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ISOTOPES WELL SUITED FOR IN-FLIGHT ISOTOPIC
SEPARATION**

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Projectile fission at relativistic velocities:
a novel and powerful source of neutron-rich isotopes
well suited for in-flight isotopic separation

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Abstract

Projectile fission of ^{238}U was investigated at a bombarding energy of 750 A·MeV using a Pb target. Forward emitted fragments from ^{80}Zn up to ^{155}Ce were analyzed with the Fragment Separator (FRS) and unambiguously identified by their energy-loss and time-of-flight. The magnetic selection of the largest momenta acted as a trigger of the low-energy fission component. More than forty new nuclear species were identified. The related isotopic production cross-sections are presented.

Since the discovery of fission, chemists and physicists have been devoting a steady effort to understand the process and to separate the numerous fragments. Even if it is not yet possible to describe fission by a unique theory, the process itself is rather well understood [1]. More than 400 new isotopes became accessible through fission for spectroscopic investigations [2]. New regions of deformation at $N > 60$, shape isomerism as in ^{98}Y , and the double shell closures far from stability in ^{132}Sn and ^{78}Ni exemplify the widened field of spectroscopic studies opened by this process. Many of the neutron-rich isotopes produced in fission are involved in the astrophysical r-process. Their basic nuclear properties are necessary for calculations of the mass abundance distribution in the solar system and for constraints on the physical parameters of the r-process like temperature or neutron flux.

Despite improvements in target handling, spectrometers, and detection systems, the direct separation and investigation of fission fragments by recoil-in-flight separation remains difficult due to the broad ionic charge state distribution of the fission products [3]. The direct separation is even impossible for the heavy group of fission fragments due to limitations of the Z resolving power at low energies. Furthermore, studies of extremely low production yields are difficult to perform, since recoil spectrometers have rather small solid angles, while the fragments are emitted isotropically [4].

Acceleration of a ^{238}U beam to relativistic energies by using the heavy ion synchrotron SIS at GSI provides new prospects for the study of the fission process. The use of inverse kinematics at high energies to induce fission in peripheral collisions has three main advantages : 1) The momenta of the fragments are focussed into a small cone centered around the beam direction and their velocities are almost constant. Thus, fragments are efficiently transmitted through a separator. 2) The fragments are completely stripped of electrons, i.e. their charge state equals their atomic number and ambiguities when dealing with different charge states can be totally avoided. 3) At these high velocities, fragments can be identified by $B\rho$ - ΔE -ToF techniques.

In this experiment, a beam of $2 \cdot 10^5$ U ions/s accelerated up to 750 A·MeV impinged on a 1.25 g/cm² thick lead target. The high Z target was chosen to favour fission following electromagnetic processes. Reaction products were analyzed with the fragment separator FRS [5] which was operated in the achromatic mode. Energy loss of the separated fragments, which is characteristic for their nuclear charge Z , was measured in a four-stage MUSIC ionization chamber [6] at the exit of the FRS. For each fragment the deflection radius ρ in the last two dipoles was extracted from the position of the trajectory in the intermediate and in the final focal planes measured with two plastic scintillator counters. Thus for a specific Z , the momentum $p = \beta\gamma Ac$ was extracted from the rigidity $B\rho$, βc being the velocity, and γ the Lorentz factor. The value of $\beta\gamma$ was determined from the time of flight over the 37 meters between the two plastic scintillators. The mass A was calculated from the ratio of p to $\beta\gamma$. For fragments of similar velocities, the magnetic deflection separates isotopes according to the mass-to-charge ratio, A/Z . A survey of production rates in terms

of A/Z was obtained by successive tunings of the magnetic fields.

The velocity of fission fragments in the laboratory is the relativistic addition of projectile and fission velocities, β_0 and β_F . In the projectile frame the momenta of both fragments are in opposite directions. In the following they are assumed to be isotropically distributed. The vectors populate a diffuse sphere, the radius of which changes slightly with the mass partition. In the laboratory system, these momenta are distributed in a cone with an opening angle of $\beta_F/\beta\gamma$, i.e. $\simeq 30$ mr (see fig. 1). Within the 15 mr angular aperture of the FRS only fragments emitted either forward or backward are transmitted. The velocities of forward-emitted fragments are higher by about 4% than the projectile velocity. Only an exothermic process like fission does increase the fragment velocity. It should be noted that neutron-rich isotopes with N/Z values larger than in ^{238}U can be produced only if the excitation energy is low and only very few neutrons are evaporated. Therefore, low energy fission can be selectively observed without background from other reactions in forward direction at the largest longitudinal momenta. In the following we shall consider only those fragments.

The energy loss in the 5 mm thick plastic scintillator counter located in the intermediate dispersive focal plane introduces a slight Z selectivity, thus, only a limited number of elements ($\simeq 20$) is optimally transmitted. The FRS was tuned on the light group of fission fragments ($Z=40$), the heavy fragments ($Z > 50$) as well as the very light ones ($Z < 30$) were cut and beyond $Z=58$ no fission fragments were detected. A thick target was chosen in order to maximize the count rates. It causes an energy broadening depending on the depth where fission takes place in the target. Converted to momentum this “location straggling” width reaches 3% for the smallest, $Z=30$, and 2% for the largest atomic number, $Z=58$. It dominates over the small spread due to the fission process and the 15 mr angular aperture of the FRS (Fig.1). Taking into account the 2% FRS momentum acceptance, the momentum widths of the lightest and heaviest fragments are 3.8% and 3.1% FWHM, respectively. The angular aperture of the FRS and the target thickness reduce the transmission through the FRS: Values of 2.8% for Sr-fragments and 5% for Te-fragments were calculated by using the Monte-Carlo simulation code MOCADI [7] with fission kinematics included.

The element distribution of separated fragments measured at a magnetic rigidity of 3% above that of the projectile, $(B\rho)_0$, is shown in fig. 2a. The highest yields were transmitted at this rigidity. The double-humped Z distribution well-known from low energy fission can be seen, each element being nicely separated. By correcting the energy loss signals for their velocity dependence the resolving power $Z/\Delta Z$ increases to 140. The Z calibration was extrapolated from the U energy loss and confirmed by finding the well established enhanced yields for Zr and Te. This spectrum, not corrected for the FRS transmission, reveals clearly several features of the underlying fission process. A significant odd-even effect (5%) and a shell effect, which enhances the yields for the complementary pair Te ($Z=52$) and Zr ($Z=40$) at $N=82$, indicate a low excitation energy of the fission process. However, the filling of

the valley by two orders of magnitude as compared to the thermal neutron fission of ^{235}U can be attributed only to fission occurring with a higher excitation energy. Two reaction mechanisms at least contribute to the elemental distribution measured: electromagnetic fission induced by virtual photons with energies in the giant resonance region (8 to 12 MeV) and a low energy nuclear fission process filling the valley (20 to 25 MeV).

In order to achieve adequate mass resolution, an absolute accuracy of 10^{-3} is required for the ToF signal measurement. The nominal field of the magnets is known with a precision of 10^{-4} . Radius and length of each of the trajectories must be determined with a similar precision. Therefore an event-by-event determination of the position of the particle in the intermediate and final focal planes and of the angle at the exit of the FRS was performed. A timing precision of 200 ps over a flight time of 150 ns provides a mass resolution of $\Delta A/A = \gamma^2 \cdot \Delta t/t = 1/250$, as illustrated in the following. The mass is determined numerically from Z , $B\rho$ and the ToF values. The calibration, deduced from $B\rho$ and ToF parameters for ^{238}U , is confirmed by finding the well-established enhanced yields for the $N=82$ nuclei, ^{134}Te and ^{132}Sn , at rigidities of $1.05 \cdot (B\rho)_0$ and $1.08 \cdot (B\rho)_0$, respectively. Due to the momentum spread mentioned before, 5 to 6 masses of each element can be transmitted simultaneously in the FRS momentum acceptance of 2%. Since forward-emitted fission fragments have nearly equal velocities, higher $B\rho$ leads to a selection of larger A/Z ratios. The largest $B\rho$ setting for which fragments were still detected with sufficient intensity was limited by the rather low ^{238}U beam intensity and amounted to $1.08 \cdot (B\rho)_0$. On the corresponding ΔE -ToF scatter-plot shown in fig. 3 many new neutron rich isotopes can be observed. The most abundant one observed is the doubly magic ^{132}Sn with a count rate of about 1 event/s.

The most neutron-rich isotopes are found at the smallest velocities for a given $B\rho$ setting, thus they appear on the left-hand side of the frontier line of known isotopes [8] as indicated in fig. 3.

In order to prove the existence of a new isotope based on low statistics data, one has to take into account that related events should be concentrated around the expected $\beta\gamma$ value and that a realistic fall-off of the mass yields is required for each element. Note that isotope signals can be superimposed on a non-zero background. After applying these criteria many new isotopes could be identified and are listed in table 1. The count rate was transformed to production cross-sections by using FRS transmission values calculated with the Monte-Carlo code MOCADI [7]. In this first run, with only about 10 hours of data taking the lower limit of measurable cross-section was about $1 \mu\text{b}$.

As an example the mass distribution of Y is shown in fig. 2b in order to illustrate those findings: The three new isotopes $^{103,104,105}\text{Y}$ are clearly separated and show a reasonable fall-off in yield. The ratio N/Z of the heaviest isotopes is 1.70, a value well above the value for ^{238}U . The fact that so many new neutron-rich isotopes were found requires that the nuclear charge dispersion of the processes involved has to be larger than or at least equal

to that of thermal fission [9]. In the region of symmetric fission, where yields are much higher than in low energy fission, three new isotopes were discovered for Nb, Mo, Ru, Rh, and Pd, respectively. Note that previous studies in this region were performed with the IGISOL facility by using 20 MeV proton-induced fission of ^{238}U [10]. The new nuclear species observed in our measurement fill the gap of elements not accessible by chemistry or by ISOL ion source techniques.

It should be noted that our experiment reveals not only a new landscape of fragments, hidden until present, but represents also the first ΔE -ToF isotopic identification of heavy fission fragments. Such a direct separation leads to accurate relative fission yields, free from any chemical bias. Moreover, the 300 ns required for the ions to pass through the FRS are shorter than any β decay half-lives so the measured yields cannot be influenced by fast β^- -decays.

Since fission fragments can be separated now the same way as any other projectile fragments, a new class of neutron-rich high energy secondary beams can be made available in addition to the fragmentation products used up to now.

We have shown that by studying ^{238}U fission in inverse kinematics all fragments can be separated by the FRS and identified unambiguously. A large number of isotopes can be measured simultaneously. Hence, the experimental method described represents also a novel and very efficient way to study both qualitatively and quantitatively all processes which take place in collisions of 1 GeV protons on actinide targets in incineration reactions [11] or on thorium targets in energy-producing subcritical accelerator devices [12]. For such reactions, the low laboratory velocities of the fragments have up to now prevented a comprehensive study of the related cross-sections.

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Figure captions

Fig. 1a: Schematic view of the FRS magnets with the detectors.

Fig. 1b: Momentum diagram of fission in relativistic inverse kinematics. The part of the fission fragment momentum distribution that is accepted by the FRS is indicated by the filled areas.

Fig. 2a: Elemental yields measured in 750 A.MeV U projectile fission on Pb at 1.03 times the projectile magnetic rigidity, $(B\rho)_0$. This setting corresponds to a maximum of the transmission for the most abundant isotopes.

Fig. 2b: Measured mass spectrum of Y isotopes at $1.08 \cdot (B\rho)_0$. The 3 new isotopes are indicated.

Fig. 3: ΔE -ToF scatter plot of fission fragments observed at $1.08 \cdot (B\rho)_0$. $3 \cdot 10^5$ events were recorded during the 10 hours irradiation. The full line indicates the present limit of known isotopes [8]. On the left of it, many of the new isotopes are clearly visible. Some isotopes are indicated for orientation.

Table Caption

Table: List of the new isotopes and of the number of counts observed at a setting of the FRS corresponding to 1.08 times the magnetic rigidity of the primary beam. Isotopes with less than 6 events registered are given in brackets. The cross-sections have been evaluated as described in the text. Their uncertainties amount to $\simeq 40\%$ and include both statistical errors and the uncertainty of the calculated transmission. For the last five isotopes the uncertainty of the transmission precluded a reasonable estimation of their cross-sections.

2 a)

2 b)

'X021.NEWXL.DATA(B1)' 'X021.NEWXL.DATA(C)' 'X021.NEWXL.DATA(D)' 'X021.NEWXL.DATA(E)' 'X021.NEWXL.DATA(F)'

Isotope	Counts	$\sigma(\mu\text{b})$	Isotope	Counts	$\sigma(\mu\text{b})$	Isotope	Counts	$\sigma(\mu\text{b})$	Isotope	Counts	$\sigma(\mu\text{b})$
⁸⁶ Ge	10	7	¹¹¹ Mo	135	66	¹²² Pd	79	25	(¹⁴² Te)	5	
⁸⁸ As	51	31	¹¹² Mo	29	15	¹²³ Pd	12	4	¹⁴³ I	49	15
⁸⁹ As	8	5	(¹¹³ Mo)	3	2	¹²⁵ Ag	119	37	¹⁴⁴ I	7	3
⁹⁰ Se	409	240	¹¹⁴ Tc	53	24	¹²⁶ Ag	19	6	(¹⁴⁷ Xe)	4	1
⁹⁶ Kr	155	88	¹¹⁵ Tc	18	9	¹³⁵ Sn	193	63	¹⁵¹ Ba	13	4
¹⁰³ Y	972	490	¹¹⁶ Ru	168	63	¹³⁶ Sn	34	12	¹⁵¹ La	106	>52
¹⁰⁴ Y	94	48	¹¹⁷ Ru	30	11	(¹³⁷ Sn)	5	2	¹⁵² La	20	
¹⁰⁵ Y	12	7	(¹¹⁸ Ru)	3	2	¹³⁷ Sb	548	164	(¹⁵³ La)	5	
¹⁰⁶ Zr	336	165	¹¹⁸ Rh	313	160	¹³⁸ Sb	50	16	(¹⁵³ Ce)	2	
¹⁰⁷ Zr	38	21	¹¹⁹ Rh	82	30	¹³⁹ Sb	8	3	¹⁵⁴ Ce	9	
¹⁰⁸ Nb	413	210	¹²⁰ Rh	13	5	¹³⁹ Te	1549	540	(¹⁵⁵ Ce)	4	
¹⁰⁹ Nb	81	41	(¹²¹ Rh)	3	1	¹⁴⁰ Te	264	78			
¹¹⁰ Nb	10	6	¹²¹ Pd	211	150	¹⁴¹ Te	39	13			

Table 1