Discovery of Rare Neutron-Rich Zr, Nb, Mo, Tc, and Ru Isotopes in Fission: Test of β Half-Life Predictions Very Far from Stability

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In experiments employing fast ion-guide-based on-line mass separation we have identified six new, rare nuclides of highly refractory elements produced in the fission of ^{238}U by 20-MeV protons. The new isotopes ^{105}Zr, ^{107}Nb, ^{109}Mo, ^{110}Mo, ^{113}Tc, and ^{115}Ru were identified through (β, $K\alpha$ x-ray) coincidences and β-delayed γ decay. The observed β half-lives range from 130 ms to approximately 1 s. All our recent β half-life data are compared with predictions of the deformed quasirandom-phase-approximation model. The main uncertainty is found to be the inaccuracy of the decay energy predictions.

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Low-energy nuclear fission is still an efficient way to produce new neutron-rich, medium heavy nuclei. Nuclei with an excess of as many as 14 neutrons, as compared with the nearest stable isotope, have been recently produced both in asymmetric $n_{\rm th}$-induced fission of ^{239}Pu [1] and in symmetric p-induced fission of ^{238}U [2]. The effect of a large neutron excess on the nuclear structure and β decay strength as well as its role in modeling the astrophysical r process are of central importance in these studies.

The aim of the present Letter is to provide new information obtained at the IGISOL (Ion Guide Isotope Separator On-Line) facility on the existence and decay of new neutron-rich nuclides with $40 \leq Z \leq 44$ and to discuss their β half-lives in the frame of the deformed quasirandom-phase-approximation (QRPA) model of Möller and Randrup [3]. Generally our knowledge of neutron-rich nuclei produced in fission is based on radioactivity measurements that are often associated with limited efficiency and/or selectivity. In our search for new isotopes we have utilized the internal conversion process following beta decay after fission [4]. This process is important for fission products with $A=100$-120 and 140-160 and particularly for the doubly even or odd nuclei, whose β decays often result in emission of characteristic K x rays of the daughter. This Z identification of the decaying nucleus together with the mass analysis provide unique isotopic identification.

Neutron-rich nuclei were produced in fission reactions induced by 20-MeV protons on four 20-mg/cm^2-thick uranium targets. Energetic fission fragments were slowed down, thermalized in helium, and transported by the ion guide for acceleration and mass analysis. The 40-keV beam of radioactive ions was then implanted on a catcher tape for measuring their radioactive decays. The method is chemically nonselective and allows fast separation (≈ 1 ms) of all fission products with nearly equal transmission [5]. In the present study, a typical intensity of 10^3 ions/s per mass number was obtained after mass separation for highly refractory elements between Zr and Rh. Nuclei of these elements with half-lives below 1 s are not available in ion-source-based conventional on-line isotope separators. The fission yield sensitivity of the whole setup was about 10^{-6}, which corresponds to a production rate of about 0.1 ion/s, as normalized to the 1-μA proton beam intensity.

To overcome the problem of poor efficiency and to utilize all the decay channels leading to x rays, we have developed an efficient system to record β decays in coincidence with low-energy photons. The setup consists of a 10-mm-thick planar Ge detector with an area of 10 cm^2 placed in close geometry behind the catcher tape and two 0.9-mm-thick plastic detectors placed on both sides of the implantation point for detecting β particles. This system had a nearly 10% coincidence efficiency for β-coincident K x rays. Conversion-electron-triggered events were eliminated by using 100 mg/cm^2 plastic as absorber between the source and the electron transmission detectors. High-energy γ rays were detected simultaneously with a 50% Ge detector placed behind one of the thin plastic detectors.

Figure 1 shows six examples of β-gated photon spectra of fission products, which were recorded for $A=105$, 107, 109, 110, 113, and 115 activities. The energy range is limited to 15-26 keV to highlight the region of the characteristic K x rays of interest. These mass numbers were selected as the most promising candidates for observing new isotopes. Each spectrum in Fig. 1 required about 30-50 mC of 20-MeV proton beam. The beam intensity in these experiments was typically only 300 nA. The cyclotron and the separator beams were pulsed in a synchronous way. Activity was recorded continuously during the beam-on and -off periods and the collection tape was moved 40 mm in the beginning of each new cy-

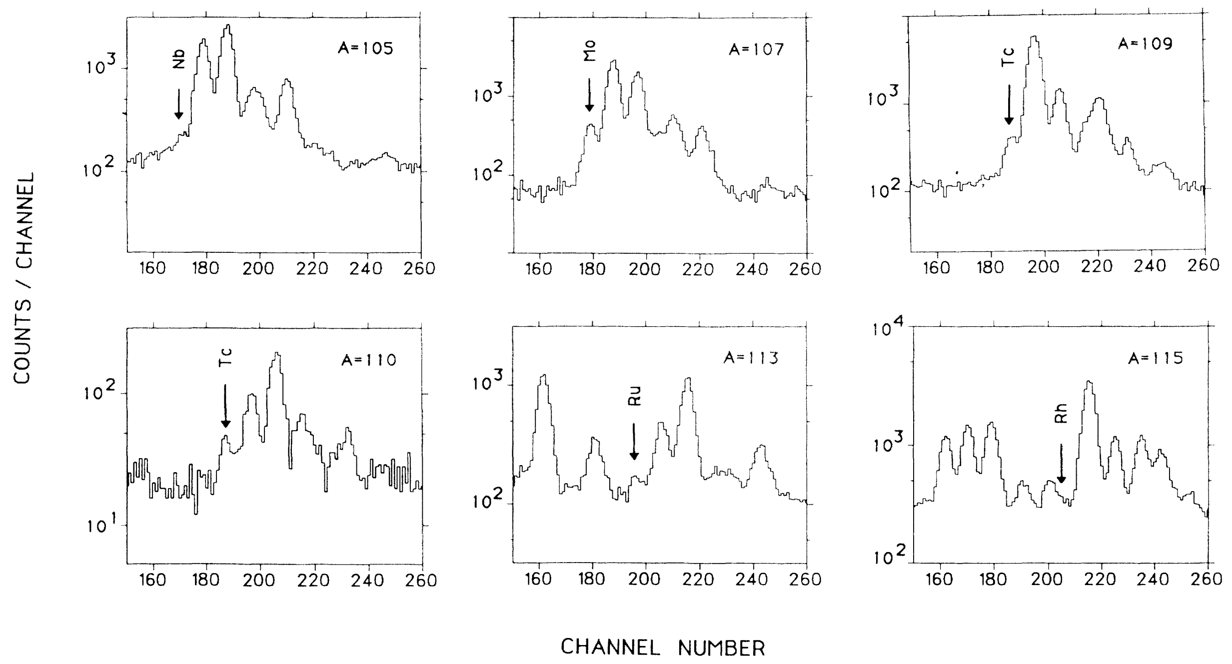

FIG. 1. Spectra of low-energy photons in coincidence with β particles as observed in the focal plane of the IGISOL system. The energy gain was 0.1 keV channel. The arrows for the elements denote the $K\alpha$ x-ray energy of the daughter nucleus following the β decay of a new isotope.

cle. The data were stored in the list mode with the time of occurrence associated with each event.

The arrows in Fig. 1 mark the positions of the expected characteristic $K\alpha$ peaks of the daughters of the new isotopes. The data show clearly $K\alpha$ peaks associated with β decay of the new isotopes ^{107}Nb, ^{109}Mo, ^{110}Mo, and ^{113}Tc. Only a slight indication of ^{105}Zr is seen in the $A=105$ spectrum. However, at this mass number we detected a 127.9-keV γ transition in coincidence with β particles giving further evidence for ^{105}Zr; a γ ray with the same energy was recently found to deexcite the first excited state of ^{105}Nb in spontaneous fission of ^{248}Cm [6]. Spectra for $A=113$ and 115 show also the characteristic $K\alpha$ and $K\beta$ x rays of $A=97$ and 99 fission products produced as oxides. It is evident that in the case of $A=115$ the $K\beta$ line of ^{99}Mo, the decay product of ^{99}Nb, prevents us from seeing distinctly the $K\alpha$ line of Rh.

As an example, the time distribution of the β-gated ^{107}Mo $K\alpha$ x rays is shown in the top part of Fig. 2. In all cases the half-lives were obtained from the combined growth and decay curves by using a least-squares fitting procedure on a single-component growth and decay. Analyses based on the β-gated K x rays could not be made for ^{105}Zr and ^{115}Ru due to poor statistics. Only an approximate half-life of ~ 1 s could be extracted for ^{105}Zr from the ratio of the intensities measured during the growth and decay periods. In case of $A=115$ a 292.8-keV γ ray was observed in coincidence with both $K\alpha$ x rays of Rh and β particles. The β-gated γ spectrum

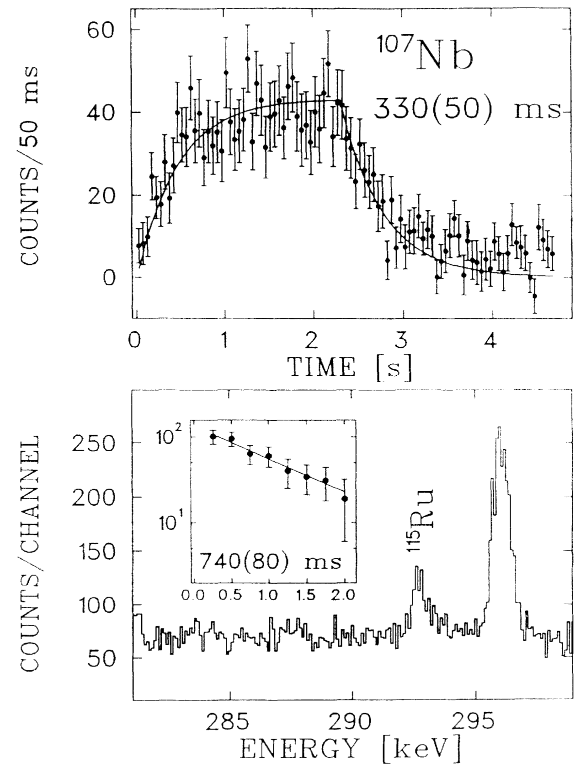

FIG. 2. Top: Time distribution of β-gated $K\alpha$ x rays of ^{107}Mo, the daughter of ^{107}Nb. Bottom: β-gated γ-ray spectrum for $A=115$ activities. Inset: The decay of the 292.8-keV peak due to the β decay of ^{115}Ru.

for $A = 115$ is shown in the bottom part of Fig. 2 with the inset showing the decay of the 292.8-keV peak. The summary of the results for the new isotopes observed in this work is given in Table I.

Our newest half-life data, consisting of 24 n-rich nuclides from zirconium to palladium, provide a stringent test for model calculations involving β decay. A β half-life value depends on decay energy, decay strength, and its distribution. To obtain the strength we have performed calculations using the deformed QRPA model of Möller and Randrup [3]. In this particular calculation the Nilsson wave functions and single-particle energies were used as a starting point. Additionally, the BCS pairing with a global gap energy of $12/\sqrt{A}$ was used. A residual Gamow-Teller (GT) interaction is treated in the random-phase approximation (RPA) with a coupling constant $\chi_{\mathrm{GT}} = 23/A$ MeV. No "universal" GT quenching factor is included in the final half-life value. The deformation for the half-life calculation is obtained from the mass models of Möller and Nix [7], von Groote *et al.* [8], or Hilf *et al.* [9]. The same models were also used to calculate the corresponding Q_β values. Figure 3 shows a comparison between the theoretical and experimental half-lives for 24 β^- decays from Zr to Pd when using the Q_β values from the mass model of Möller and Nix. The half-life value of 0.5 ± 0.1 s for ^{120}Pd is also included here [10]. The experimental values not explicitly given in this Letter can be found from references in Refs. [2] and [5]. Although the scattering of the values is large we can deduce from the average value of 2.1 for the ratio of the theoretical to experimental half-lives that the Möller-Nix model is clearly better than the other two models; they both show a ratio of about 10. This is clearly due to the underestimation of the Q_β values leading to an increase in the half-lives.

There are two additionally interesting general features of Fig. 3 which need further discussion. First, for a given Z the scattering of the $T_{1/2}$ ratios is relatively small, except for Pd isotopes. When going from $^{117}\mathrm{Pd}_{71}$ to $^{119}\mathrm{Pd}_{73}$, for the predicted oblate deformation, the change of the neutron valence orbital allows a $\nu[501\,\frac{3}{2}]$-$\pi[330\,\frac{1}{2}]$ GT ground-state transition, which results in a "short" model $T_{1/2}$. Only a very small change in deformation will place the valence proton into the $\pi[402\,\frac{3}{2}]$ orbital, which makes the ground-state transition first-forbidden and results in a "longer" model $T_{1/2}$. Second, there is a distinct difference in the model $T_{1/2}$ of even- and odd-Z isotopes. Whereas for odd Z, the predicted half-lives are too short (except for ^{107}Nb), they are too long in the case of even Z. Apart from the Q_β effect for the Ru isotopes discussed below, this can be attributed to the excitation energies of the lowest 1^+ states of the $\nu g_{7/2}$ and $\pi g_{9/2}$ parentage in the odd-odd daughter nuclei populated via the allowed beta decay. These 1^+ states are calculated in the RPA model to be about 500 keV too high. This has already been observed earlier, e.g., in the case of ^{96}Y, and can also be seen from a comparison with the experimental level schemes of ^{110,112,114}Rh [11,12]. The reason lies in the known enhanced neutron-proton residual interaction between the $\nu g_{7/2}$-$\pi g_{9/2}$ partner orbitals, which is not taken into account in the present version of the QRPA model. If, however, one wants to compensate for this effect, one can simply add 500 keV to the β decay energies, which reduces the $T_{1/2}$ ratios for even Z nuclei by about a factor of 2.

The region under study presents a challenge for the nuclear structure calculations due to the varying nuclear shapes and coexistence [13]. The Möller-Nix model predicts oblate deformation for Tc, Ru, Rh, and Pd ground states with $\varepsilon_2 \approx -0.25$ and $\varepsilon_4 \approx 0.08$, but prolate deformation for Zr and Nb. The other two mass models [8,9] assume prolate deformations for all nuclei studied in this work. Recent analyses of the β decays of ^{110}Ru, ^{112}Ru [11], and ^{114}Ru [12] with the macroscopic model [14]

TABLE I. New isotopes identified at IGISOL facility from proton-induced fission of ^{238}U.

Nuclide	$T_{1/2}$ (ms)	Method of identification
^{105}Zr	1000^{+1200}_{-400}	K x ray and γ_{128} with β
^{107}Nb	330 ± 50	K x ray with β
^{109}Mo	530 ± 60	K x ray with β
^{110}Mo	250 ± 100	K x ray with β
^{113}Tc	130 ± 50	K x ray with β
^{115}Ru	740 ± 80	γ_{293} with β and K x ray

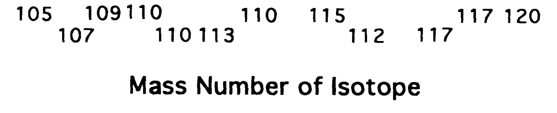

FIG. 3. The ratio of the theoretical and the experimental half-lives for all the new isotopes observed at the IGISOL facility. Theoretical calculation is based on the deformed QRPA approach of Möller and Randrup [3]. The Q_β values are calculated using the model of Möller and Nix [7]. The lower values for even Ru isotopes are obtained when using the experimental values for Q_β [11,12].

also suggest oblate deformation for these nuclei. The only cases in Fig. 3 whose experimental Q_β values are known are ^{110,112,114}Ru. These values are about 1 MeV higher than those predicted by Möller and Nix. Taking this into account reduces the $T_{1/2}$ ratios by a factor of about 2 and leads to considerably better agreement between theory and experiment. Based on the presented data, it has become obvious that the main problem in β half-life predictions lies in the uncertainty of binding-energy calculations and is not necessarily solved by the renormalization of the coupling constant χ_{GT} for different Z, as was done by Staudt *et al.* [15].

Summarizing, we conclude that it is important to continue to measure decay energies (Q_β) and deduce more accurate neutron binding energies (B_n) and deformations to remove uncertainties in half-life predictions of medium-mass neutron-rich nuclei. The role of these measurements may become important in explaining, for example, the severe underproduction of $A \approx 110-120$ elements in the recent steady-flow *r*-process calculations of Thieleman *et al.* [16].

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