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PROPERTIES OF SOME NEUTRON-DEFICIENT CESIUM ISOTOPES

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July, 1949

Berkeley, California

Thesis of

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to be submitted to the

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PROPERTIES OF SOME NEUTRON-DEFICIENT CESIUM ISOTOPES

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July, 1949

ABSTRACT

The discovery of two new neutron-deficient cesium isotopes, Cs^{127} and Cs^{129}, together with some observations on a 30 minute cesium activity is reported. The following radiation characteristics are reported:

Cs^{127}, half-life 5.5 ± 0.5 hours, positrons of maximum energy 1.2 Mev. Annihilation Radiation observed: 0.51 Mev gamma rays. Probable internal conversion electrons of energy 0.35 ± 0.05 Mev. Genetically produces Xe^{127} daughter. No harder gamma-rays.

Cs^{129}, half-life 31 ± 1 hours, no detectable positrons. K-capture. No hard gamma-rays, but probable emission of a 0.5 Mev gamma-ray. 0.35 ± 0.05 Mev conversion electrons.

Cs^{130}, half-life 30 minutes. K-capture. Emits 0.5 Mev gamma-ray. Mass number presumed from literature reference, and yields.

Proof for the genetic relationship $Cs^{127} \xrightarrow{\beta^+} Xe^{127}$ is given by both repurification experiments and direct-vacuum line isolation of the daughter radioxenon with identification of its 34 day half-life.

Mass spectrograph proof for the mass assignments is given, showing that mass number 127 corresponds to the 5.5 hour positron-emitter and mass number 129 corresponds to the 31 hour K-capture cesium isotope.

PROPERTIES OF SOME NEUTRON-DEFICIENT CESIUM ISOTOPES[1]

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This research was launched to explore the light mass region of cesium isotopes for possible new radioactivities and to extend this knowledge of the light region somewhat further. The recent completion of a thermally-ionizing mass spectrograph instrument at the Radiation Laboratory, coupled with the availability of high energy alpha-particles from the 184-inch cyclotron, made the choice of cesium as the element to be investigated most appropriate, since cesium is very easily ionized on a thermal filament source in the mass spectrograph.

Two new, neutron-deficient cesium isotopes have been discovered by bombarding stable I^{127} (100% abundance) in the form of ammonium iodide with 60 Mev alpha-particles in the 184-inch cyclotron for periods of from one to four hours. The ammonium iodide was wrapped in 0.001-inch aluminum foil and bombarded mounted on water-cooled copper blocks. Many 60 Mev bombardments were performed as well as one at 36 Mev on the 60-inch cyclotron.

The cesium radioactivities induced were isolated from the target material by use of the following chemical procedure, which on certain bombardments was abridged with no change in results:

NH_4I target. 20 μg Cs carrier, few drops each of concentrated HNO_3 and concentrated HCl added. Ignited to evanescence of all visible iodine and ammonium salts.

Volatile Matter
Xe, I_2, F, NH_3, etc.

Residue
Taken up in dilute HNO_3; precipitated thrice with $AgNO_3$ and HCl to scavenge any remaining halogen impurities.

Solution Remaining from AgCl Precipitations

Tellurium carrier added and then excess $SnCl_2$ (The Te step was subsequently omitted with no change in results).

Precipitate
Te

Solution Remaining

Made alkaline with concentrated NH_4OH and $FeCl_3$ solution added. $Fe(OH)_3$ was twice precipitated as scavenger for any remaining impurities.

Precipitate
$Fe(OH)_3$

Final Solution
Radioactive Cesium

Although the above chemical technique yielded sufficient cesium activity for all experimental purposes except mass spectrograph work, it became apparent that an appreciable loss of cesium radioactivity was experienced in the open crucible ignition to expell ammonium salts, since CsCl has considerable volatility at 1000° K. To confirm this, a tracer cesium run was made with the ignition procedure. Some ammonium iodide was added to several thousand counts/minute of tracer cesium radioactivity, a few drops of concentrated nitric acid and concentrated hydrochloric acid and 20 μg of cesium carrier introduced, and the whole ignited to

evanescence of ammonium salts in a porcelain crucible. The residue was again counted on a plate and a loss factor of 53% was obtained; this figure would be much higher on a target run. To prevent this loss and thereby isolate sufficient cesium activity for mass spectrograph runs, an iodine sublimating apparatus was employed. Figure 1 shows the cesium sublimator used to isolate radioactive cesium from target ammonium iodide. Carbon dioxide snow was placed in the condensor a. Freshly-bombarded ammonium iodide together with a drop of concentrated nitric acid and a few drops of concentrated sulfuric acid is introduced at c. The sublimator is then closed and a low vacuum pulled at b, heat being applied at c until complete decomposition results, as evidenced by the lack of iodine color of the solution remaining at c. The condenser becomes coated with free iodine and ice. The decomposition occurs in less than one minute, and in three minutes the solution may be withdrawn, nearly free from extraneous matter, for further chemical treatment. This sublimating procedure replaces the open-crucible ignition step in the previously described chemical procedure; the result is an appreciable gain in total activity finally isolated in the cesium fraction over the yield from the ignition method.

The gross decay curve, Figure 2, always reproducible with 60 Mev alpha-bombardments, was obtained from an argon-filled Geiger counter and was resolved into a 5.5 hour, a 31 to 32 hour, and a long 20-day activity. It subsequently developed that this long 20-day tail was in reality gaseous xenon daughter of 34 day reported half-life[2] slowly diffusing through the cellulose tape covering the sample, giving the appearance of a 20-day activity. Occasionally, the chemical procedure was finished in time to allow observation of a half-hour cesium activity.

A crude magnetic spectrometer ("bender") was used to aid in characterizing the radiations emitted from the two new cesium activities. It was found that the 5.5 hour cesium emitted positrons of maximum energy 1.2 Mev, checked reasonably

Figure 1: Sublimator used to separate radioactive cesium from ammonium iodide target material. Target NH_4I and HNO_3 with H_2SO_4 and carrier Cs if desired are introduced at c. CO_2 snow is added to the condenser a and a low vacuum pulled at b, while bunsen burner heat is applied at c. In three minutes complete decomposition is attained and the solution at c, free from iodine color, is ready for further chemical use or mass spectrograph runs.

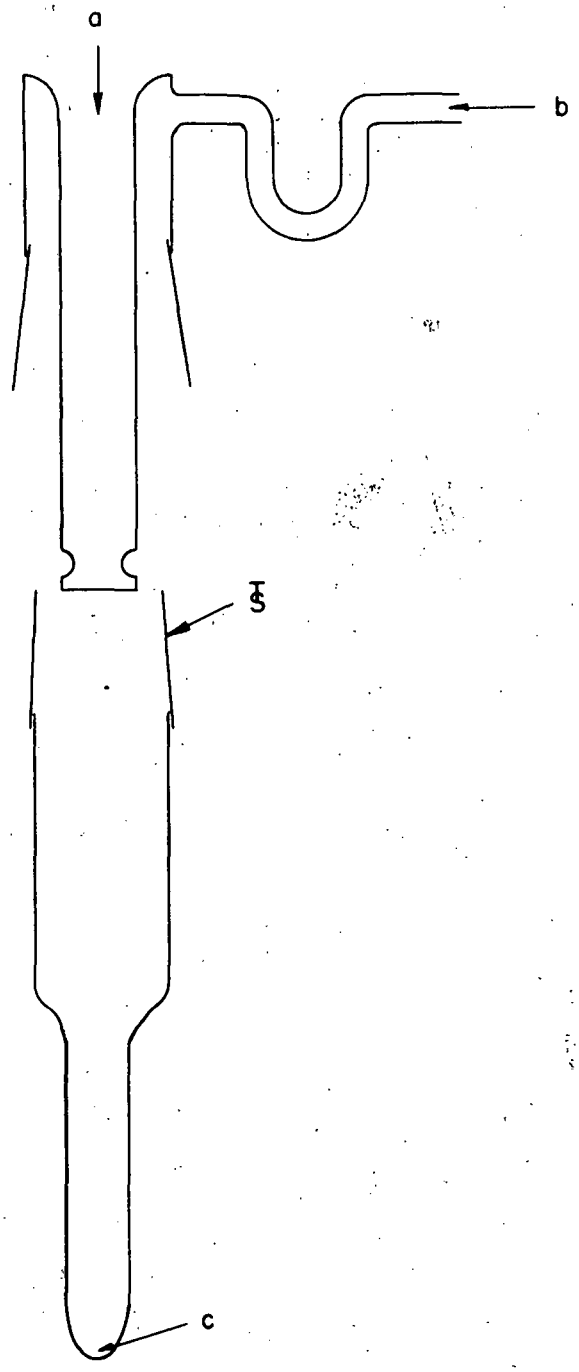


FIG. 1

Figure 2: Gross decay curve of cesium radioactivities induced by 60 Mev alpha-particles on ammonium iodide. The long 20-day tail is due to Xe^{127} of 34 days reported half-life[2] slowly diffusing through the cellulose tape covering the sample.

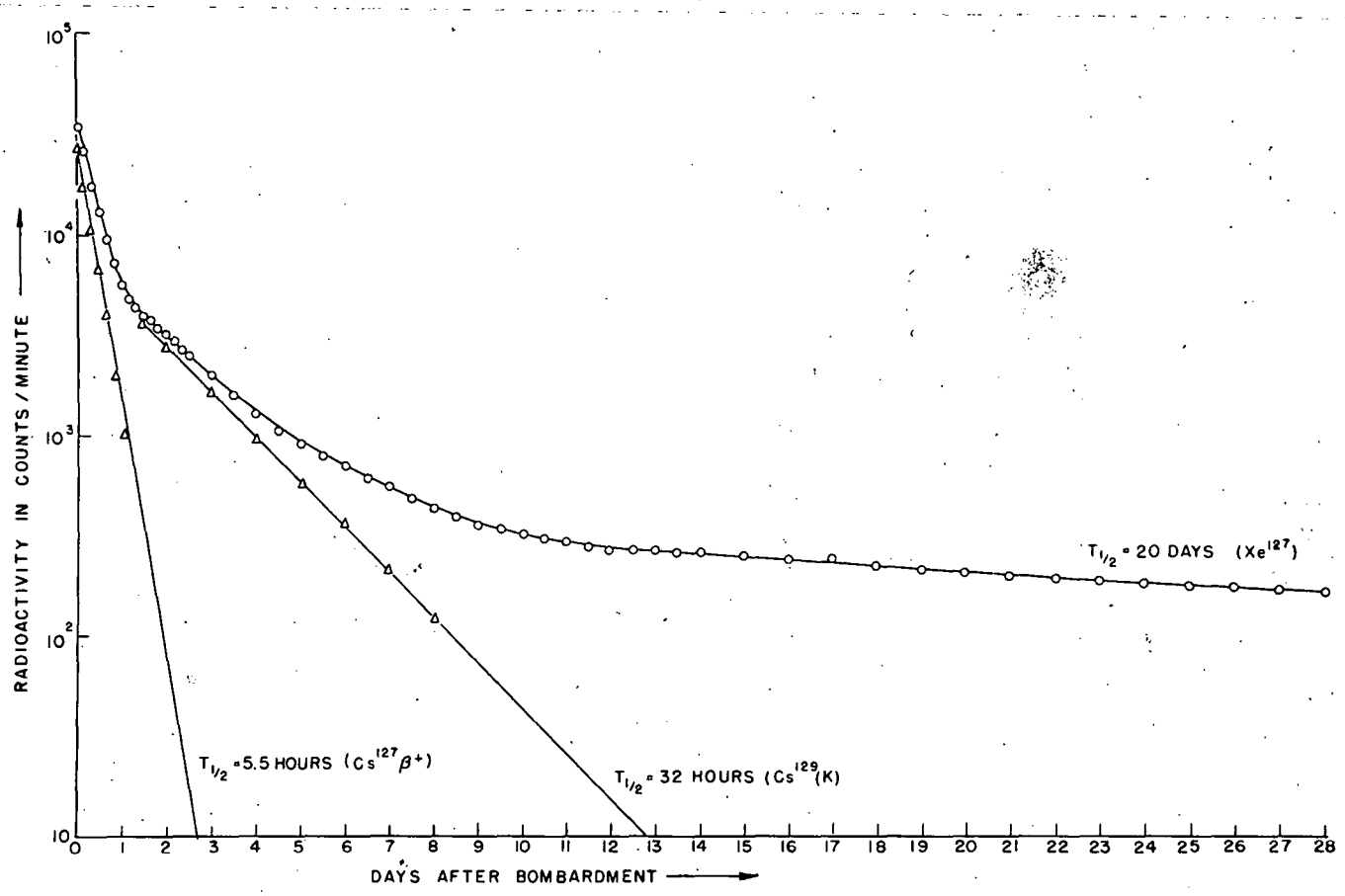

RADIOACTIVITY IN COUNTS / MINUTE
DAYS AFTER BOMBARDMENT
$T_{1/2}$ = 20 DAYS (Xe^{127})
$T_{1/2}$ = 5.5 HOURS ($Cs^{127}\beta^+$)
$T_{1/2}$ = 32 HOURS (Cs^{129}(K)

FIG. 2

well by values obtained from aluminum and aluminum-plus-beryllium absorption curves, one of which is shown in Figure 3. These absorption curves give maximum positron energies for Cs^{127} ranging from 0.94 to 1.19 Mev. The activity of these positrons was followed on the magnetic spectrometer overnight and found to decay with the 5.5 hour half-life shown below to correspond to Cs^{127}. The 31 hour half-life, shown below to be Cs^{129} was found to emit internal conversion electrons of 0.33 Mev energy by the crude magnetic spectrometer, and this value is roughly checked by aluminum and aluminum-plus-beryllium absorption curves, Figure 4, taken after the 5.5 hour activity had effectively died out and when only Cs^{129} remained active. No positrons could be detected emanating from the 31 hour electron-capturing isotope, even with a rather sensitive counting rate meter installed in the magnetic spectrometer, so that it appears likely that 31 hour Cs^{129} is a pure electron-capturing isotope. The 5.5 hour positron-emitter, Cs^{127}, showed 0.51 Mev gamma-rays due to annihilation radiation on hard lead absorption measurements, and no evidence for gamma-rays of energy higher than this was obtained from these lead absorption curves. A mixture of two cesium activities, 30 minutes and 31 hours, was placed in an electromagnetic radiation counter and covered with 196.4 mg/cm^2 beryllium absorber. This counter was fitted with a permanent alnico magnet of 1600 gauss, and xenon-filled Geiger tube, of such geometrical arrangement that all charged particles of energy less than 2.4 Mev would be bent away from the tube and not counted. A decay curve from this counter exhibited 30 minute and 31 hour x-rays, with some gamma-radiation. This indicates that the 30 minute and 31 hour cesium isotopes are electron-capturing nuclei, but little can be said for 5.5 hour cesium since it was formed in lowered yield in this particular bombardment due to thick target. This target consisted of a 1/8-inch diameter, 3 mil platinum tube containing a solid rod of free iodine. This constitutes a rather "thick" target to 60 Mev alpha-particles, leading to an average bombardment energy

Figure 3: Aluminum and aluminum-plus-beryllium absorption curves of a mixture of 5.5 hour and 31 hour cesium activities. Curve A is a straight aluminum absorption curve, while curve B is an aluminum absorption curve with 196.4 mg/cm^2 beryllium absorber interposed between sample and counter. Curve B represents electromagnetic radiation only. Curve A exhibits a line of conversion electrons which is seen near the beginning of the curve.

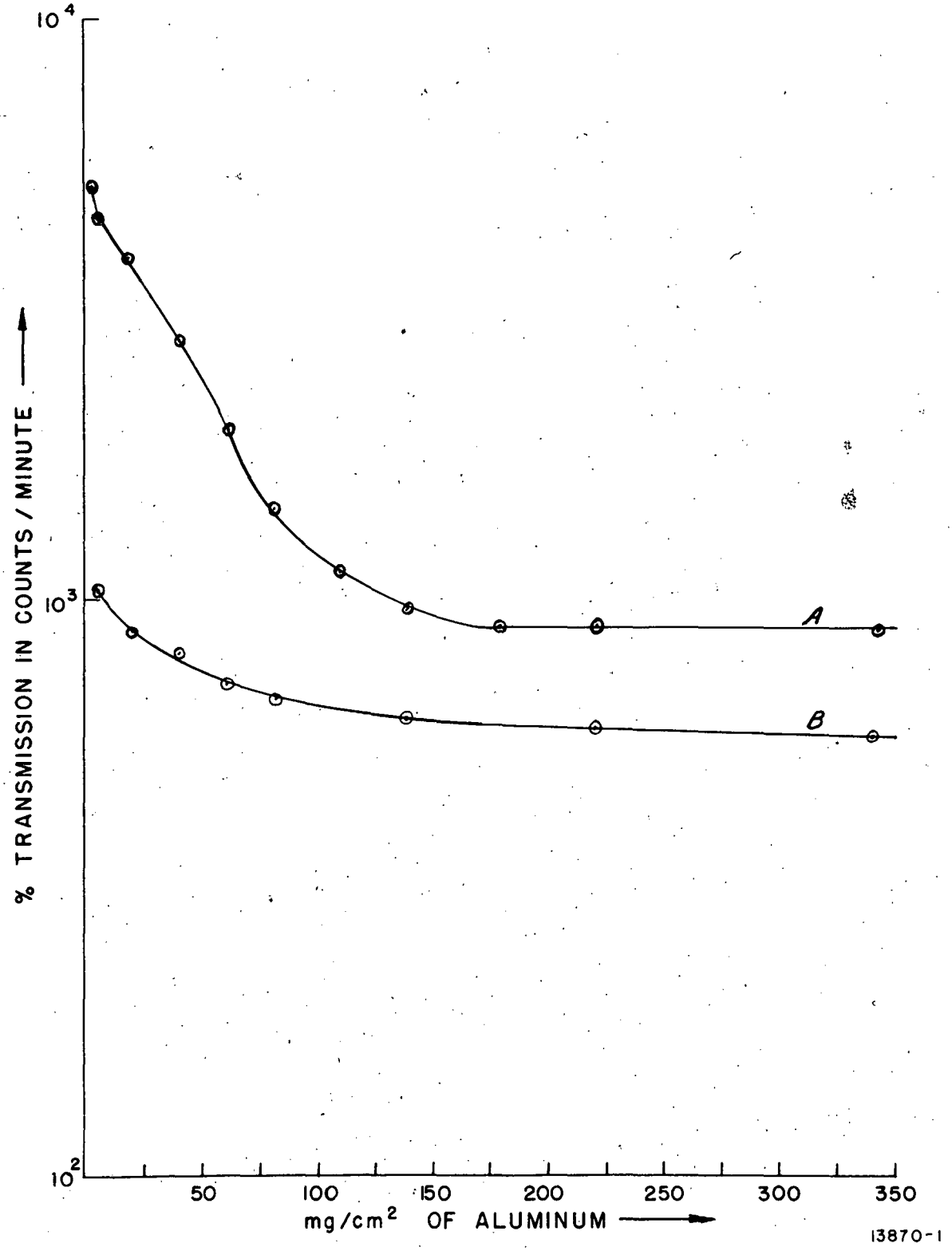

10^4
10^3
10^2
% TRANSMISSION IN COUNTS / MINUTE
A
B
50
100
150
200
250
300
350
mg/cm² OF ALUMINUM
13870-1

FIG. 3

Figure 4: Cesium absorption curve after 4 days of decay, so that Cs^{129} only is active. Curve A is absorption in aluminum only; curve B is aluminum absorption with 90.5 mg/cm^2 beryllium interposed; curve C is that of conversion electrons; and curve D is L x-rays of energy of about 4 Kev roughly.

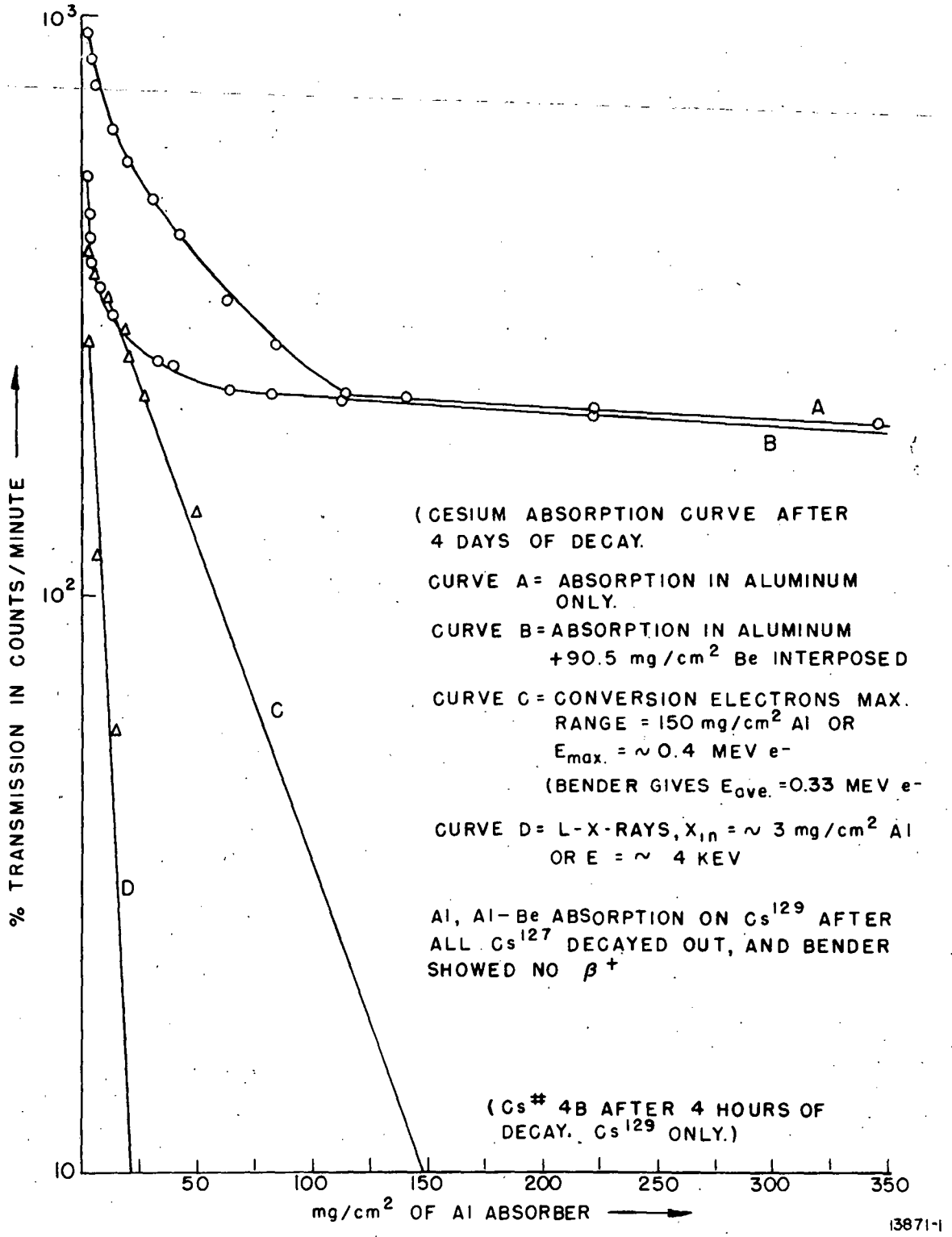

10³
10²
10
% TRANSMISSION IN COUNTS/MINUTE
A
B
C
D
(CESIUM ABSORPTION CURVE AFTER 4 DAYS OF DECAY.
CURVE A = ABSORPTION IN ALUMINUM ONLY.
CURVE B = ABSORPTION IN ALUMINUM +90.5 mg/cm² Be INTERPOSED
CURVE C = CONVERSION ELECTRONS MAX. RANGE = 150 mg/cm² Al OR E_max. = ~0.4 MEV e⁻
(BENDER GIVES E_ave. = 0.33 MEV e⁻
CURVE D = L-X-RAYS, X_in = ~ 3 mg/cm² Al OR E = ~ 4 KEV
Al, Al-Be ABSORPTION ON Cs¹²⁹ AFTER ALL Cs¹²⁷ DECAYED OUT, AND BENDER SHOWED NO β⁺
(Cs# 4B AFTER 4 HOURS OF DECAY. Cs¹²⁹ ONLY.)
50
100
150
200
250
300
350
mg/cm² OF Al ABSORBER
13871-1

FIG. 4

less than 60 Mev, so that the yield of Cs^{127} was very much lowered relative to the yields of 30 minute and 31 hour cesium isotopes. This experiment, and the bombardment at 36 Mev mentioned subsequently, lead one to consider that the 30 minute K-capture cesium activity is, in the absence of other evidence, Cs^{130}, rather than Cs^{128}, on the basis of previous work[3].

A soft lead absorption curve, Figure 5, was taken with both cesium activities, 5.5 hour and 31 hour, present with 599 mg/cm^2 beryllium interposed between sample and Geiger tube; the presence of a 31 Kev K x-ray is observed in addition to 0.51 Mev. annihilation radiation. Hard lead absorption curves showed no evidence of gamma-rays harder than 0.51 Mev emitted by any cesium isotope studied. It was observed that the 30 minute cesium activity also emitted a gamma-ray of 0.51 Mev, but no experiments have been performed as yet to determine whether the 30 minute activity also emits positrons. In aluminum and aluminum-plus-beryllium absorption curves, in pure beryllium absorption curves, and in x-ray counting, L-x-rays have been observed from the 31 hour activity, after all other cesium activities had decayed out.

It is interesting to note that when ammonium iodide was bombarded with 36 Mev alpha-particles on the 60-inch cyclotron, the cesium radioactivities isolated had no observable positrons on the crude magnetic spectrometer and exhibited a gross decay curve having only 30 minute and 31 hour cesium activities, while the 5.5 hour positron-emitter was conspicuously absent. This decay curve is shown in Figure 6. Evidently, 36 Mev is below the practical threshold for production of observable quantities of Cs^{127}, a fact consistent with present ideas on excitation functions for (α,xn) nuclear reactions.

To make the mass number assignments of these new light cesium isotopes, both mass spectrograph and daughter xenon isolation experiments were performed. The proof for the genetic relationship $Cs^{127} \xrightarrow{\beta^+} Xe^{127}$ will be discussed first.

Figure 5: Soft lead absorption curve taken in the presence of 5.5 hour and 31 hour cesium activities. Curve A is the experimental curve, with 599 mg/cm^2 Be interposed. Curve B is a 31 Kev K x-ray resolved from Curve A.

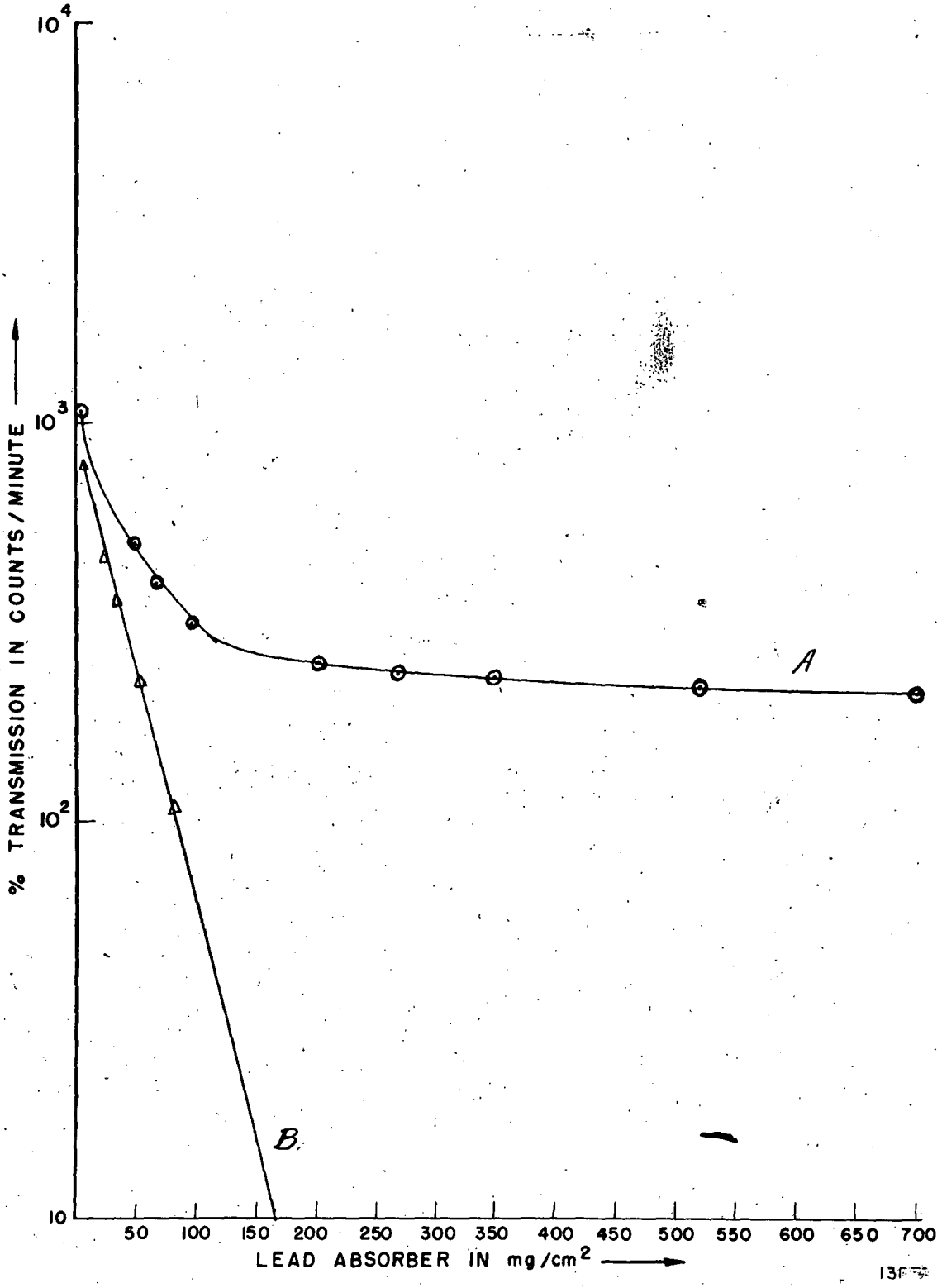

10^4
10^3
10^2
10
% TRANSMISSION IN COUNTS / MINUTE
A
B
50
100
150
200
250
300
350
400
450
500
550
600
650
700
LEAD ABSORBER IN mg/cm^2

FIG. 5

Figure 6: Gross decay curve of cesium radioactivities induced by bombarding NH_4I with 36 Mev alpha-particles on 60-inch cyclotron for one hour. The presence of 30 minute and 31 hour activities is noted, but the absence of the 5.5 hour activity induced at higher energies and the absence of the long 20 day tail of xenon daughter diffusing through cellulose tape cover are conspicuous.

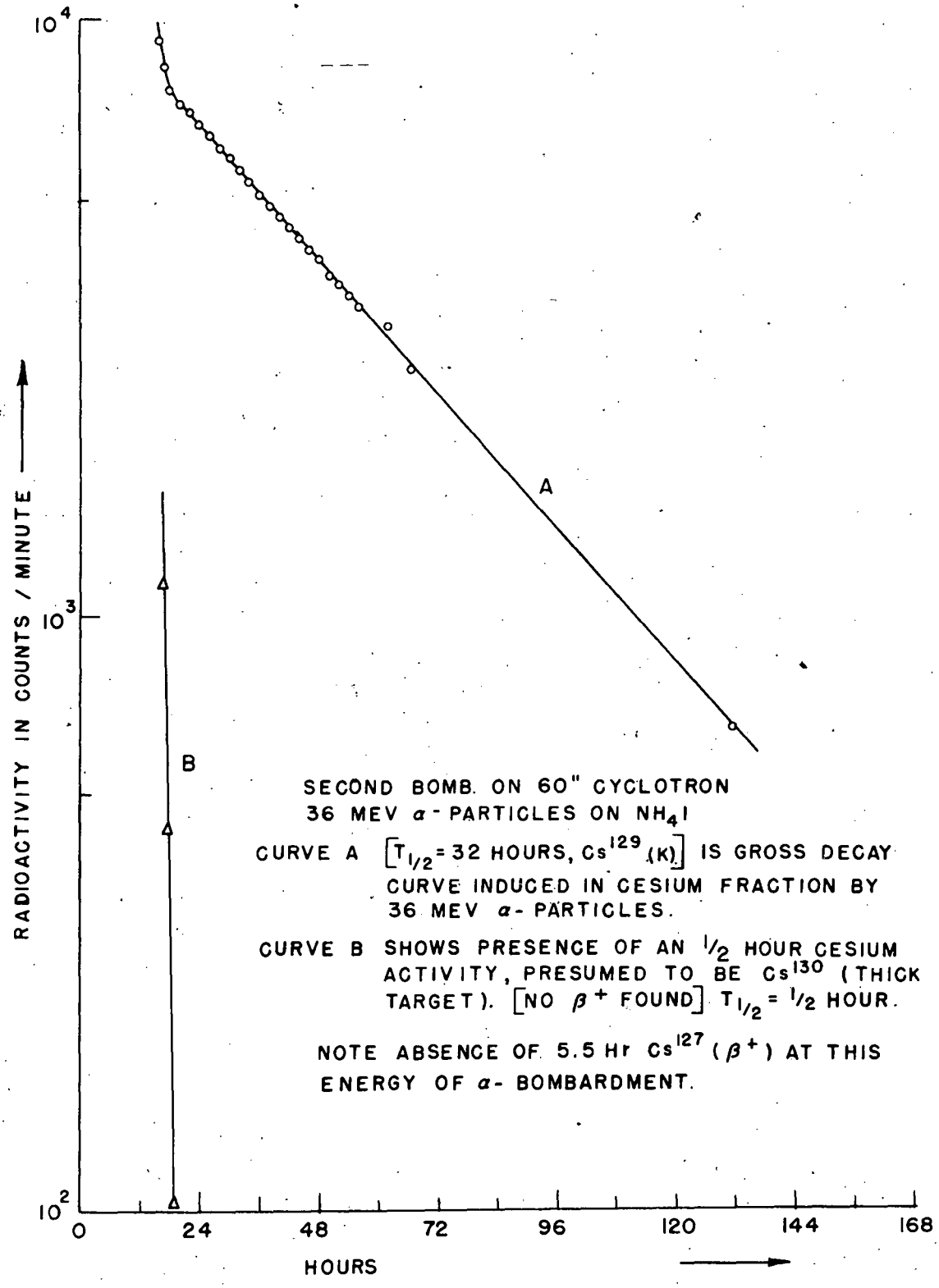

10^4
10^3
10^2
RADIOACTIVITY IN COUNTS / MINUTE
A
B
SECOND BOMB. ON 60" CYCLOTRON
36 MEV α - PARTICLES ON NH_4I
CURVE A [$T_{1/2}$ = 32 HOURS, Cs^{129} (K)] IS GROSS DECAY CURVE INDUCED IN CESIUM FRACTION BY 36 MEV α - PARTICLES.
CURVE B SHOWS PRESENCE OF AN 1/2 HOUR CESIUM ACTIVITY, PRESUMED TO BE Cs^{130} (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. 6

It was early suspected that the apparent 20-day tail (Figure 2) in the gross cesium decay curves was perhaps the previously reported 34 day Xe^{127}, gradually diffusing through the cellulose tape covering the samples; to test if this were derived only from the 5.5 hour cesium positron-emitter and not from the 31 hour K-capturing activity, repurification experiments were performed in which a sample of an initial mixture of the two activities was dissolved in 1:1 HCl after the 5.5 hour cesium isotope had effectively decayed out, but while the 31 hour cesium was still very active. The gaseous xenon daughter radioactivity was expelled by boiling, the activity again placed on a sample plate, covered with cellulose tape, and the gross decay followed and compared with a similar sample which was not so repurified. These repurification experiments are shown in Figure 7, where curve A represents the decay of the cesium sample thus repurified and where curve B shows another similar sample from the same bombardment which was not repurified and consequently tailed out to the long 20-day activity characteristic of 34-day daughter Xe^{127} diffusing through the cellulose tape. This experiment established that the xenon daughter was growing from the 5.5 hour positron-emitter, rather than from the 31 hour K-capturing isotope.

To establish the fact that this xenon daughter was actually the 34 day Xe^{127} reported in the literature to emit a 0.9 Mev gamma-ray[2], a bombardment of freshly-isolated cesium, containing both 5.5 hour and 31 hour activities, was placed in an apparatus on a vacuum line. This apparatus, Figure 8, consisted of three 5 cc pyrex glass bulbs, each of which was fitted with a stopcock f, and a connecting tube g into the bottom of which the cesium sample was introduced. A plug of glass wool c and a circular loop in the glass tube connecting each bulb to its stopcock prevented the possibility of any non-gaseous radioactivity from being mechanically carried into the bulbs d. The bulbs and apparatus were evacuated at a to 10^{-3} mm of Hg after introducing the cesium sample, and when evacuated, the

Figure 7: Repurification experiments showing that the long 20-day tail is due to xenon daughter which grows from the 5.5 hour positron-emitting cesium only, and not from the 31 hour isotope. Curve A shows a sample from which the xenon was expelled by boiling after all 5.5 hour activity had effectively died, while curve B shows a gross decay of a similar sample not so repurified. The time axis of both samples is identical.

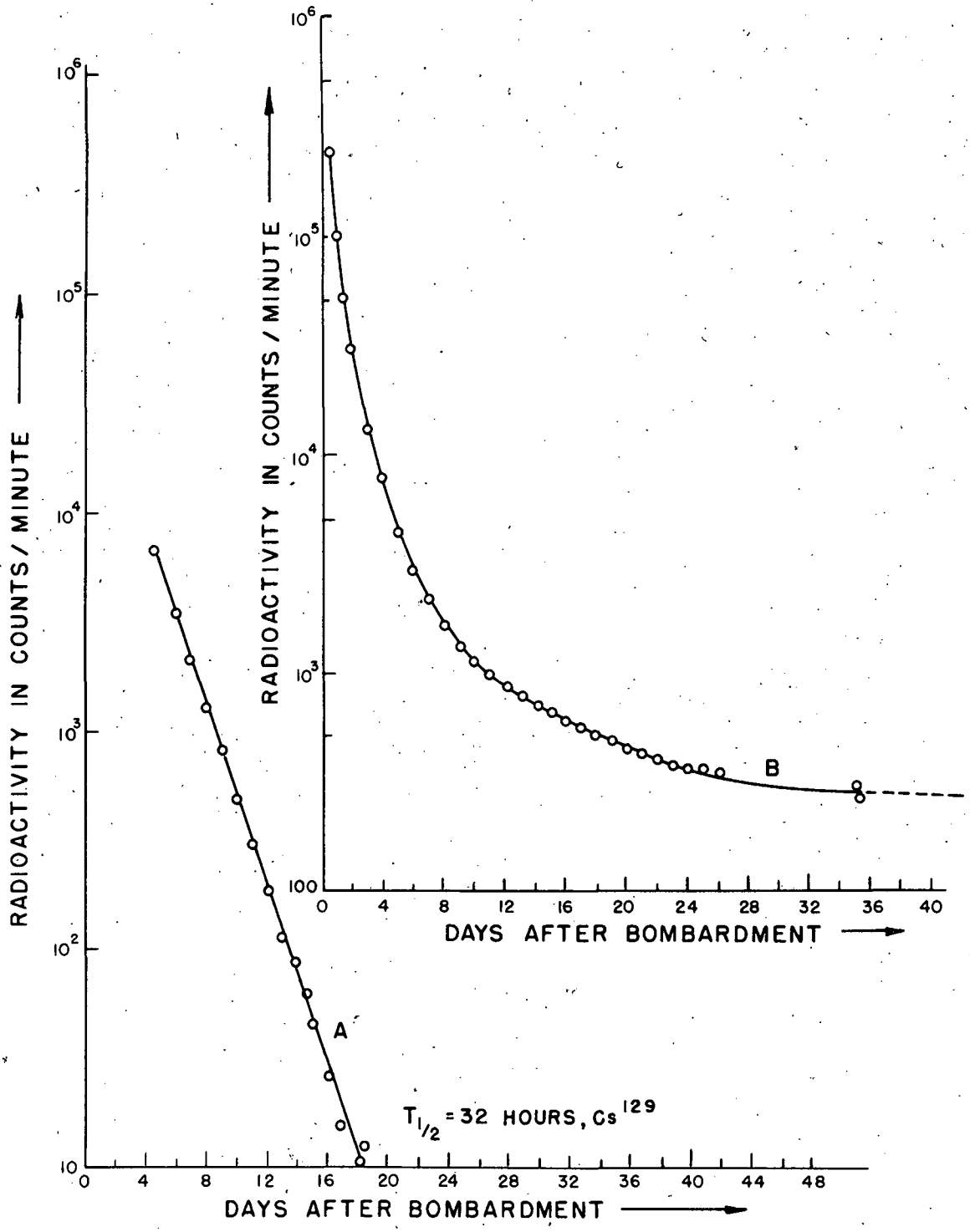
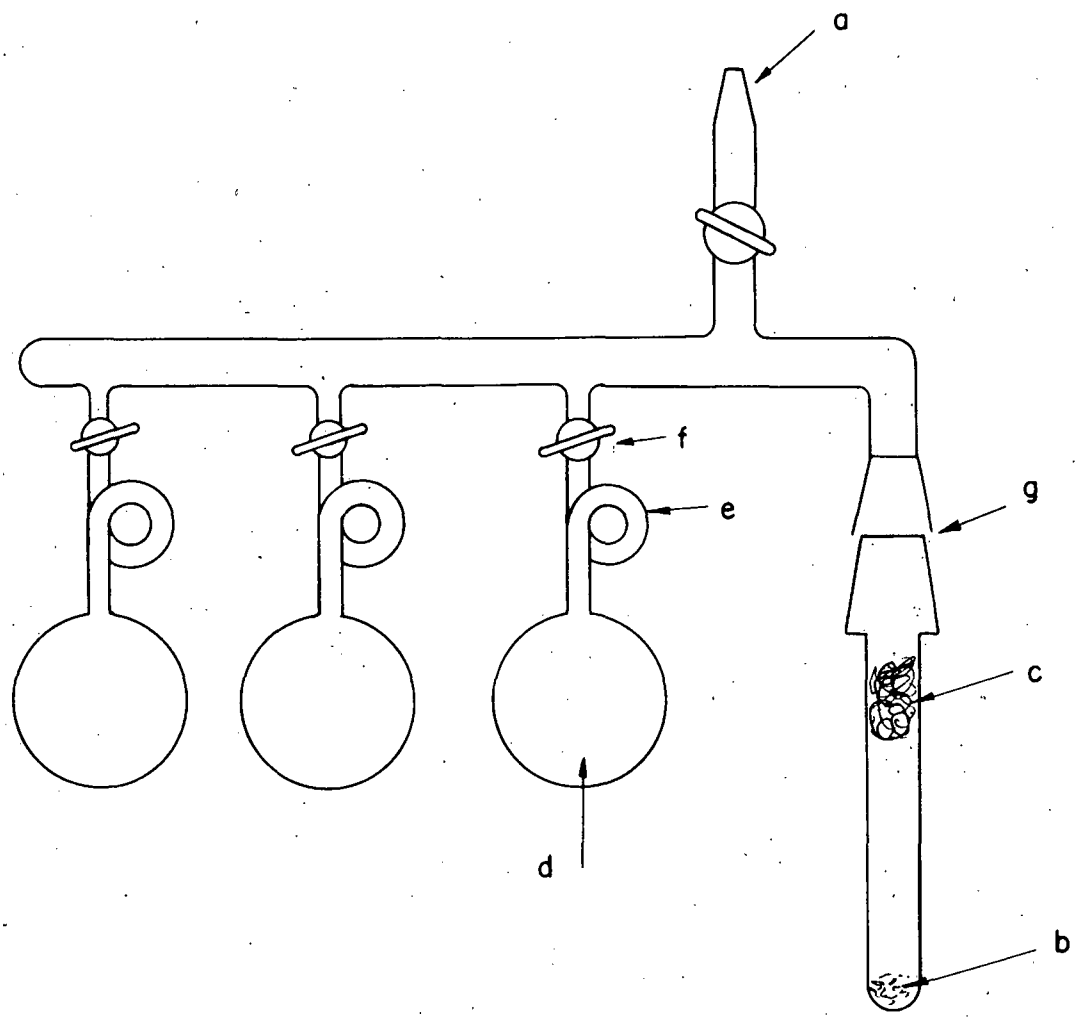

RADIOACTIVITY IN COUNTS / MINUTE
DAYS AFTER BOMBARDMENT
A
B
$T_{1/2}$ = 32 HOURS, Cs^{129}

FIG. 7

Figure 8: Direct vacuum-line isolation of daughter radioxenon from cesium. High vacuum is pulled at a. The cesium sample is introduced at b in a connecting tube g. The gaseous xenon daughter is collected in pyrex bulbs d, fitted with circular loops e and stopcocks f. To reach the bulbs, the xenon must filter through a plug of glass wool c. At the time of collection, b is flamed gently while d is cooled in a beaker of liquid nitrogen, then quickly sealed off below the loop.



ISOLATION OF DAUGHTER RADIOXENON
Xe^{127} FROM CESIUM Cs^{127}

FIG. 8

apparatus was closed off from the rest of the vacuum line. The first bulb was opened to receive any radioxenon which might emanate from the cesium sample. After 5.5 hours, the cesium sample was heated with a flame gently to expel gases while the first bulb was cooled with a beaker of liquid nitrogen in order to collect the maximum yield of any daughter radioxenon in the bulb. The bulb was then quickly sealed off below its stopcock from the rest of the apparatus, mounted on a suitable counting card and counted under a Geiger counter. About 109 counts/minute were observed, due to the reported 0.9 Mev gamma ray emitted by Xe^{127}; the walls of the bulb were too thick to permit transmission of very much of any less penetrating radiation. This corresponds to almost 30,000 actual disintegrations/minute of radioxenon, assuming 20% geometry and 2% counting efficiency of the 0.9 Mev gamma ray, representing an excellent yield of daughter xenon under conditions prevailing in this experiment. The decay of the activity in this bulb was followed for about six weeks and gave a half-life of 30 to 35 days. The apparatus was reevacuated after the 5.5 hour activity had effectively died out, and the second and third bulbs, exposed to collection of xenon after the 5.5 hour activity had died, gave no counts. This experiment was repeated on two other bombardments with similar results.

This direct vacuum-line isolation of radioxenon daughter established that not only did the xenon emanate from the 5.5 hour cesium alone, but that the 5.5 hour activity must be Cs^{127} if the assignment of the 34 day xenon daughter activity to Xe^{127}, based on $I^{127}(p,n)Xe^{127}$ reaction with 5 to 6 Mev protons, was correct[2].

After many attempts to obtain mass spectrograph proof of the mass numbers of the 5.5 hour and 31 hour cesium activities, loss factors for cesium salts were run on the mass spectrograph[4] to determine which salt would be most apt to yield successful mass spectrograph results from a thermally-ionizing source. The interesting results are shown in Table I.

Table I

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

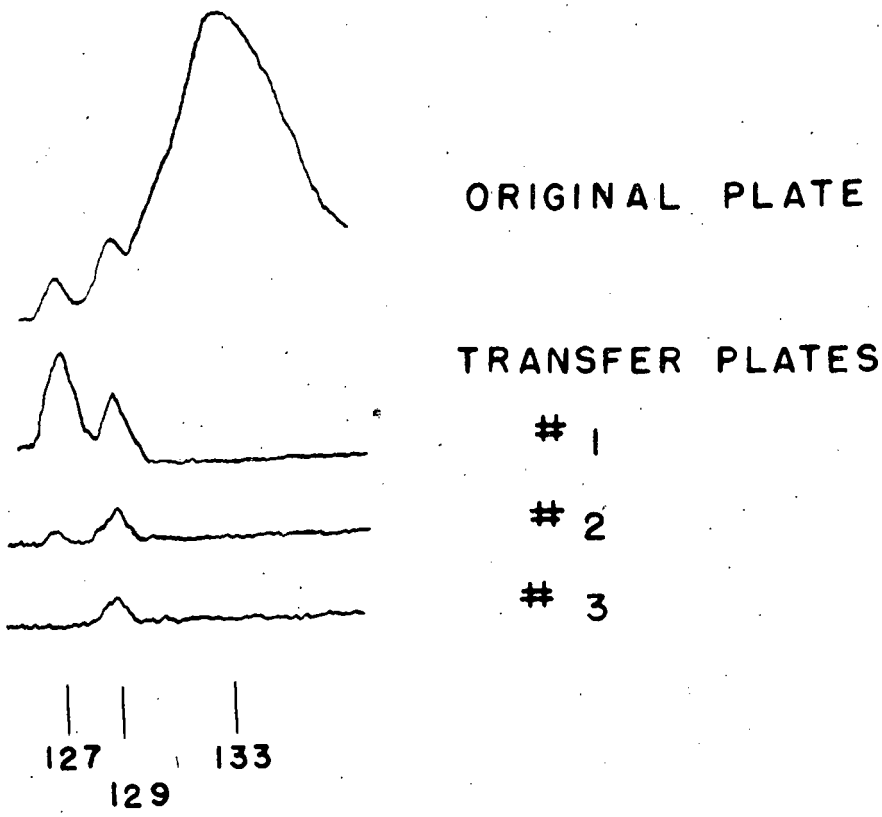
Since the geometrical loss factor on the mass spectrograph is of the order of 50, with cesium sulfate at least 25% of the cesium is ionized. One expects 100% ionization under optimum conditions, because of the very low ionization potential (3.87 volts) of cesium. The very low ionization of the chloride 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 sublimator combined with especial care to prepare practically weightless samples of the sulfate of the radioactive cesium isotopes, instead of the previously-tried chloride, 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 9, which consists of microphotometer tracings of the original mass spectrograph plate and three transfer plates made at designated times after bombardment.

These results conformed exactly to all previous evidence and predictions concerning Cs^{127} mass assignment. Mass spectrograph runs with excellent transfer plates were repeated in a total of three bombardments. Although the 30 minute

Figure 9: Microphotometer tracings of mass spectrograph plates showing Cs^{127}, Cs^{129}, and stable Cs^{133}. Transfer plate #1 was exposed to the original plate, emulsion to emulsion, immediately after removal from the mass spectrograph and remained on for 24 hours, when the crude magnetic spectrometer showed very few remaining positrons; then the transfer plate #2 was exposed to the original plate for about 48 hours, when the crude spectrometer showed no positrons but only conversion electrons; then transfer plate #3 was exposed for another 72 hours, approximately.

This proves that the positron-emitter with the 5.5 hour half-life is mass 127, and the 31 hour K-capture isotope is mass 129.



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

FIG. 9

cesium activity could not be observed on the mass spectrograph, one tentatively assigns it to Cs^{130} on the basis that it was first made[3] with low energy alpha-particles on iodine, and in the absence of evidence to the contrary, it is assumed that our 30 minute cesium activity, which is formed both at 60 Mev and 36 Mev, is possibly the same isotope.

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(1) This paper is based on work performed under the auspices of the United States Atomic Energy Commission.

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(3) J. R. Risser and R. N. Smith, private communication to Professor Seaborg from K. Lark-Horowitz.

(4) These loss factors were run by Mr. F. L. Reynolds of the Radiation Laboratory.