

On-line mass-spectrometric measurement of rubidium and cesium isotopic distributions in ^{11}B - and ^{22}Ne -induced fission of ^{238}U

M. de Saint-Simon, L. Lessard,* W. Reisdorf,[†] L. Remsberg,[‡] C. Thibault, E. Roeckl,[†] and R. Klapisch
*Laboratoire René Bernas du Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse,
 B. P. No. 1, 91406 Orsay, France*

I. V. Kuznetsov, Yu. Ts. Oganessian, and Yu. E. Penionshkevitch
*Laboratory of Nuclear Reactions of the Joint Institute for Nuclear Research, Dubna,
 Union of Soviet Socialist Republics
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Using an on-line mass spectrometer we have measured isotopic distributions of rubidium and cesium in the fission of ^{238}U by ^{11}B (85 MeV) and ^{22}Ne (132 and 162 MeV).

[NUCLEAR REACTIONS, FISSION $^{238}\text{U}(^{11}\text{B},f)$, $^{238}\text{U}(^{22}\text{Ne},f)$; measured Rb, Cs isotopic distributions with mass spectrometer; deduced first moments, variances, structure.]

I. INTRODUCTION

In the fission of nuclei induced by heavy ions, typical excitation energies of the compound nuclei are generally in excess of 50 MeV. Under these circumstances one expects that mass and charge distributions of fission fragments follow smooth regular trends, since shell effects should be washed out. The understanding of these high energy processes should be considerably simplified as compared to the situation at low energy, where the well known phenomenon of mass asymmetry of the fragment distributions exists in the actinide region.

On the other hand, fission induced by heavy ions is characterized also by a number of new interesting features; first, the compound nuclei have large angular momenta, and second, one also observes fission events following direct reactions, rather than fusion, with the target nuclei.

Yields of various fission products in heavy-ion-induced reactions have been determined in the past,^{1,2} using radiochemical methods. These data comprise a large variety of targets, including ^{238}U , and of heavy-ion beams. Generally, because of half-life limitations, only parts of the isotopic distributions published represent independent yields (as opposed to cumulative yields). Missing independent yields are estimated using interpolatory schemes as well as a variety of semitheoretical rules. Uncertainties in these procedures limit the accuracy with which first moments and above all variances of the isotopic distributions can be determined.

In past years a method has been developed³

which is based on mass spectrometry to separate and detect selectively isotopes of alkali reaction products. Complete independent yields of these isotopes can be obtained with high relative precision. This allows us to determine the first moment and the variance of these isotopic distributions with high accuracy and to see if any structure exists in the yields. It seemed to us that such complete and independent isotopic fragment distributions could be of interest in clarifying some of the mechanisms involved in heavy-ion-induced fission. In this paper we present an application of the mass spectrometric technique to obtain rubidium and cesium isotopic distributions in ^{11}B - and ^{22}Ne -induced fission of ^{238}U . In a companion paper⁴ a theoretical analysis of the data will be presented.

II. EXPERIMENTAL TECHNIQUE

A. Apparatus and beams

The method is an adaptation to heavy-ion reactions of a mass spectrometric technique that has been used^{3,5} to measure Rb and Cs isotopic distributions in proton- and neutron-induced fission. Detailed descriptions of the apparatus have also been given in Ref. 6.

The target was a metallic uranium layer of 1 mg/cm² deposited by an electromagnetic mass separator on a thin graphite foil (2.8 mg/cm²) made from bulk nuclear purity graphite by abrasion. The target was located inside a 30 mm long and 16 mm diameter cylindrical graphite oven heated at about 1500°C by dc current. Fission recoils were stopped in thin graphite foils placed on both sides of the target inside the oven. The assembly

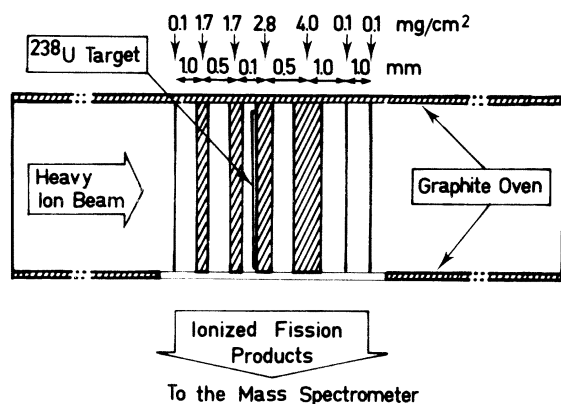


FIG. 1. Schematic view of the target-oven assembly used for $^{238}\text{U}(^{22}\text{Ne},f)\text{Rb, Cs}$. The very thin foils of carbon (0.1 mg/cm^2) are thermal shieldings. The two thicker catcher foils upstream of the target are made of highly uniform pyrolytic graphite. The 2.8 mg/cm^2 target backing as well as the 4 mg/cm^2 foils are downstream catchers made of bulk graphite bars by abrasion.

is shown in Fig. 1. Two highly selective properties of alkali reaction products were used: they diffuse fast out of the heated graphite stoppers and, because of their low ionization potential, they are ionized on the hot graphite surface. Then the alkali ions are extracted from the oven through a thin slit by a 10 kV accelerating potential and mass analyzed by an inhomogeneous magnetic sector having a mean radius of 35 cm . A typical resolving power is 500 at 10% of the peak height, while the total efficiency is 2% . A triangular modulation of the accelerating voltage (40 c/s) allows a scan over a three-mass interval. Counting rates for the three masses are stored in a multiscaler memory having a channel sweep synchronized with the modulating voltage. From the resulting mass scans relative cross sections of the various isotopes are obtained.

The ^{11}B and ^{22}Ne beams were extracted from the 310 cm cyclotron of the Joint Institute for Nuclear Research. The beam energy was degraded by aluminum foils and measured by observing elastically scattered ^{22}Ne from $50\text{ }\mu\text{g/cm}^2$ gold foil with a calibrated⁷ and collimated surface barrier detector located at 40° relative to the incident beam. The average energies on target after correction for energy losses⁸ in various windows and in the graphite foils upstream of the target were $85 \pm 2\text{ MeV}$ and $162 \pm 3\text{ MeV}$ for ^{11}B and ^{22}Ne , respectively. Some data were also obtained with 132 MeV (on target) ^{22}Ne beams. The beam intensity was monitored by a Faraday cup; typical charge currents were $0.8\text{ }\mu\text{A}$ on target.

B. Experimental procedure

Before each major run the optimum temperature for the oven was determined fulfilling two important criteria: (1) The continuous beam production of reaction products was reasonably high as compared to natural contaminations (mainly Ba and Sr isotopes as well as natural Cs and Rb). (2) The time of diffusion of alkali isotopes produced in the reaction was short enough. This was checked by recording the ion current as a function of time after irradiation by a chopped beam. The experimental diffusion curve may be reproduced by a sum of time-dependent exponential functions. Under optimum conditions an important fraction of the ions leaves the oven within several tenths of a second.

The runs were then started with pulsed-ion beams to give an irradiation lasting 1 s every 4 s . During each beam cycle two mass spectra were stored in different subgroups of the multiscaler memory: spectrum A was recorded during and immediately after the irradiation, spectrum B just before the beginning of the next beam burst. These two kinds of spectra are shown in Fig. 2. The peaks in spectrum B are due to slowly diffusing components of the reaction products, cumulative effects (e.g., at mass 135), and the natural contaminants mentioned earlier. The difference spectrum (A - B) gives the relative independent yields directly.

For natural masses even when the energy of the heavy-ion beam was below the Coulomb barrier, we sometimes noticed the appearance of a nonzero difference spectrum. These parasitic peaks were traced to the temporary and local rises in temperature of the target oven assembly under the in-

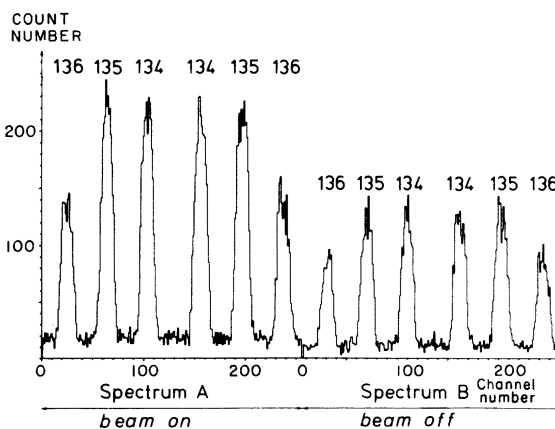


FIG. 2. Mass spectra of $^{134,135,136}\text{Cs}$ obtained in the reaction $^{238}\text{U}(^{22}\text{Ne},f)\text{Cs}$. The symmetrical shape of mass spectra is due to the triangular high voltage modulation. The spectrum A is recorded during bombardment of the target; the spectrum B is recorded 3 s later when the heavy-ion beam is turned off.

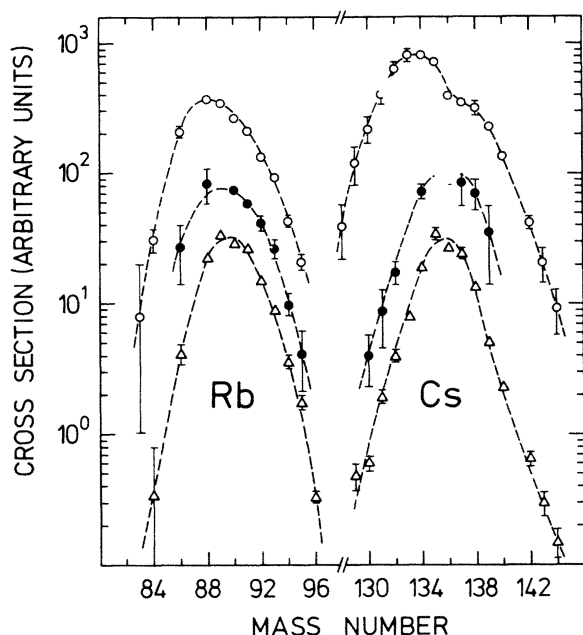


FIG. 3. Isotopic distributions of Rb and Cs in the fission of ^{238}U induced by ^{11}B ($E_{\text{lab}}=85$ MeV, triangles) and ^{22}Ne ($E_{\text{lab}}=132$ MeV, closed circles, and 162 MeV, open circles). The dashed lines are arbitrary curves drawn to guide the eye.

fluence of the beam that increases the extracted ion current of natural element isotopes such as Ba. This problem was minimized by cleaning of the oven heated at 1800°C with the high voltage turned on for several days just before a run. For the data presented here the parasitic counting rates were relatively low and could be corrected by comparing mass spectra differences obtained above and below the Coulomb barrier.

III. RESULTS AND DISCUSSION

A number of corrections have been applied to the data:

a. Corrections for radioactive decay. They are obtained using diffusion curves of long-lived isotopes and known half-lives. They were only noticeable for isotopes with half-lives shorter than 10 s, such as $^{92-96}\text{Rb}$ and $^{142,143}\text{Cs}$. Typically, the correction was 10% for a half-life of 1 s.

b. Correction for cumulative products. This contribution is due to decay of slowly diffusing nuclear products as noble gases which decay to alkali elements. The correction depends upon production cross sections and half-lives of alkali isotopes and their parents, and upon the timing of the experiment. The corrections are estimated assuming the isotopic Xe (or Kr) distributions are the same as Cs (or Rb) except for an overall mass displacement of about A/Z masses (A, Z mass and

charge of the fissioning nucleus). The corrections of this kind are always lower than 20%.

c. Normalization of the results. Experimental mass scanning was three or four masses. In order to obtain a complete isotopic distribution we had to adjust mass spectra together. This was done using two methods. First, adjacent groups of masses were measured in such a way that there was at least one overlapping mass. The yield of this common mass number was then used for relative normalization. This procedure eliminates efficiency variations of the mass spectrometer, but has the disadvantage of large uncertainties in the yields at both ends of the distribution because of propagation of errors. Second, yields for each mass number were reduced to the same integrated beam charge. This method is limited because of uncertainties in beam charge measurement. The agreement observed between the two methods implies that the efficiency of the mass spectrometer was constant within 20%.

Relative cross sections obtained for Rb and Cs isotopes in the fission of ^{238}U induced by 85 MeV ^{11}B and 132 and 162 MeV ^{22}Ne are presented in Fig. 3. Error bars correspond to statistical errors only. A summary of the first moments A_s and the variances σ_s^2 of the measured isotopic distributions is given in Table I.

Since a more detailed analysis will be given in the companion paper⁴ only a few comments will be given here. Both from Table I and Fig. 3 it is clearly visible that the variances of the isotopic distributions for ^{22}Ne induced fission (at 162 MeV) are substantially larger than for ^{11}B induced fission. This is in good agreement with the systematic trend as a function of the parameter Z^2/A found by Karamyan *et al.*¹ For $Z^2/A > 37$ these authors observed a strong increase in both the mass and charge variances of the fission fragment yields which could not be reproduced with standard statistical theories for fission. (For $^{11}\text{B} + ^{238}\text{U}$ and $^{22}\text{Ne} + ^{238}\text{U}$, the value of Z^2/A is 37.8 and 40.0, respectively). In this context it is interesting to note, in the present data, the occurrence of a well developed hump at the heavy side of the Cs distribution from ^{22}Ne induced fission (Fig. 3). This suggests that at least two fissioning mechanisms are present, fission following complete fusion and fission following direct reactions⁹ between projectile and target. The latter mechanism will tend to yield more neutron-rich fragments for two reasons. First, the initial mass to charge ratios of typical "quasiuranium" nuclei is somewhat higher than that of the compound nucleus (for Cs fragments one expects on this ground an average difference of two mass units). Second, the fission fragments from compound nuclear fission are estimated to be

TABLE I. Summary of results. A_s and σ_s^2 are the first moment and variance of the isotopic distributions.

System	E_{lab} (MeV)	$A_s(\text{Rb})$	$\sigma_s^2(\text{Rb})$	$A_s(\text{Cs})$	$\sigma_s^2(\text{Cs})$
$^{11}\text{B} + ^{238}\text{U}$	85.1	89.84 ± 0.05	3.84 ± 0.16	135.72 ± 0.04	3.95 ± 0.14
$^{22}\text{Ne} + ^{238}\text{U}$	132.0	89.39 ± 0.30	4.23 ± 0.40	135.80 ± 0.4	4.26 ± 0.9
	162.5	88.82 ± 0.07	5.41 ± 0.22	134.36 ± 0.09	8.27 ± 0.29

more highly excited (and hence to emit more neutrons) than those from fission following direct reactions for the systems and the energies of interest here.⁹

The accuracy and completeness of the present experimental isotopic distributions make it possible, better than has been previously possible from such data, to test these qualitative arguments quantitatively.⁴ The data presented here, if extended to argon and heavier beams, would appear to be a useful complement to the data of references.^{1,2}

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*France-Québec Postdoctoral Fellow 1972-1974. Present address: Foster Radiation Laboratory, McGill University, Montréal, Canada.

†Also at Gesellschaft Für Schwerionenforschung, Postfach 541, 6100 Darmstadt 1, West Germany.

‡Guggenheim Fellow 1973-1974. Present address: Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973.

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