Identification of New Isotopes $^{125}$Pd and $^{126}$Pd Produced by In-Flight Fission of 345 MeV/nucleon $^{238}$U: First Results from the RIKEN RI Beam Factory

Tetsuya OHISHI, Toshiyuki KUBO, Kensuke KUSAKA, Atsushi YOSHIDA, Koichi YOSHIDA, Naoki FUKUDA, Masao OHTAKE, Yoshiyuki YANAGISAWA, Hiroyuki TAKEDA, Daisuke KAMEDA, Yoshitaka YAMAGUCHI, Nori AOI, Ken-ichiro YONEDA, Hideaki OTSU, Satoshi TAKEUCHI, Takashi SUGIMOTO, Yosuke KONDO, Heiko SCHEIT, Yasuyuki GONO, Hiroyoshi SAKURAI, Tohru MOYOBAYASHI, Hiroshi SUZUKI,1, Masafumi MATSUSHITA, Kazuo IEKI, Takamasa KUBOKI, Takayuki YAMAGUCHI, Takeshi SUZUKI, Akira OZAWA, Tetsuaki MORIGUCHI, Yusuke YASUDA, Takashi NAKAMURA, Tomoyuki SHIMAMURA, Yoshihisa NAKAYAMA, Hans Geissel, Helmut WEICK, Jerry A. NOLEN, Oleg B. TARASOV, Anthony S. NETTLETON, Daniel P. BAZIN, Bradley M. SHERRILL, David J. MORRISSEY, and Wolfgang MITTIG

RIKEN Nishina Center, RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198
1Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033
2Department of Engineering Science, Osaka Electro-Communication University, 18-8 Hatsucho, Neyagawa, Osaka 572-8530
3Department of Physics, Rikkyo University, 3-34-1 Nishi-Bebakuro, Toshima-ku, Tokyo 171-8501
4Department of Physics, Saitama University, 255 Shimo-Okubo, Saitama 338-8570
5Institute of Physics, University of Tsukuba, 1-1-1 Ten'noudai, Tsukuba, Ibaraki 305-8571
6Department of Physics, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo 152-8551
7Department of Science and Technology, RIKEN, Wako 351-0198
8Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research (JINR), Dubna, 141980, Moscow Region, Russian Federation
9Grand Accelerateur National d’Ions Lourds (GANIL), Bd Henri Becquerel, BP 55027, 14076 CAEN Cedex 05, France

A search for new isotopes using in-flight fission of a 345 MeV/nucleon $^{238}$U beam has been carried out in the commissioning experiment of the next-generation in-flight radioactive isotope beam separator BigRIPS at the RI Beam Factory at the RIKEN Nishina Center. Two neutron-rich palladium isotopes $^{125}$Pd and $^{126}$Pd were observed for the first time, which demonstrates the great potential of the RIKEN RI beam factory.

KEYWORDS: nuclear reaction Be($^{238}$U, x) E = 345 MeV/nucleon, in-flight fission, new isotopes $^{125}$Pd and $^{126}$Pd, in-flight RI beam separator

LETTERS

DOI: 10.1143/JPSJ.77.083201-1

A next-generation radioactive isotope (RI) beam facility called the RI Beam Factory (RIBF) has been constructed and recently commissioned at the RIKEN Nishina Center that promises to advance the study of exotic nuclei to a great extent. The new superconducting in-flight RI beam separator BigRIPS is a major experimental device at the RIBF for RI beam production based on the in-flight separation technique and for research with exotic nuclei. Thanks to its large acceptances, not only projectile fragmentation of various heavy-ion beams but also in-flight fission of fissionable beams can be efficiently used as a production reaction in the BigRIPS separator. Figure 1 shows a schematic layout of the BigRIPS separator along with the IRC and SRC cyclotrons and the ZeroDegree spectrometer. The cyclotrons at the RIBF can accelerate all heavy ions up to approximately 400 MeV/nucleon, including very heavy elements such as uranium, with the goal of reaching an intensity of 1 pμA ($6 \times 10^{12}$ particles/s). In-flight fission of fissionable beams, such as a $^{238}$U beam, is known as an excellent mechanism for producing a wide range of neutron-rich exotic nuclei far from stability. This was well demonstrated by experiments at GSI, in which more than a hundred new isotopes were identified in a single experiment. The BigRIPS separator has been designed with large acceptances, to take advantage of the high production cross sections of neutron-rich isotopes from these reactions. The full angular acceptances of 80 mrad (horizontal) and 100 mrad (vertical), and the full momentum acceptance of 6% allows efficient RI beam production with in-flight fission, in which fission fragments are produced with large spreads in both angle and momentum. Due to its large acceptance, the BigRIPS separator has an approximately 50% efficiency for the collection of these fission fragments.

Another important feature of the BigRIPS separator is its two-stage structure, which allows delivery of tagged RI beams, or use as a two-stage separator. In the tagging mode, which may also be called a separator-spectrometer mode, the first stage is used to produce and separate RI beams with a wedge energy degrader, while the second works as a spectrometer to analyze and identify those RI beams. The momentum resolution of the second stage has been designed to be high enough to identify RI beams without measuring their total kinetic energies, even though they are produced in several charge states in our energy domain. In the two-stage
separator mode, the energy degrader is used at both of the stages in order to further purify the RI beams. The BigRIPS separator is composed of 14 large-aperture superconducting quadrupole triplets (STQ) and 6 room-temperature dipoles. They are labeled as STQ1–STQ14 and D1–D6 in Fig. 1, respectively. There are seven foci in the beam line, indicated as F1–F7. The BigRIPS separator includes the components from the production target position (F0) to F2, while the second stage spans those from F3 to F7. The two STQs, located between F2 and F3, are used as a matching section. Each focus is contained in a focal plane chamber to accommodate various beam-line detectors used for the diagnostic and particle identification.

The commissioning of the BigRIPS separator started in March 2007. The first RI beams were successfully produced in late March using $^{86}$Kr and $^{238}$U primary beams. Then in a follow-up experiment in May 2007, after evaluating the performance of the ion optics in the BigRIPS separator, the detector performance and the particle identification system, some beam time was devoted to search for new isotopes using in-flight fission of a $^{238}$U beam. During these measurements we reached easily the frontiers of known isotopes which were obtained experimentally with the RI beams and that took into account the effects of measured fringe fields of the magnets. The experimental conditions to search for new isotopes, such as the target thickness and the $B_\rho$ setting of the BigRIPS separator, were determined based on detailed simulations with the codes LISE++9) and MOCADI.10) The measurements were made at several $B_\rho$ settings ranging from 7.0 to 7.6 Tm in order to determine the production yields of neutron-rich isotopes near the known limits.

The particle identification (PID) was performed by measuring the time of flight (TOF), energy loss ($\Delta E$), and magnetic rigidity ($B_\rho$) of the fragments with the beam-line detectors. The TOF was recorded between two plastic scintillation counters placed at the F3 and F7 achromatic foci in the second stage. The scintillation counters were 0.2 mm thick and $100 \times 100$ mm$^2$ in area, and the mean flight path was 46.98 m. The $\Delta E$ was measured at F7 with two 0.35-mm-thick silicon detectors with an active area of $50 \times 50$ mm$^2$. The $B_\rho$ measurement was made by trajectory reconstruction from the positions and angles of the fragments measured in two position-sensitive PPACs11) at the F3 achromatic focus and two more at the F5 dispersive focus. The effective size of the PPACs was 240 mm (horizontal) and 150 mm (vertical). First-order ion-optical transfer matrices, which were obtained experimentally with the RI beams and the primary uranium beam, were used to reconstruct the trajectory. The $B_\rho$ value of the fragments on the central trajectory (central $B_\rho$) was determined by the magnetic fields of the dipoles measured by NMR probes and the central trajectory radii of the dipoles deduced from the magnetic field-map data. This method provided the high-resolution $B_\rho$ measurement of each fragment as described below.

The calibration of the TOF and $\Delta E$ measurements was made using the central $B_\rho$ value of the fragments. The small energy loss in the PPACs at F5 was taken into account in deducing the velocities of the fragments. The TOF values of the present measurements were around 230 ns. The mean-square (rms) resolution achieved in the present TOF and $B_\rho$ measurements was typically 40 ps and 0.03%, respectively. These values are the estimates for $^{104}$Zr fragments produced at a $B_\rho$ setting of 7.395 Tm.

Pile-up events were rejected by comparison of the pulse-heights in the two silicon detectors at F7 and the plastic scintillation counter at F7. Inconsistent events were also
searched for new isotopes. The resolution was 0.55%, and the relative rms was 0.041% for Pd isotopes.

Back mode, and the overall full-energy-peak efficiency was measured to be 3.5% for 1-MeV γ rays. In the present measurement we observed delayed γ rays from 96Rb that confirmed the PID. Figure 3 shows the γ ray energy spectrum gated on 96Rb events in the PID plot and the time span from 0.3 to 15 µs after the implantation. The γ ray peaks at 92.1, 116.3, 122.3, 184.8, 240.2, 300.0, 366.4, 370.0, and 461.2 keV originated from the decay of the isomeric state in 96Rb with an excitation energy of 1135 keV and a half-life of 2.0 µs. The lower panel of Fig. 2 shows an enlarged view of the PID plot, in which the fully-stripped 96Rb, 122Pd, and 125Pd isotopes are labeled. The new isotope 125Pd can be clearly identified. The neighboring events correspond to hydrogen-like 122Pd fragments.

Figure 4 shows the projected one-dimensional A/Q spectrum of Pd isotopes, along with the two-dimensional Z versus A/Q plot expanded around the region of the new Pd isotopes. The events that did not change their charge state at the F3 and F5 PPACs were selected by gating on the reconstructed Bρ spectra. The width of this gate was ±1% and matched the setting of the separator acceptance. The A/Q spectrum in Fig. 4 was obtained by gating the PID plot with a Z gate set to 46 ± 0.38(1.5σ), where σ represents the absolute rms Z resolution. The peaks for the fully stripped (Q = Z), hydrogen-like (Q = Z − 1), and helium-like (Q = Z − 2) ions are well separated from each other and the new isotope 125Pd is clearly identified at A/Q = 2.717. The peak centroid of this fully-stripped 125Pd is separated by 6.2σ from the neighboring 125Pd35, where σ, the absolute rms A/Q resolution, is 0.0011. The total yield of 125Pd is 22 counts. The new isotope 125Pd was also identified in both the A/Q spectrum and the Z versus A/Q plot in Fig. 4, although its total yield is only 3 counts. The integrated beam dose and the total irradiation time during the measurement were 3.6 × 1012 particles and approximately 25 h, respectively.

Figure 5 shows the measured production rates of the fully-stripped Pd isotopes along with the predictions from the LISE++ simulation in which the production cross sections of fission fragments were calculated with the LISE++ Abrasion–Fission (AF) model. This AF model relies on the so-called three excitation energy model in which three nuclei, 236U, 226Th, and 220Ra, are chosen to represent all the
fissile nuclei created in the abrasion–ablation stage and then the fission fragment distribution is calculated based on the semi-empirical model of ref. 5. The standard parameters of this model, such as excitation energies and production cross sections of the fissile nuclei, that reproduced the experimental data of the $^{238}\text{U} + \text{Be}$ reaction at 750 MeV/nucleon\(^7\) were used in the Monte Carlo mode to track the fission fragments produced in the target. The measured production rates smoothly decrease with increasing mass number, and are fairly well reproduced by the LISE++ predictions, although they are somewhat overestimated. The production cross sections of Pd isotopes have been estimated using the cross sections from the LISE++ AF model and the ratio of measured and predicted rates. The estimates for $^{124}\text{Pd}$, $^{125}\text{Pd}$, and $^{126}\text{Pd}$ isotopes are 80, 7, and 0.7 nb, respectively, and are consistent with the experimental results at GSI for $^{124}\text{Pd}$ (32 nb).\(^7\)

In summary, we have performed the first experiment at the RIBF using the BigRIPS in-flight RI beam separator and a search for new neutron-rich isotopes was carried out using the in-flight fission of a $^{238}\text{U}$ beam at 345 MeV/nucleon. We observed the production of two new isotopes $^{125}\text{Pd}$ and $^{126}\text{Pd}$, even though the uranium beam intensity was far from the goal for the RIBF and the total observation time was only about a day. This discovery demonstrates not only the performance of the BigRIPS separator, but also marks the launch of this new-generation RI beam facility.

**Acknowledgements**

This experiment was carried out under Program No. NP0702-RIBF20 at the RI Beam Factory operated by

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