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Identification of more than a 100 new isotopes from ^{238}U projectile fission and beams of neutron-rich nuclei at BRENDA

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Abstract

Projectile fission of ^{238}U was investigated at a bombarding energy of 750 A-MeV using Pb and Be targets. The fully stripped forward emitted fragments from Ti to Cs 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 a hundred new nuclear species were identified including the ^{78}Ni , for which a cross-section of 300 pb was measured.

1 Introduction

On the nuclear chart large empty regions remain unexplored. They are located on the neutron-rich side of the valley. The investigation of this wild land becomes a challenge for experimentalists. N-rich nuclei of intermediate masses are of interest for nuclear physics as well as for astrophysics. Collective effects due to large asymmetries between proton and neutron numbers can only show up in this region. How the neutrons in excess are coupled to the core or how they are incorporated in the bulk of the nuclear matter is an open question. Nuclear properties of neutron-rich isotopes have to be known to reproduce the mass abundance distribution in the solar system resulting from the r-process and to constrain the conditions of the nucleosynthesis. Fission has been the most efficient nuclear process to produce and study neutron rich species of intermediate mass. More than 400 new isotopes were made

accessible for spectroscopic investigations owing to fission [1]. 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.

Despite improvements of the 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 fission products. 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 angular aperture, while the fragments are emitted isotropically. The present step between target fission and projectile fission is similar to the one between target spallation and projectile fragmentation.

2 Experiment

Acceleration of a ^{238}U beam up to relativistic energies by using the heavy ion synchrotron SIS at GSI offers a new way to investigate fission. The use of inverse kinematics at high energies to induce fission in peripheral collisions provides three main advantages:

1) The fragments are focused into a narrow cone centered around the beam direction and their velocities are almost constant. Thus, fragments are efficiently transmitted through a spectrometer.

2) The fragments are totally ionized, i.e. their charge state equals their atomic number and ambiguities when dealing with different charge states are avoided.

3) At these high velocities, fragments can be identified by $B\rho$ - ΔE -ToF techniques.

The thickness of the target can be increased up to $\text{gr}\cdot\text{cm}^{-2}$ however it does not compensate the fact that cross sections are much lower than in thermal neutron induced fission.

Two experiments were performed with a ^{238}U beam at 750 A·MeV energy; the first with a beam intensity of $2 \cdot 10^5$ U ions/s and the second with 10^7 U ions/s. The beam impinged on Be or Pb targets of 1 g/cm² and 1.25 g/cm² thickness respectively. The Pb target with a high Z was chosen to enhance the contribution of electromagnetic fission.

Fission products were analyzed with the fragment separator FRS [2] operated in the achromatic mode. Energy loss of the separated fragments, characteristic of their nuclear charge Z , was measured in a four-stage MUSIC ionization chamber [3] at the exit of the FRS. Event-by-event, energy loss, time of flight, positions of the ion at intermediate and final focus were measured. The mass A was determined from

$$B\rho = \beta\gamma Ac/Z$$

with an accuracy of 1/250. For fragments of similar velocities, the magnetic deflection separates isotopes according to the mass-to-charge ratio, A/Z . Therefore, a survey of fission production rates in terms of A/Z is obtained by scanning the magnetic fields over the appropriate rigidity range (32%).

The velocities of forward (or backward)-emitted fragments are higher (or lower) by about 5% as compared to the projectile velocity. They are selected within the angular and momentum aperture of the FRS as shown in Fig.1. Note that only an exothermic

process like fission can increase the velocity of the fragment as compared to projectile. Neutron-rich isotopes with A/Z values equal or larger than in ^{238}U can be produced only when very few or no neutrons are evaporated, and therefore at low excitation energy. Low energy fission is selected at the largest longitudinal momenta.

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

Due to the 5 mm scintillator in the dispersive focal plane only a certain range of elements was simultaneously transferred. The FRS was tuned on the light group of fission fragments ($Z=40$), therefore the heavy fragments ($Z > 50$) as well as the very light ones ($Z < 30$) were poorly transmitted. Beyond $Z=58$ fragments could not be detected. The entrance angle and momentum aperture of the FRS introduce cuts in the phase space of fission fragments. For a target thickness of $\approx 1 \text{ g/cm}^2$, transmissions of 2.8% for Sr-fragments and 5% for Te-fragments were calculated by using the Monte-Carlo simulation code MOCADI [4].

3 Fission Results

The element distribution of separated fragments measured on the Pb target at a magnetic rigidity of 4% above that of the projectile, $(B\rho)_0$, is shown in fig. 2a.

Fission fragments produced with the highest yields were transmitted at this rigidity. A double-humped Z distribution typical of low energy fission of U is obtained, each element being nicely separated. The well established features of low energy fission process are observed as a significant odd-even effect (5%) and a shell effect at $N=82$, which enhances the yields for the complementary pair Te ($Z=52$) and Zr ($Z=40$). On the corresponding scatter-plot ΔE -ToF (Fig. 2b), masses are also separated. The pair of isotopes ^{134}Te - ^{100}Zr , found with the largest yields indicates an evaporation of four neutrons. In the region of symmetry, ^{116}Pd appears with the largest yield and fission then is associated with the emission of six neutrons. The fragment production was integrated over the velocity distribution (or $B\rho$).

Fig. 2a Elemental yields measured in 750 A.MeV U projectile fission on Pb at 1.04 times the projectile magnetic rigidity, $(B\rho)_0$. This setting corresponds to a maximum of the transmission for the most abundant isotopes.

Fig. 2b ΔE versus A/Z scatter-plot for the same measurement.

a) Cross-sections and yields

Production yields and cross-sections were derived after correction for the transmission through the FRS. Values of 2.1 ± 0.2 barn and 0.12 ± 0.04 barn were found for cross-sections of ^{238}U low excitation fission on Pb and Be respectively. It is only a fraction of the TOTAL fission process for which cross-sections were measured in another stage of the experiment [5]. They were found to be 3.54 barn for the U/Pb system -compatible with an independant measurement

performed at GSI [6]- and 1.03 barn for the U/Be system. The distributions of yields normalized to 200% are reported on Fig. 3. On Pb, elemental yields are compared with thermal neutron induced fission yields on ^{235}U . [7].

Fig. 3 Elemental distribution of ^{238}U fission yields for Be and Pb targets. The yields obtained with thermal neutrons on ^{235}U is given for comparison.

The filling of the valley by two orders of magnitude as compared to the thermal neutron induced fission and the smaller odd-even effects in the symmetry region can be attributed only to fission occurring with a higher excitation energy. The yields are also larger for large asymmetry. 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 15 MeV) and a nuclear fission process filling the valley (20 to 25 MeV). On Be, the elemental yield distribution is asymmetric, in contrast with a pure "nuclear" fission although Coulomb fission contributes only to a small extent 0.03 barn [5]. Yields for the production of elements with $Z < 30$ or elements in the valley are found to be larger on Be than on Pb target.

b) Very neutron-rich nuclei

Since forward-emitted fission fragments have nearly equal velocities [9], higher values of $B\rho$ select larger A/Z ratios. The largest $B\rho$ setting for which fragments were still detected with sufficient intensities in the first experiment, - limited by the ^{238}U beam intensity - amounted to $1.08 \cdot (B\rho)_0$. In the corresponding ΔE -ToF scatter-plot more than 50 new neutron rich isotopes were discovered [10]. The most abundant isotope observed for this setting was the doubly magic ^{132}Sn counted with a rate of about 1 event/s for a beam intensity of $10^5/\text{s}$.

By decreasing the rigidity, the backward emitted fragments are selected. In Fig. 4 for a

rigidity of $0.94 \cdot (B\rho)_0$, very n-rich fission fragments backward emitted ($A/Z = 2.55$) are selected. They are transmitted together with lighter fragments emitted forwardly from secondary fission.

Fig. 4 Scatter plot of fragments at $0.94 \cdot (B\rho)_0$.

In this plot events from U-fission at low excitation energy emitted backward are observed and fission from excited U-fragments - with lower A/Z - and larger velocities.

The velocity distribution of these lighter fragments demonstrates that they are due to fission of excited U-fragments (see Fig. 5 and Fig. 1). The U-fragments are produced by the abrasion of several nucleons in more central collisions. In contrast the distribution of fragmentation products is centered around the projectile velocity. The A/Z ratio for those last fragments is even lower; On the neutron deficient side, isotopes of ^{112}Te , ^{97}Pd and ^{84}Zr are observed which belong to the "corridor of fragmentation" residues.

Fig. 5 Velocity distributions of fission and fragmentation products for Pd isotopes. Note the large mass range from 122 to 99.

4 Discovery of new n-rich isotopes including ^{78}Ni

Until today the most neutron-rich Ni isotopes were produced either by low-energy fission or by projectile fragmentation. The isotopes of $^{70-74}\text{Ni}$ were discovered [11] in very asymmetric thermal-neutron induced fission of ^{235}U . Production of $^{75-76}\text{Ni}$ was investigated by ^{86}Kr induced projectile-fragmentation at 500 A-MeV, and their half-lives were determined [12, 13].

A 1 g/cm² Be target was selected to separate more n-rich isotopes of elements around Ni using fission of U-projectiles since the yield for asymmetric fission was found higher [8] and the luminosity is larger as compared to a Pb target with a similar location straggling. The field setting of the FRS was calculated with the velocity $\beta_f = 0.054$ deduced from the mean fission kinetic energy measured in $^{235}\text{U}(n_{th}, f)$ [14] and the rigidity was calculated to be $B\rho = 1.16 \cdot (B\rho)_0$. The momentum width of Ni fragments reached 4 %, due to the large difference in the stopping power in the target between U and Ni. The transmission was calculated to be 1.6 % using the Monte-Carlo simulation and it results an effective luminosity of $2 \cdot 10^{28} \text{cm}^{-2} \text{s}^{-1}$.

In the corresponding scatter plot $\Delta E - A/q$, isotopes of twenty elements were clearly separated. An ionization chamber located in the dispersive focal plane was delivering ΔE signals to exclude a possible change in the atomic charge state at any ends of the flight path used to characterize the isotope. For a total dose of 10^{13} U ions delivered in 132 h, three events were assigned to the isotope ^{78}Ni . The count rate of 3 events in 132 hours is compatible with the rates extrapolated from ^{77}Ni and ^{76}Ni counting and with the gaussian fall-off obtained for other isotopic distributions. A neutron odd-even effect is seen in the production of very n-rich nuclei. The striking drop between $^{79}\text{Cu}_{50}$ and $^{80}\text{Cu}_{51}$ points to the shell closure at $N=50$ [15].

Fig. 6 Mass spectra of the elements selected at $B\rho = 1.16 B\rho_0$ for a setting centered on ^{78}Ni . The mass numbers are indicated for the isotopes discovered with U-projectile fission.

The spectra shown on Fig.6 were obtained by projecting events collected during this long measurement on the A/q axis. In the region from Ti to Nb fifty seven new nuclei are identified. For other elements, three to five new neutron-rich isotopes are discovered for each element between Ge and Pd. Production cross sections were calculated. For ^{78}Ni a value of 0.3 (1) nb is obtained. It is the present lower value of cross-section which can be measured.

Finally referring to the last edition of the chart-of-nuclei [16] more than 100 new isotopes were observed in the two measurements. In the first run with the FRS tuned on very n-rich isotopes more than 50 new nuclei were discovered during about 10 hours of data taking. A limit in A/Z of 2.68 and cross-sections as low as 1 μbarn were reached. In the second experiment with a beam a hundred times more intense and a counting time of 130 h the lower limit of the cross-section was pushed down by a factor of 3000 and the ratio A/Z increased up to 2.78.

Fig. 7 Chart of isotopes with the new isotopes discovered during the two experiments of U projectile fission. The r -process limit indicated on the plot [17] is reached up to bromine and between Sn and Cs.

In the region of symmetric fission, where yields are much higher than in low energy fission, altogether five isotopes were discovered for each element Nb, Mo, Tc, Ru, Rh and Pd. Note that previous studies in this region were performed with the IGISOL facility by using 20 MeV proton-induced fission of ^{238}U [18]. The new nuclear species observed in our measurement fill the gap of elements not accessible by chemistry or by ISOL ion-source techniques. Isotopes located along the limits of the r -process are now accessible up to Br (see Fig. 7) and between Sn and Cs.

It has to be stressed that our experiment not only reveals a new landscape of fragments,

hidden until now, but it represents also the first ΔE -ToF isotopic identification of any heavy fission fragments. Such a direct separation provides the way to accurately measure relative fission yields, free from any chemical bias. Moreover, the 300 ns required for the ions to travel through the FRS are shorter than any β decay half-lives thus the measured yields cannot be biased by β^- -decay.

Since fission fragments can be separated the same way as any other projectile fragments - ^{73}Ni was selectively implanted-, a new class of very neutron-rich high energy secondary beams are available in addition to the fragmentation products used until now. They can be selectively implanted in order to measure β -decay half-lives [19]. Even though the counting rates are still low at the present beam intensities, one can expect an increase by a factor of 10^3 with the new injector and improvements in the angular transmission in the FRS. Today, BREND A is the only facility where extremely n-rich beams are available.

5 Conclusions

We have shown that by studying collisions of relativistic ^{238}U all fragments resulting from fission (500) and from fragmentation (1500) can be separated in flight by the FRS and identified unambiguously. With each setting of the FRS, a hundred and fifty isotopes are simultaneously analyzed.

The experimental method described here represents a novel and efficient way to study both qualitatively and quantitatively the processes which take place in collisions of 1 GeV protons on actinide targets in incineration reactions or on Pb targets in energy-producing subcritical accelerator devices. Up to now, in spallation or fission of target nuclei, the low velocities of the fragments have prevented a comprehensive study of the processes.

A number of new neutron rich isotopes were discovered with emittances and velocities appropriate for secondary reaction studies. Total cross-sections and Coulomb excitation or dissociation can already be measured which are relevant for nuclear spectroscopy and for astrophysics. The mass excess of the new isotopes can be determined using the ESR as a high resolution spectrometer since most of the expected half-lives are short.

Parallel to the efforts to increase the U-beam intensity, new experimental setups aiming at nuclear structure studies are to be initiated.

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