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Production of new neutron-rich isotopes of heavy elements in fragmentation reactions of ^{238}U projectiles at 1A GeV

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The production of heavy neutron-rich nuclei has been investigated using cold-fragmentation reactions of ^{238}U projectiles at relativistic energies. The experiment performed at the high-resolving-power magnetic spectrometer Fragment Separator at GSI made it possible to identify 40 new heavy neutron-rich nuclei: ^{205}Pt, $^{207-210}$Au, $^{211-216}$Hg, $^{214-217}$Tl, $^{215-220}$Pb, $^{219-224}$Bi, $^{223-227}$Po, $^{225-229}$At, ^{230,231}Rn, and ^{233}Fr. The production cross sections of these nuclei were also determined and used to benchmark reaction codes that predict the production of nuclei far from stability.

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The possibility to extend the present limits of the chart of the nuclides provides unique opportunities for investigating the nuclear many-body system with extreme values of isospin and most of the stellar nucleosynthesis processes leading to the production of the heaviest elements in our universe [1]. This is the reason why presently, several new-generation in-flight radioactive-beam facilities are being commissioned, built, or designed [2–4]. These facilities will take advantage of two reaction mechanisms, fission and fragmentation, for producing nuclei far from stability.

Fragmentation reactions of ^{48}Ca beams have been used to produce light neutron-rich nuclei [5] and reach the heaviest known nuclei at the neutron drip line [6]. Fission reactions have proven to be an optimum reaction mechanism for the production of medium-mass neutron-rich nuclei [7,8]. Recently, intense beams of ^{238}U at the new Radioactive Ion Beam Factory in RIKEN made possible the production of 45 new medium-mass neutron-rich nuclei in in-flight fission reactions [9]. The next step could be the fragmentation of beams of neutron-rich fission fragments such as ^{132}Sn to produce extremely neutron-rich nuclei as proposed in Ref. [10].

However, the access to heavy neutron-rich nuclei—in the "northeast" region of the chart of nuclides—seems to be still a real challenge. Indeed, the heaviest known isotopes in this region are still located relatively close to the β stability line. A few years ago, it was proposed to use fragmentation reactions of heavy stable projectiles such as ^{238}U or ^{208}Pb at relativistic energies to populate that region of the chart of nuclides [11,12]. The idea behind it was to take advantage of the complete noncorrelation of the nucleons removed from the projectile, leading to a large distribution in N/Z of the projectile prefragments, and the large range in excitation energy gained per abraded nucleon during this process. Both effects should be sufficient to populate cold-fragmentation reaction channels where the incident projectiles lose mostly protons while the excitation energy gained in the process is rather low. The extreme case for these reactions involves the proton-removal channels where the projectiles lose only protons, and the excitation energy gained is below the particle-evaporation threshold.

To test this idea, we performed an experiment at GSI Darmstadt where a ^{238}U beam was accelerated at 1A GeV with the SIS synchrotron with an intensity around 10^8 ions/s. The fragmentation residues produced in collisions with a 2500 mg/cm^2 beryllium target were analyzed with the high-resolving-power magnetic spectrometer Fragment Separator (FRS) [13]. This is a zero-degree magnetic spectrometer with two symmetric sections to preserve the achromatism of the system. The spectrometer is characterized by a resolving power $B\rho/\Delta(B\rho) \approx 1500$, a momentum acceptance $\Delta p/p \approx 3\%$, and an angular acceptance around its central trajectory $\Delta\theta \approx \pm$ 15 mrad. A profiled energy degrader placed at the intermediate image plane was fundamental for the separation of the transmitted nuclei [14] and the identification of atomic charge states, as explained in what follows. The nuclei traversing the spectrometer were identified by determining their magnetic rigidity, velocity, and atomic number. The magnetic rigidity was derived from the positions of the nuclei along the dispersive coordinate at the intermediate and final image planes. These positions were measured with two plastic scintillators covering both image planes and providing the arrival times of the induced signals at both ends of the scintillation plate. These two scintillators provided also

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the time of flight of the ions between both image planes (≈35 m). Finally, two multisampling ionization chambers placed at the end of the spectrometer provided the identification of the residual nuclei in atomic number from the registered energy loss. A description of the identification method for these nuclei can be found in Refs. [15,16].

The unambiguous identification of neutron-rich nuclei requires the separation of the different charge states of the nuclei transmitted through the spectrometer. Those nuclei changing their charge state while traversing the matter located at the intermediate image plane (plastic scintillator and energy degrader) can be easily identified by their change in magnetic rigidity. However, the identification of non-fully stripped nuclei preserving their charge state along the spectrometer is a challenging problem. Hydrogenlike nuclei have almost the same mass over ionic charge state ratio A/q as fully stripped nuclei of the same element with three more neutrons. Because the cross section of the most neutron-rich isotope is, on average, around two orders of magnitude smaller than the contaminant, contributions of hydrogenlike nuclei of the order of few percent represent a large contamination for the identification of the most neutron-rich isotopes. This problem is particularly important in the region of interest of this work because the fraction of non-fully stripped nuclei increases considerably with the atomic number of the nuclei [17].

The key parameter in the present experiment for overcoming this problem was the relativistic energy of the nuclei under investigation, reducing drastically the fraction of non-fully stripped nuclei [17]. For example, using a $1A$ GeV ^{238}U beam, the expected fraction of fully stripped ^{226}Po nuclei produced in a beryllium target having a thickness equivalent to 20% of the range of the projectile and a 120 mg/cm^2 niobium stripper is 89%. At $500A$ MeV that fraction would be only 58% [18]. Moreover, we used two multisampling ionization chambers at the final image plane with a 100 mg/cm^2 niobium stripper foil in between, further reducing the amount of non-fully stripped nuclei. As explained in Ref. [11], combining the difference in magnetic rigidity in the two sections of the spectrometer and the difference in energy loss in both ionization chambers, we could separate the charge states of the transmitted nuclei and, in particular, most of the hydrogenlike nuclei that preserve their charge state along the experimental setup [15]. The final absolute contamination of the remaining hydrogenlike nuclei to the corresponding $A+3$ isotopes was estimated to be, at most, 40%.

To search for new heavy neutron-rich nuclei, we tuned the FRS magnets for centering the nuclei ^{227}At, ^{229}At, ^{216}Pb, ^{219}Pb, and ^{210}Au along its central trajectory. Combining the signals recorded in these settings of the FRS and using the analysis technique previously explained, we were able to identify 40 new neutron-rich nuclei with atomic numbers between $Z = 78$ and $Z = 87$: ^{205}Pt, $^{207-210}$Au, $^{211-216}$Hg, $^{214-217}$Tl, $^{215-220}$Pb, $^{219-224}$Bi, $^{223-227}$Po, $^{225cd-229}$At, ^{230,231}Rn, and ^{233}Fr. In Fig. 1 we present the identification matrix obtained for all the observed isotopes of elements between platinum and francium. The criteria followed for accepting the identification of a nucleus observed for the first time is a number of events compatible with the corresponding mass and atomic number, located in the expected range of positions at both image planes of the spectrometer. Moreover, this number of events should also be compatible with a probability larger than 95% that at least one of the counts does not correspond to a charge-state contaminant. Random background cannot be fully excluded; therefore, one should take with caution the identifications of ^{217}Tl and ^{216}Hg because the observed events do not seem to properly scatter around the expected values of mass and atomic number. The left panel in this figure corresponds to the scatter plot where we represent atomic number versus the ratio mass over nuclear charge, which allows us to identify the nuclei measured in the experiment. In this figure the horizontal and vertical thick-solid lines show the present limits of the chart of nuclides. Therefore, all nuclei located to the right of these lines were previously unobserved. The production of ^{203}Pt and ^{204}Pt in the fragmentation of ^{208}Pb projectiles at $1A$ GeV was previously reported by some of us [19]. In the right panel of the figure we represent the corresponding mass-over-nuclear charge distribution for the most neutron-rich isotopes of elements between platinum and francium we have measured. This plot clearly shows the resolving power and the unambiguous isotopic identification we obtain. We indicate the previously unknown isotopes by their mass number.

We could also determine the production cross sections of these nuclei normalizing the production yields to the integrated beam current and the number of target nuclei. The beam current was continuously measured during the experiment using a secondary-electron monitor placed before the reaction target. This monitor was carefully calibrated during the experiment using a plastic scintillator [15]. The measured yields were also corrected by the losses inherent to the experimental technique used in this work. The main corrections were attributable to the limited momentum acceptance of the spectrometer, in particular, for those nuclei with magnetic rigidities far from the central value defined by the FRS tune, the fraction of non-fully stripped nuclei, and secondary reactions in all layers of matter placed along the spectrometer. Smaller corrections were attributable to the acquisition dead time and detectors efficiency.

Particular attention was paid to the evaluation of the uncertainties associated with the measured cross sections. Statistical uncertainties were below 10%, except for the two most neutron-rich nuclei measured per element with much smaller statistics. The systematic uncertainties were associated with the different corrections applied to the measured yields, and their magnitude was carefully evaluated around 20%.

In Fig. 2 we represent the isotopic distributions of the cross sections measured in this work. The error bars are shown when larger than the data points. Those points in the figure surrounded by a square correspond to the 40 new isotopes discovered in this experiment. In a few days' measurements we were able to reach cross sections as low as 100 pb. As expected, the production cross sections decrease drastically with the neutron number. On average, an additional neutron decreases the production cross section by about a factor 4.

In the same figure we also compare the measured cross sections with predictions obtained with the codes EPAX [20] and COFRA [11,21]. EPAX is a well-known parametrization of measured cross sections and COFRA is an analytical version

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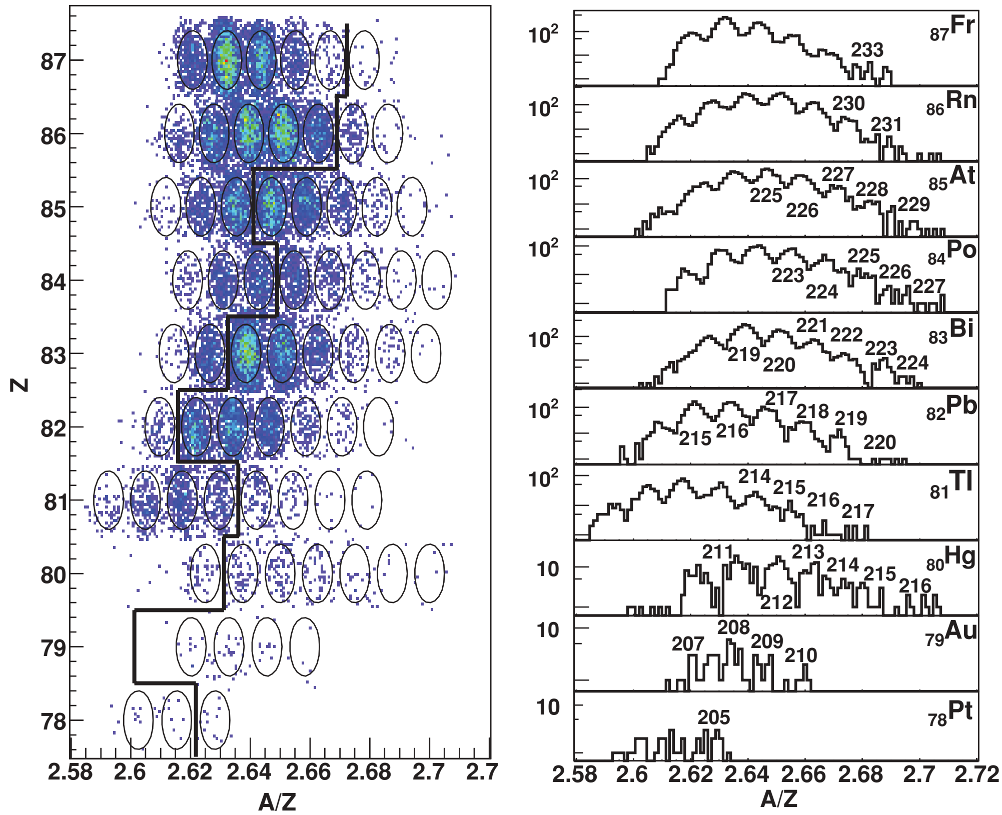

FIG. 1. (Color online) (Left) Identification plot of all nuclei produced in this work; see text for details. (Right) A/Z distribution of the recorded events for elements between Pt and Fr. Previously unknown nuclei are indicated by their mass number.

of the abrasion-ablation fragmentation model of Gaimard and Schmidt [22]. The COFRA code uses a full description of the abrasion stage of the collision while the ablation stage considers only the evaporation of neutrons, which is well adapted to the present case. This simplification makes possible an analytical formulation of the deexcitation stage that reduces considerably the computation time. However, COFRA includes the description of the uncorrelated abrasion of protons and neutrons, according to a hypergeometrical distribution [23], in the large range in excitation energy per abraded nucleon gained by the prefragments, owing to the random hole creation in the Fermi distribution of the nucleons inside the nucleus [11]. Although fragmentation produces mainly neutron-deficient nuclei, the aforementioned fluctuations in the abrasion process lead to less probable reaction channels where mostly protons are abraded and the excitation energy gained in the process is low enough to minimize neutron evaporation. Those reaction channels are thought to be responsible for the production of the most neutron-rich nuclei observed in this work.

The benchmarking of both calculations yields the following conclusions. The EPAX code describes rather well the production of residual nuclei relatively close in mass number to the projectile; however, it overpredicts the production cross sections of neutron-rich residual nuclei produced in midperipheral collisions where the projectile loses a larger number of nucleons. Similar conclusions have been obtained in other works [24]; EPAX describes rather well the production cross sections of neutron-deficient nuclei in fragmentation reactions but it overestimates the production of neutron-rich nuclei relatively far in mass number to the initial projectile. Because the parameters used in EPAX were obtained from fits to measured cross sections, the lack of data in the region covered by the present work might explain this finding.

COFRA provides an overall good description of the cross sections of neutron-rich nuclei produced in fragmentation reactions induced by ^{238}U projectiles. A more detailed analysis indicates a slight overestimation of the cross sections of nuclei close to the projectile (e.g., francium isotopes) and far from the projectile (e.g., gold and platinum isotopes). Since francium isotopes were transmitted at the edge of the spectrometer acceptance, problems in the evaluation of their transmission could explain the observed underestimation. For the second case (gold and platinum isotopes), it has been indicated that the overestimation of the cross sections obtained with COFRA could be attributable to the depopulation of these neutron-rich nuclei by fission reactions not included in the code [25]. However, the fact that this slight overestimation of the cross sections of neutron-rich residual nuclei has also been observed in the fragmentation of nonfissile projectiles such as ^{136}Xe [24] rules out this conclusion.

In this work we have investigated the production of heavy neutron-rich nuclei using fragmentation reactions of relativistic ^{238}U projectiles. The use of an intense beam and a high-resolving-power magnetic spectrometer made it possible to produce in few days a large number of neutron-rich isotopes of elements between platinum and francium with cross sections

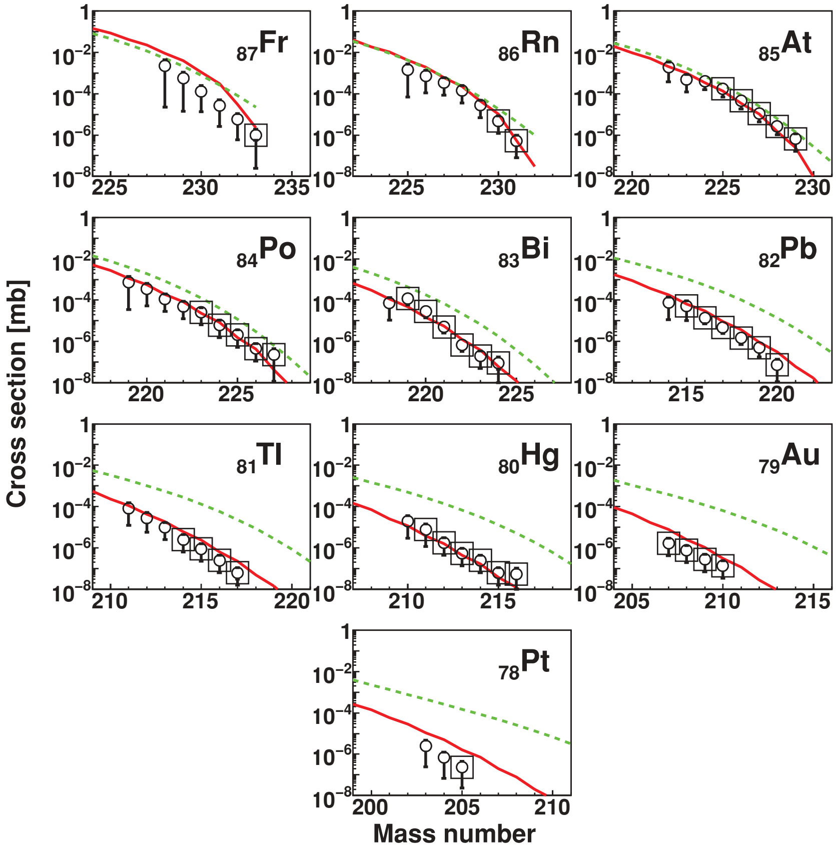

FIG. 2. (Color online) Isotopic distributions of the production cross sections of heavy neutron-rich nuclei determined in this work. Nuclei observed for the first time are surrounded by a square symbol. The experimental measurements are compared with the predictions obtained with the code COFRA (solid line) and EPAX (dashed line).

as low as 100 pb. In particular, we identified for the first time 40 new neutron-rich isotopes of the elements ^{205}Pt, $^{207-210}$Au, $^{211-216}$Hg, $^{214-217}$Tl, $^{215-220}$Pb, $^{219-224}$Bi, $^{223-227}$Po, $^{225-229}$At, ^{230,231}Rn, and ^{233}Fr. It was also shown that a key parameter for the unambiguous identification of these new nuclei was the high energy of the projectiles, reducing drastically the contamination of atomic charge states. The production cross sections of these nuclei were also determined with good accuracy. These cross sections were used to benchmark the two most widely used codes for estimating the production cross sections of neutron-rich residual nuclei in fragmentation reactions. While EPAX clearly overestimates the production of residual nuclei far in mass number to the initial projectile, the COFRA code provides an overall good description of the cross sections. The description of the data with this code confirms the large fluctuations in the number of abraded protons and neutrons and in the excitation energy gained by the prefragments. These large fluctuations make it possible to populate cold-fragmentation reaction channels leading to the production of the most neutron-rich nuclei. These results pave the way for a considerable extension of the northeast limit of the chart of nuclides expected with the new generation of radioactive-beam facilities.

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