Mass Spectrometric Studies of the Interaction of Gold with 3.0-Bev Protons*

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Gold targets have been irradiated with 3.0-Bev protons and the yields of cesium, barium, and rubidium nuclides investigated by mass spectrometry. A high-sensitivity surface-ionization technique, using a multiple filament source arrangement and capable of determining (within 5%) 10^8 alkali metal atoms, was developed. The separation of products from the gold target was accomplished by extracting the gold into diethyl ether. The contamination level of stable Cs^{133} in the gold foil and reagents was determined by isotope dilution methods to be $\sim 9\times 10^9$ atoms. Ratios of production cross sections for neutron-deficient cesium isotopes and their barium parents were calculated from mass spectra. Cross sections were placed on an absolute scale by a radiochemical determination of the Cs^{127} cross section. No cesium isotopes with $A>128$ are formed in appreciable independent yield. An upper limit of 0.002 mb was placed on cross sections for Cs^{132} and the neutron-excess cesium isotopes. No stable cesium resulting from nuclear reactions was detected above the natural contamination background. Ratios of production cross sections for rubidium radioisotopes were determined. The natural contamination of rubidium was too large to place a meaningful limit on stable rubidium production.

RADIOCHEMICAL studies of products resulting from the bombardment of heavy elements with 340-Mev to 3-Bev protons have been reported in recent years.[1-4] In obtaining a mass-yield curve for these high-energy nuclear reactions, it has been necessary to interpolate the yields of isotopes which cannot be measured radiochemically. Among these are the stable nuclides, the very long-lived nuclides, and those radionuclides whose decay schemes are not firmly established. It therefore seemed worthwhile to develop mass-spectrometric techniques to supplement the radiochemical measurements. In this paper the mass-spectrometric determination of nuclides produced in the interaction of heavy nuclei with 3-Bev protons accelerated in the Brookhaven Cosmotron is reported.

Two aspects of this problem may be considered independently. The first involves the development of instrument sensitivity permitting the detection and measurement of nuclides at a level of 10^8 atoms per mass, the level expected in Cosmotron irradiations. The second part was to devise chemical techniques of separation which permit isolation of the desired reaction products from the bulk of the target material with minimum contamination by naturally occurring nuclides. The decision to investigate the yields of cesium isotopes by surface ionization was based on the low-ionization potential of cesium, the low abundance in nature, and the volatility of cesium salts. Gold was used as the target material because it was assumed that the cesium contamination would be less than in other convenient target materials.

EXPERIMENTAL

A 6-inch 60° sector General Electric mass spectrometer was adapted for these surface ionization studies. A Dumont *SP* 102 electron multiplier, modified by replacement of the ninth and tenth dynodes with a shielded collector mounted on an independent lead, was used as the ion detector. The output of the multiplier was amplified by a vibrating reed electrometer using a 3×10^9-ohm input resistor. Data were recorded on a ten-millivolt Brown recorder. The dynode voltage used was 450 volts per stage. The noise level on the recorder with this arrangement of equipment was less than 0.1 millivolt. The estimated sensitivity at the collector for Cs^+ ions was approximately 3×10^{-18} ampere for one scale dividion of peak intensity.

The ion source was obtained by replacing the electron impact ionization chamber of the original spectrometer ion source with a filament which can easily be positioned in a plane identical to the plane of origin of gaseous ions formed by electron impact in the original source. No attempt was made to obtain a precise location of the filament after experiments showed that reasonably constant sensitivity could be attained by compensating for slight deviations from the optimum by varying drawout and focus voltages in the ion source. The filament assembly was mounted in a ball joint which was fitted with hollow Covar sleeves into which the filament leads were waxed. This permitted simple and rapid positioning of the filament.

Efficiency of ionization from the platinum filament was found to vary widely with the chemical history of

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[1] W. F. Biller, University of California Radiation Laboratory Report UCRL-2067 (unpublished).

[2] Vinogradov, Alimarin, Baranov, Lavrukhina, Baranova, Pavlotskaya, Bragina, and Yakovlev, *Proceedings of the Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, Moscow, July 1–5, 1955;* Session of Division of Chemical Sciences (Akadmiia Nauk, S.S.S.R., Moscow, 1955), p. 97 and p. 132. [English translation by Consultants Bureau, New York, U. S. Atomic Energy Commission Report TR-2435, 1956, Part 2, pp. 65 and 85.]

[3] Murin, Preobrazhensky, Yutlandov, and Yakimov, *Proceedings of the Conference of the Academy of Sciences of the U.S.R.R. on the Peaceful Uses of Atomic Energy, Moscow, July, 1955* (Akademiia Nauk, Moscow, 1955), p. 160. [English translation by Consultant Bureau, New York, U. S. Atomic Energy Commission Report TR-2435, 1956, Part 2, p.101.]

[4] Wolfgang, Baker, Caretto, Cumming, Friedlander, and Hudis, Phys. Rev. **103**, 394 (1956).

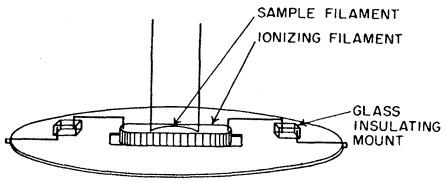

FIG. 1. Multiple-filament surface ionization source.

the sample. Source efficiency was assumed to be the sole variable in the over-all efficiency of the process as long as the multiplier showed no signs of a high noise level. Multiplier gain was checked in the earlier stages of this work and found to be relatively constant for long time periods. Source efficiency was measured in units of second-divisions of integrated peak intensity over the running period. It was found that a flux of $(NH_4)_2SO_4$ which was carefully evaporated in a preheat period gave the optimum sensitivity with approximately 1000 second-divisions for a sample of 1×10^8 atoms of Cs^{131}. This corresponds to a very low ionization efficiency if reasonable estimates of the lower limits of ion collection efficiency (i.e., $\sim1\%$) are made.

Efficient surface ionization requires intimate contact of atoms of low-ionization potential with a surface of high work function. The geometric surface area of filaments used in this work could be covered with a monolayer containing roughly 10^{13} atoms. Thus a small fraction of a microgram of a nonvolatile impurity in the solution finally evaporated on the filament could easily insulate the cesium salt from the filament and thus lower the work function of the surface. The level of alkali contamination estimated from the isotope dilution determinations of cesium and the relative abundances of alkalis in nature indicated that the alkali metal contamination was sufficient to destroy effective sensitivity in all preparations tested. Thus, the highest sensitivity could not be achieved with single-filament surface ionization techniques.

A multiple-filament assembly based on principles described by Inghram and Chupka[5] was designed and is shown in Fig. 1. In view of the relative dimensions of the source filament and the ionizing filament, it was possible to contact better than 70% of the total evaporated materials with the hot walls of the ionizing filament. Optimum sensitivity of the order of 50 000 second-divisions was obtained with samples of 1×10^8 atoms of Cs^{131} with this filament assembly. Furthermore, much of the variability of sensitivity disappeared. There was still the necessity of a careful preheat period to preserve the best sensitivity.

Samples were run by using a Varian Associates nuclear fluxmeter to determine the magnetic field and a Leeds & Northrup type K potentiometer to measure the ion accelerating voltage. The filaments were heated with current regulated by a Sorenson Model 1001 regulator and which was fed through variacs and step-down isolation transformers. Spectra were taken by scanning magnetically. Masses were identified by measurement of the ion accelerating voltage required to focus ions at a constant magnetic field. Independent mass identification was obtained from the observed decay of some of the shorter lived cesium isotopes. For example, in run 2 (Table I), the decay in the ratio Cs^{129}/Cs^{133} corresponds to a half-life for Cs^{129} of roughly 29 hours, which agrees with the literature value of 31 hours.[6]

In the determination of mass spectrometer sensitivities at a level of 10^8 atoms, it is imperative that a radioactive isotope or enriched stable isotope be used as the calibrating nuclide. Stable isotopes in their natural abundance are unsuitable for determining sensitivities at this level because natural contamination will usually contribute and yield a sensitivity which is erroneously high. For example, triply-distilled water, the last distillation being done in quartz, contained approximately 2×10^8 atoms of cesium per ml, as determination by an isotope dilution analysis. Therefore, in successive dilutions of a Cs_2SO_4 solution with "pure" water, a point is reached where the diluting water contributes almost as much cesium as the solution being diluted. A radioactive isotope has the further advantage of decaying with a given half-life. By observing this decay in the mass spectrometer, one can monitor the contribution of other elements to the peak heights at the mass of the radioactive isotope. Also, with a 9.7-day activity such as Cs^{131}, one can easily aliquot samples containing as few as 6×10^5 atoms.

Carrier-free Cs^{131} was prepared by irradiating a 0.5-gram $BaCO_3$ sample in the Brookhaven reactor for 4 days. The sample was milked for cesium immediately after irradiation and this first milking discarded. Cesium-131 was then allowed to grow in from 11.6-day Ba^{131} and the sample was again milked for cesium after a few days of growth. The cesium fraction was a pure 30-kev x-ray emitter as determined through scintillation spectrometer studies and, over a decay factor of thirty, the half-life was observed as 9.7 days, in good agreement with the literature value for Cs^{131}.[6] A sample of Cs^{131} was assayed by counting the K x-rays on a scintillation spectrometer using an accurately machined 3×3-in. NaI crystal in a 1.47% geometry. The absolute disintegration rate was computed from the count rate, after corrections for fluorescence yield,[7] K/L capture ratio,[8] and aluminum absorption were applied. This calibrated Cs^{131} sample served as a standard for all Cs^{131} samples counted subsequently. The uncertainty in the assay was estimated as $\pm5\%$.

In order to determine the yield of stable Cs^{133} in the nuclear reaction, it was necessary to analyze the gold

[5] M. G. Inghram and W. A. Chupka, Rev. Sci. Instr. **24**, 518 (1953).

[6] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[7] E. H. S. Burhop, J. phys. radium **16**, 625 (1955).

[8] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).

target and reagents for naturally occurring cesium contamination to determine the background level. An isotopic dilution technique, utilizing 9.7-day Cs^{131} as the isotopic diluent, was used for this analysis. The isotope dilution technique involved adding an aliquot of tracer solution with a known Cs^{133}/Cs^{131} ratio ($\sim$1) and a known number of Cs^{131} atoms to a blank run with an unirradiated gold foil. The foil was then treated as in a "hot" run and the resultant Cs^{133}/Cs^{131} ratio was measured. The difference between this ratio and the tracer ratio is a measure of the Cs^{133} atoms in the gold foil and reagents.

The separation technique originally attempted involved the ion-exchange column method employed by Hyde.[9] This "carrier-free" separation utilized a relatively large volume of solution for column washings and also used silicotungstic acid, a nondistillable reagent. Blank runs using the column separation showed a contamination of the order of 10^{11} atoms. Thus, it was imperative that an extraction method or some physical method which kept reagents and handling to a minimum be employed. In this case, an extraction method was used. The gold target was dissolved in 0.5 ml of aqua regia. The solution was evaporated and subsequently made up to 0.5 ml 6N HCl. The gold in this solution was extracted with two 0.5-ml portions of diethyl ether. All reagent solutions were redistilled in quartz. Tracer experiments showed that cesium and barium remained in the aqueous phase under these extraction conditions. Blank runs on the extraction procedure using unirradiated gold foils showed a cesium contamination of 8.7×10^9 atoms.

Irradiations were carried out in the circulating beam of the Brookhaven Cosmotron using the "shutter" target arrangement which has been described previously.[4] The gold target was 0.5-mil foil, 0.50 in. long and 0.25 in. high. No monitor was used in irradiations for mass spectrometric analysis because the added thickness would reduce the number of traversals through the target that each proton makes. The extracted aqueous solution was evaporated to dryness and then taken up in a drop of water and evaporated with an ammonium sulfate flux of 100 μg on a 0.5-mil platinum filament, 1 cm long and 20 mils wide. The filament was then placed in the spectrometer to form the multiple filament arrangement as pictured in Fig. 1.

A radiochemical determination of the Cs^{127} production cross section was carried out in order to put all cross section ratios taken from mass spectrometric data on an absolute basis. The target foil and aluminum foil monitor were irradiated in the Cosmotron in a manner described previously.[4] The Na^{24} produced in the aluminum by the $Al^{27}(p,3pn)Na^{24}$ reaction was determined by β counting in a standard geometry and was used as a measure of the beam intensity. The cesium in the gold foil was separated with carrier present according to the procedure outlined by the Los Alamos group.[10] The sample was counted with a thin NaI crystal using a single channel analyzer set to count only the 30-kev x-rays. The resolved decay curve gave a 6.30-hr activity over 4 half-lives and this value is to be compared with the reported[11] half-life of 6.3 hr for Cs^{127}.

TABLE I. Mass spectra of cesium isotopes produced in Cosmotron irradiations.[a]

Cosmotron Run No.	τ (hr)	t (hr)	Cs^{125} 45-min	Cs^{127} 6.3-hr	Cs^{129} 31-hr	Cs^{131} 9.7-day	Cs^{132} 6.2-day	Cs^{133} stable	$Cs^{>133}$
1	1.2	2.75[b]	<20	~100	100				
		8.50[b]			100	5.88			
2	4.7	49[c]			100	36.2	<0.5	410	<0.5
		72[c]			100	186	<0.3	708	<0.3
3	2.33	8.0		44	100	<5		645	

[a] The peak heights in an individual mass spectrum are normalized to $Cs^{129}=100$ for that spectrum. The precision of peak height ratios in runs 1 and 3 is 15%. The precision in run 2 is 5%. No correction is made for isotopic fractionation in the evaporation process.
[b] Cs was completely burned off the filament at 2.75 hours. No Ba was burned off. Run at 2.75 hours showed poor sensitivity.
[c] Only a fraction of Cs was burned off at $t=49$ hours. The filament was reheated at $t=72$ hours and the decay of Cs^{129}/Cs^{133} over this 23-hour period was observed.

RESULTS

The mass spectra for three Cosmotron irradiations of gold with 3.0-Bev protons are given in Table I. The mass spectra were used to obtain ratios of cross sections for the production of cesium isotopes. The radiochemical determination of the Cs^{127} production cross section then allowed all cross sections to be put on an absolute scale.

Since peak ratios in a mass spectrum are equal to the ratios of cesium atoms in a sample for corresponding mass numbers, neglecting the effect of isotopic fractionation in the evaporation process, we can calculate the number of cesium atoms of each mass number present in a sample at any given time. For a given isobaric pair of neutron-deficient cesium and barium isotopes, the rates of production during a bombardment are

$$\begin{aligned} dN_{Cs}/d\tau &= In\sigma_{Cs}+\lambda_{Ba}N_{Ba}-\lambda_{Cs}N_{Cs}, \\ dN_{Ba}/d\tau &= In\sigma_{Ba}-\lambda_{Ba}N_{Ba}, \end{aligned} \tag{1}$$

where I is the proton beam intensity, n is the number of atoms per cm^2 in the target, σ_{Ba} and σ_{Cs} are the production cross sections, λ_{Ba} and λ_{Cs} are the decay constants, and N_{Ba} and N_{Cs} are the numbers of atoms present. By solving these simultaneous differential equations and correcting for decay of cesium after bombardment and for decay of the barium present at the end of bombardment, we obtain the number of cesium atoms at any time t after a bombardment of length τ.

[9] E. K. Hyde, J. Am. Chem. Soc. **74**, 4181 (1952).

[10] J. Kleinberg *et al.*, Atomic Energy Commission Report LA-1566, February 1, 1953 (unpublished).

[11] H. B. Mathur and E. K. Hyde, Phys. Rev. **95**, 708 (1954).

$$N_{Cs}=In\left\{\frac{\sigma_{Cs}+\sigma_{Ba}}{\lambda_{Cs}}(1-e^{-\lambda_{Cs}\tau})e^{-\lambda_{Cs}t}+\frac{\sigma_{Ba}}{\lambda_{Cs}-\lambda_{Ba}}\times[e^{-\lambda_{Cs}(\tau+t)}+e^{-\lambda_{Ba}t}-e^{-\lambda_{Cs}t}-e^{-\lambda_{Ba}(\tau+t)}]\right\}. \quad (2)$$

The ratio $\sigma_{Ba}^{129}/\sigma_{Ba}^{131}$ was computed from the data of run 1. The half-life of Ba^{129} was taken as 2.4 hours.[12] Since all the cesium was burned off the filament at $t=2.75$ hours, all cesium seen in the spectrum at $t=8.50$ hours must have grown in from barium during the time between the two spectrometer runs. It can also be seen from the Cs^{129}/Cs^{131} ratio in run 3 and the subsequent decrease of that ratio with time in run 2 that $\sigma_{Cs}^{131}\ll\sigma_{Ba}^{131}$. Since σ_{Ba}^{129} is known relative to σ_{Ba}^{131}, σ_{Cs}^{129} can be determined relative to σ_{Ba}^{129} by equating the ratio of Eq. (2) for the respective mass numbers, 129 and 131, with the experimentally determined ratio from run 2 at $t=49$ hours. The result is $\sigma_{Cs}^{129}/\sigma_{Ba}^{129}=0.03$. The principal error in this calculation is probably the error in the Cs^{129}/Cs^{131} ratio used in run 1 to determine the $\sigma_{Ba}^{129}/\sigma_{Ba}^{131}$ ratio. Making the assumption that all Cs^{129} grows in from Ba^{129} and its precursors, the $\sigma_{Ba}^{127}/\sigma_{Ba}^{129}$ ratio can be determined by equating the Cs^{127}/Cs^{129} ratio determined in run 3 with the ratio of Eq. (2) for mass numbers 127 and 129. This calculation assumes that all Cs^{127} formed grows in from the 12-min Ba^{127}. Because of the short-life of Ba^{127}, the 127-chain cross section is increased by only 4% if one makes the alternate assumption that all the Cs^{127} is formed directly. Thus, the relative production cross sections for mass numbers 127, 129, and 131 are determined. The radiochemical experiment yielded the absolute production cross section for mass 127 as $\sigma_{Cs}^{127}=4.3$ mb. This cross section was calculated by using an aluminum monitor cross section of 8.1 mb.[13] Current experiments indicate the monitor cross section at 3.0-Bev bombarding energy to be 30% larger.[14] Revision of the cesium and barium cross sections in this research awaits the completion of those experiments.

An attempt was made to determine the cross section for the stable Cs^{133}. Blank runs on unirradiated gold foils were run to determine the level of cesium contamination in the foil and reagents. Four blank runs were made by the isotope dilution method described above with the result that the cesium background was $(8.7\pm0.3)\times10^9$ atoms. By varying the size of the foil, but keeping the volume of reagents constant, it was found that a gold foil of target size contained 4.7×10^9 atoms of cesium and the reagents 4.0×10^9 atoms of cesium. If the number of Cs^{129} atoms produced can be calculated, the ratios Cs^{129}/Cs^{133} in runs 2 and 3 can be used to calculate the number of Cs^{133} atoms and this value compared with that of the blanks. Since no monitor foil was used, an estimate of the beam intensity was made by multiplying the circulating beam intensity (as measured by an electromagnetic monitor) by 120 to take into account the estimated number of traversals of each proton through the target foil. This estimate of the number of traversals is made from a run in which the beam intensity as determined by an aluminum monitor was compared with the circulating beam monitor of the Cosmotron. The data of run 2 give a value of 6.0×10^9 atoms of Cs^{133} whereas the data of run 3 give a value of 8.1×10^9 atoms. That these values are lower than the blanks is attributed to the uncertainty in beam intensity and possible uneven distribution of cesium in the gold. It is reasonable to believe that the number of traversals could not be much greater than the assumed value of 120. Therefore, the Cs^{133} in an irradiated foil must be close to that found in the blank runs. Since Cs^{133} is shielded from the neutron-deficient side by long-lived Ba^{133} and no neutron-excess isotopes are observed, any Cs^{133} formed would be formed in independent yield. Our conclusion that no Cs^{133} is formed directly is consistent with the fact that no other cesium isotopes from Cs^{129} up in mass number are formed directly.

Run 1 was an attempt to measure the 45-min Cs^{125}. The filament was heated 2.75 hours after the end of the bombardment; this represented the minimum time required for chemistry, mounting, and mass spectrometer pump-out. The noise level was higher than usual because of insufficient pump-out and the curtailment of the preheat period required for the volatilization of the flux and the outgassing of the filament. However, an upper limit was set for the Cs^{125}/Cs^{129} ratio. Upper limits were also placed on the cross sections for Cs^{132} and the neutron-excess cesium isotopes. These limits were taken from run 2 since the target was allowed to sit for 49 hours, a period sufficient to allow 90% of the chain yield of mass 135 to grow into Cs^{135}.

Several scans of the rubidium mass spectrum were made in runs 1 and 2. Ratios of the production cross sections for Rb^{83}, Rb^{84}, and Rb^{86} were observed. Rb^{84} and Rb^{86} are shielded isotopes and therefore must have been formed directly in the nuclear reaction. Rb^{83}, on the other hand, was observed to grow in from Sr^{83} in a run where all active rubidium nuclides were burned off and the spectrum run again two days later. This spectrum showed a Rb^{83} peak, but no Rb^{84} or Rb^{86} peak. The relative cross sections for direct and cumulative chain production cannot be calculated from the data at hand. Estimates of absolute cross sections for the rubidium nuclides can be made if one assumes the ratio of natural rubidium contamination to natural cesium contamination in the foil and reagents to be the same as the Rb/Cs ratio found in igneous rocks. This ratio is ~55.[15] This assumption is based on the fact

[12] W. Henkes (private communication).
[13] Friedlander, Hudis, and Wolfgang, Phys. Rev. **99**, 263 (1955).
[14] J. B. Cumming and G. Friedlander (private communication).
[15] V. M. Goldschmidt, *Geochemistry* (Oxford University Press, London, 1954), pp. 163, 170.

that no alkali metal reagents were used in the separation procedure. The $(Rb^{85}+Rb^{87})/Cs^{133}$ ratio of peak heights at the time of measurement was 22.7. The experimental ratio is not equal to the natural-abundance ratio because the rates of evaporation of the alkali salts from the filament will differ and therefore, the Rb/Cs ratio will be a function of the fraction of the sample burned off. Since the fraction burned off is unknown, the above assumption cannot be verified. If the peaks in the rubidium scan are normalized to a Rb/Cs ratio of 55, the Rb^{83}, Rb^{84}, and Rb^{86} peak heights can be compared with the Cs^{129} peak height of the cesium scan. Since the cumulative yield for Cs^{129} is known, the rubidium yields can be calculated and those yields are shown in Table II. The ratios of rubidium cross sections to each other do not rely on the validity of the above assumption.

TABLE II. Production cross sections for nuclides from the interaction of 3-Bev protons with gold.

Nuclide	Type of yield[a]	Cross section (mb)
Cs^{125}	C	<12
Cs^{127}	C[b]	4.3
Cs^{129}	I	<0.4
Cs^{131}	I	<0.2
Cs^{132}	I	<0.002
$Cs^{134, 136}$	I	<0.002
$Cs^{135, 137}$	C	<0.002
Ba^{129}	C	4.4
Ba^{131}	C	5.3
Rb^{83}	C-I	>2.5[c]
Rb^{84}	I	1.2
Rb^{86}	I	0.5

[a] C denotes cumulative yield of mass chain decaying to given nuclide; I denotes independent yield of a given nuclide.
[b] This value was determined radiochemically and served as the basis for converting cross-section ratios to absolute cross sections.
[c] Burn-off experiment indicated a chain-yield contribution. This lower limit is based on the total yield being an independent yield.

CONCLUSION

As shown in Table II, the yields for cesium radioisotopes near stability and on the neutron-excess side are very small. Although the experimental upper limit for Cs^{133} production is not as low as the limits set for the radioisotopes, one hardly expects any specificity for Cs^{133}. The experimental data in the rubidium region are less conclusive since the natural contamination is such that it would mask out even abnormally large yields of stable Rb^{85} and Rb^{87}. However, the yields of Rb^{86} and Rb^{84} are smaller than the cumulative yield of Rb^{83} and the yields given in Table II indicate a continuous rise in cross section to the neutron-deficient side. This research points to the conclusion that given sufficient data, one can interpolate yields of products which are themselves not easily determined.

It is seen that the cesium nuclides produced in independent yield must have a neutron to proton ratio, N/Z, $\leqslant 1.34$, the ratio for Cs^{129}. All cesiums with $N/Z>1.34$ are definitely not formed in independent yield. The independent yields for the shielded nuclides in the rubidium spectrum indicate that the primary products in this region are of $N/Z\leqslant 1.32$. It is likely that all primary products in the region between rubidium and cesium have $N/Z\leqslant 1.34$. These results are consistent with the mechanism postulated by Goeckermann and Perlman.[16] That mechanism involves neutron evaporation from highly excited nuclei followed by fission with an unchanged charge distribution. However, the other extreme mechanism of fission followed by neutron evaporation from highly excited fission fragments cannot be ruled out.

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[16] R. H. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949).