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NEUTRON-DEFICIENT CESIUM ISOTOPES

Richard W. Fink, Frederick L. Reynolds and D. H. Templeton

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ABSTRACT

The new isotopes Cs^{127} and Cs^{129} have been produced from iodine by irradiation with high energy helium ions. Their mass assignments were determined with a mass spectrograph. Cs^{127} decays with 5.5 ± 0.5 hour half-life with emission of positrons (1.2 Mev maximum energy), giving rise to the daughter activity 34-day Xe^{127}. Cs^{129} decays by electron capture with half-life 31 ± 1 hours. A 30-minute activity, presumed to be Cs^{130}, was also produced, but its mass assignment is uncertain.

NEUTRON-DEFICIENT CESIUM ISOTOPES

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The successful completion of a thermally-ionizing mass spectrograph in this laboratory and the availability of high-energy helium ions from the Berkeley 184-inch cyclotron have led us to investigate neutron-deficient isotopes of alkali metals, which are relatively easy to ionize. Some results for rubidium isotopes have already been reported.[1] This paper is concerned with two new cesium isotopes.

(1) F. L. Reynolds, D. G. Karraker and D. H. Templeton, Phys. Rev. 75, 313 (1949).

The cesium isotopes of mass 131 and greater have been studied extensively.[2]

(2) For references, see Table of Isotopes, G. T. Seaborg and I. Perlman, Revs.Mod. Phys. 20, 585 (1948); also, L. Yaffe, M. Kirsch, S. Standil and J. M. Grunlund, Phys. Rev. 75, 699 (1949); N. Sugarman, Phys. Rev. 75, 1473 (1949); J. L. Meem and F. Maienschein, Phys. Rev. 76, 328 (1949); and J. S. Osoba, Phys. Rev. 76, 345 (1949). The assignment of the 10-day cesium to mass 131 has now been confirmed with the mass spectrograph by D. G. Karraker, F. L. Reynolds and D. H. Templeton, UCRL Report-285, Feb. 11, 1949, not yet published.

The only report of lighter isotopes seems to be a 30-minute period produced from iodine with helium ions, presumably of 16 Mev energy from the Purdue University cyclotron.[3]

(3) "J. R. Risser and R. N. Smith, private communication from K. Lark-Horowitz", quoted from G. T. Seaborg and I. Perlman, loc. cit.

Iodine Bombardments

Stable I^{127} (100% abundance) was bombarded in the form of ammonium iodide with 60 Mev helium ions in the Berkeley 184-inch cyclotron for periods of from one to four hours. The ammonium iodide was wrapped in 0.001-inch aluminum foil and irradiated mounted on a water-cooled copper block on the cyclotron probe. Elemental iodide sealed in a platinum capsule was also bombarded. Ammonium iodide was also bombarded with 36 Mev helium ions from the Crocker 60-inch cyclotron.

The cesium radioactivities induced were isolated from the target material by chemical procedures[4] designed to remove Xe, I, Te, Sb, Sn and most other elements

(4) For details, see R. W. Fink, M. S. Thesis, University of California, Berkeley, 1949.

except alkali metals. Carrier cesium was added in some cases, but it was found that very little could be tolerated in samples destined for the mass spectrograph. It was found that sufficient Cs^{133} to serve as a reference line on the mass spectrum was provided by 0.1 μg. Although the first chemical technique yielded sufficient cesium activity for all experimental purposes except mass spectrograph work, it was found that ignition of cesium chloride in an open dish to remove ammonium salts resulted in serious loss of cesium due to the high volatility of CsCl.

To prevent this loss and thereby isolate sufficient cesium activity for mass spectrograph runs, the ammonium iodide was decomposed in moderate vacuum by gentle heating with a drop of nitric acid and a few drops of sulfuric acid. A cold finger cooled with carbon dioxide snow near the sample condensed the iodine and water which escaped. Within three minutes the residue solution could be removed, containing most of the cesium activity and relatively little of the target material. This procedure resulted in substantially better yields of activity.

Cesium Radioactivities

In Figure 1 is shown the decay curve for the cesium radioactivity from a 60 Mev alpha bombardment of ammonium iodide. The sample, mounted as cesium chloride and covered with cellulose tape, was counted with an argon - alcohol Geiger tube. The decay curve is resolved into 5.5-hour, 31-hour, and 20-day components. When the chemical procedure was completed rapidly, enough, a half-hour period was also observed. With 36 Mev helium ions, only the half-hour and 31-hour activities were observed in the decay curve, Figure 2.

After the 5.5-hour cesium was dead, a portion of the cesium was redissolved, boiled to expel xenon, and evaporated on a counting disk. In Figure 3 are compared the decay curves of such a sample and a portion which was not disturbed. The 31-hour activity is not affected by this procedure. The removal of the long 20-day activity by boiling suggested that it is a xenon daughter of the 5.5 hour cesium. It cannot be the daughter of 31 hour cesium.

To establish the identity of the long-lived activity, a mixture of freshly isolated 5.5-hour and 31-hour cesium activity was placed in a small vacuum line equipped with several glass bulbs. The system was evacuated, the sample was isolated from the pumps and the bulbs. One bulb at a time was opened and cooled with liquid nitrogen, while the sample was heated. Glass wool and loops in the connecting tubing acted as a safeguard against mechanical transfer of non-gaseous material. The bulb was then sealed off and mounted on a holder for counting. The first bulb filled exhibited 109 counts/minute of activity; the walls were too thick for observation of soft radiations. This activity decayed for six weeks with a half-life of 30 to 35 days. Other bulbs filled after the 5.5-hour activity was dead yielded no observable activity.

This experiment was repeated twice with similar results. On the basis of these observations, the long-lived activity is identified as the 34-day xenon[5]

(5) E. C. Creutz, L. A. Delsasso, R. B. Sutton, M. G. White, and W. H. Barkas, Phys. Rev. 58, 481 (1940); M. Camac, Plutonium Project Report CC-2409 (Oct. 1944); R. Overstreet, L. Jacobson, K. Scott and J. G. Hamilton, Plutonium Project Report CH-379 (Dec. 1942).

assigned to Xe^{127}, produced by decay of the 5.5 hour cesium. Xe^{127} is reported to emit a 0.9 Mev gamma-ray, which is the activity counted in the glass bulb. The shorter half-life observed in cesium samples is caused by diffusion of xenon through the cellulose tape covering the sample.

Mass-spectrographic Experiments

In our mass spectrograph the sample is placed as a solid on a tungsten or platinum filament and is ionized by heat. The efficiency of the ionization for cesium has been found to be strongly dependent on the chemical state of the sample. Similar effects have been noted for some other elements, such as barium and thallium. If the photographic plate is replaced by an electrometer receiver, an integration of the current is a measure of the number of atoms ionized and passed by the slits of the apparatus. The ratio of the amount of sample placed on the filament to the amount found with the electrometer is called the "loss factor".

Experimental values for three cesium salts are listed in Table 1.

TABLE 1

LOSS FACTORS OF VARIOUS CESIUM SALTS ON THE MASS SPECTROGRAPH

Salt	Loss Factor
Cesium Nitrate, $CsNO_3$	1.4×10^6
Cesium Chloride, CsCl	8.3×10^4
Cesium Sulfate, Cs_2SO_4	180

Since the geometrical loss factor on the mass spectrograph is of the order of 50, with cesium sulfate at least 25% of the cesium is ionized. One expects 100% ionization under optimum conditions, because of the very low ionization potential (3.87 volts) of cesium. The very low ionization of the chloride and nitrate salts is probably due to greater volatility in vacuum. The presence of ammonium salts or other foreign matter operates to lower the ionization efficiency by increasing mechanical loss of chunks of material from the platinum filament. Only by increasing the yield of activity by use of the vacuum sublimation procedure combined with especial care to prepare practically weighless samples of the sulfate of the radioactive cesium isotopes, instead of the chloride used in early experiments, were successful mass spectrograph results finally attained.

Mass spectrograph plates showing dark lines at mass numbers 127, 129, and 133 (stable carrier) were obtained, and transfer plates showed the radioactivity of lines 127 and 129; after the 5.5 hour Cs^{127} activity had effectively decayed out, the radioactive line at mass 129 still gave good transfer plates. This is shown in Figure 4 which consists of microphotometer tracings of the original mass spectrograph plate and three transfer plates made at designated times after bombardment.

Radiations of 5.5 hour Cs^{127}

A crude magnetic spectrometer and absorption methods were used to examine the radiations from Cs^{127} and Cs^{129}. Positrons were found with maximum energy about 1.2 Mev. The activity of these positrons decayed with a 5.5 hour half-life. Lead absorption curves showed 0.5 Mev gamma rays due to annihilation but no harder gamma rays. Softer gammas may have been present. Negative electrons of energy of the order of 0.3 Mev and x-rays were also present, but they have not been proved to belong to Cs^{127}.

Radiations of 31-hour Cs^{129}

No positrons were observed for Cs^{129}, and it very likely decays entirely by electron capture, It emits 0.3 Mev conversion electrons, K and L x-rays, and gamma rays of about 0.5 Mev.

Radiations of the 30-minute Cesium

We have little data concerning the 30-minute activity. It emits x-rays and gamma rays of the order 0.5 Mev. We have not tested it for positrons or negative electrons. Its production with 16 Mev helium ions[3] implies that it is Cs^{130}. Our data are consistent with that assignment. Cs^{128} has not been observed, but its half-life must be 30 minutes, or less. It is conceivable that our 30-minute activity is in part due to Cs^{128}.

ACKNOWLEDGEMENTS

We wish to express our sincere appreciation to James T. Vale and B. Rossi and the crews of the 184-inch and 60-inch cyclotrons; to D. G. Karraker, for operation of the mass spectrograph on many night runs; and to Lieselotte K. Templeton, for microphotometer tracings of the mass spectrograph plates. This work was supported by the Atomic Energy Commission.

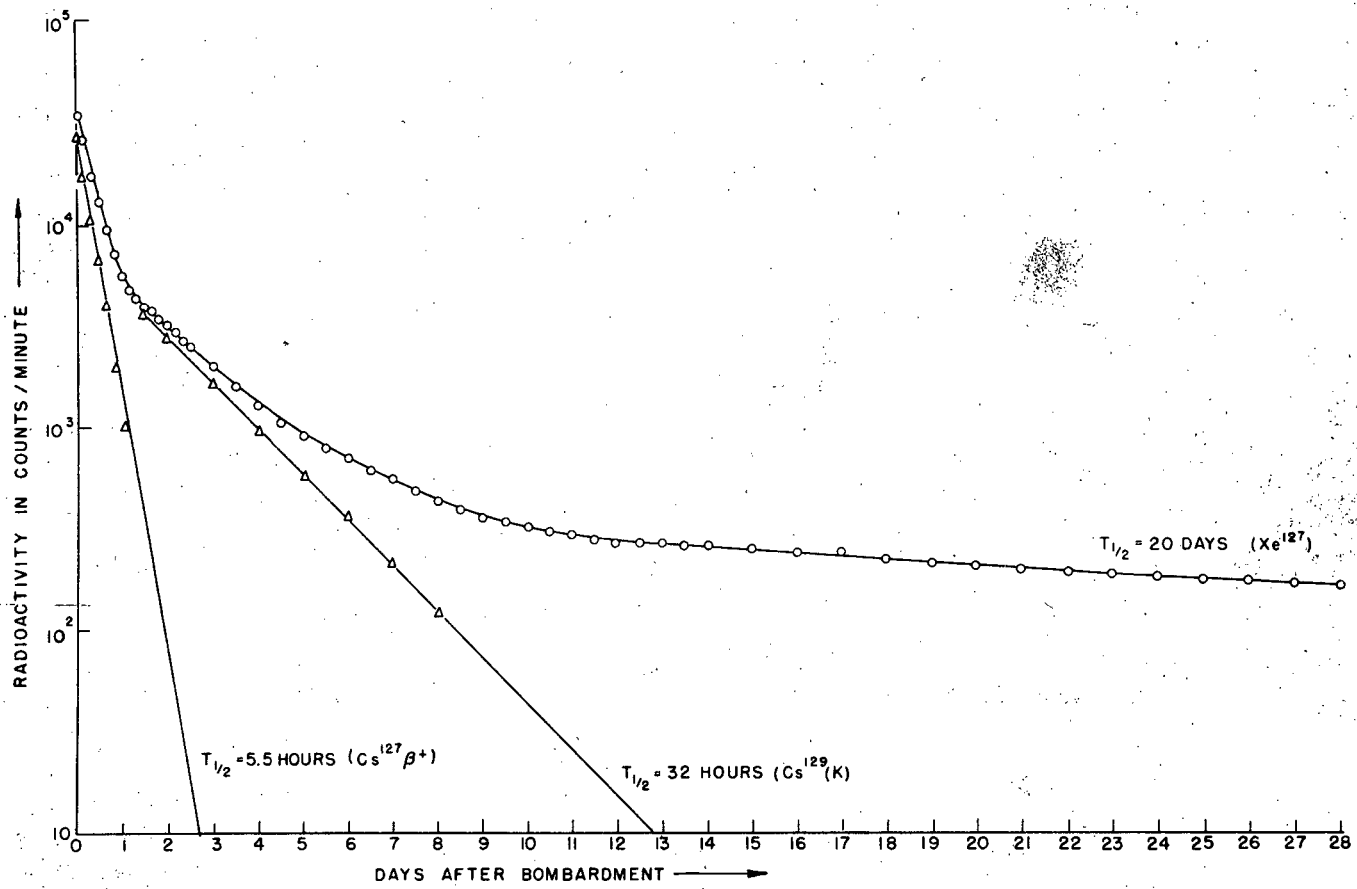

RADIOACTIVITY IN COUNTS / MINUTE
DAYS AFTER BOMBARDMENT
$T_{1/2}$ = 20 DAYS (Xe^{127})
$T_{1/2}$ = 5.5 HOURS ($Cs^{127}\beta^+$)
$T_{1/2}$ = 32 HOURS (Cs^{129}(K)

FIG. 1

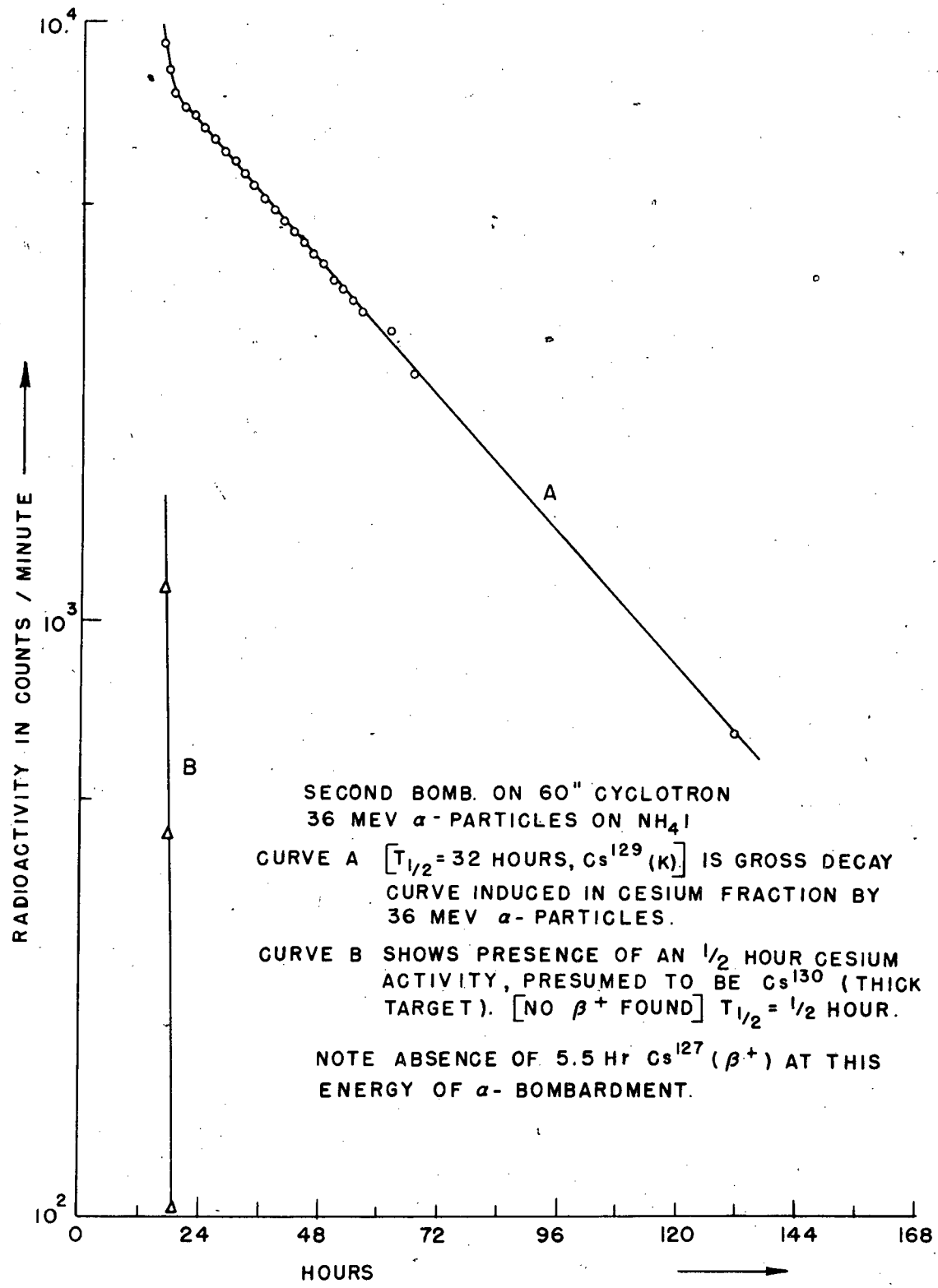

10^4
10^3
10^2
RADIOACTIVITY IN COUNTS / MINUTE
A
B
SECOND BOMB. ON 60" CYCLOTRON
36 MEV α - PARTICLES ON NH_4I
CURVE A [$T_{1/2}$ = 32 HOURS, Cs^{129} (K)] IS GROSS DECAY CURVE INDUCED IN CESIUM FRACTION BY 36 MEV α - PARTICLES.
CURVE B SHOWS PRESENCE OF AN 1/2 HOUR CESIUM ACTIVITY, PRESUMED TO BE Cs^{130} (THICK TARGET). [NO β^+ FOUND] $T_{1/2}$ = 1/2 HOUR.
NOTE ABSENCE OF 5.5 Hr Cs^{127} (β^+) AT THIS ENERGY OF α - BOMBARDMENT.
0
24
48
72
96
120
144
168
HOURS

FIG. 2

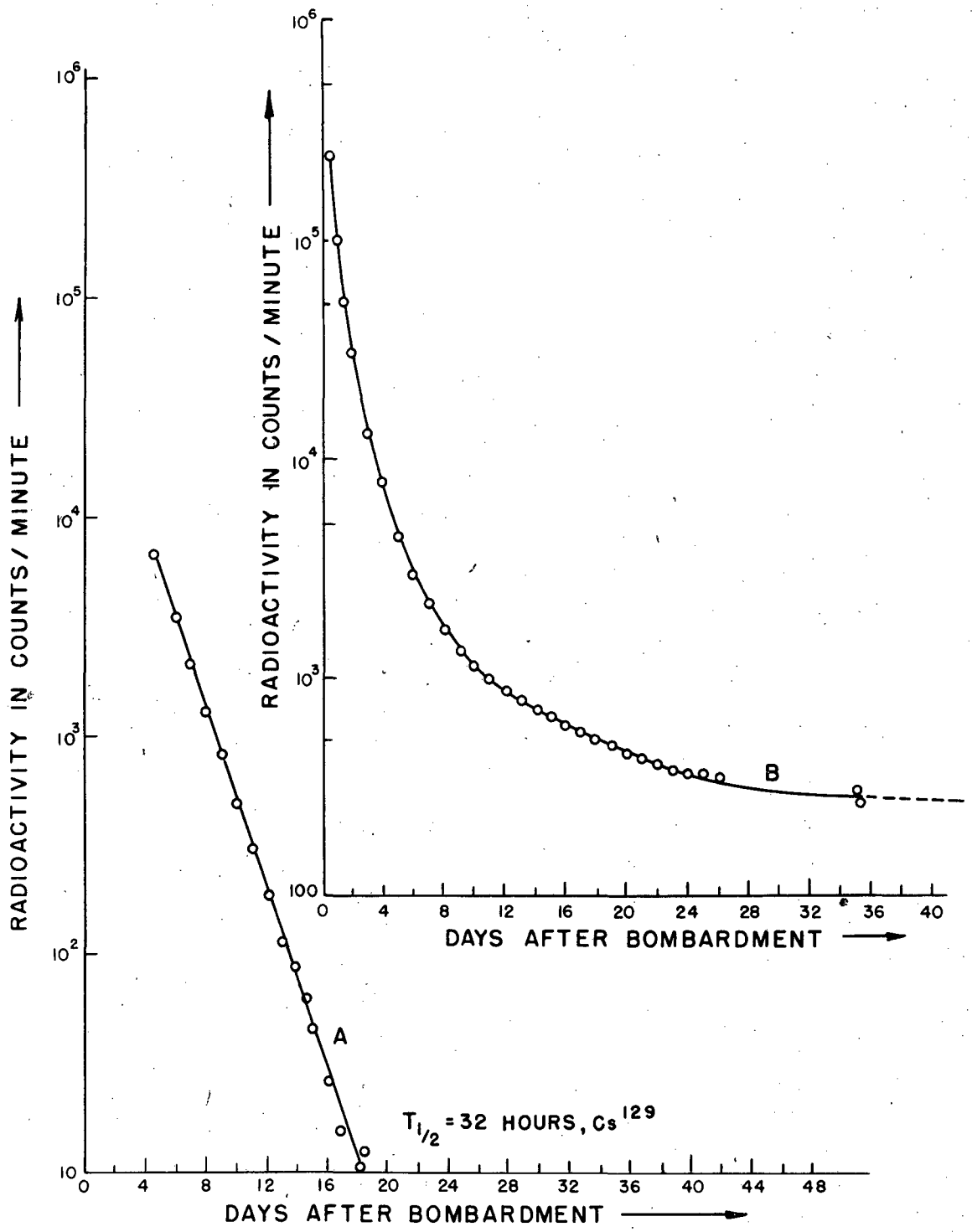
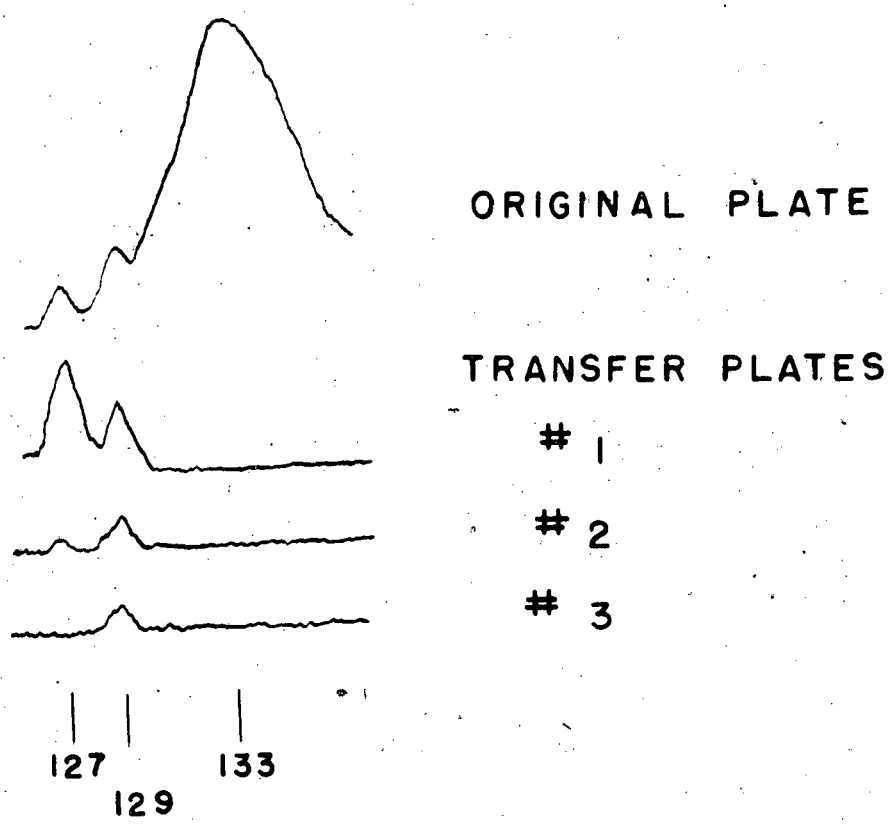

RADIOACTIVITY IN COUNTS / MINUTE
DAYS AFTER BOMBARDMENT
A
B
$T_{1/2}$ = 32 HOURS, Cs^{129}

FIG. 3



MICROPHOTOMETER TRACINGS OF MASS SPECTROGRAPH PLATES SHOWING Cs^{127}, Cs^{129}, AND STABLE Cs^{133}.

FIG. 4

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