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

H. Alvarez-Pol,1 J. Benlliure,1 L. Audouin,2 E. Casarejos,1 D. Cortina-Gil,1 T. Enqvist,3 B. Fernandez,1 A.R. Junghans,2 B. Jurado,3 P. Napolitani,2,3 J. Pereira,1 F. Rejmund,9 K.-H. Schmidt,2 and O. Yordanov1

1 Universidad de Santiago de Compostela, 15782 Santiago de Compostela, Spain
2 IPN, IN2P3-CNRS, Université Paris-Sud 11, UMR 8608, F-91406 Orsay, France
3 Forschungszentrum Dresden-Rossendorf, D-01328 Dresden, Germany
4 Université Bordeaux I, CNRS/IN2P3, CENBG, BP 120, F-33175 Gradignan, France
5 GANIL CEA/DSM-CNRS/IN2P3, BP 55027, F-14076 Caen, France

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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-resolution-power magnetic spectrometer FRS at GSI allowed to identify 45 new heavy neutron-rich nuclei: $^{205}$Pt, $^{207}$−$^{210}$Au, $^{211}$−$^{216}$Hg, $^{213}$−$^{217}$Tl, $^{215}$−$^{220}$Pb, $^{219}$−$^{224}$Bi, $^{221}$−$^{227}$Po, $^{224}$−$^{229}$At, $^{229}$−$^{231}$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. This is the reason why presently, several new-generation in-flight radioactive-beam facilities are being commissioned, built or designed. These facilities will take advantage of two reaction mechanisms, fission and fragmentation, for producing nuclei far from stability. Since a large fraction of nuclei at the proton drip-line have already been produced at the present facilities, the new ones will mostly contribute to enlarge our field of action in the neutron-rich side of the nuclear chart.

Fragmentation reactions of $^{48}$Ca beams have been used to produce light neutron-rich nuclei and reach the heaviest known nuclei at the neutron drip-line. Fission reactions have proven to be an optimum reaction mechanism for the production of medium-mass neutron-rich nuclei. 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. 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.

The access to the north-east region of the chart of nuclides seems, however, a real challenge. Indeed, the heaviest known isotopes in this region are still located relatively close to the $\beta$ 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. The idea behind was to take advantage of the large fluctuations in N/Z and excitation energy of the projectile pre-fragmentations produced in the abrasion stage of the reaction. Those fluctuations 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 are the proton-removal channels where the projectiles lose only protons, and the excitation energy gained is below the particle-evaporation threshold.

In order to test this idea, we performed an experiment at GSI Darmstadt where a $^{238}$U beam was accelerated at 1 A 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 analysed with the high-resolution-power magnetic spectrometer Fragment Separator (FRS). This is a zero-degree magnetic spectrometer with two symmetric sections in order to preserve the achromatism of the system. The spectrometer is characterised 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 $\Delta \theta \approx 15$ mrad. A profiled energy degrader placed at the intermediate image plane was fundamental for the separation of the transmitted nuclei and the identification of atomic charge states, as explained below. 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 inter-
mEDIATE and final image planes. These positions were
measured with two plastic scintillators covering both im-
age planes and providing the arrival times of the induced
signals at both ends of the scintillation plate. These two
scintillators provided also the time of flight of the ions be-
tween both image planes (≈35 m). Finally, two multi-
sampling ionisation 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 [13, 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 (plas-
tic scintillator and energy degrader) can be easily iden-
tified by their change in magnetic rigidity. However, the
identification of non fully stripped nuclei preserving their
charge state along the spectrometer is a challenging prob-
lem. Hydrogen-like 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. Since the
cross sections of the most neutron-rich isotope is, in
average, around two orders of magnitude smaller than the
contaminant, contributions of hydrogen-like nuclei of the
order of few per cent represent a large contamination for
the identification of the most neutron-rich isotopes. This
problem is particularly important in the region of inter-
rest for this work since 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 over-
coming this problem was the relativistic energy of the
nuclei under investigation, reducing drastically the frac-
tion of non fully stripped nuclei [17]. For example, us-
ing a 1 A GeV 238U beam, the expected fraction of fully
stripped 226Po nuclei produced in a beryllium target hav-
ing a thickness equivalent to 20% of the range of the pro-
jectile and a 120 mg/cm² niobium stripper is 89%. At 500
A MeV that fraction would be only 58% [13]. Moreover,
we used two multi-sampling ionisation chambers at the
final image plane with a stripper foil in between, further
reducing the amount of non fully stripped nuclei. As ex-
plained in Ref. [11], combining the difference in magnetic
rigidity in the two sections of the spectrometer and the
difference in energy loss in both ionisation chambers we
could separate the charge states of the transmitted nuclei
and, in particular, most of the hydrogen-like nuclei that
preserve their charge state along the experimental set-up
[13]. The final absolute contamination of the remaining
hydrogen-like nuclei to the corresponding A+3 isotopes
was estimated to be, at most, 40%.

In order to search for new heavy neutron-rich nuclei,
we tuned the FRS magnets for centering the following
nuclei, 227At, 229At, 218Pb, 219Pb and 210Au, along its
central trajectory. Combining the signals recorded in
these settings of the FRS and using the analysis tech-
nique previously explained we were able to identify 45
new neutron rich nuclei with atomic numbers between
Z=78 and Z=87: 205Pt, 207–210Au, 211–216Hg, 213–217Tl,
215–220Pb, 219–223Bi, 221–227Po, 224–229At, 229–231Ru
and 233Fr. In Fig[1] we present the identification ma-
trix 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 images planes of the
spectrometer and with a probability of being background
or contaminants below 5%. The left panel in this fig-
ure 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 203Pt and 204Pt in the fragmentation of 208Pb projec-
tiles at 1 A GeV was previously reported by some of us
[19]. In the right panel of the figure we represent the cor-
responding 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 identi-
fication we obtain. We indicate the previously unknown
isotopes by their mass number.

We could also determine the production cross sections
of these nuclei normalising 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 scintil-
lator [15]. The measured yields were also corrected by
the losses inherent to the experimental technique used in
this work. The main corrections were due to the limited
momentum acceptance of the spectrometer, in particu-
lar, 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 due to the acquisition dead time and
detectors efficiency.

Particular attention was paid to the evaluation of the
uncertainties associated to 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 to the different corrections applied to the
measured yields, and their magnitude was carefully eval-
uated 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
FIG. 1: (Color online) Left panel: Identification plot of all nuclei produced in this work, see text for details. Right panel: A/Z distribution of the recorded events for elements between Pt and Fr. Previously unknown nuclei are indicated by their mass number.

In the figure surrounded by a square correspond to the 45 new isotopes discovered in this experiment. In a few days measurement we were able to reach cross sections as low as 100 pbarn. As expected, the production cross sections decrease drastically with the neutron number. On average, an additional neutron decreases the production cross section 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 parametrisation of measured cross sections while COFRA is an analytical version 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 de-excitation stage that reduces considerably the computation time. Therefore, COFRA includes fluctuations in the number of abraded protons and neutrons according to a hyper-geometrical distribution [23] and in the excitation energy gained by the prefragments due to the random hole creation in the Fermi distribution of the nucleons inside the nucleus [11].

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 over-predicts the production cross sections of neutron-rich residual nuclei produced in more central collisions where the projectile loses an important number of nucleons. Similar conclusions had 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.

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 far from the projectile. It has been indicated that this effect could be due to the depopulation of these neutron-rich nuclei by fission reactions not included in the COFRA code [25]. However, the fact that this slight overestimation of the cross sections of neutron-rich residual nuclei in more central collisions has also been observed in the fragmentation of non fissile 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 as low as 100 pbarn. In particular we identified for the first time 45 new neutron-rich isotopes, $^{205}$Pt, $^{207-210}$Au, $^{211-216}$Hg, $^{213-217}$Tl, $^{215-220}$Pb, $^{219-224}$Bi, $^{221-227}$Po, $^{224-229}$At, $^{228-231}$Rn and $^{233}$Fr. It was also shown that a key parameter for the unambiguous identification of these new nuclides is...
clei 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 overestimate 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. This large fluctuations make possible to populate cold-fragmentation reactions channels leading to the production of the most neutron-rich nuclei. These results pave the way for a considerable extension of the north-east limit of the chart of nuclides expected with the new generation of radioactive beam facilities.

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