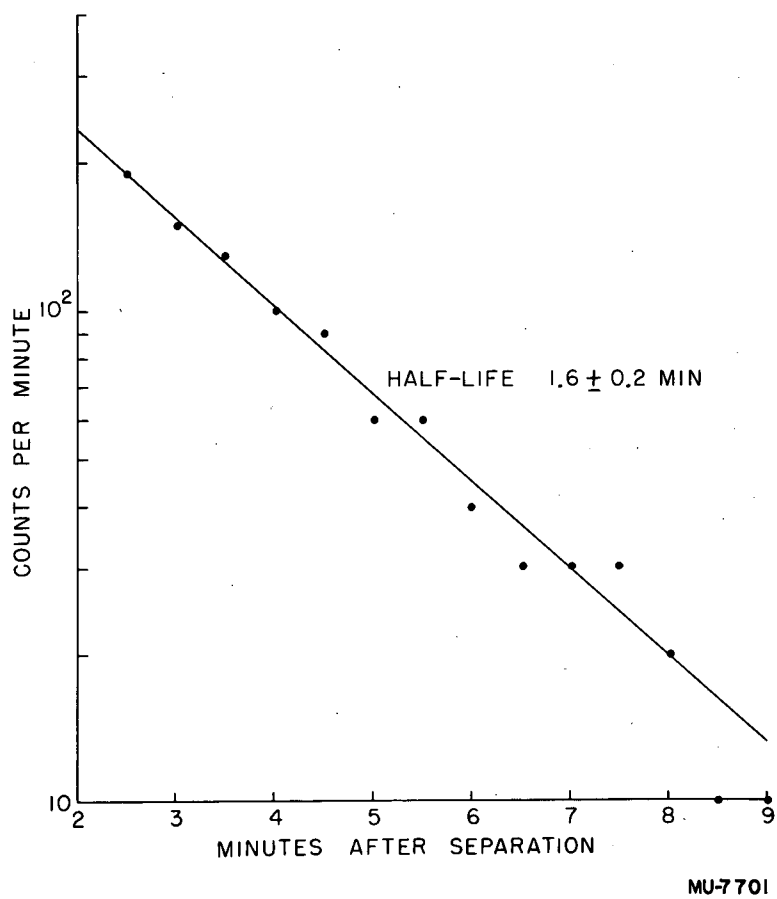
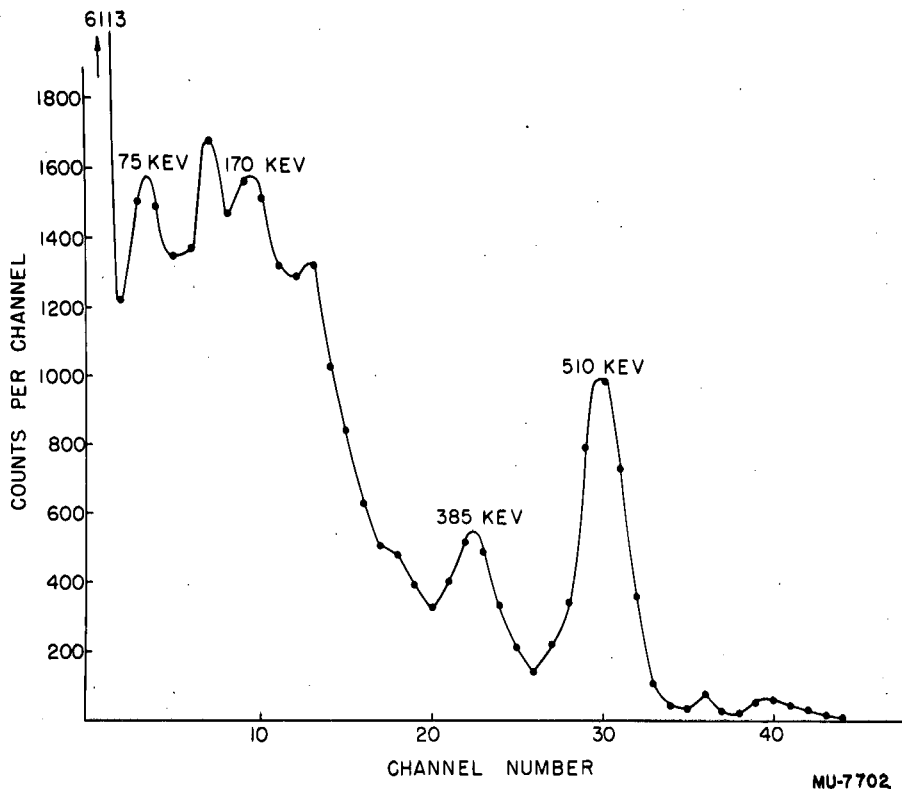


Fig. 2. Geiger counter decay curve of mass separated  $Cs^{126}$ .



MU-7702

Fig. 3. Gamma spectrum of Ba<sup>126</sup> - Cs<sup>126</sup> equilibrium mixture.

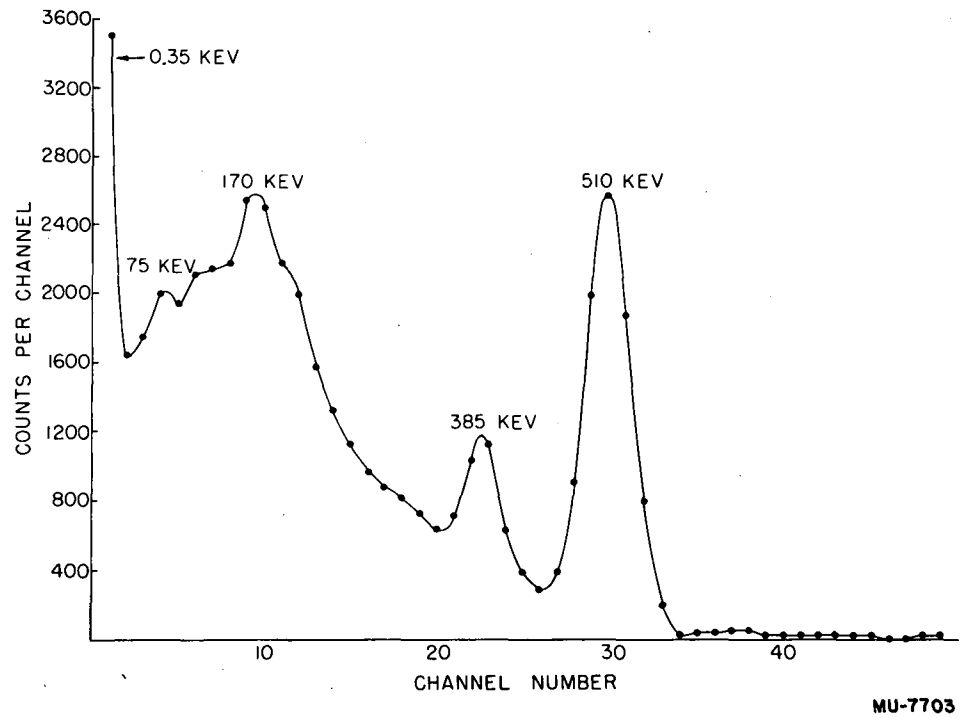
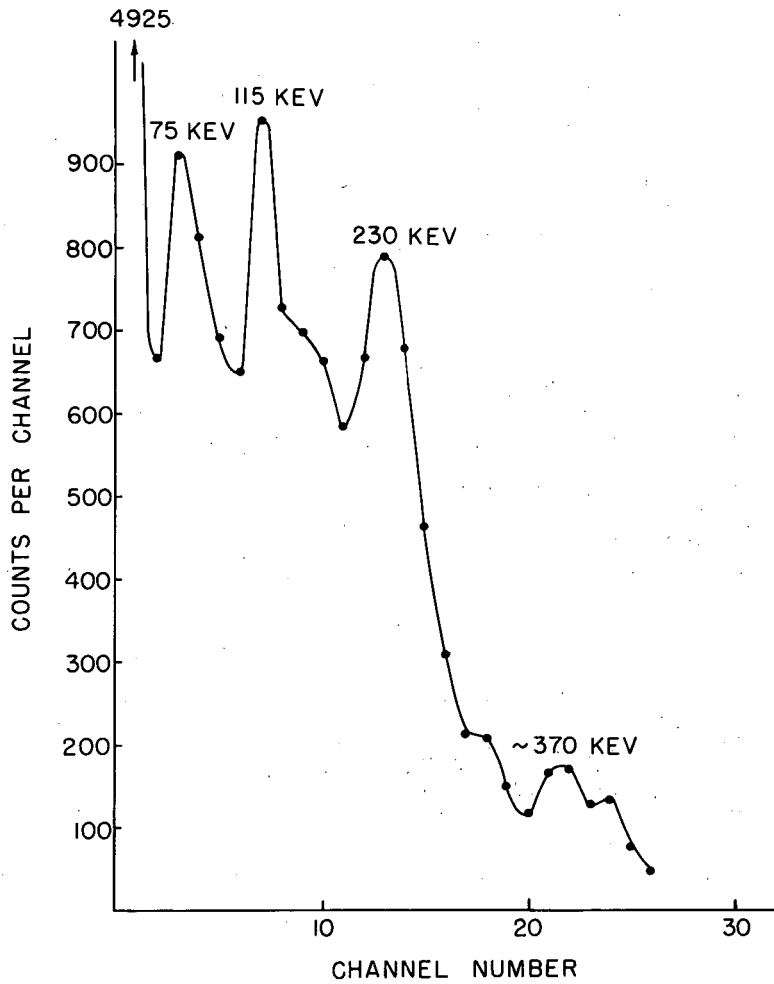


Fig. 4. Gamma spectrum of Cs<sup>126</sup>.

MU-7703





MU-7704

Fig. 5. Gamma spectrum of  $Ba^{126}$ , obtained by subtracting Fig. 4 from Fig. 3. (The 510 keV peak of Fig. 4 has been normalized to 90% of the 510 keV peak of Fig. 3).

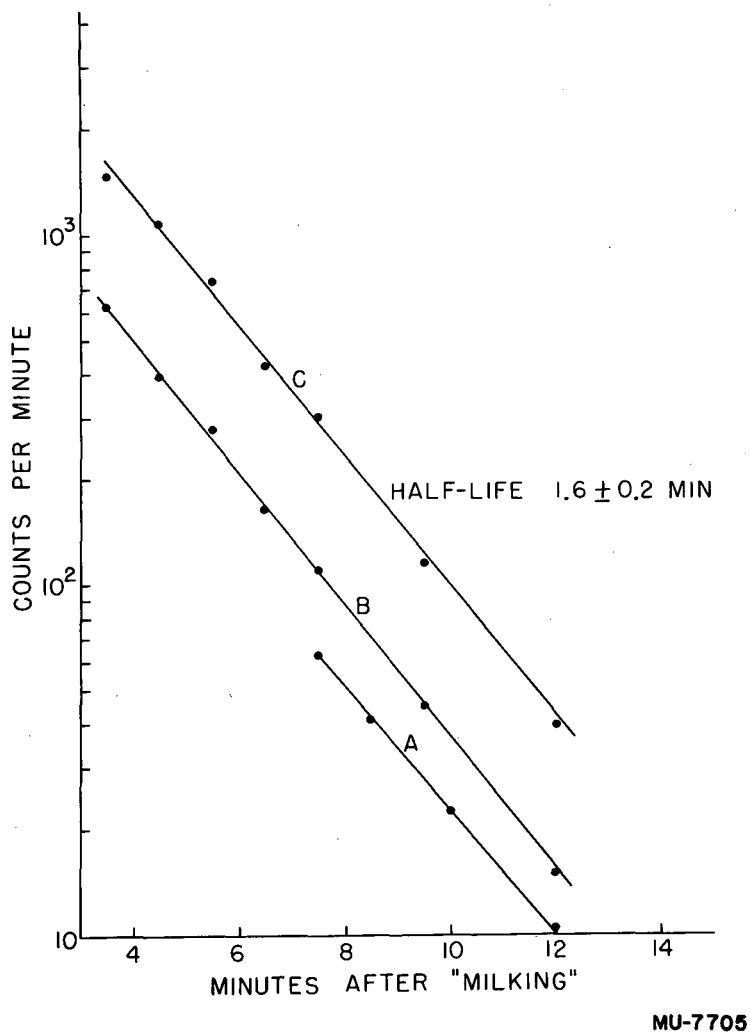
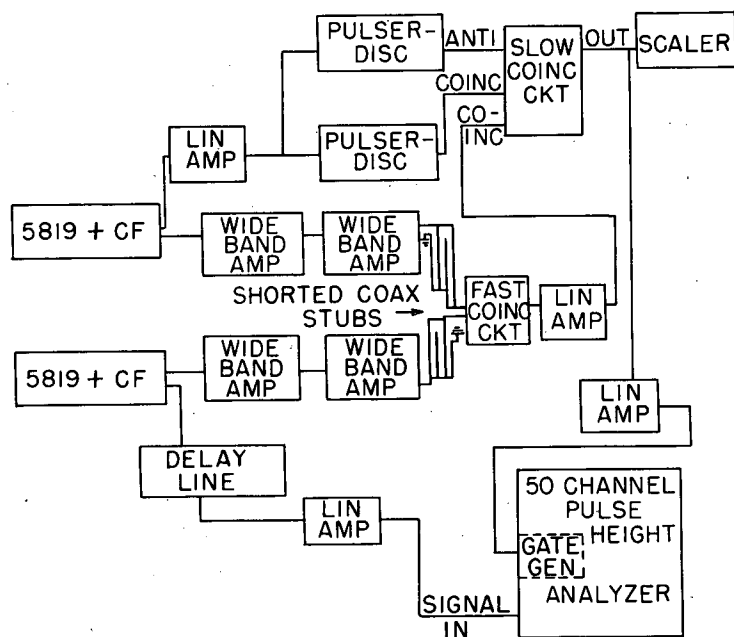
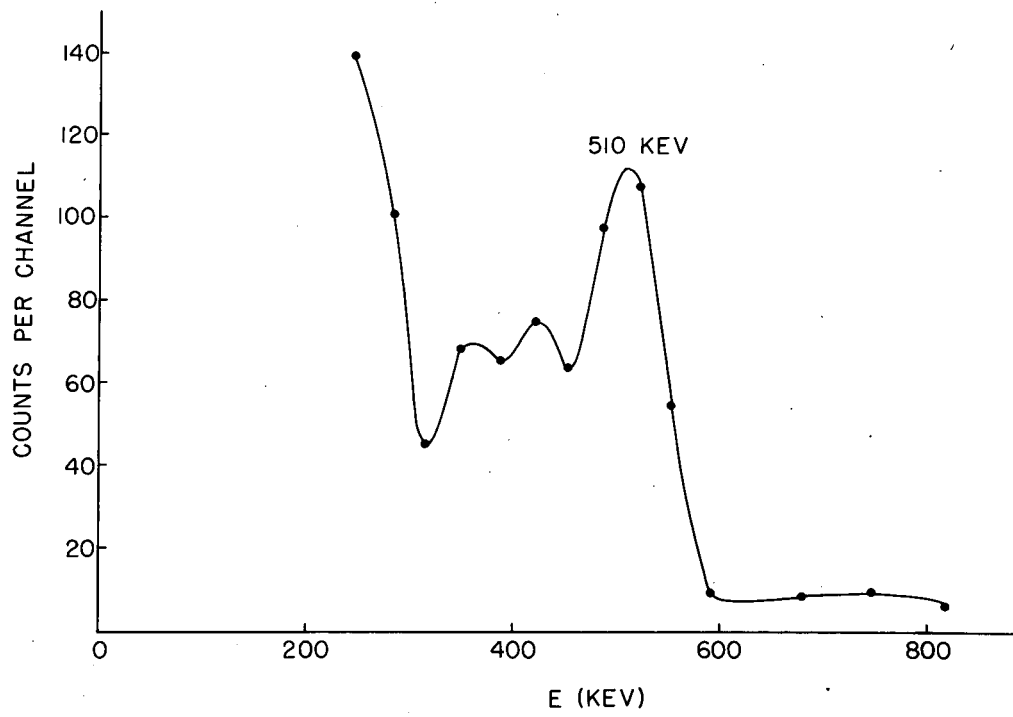


Fig. 6. Decay curves of x-rays (A), 390 keV  $\gamma$ , (B) and 510 keV  $\gamma$  (C) of pure  $Cs^{126}$ .



MU-7706

Fig. 7. Block diagram of coincidence spectrometer.



MU-7707

Fig. 8. Gamma-gamma coincidence counting rate of 385 keV  $\gamma$  as a function of gate setting.

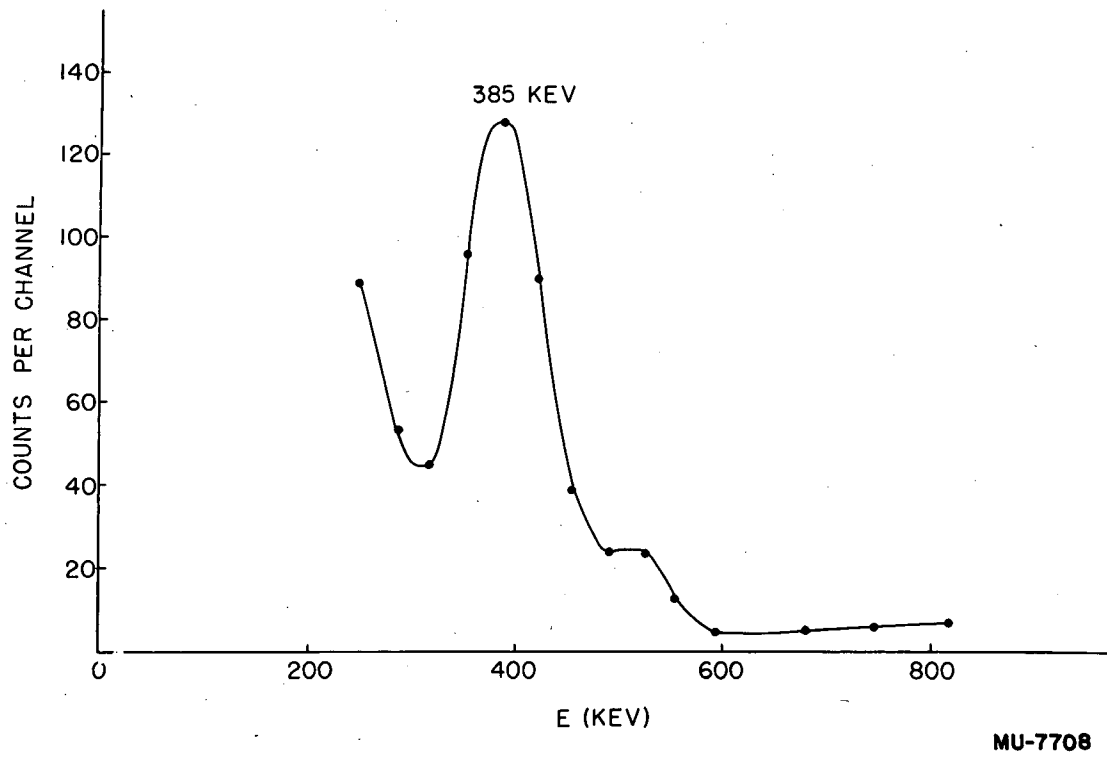
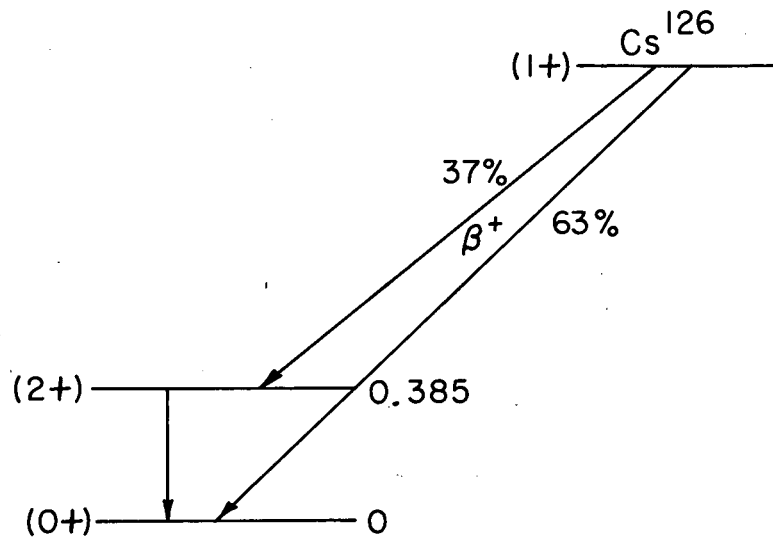


Fig. 9. Gamma-gamma coincidence counting rate of 510 keV  $\gamma$  as a function of gate setting.

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MU-7709

Fig. 10. Decay scheme of  $\text{Cs}^{126}$ .