β-Decay Half-Lives of Very Neutron-Rich Kr to Tc Isotopes on the Boundary of the r-Process Path: An Indication of Fast r-Matter Flow

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The β -decay half-lives of 38 neutron-rich isotopes from $_{36}$ Kr to $_{43}$ Tc have been measured; the half-lives of 100 Kr, $^{103-105}$ Sr, $^{106-108}$ Y, $^{108-110}$ Zr, 111,112 Nb, $^{112-115}$ Mo, and 116,117 Tc are reported here. The results when compared with previous standard models indicate an overestimation in the predicted half-lives by a factor of 2 or more in the $A \approx 110$ region. A revised model based on the second generation gross theory of β decay better predicts the measured half-lives and suggests a more rapid flow of the rapid neutron-capture process (r-matter flow) through this region than previously predicted.

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About half of the elements heavier than Fe are thought to be produced in a rapid neutron-capture process (*r*-process) nucleosynthesis, a sequence of neutron-capture and β -decay processes. Although the astronomical site and the mechanism of the r process are not yet fully understood, it is generally agreed that the process must occur in environments with extreme neutron densities. The study of the elemental distribution along the r-process path requires sensitive β -decay related information such as β -decay half-lives, β -delayed neutron-emission probabilities, and nuclear masses. In particular, determination of the time scale that governs matter flow from the r-process "seeds" to the heavy nuclei, as well as the distribution in the r-process peaks, depends sensitively on decay halflives [1,2].

Isotopes with extreme neutron-to-proton ratios in the mass region A = 110-125 have attracted special attention

since theoretical r-process yields are found to underestimate isotopic abundances observed in the predicted global abundances by an order of magnitude or more [1,3,4]. This discrepancy has been investigated using numerous mass formulas that differ mainly in the strength of the nuclear shell closures [5,6]. The results indicate that considerable improvements in the global abundances of the isotopes can be achieved by assuming a quenching of the N = 82 shell gap. The properties of most of these crucial r-process nuclei are, however, currently unknown due to their extremely low production yields in the laboratory.

A number of experimental studies on nuclei around neutron-rich krypton to technetium have been performed to investigate the region of the *r*-process path near N = 82[7-9]. In the current work, we report on a first systematic study of the β -decay properties of very exotic, neutron-rich $_{36}$ Kr to $_{43}$ Tc nuclides that contribute to the *r* process.



FIG. 1 (color online). Particle-identification plot from 8 h of accumulated beam time. The lowest-mass isotope from each isotopic chain was tagged for reference purposes. Isotopes with previously known half-lives lie to the left of the black solid line. The shaded area represents the *r*-process waiting points predicted by the ETFSI-Q mass model, within the classical *r*-process model [19].

Decay spectroscopy of very neutron-rich nuclei around A = 110 was performed at the recently commissioned RIBF facility at RIKEN. A secondary beam, composed of a cocktail of neutron-rich nuclei, was produced by inflight fission of a 345-MeV/nucleon ²³⁸U beam in a 550-mg/cm^2 Be target. The primary beam was produced by the RIKEN cyclotron accelerator complex with a typical intensity ~ 0.3 p nA at the production target position. Fragments were separated by the first stage of the BigRIPS separator [10] using a 1600-mg/cm^2 Al achromatic energy degrader. Contaminants that were not fully stripped were removed from the secondary beam using a 570-mg/cm² Al degrader placed at the second stage. Nuclei in the secondary beam were identified on an event-by-event basis measuring their A/Q and Z (see Fig. 1); these two quantities were deduced by combining the projectile time-of-flight and magnetic rigidity from BigRIPS, and from an energyloss measurement in an ionization chamber at the end of the zero-degree spectrometer (ZDS), respectively.

The nuclei transported through the ZDS were implanted in a nine-layer double-sided silicon-strip detector (DSSSD) system [11,12] with a combined thickness of 9×1 mm, which was positioned downstream from a stack of Al degraders of total thickness 6 mm. Each DSSSD was segmented into 16 strips, horizontally on the front side and vertically on the back, each with a width of 3 mm. All strips of the middle 7 were readout for reconstruction of projectile trajectories and timing of low-energy β particles using high-gain readout electronics with an energy range spanning to 2 MeV. The implantation position and



FIG. 2. Fitted decay curves for (a) ¹¹¹Nb, (b) ¹⁰⁸Zr, (c) ¹⁰⁶Y, and (d) ¹⁰³Sr. The data are presented with variable bin widths ranging from 10 to 5000 ms, with normalized counts per 10 ms. The dashed lines correspond to contributions from the parent (the shaded region), daughter, and granddaughter with mix of the β and βn branches, and background components.

timing of the identified nuclei were reconstructed, eventby-event, using their momenta and the energy deposited in each strip of the second DSSSD. Because of the relatively large energy deposition of individual nuclei in the second DSSSD, a low-gain readout system with a dynamical energy range above 4 GeV was used in parallel to the one with high gain.

The positions of an implanted nucleus and its associated β decay were deduced by calculating the average strip position on each side of the relevant DSSSD, weighted by the amplitude of the energy signal. The ion implantation rate throughout the 8 h of beam time was about 8 particles per second. A decay curve was constructed for each nuclide by measuring the correlation between heavy-ion implantations and subsequent β particles detected within 3.3 mm of each other, and in the same DSSSD layer. The maximum-likelihood analysis technique was used to extract β -decay half-lives ($T_{1/2}$) from the decay spectra; bin widths of 10 ms were adopted in all cases [9]. All necessary components were taken into consideration during the fitting procedures: the β -detection efficiency, background rate, daughter and granddaughter half-lives, including the nuclides populated by β -delayed neutron emission, and β -delayed emission branches (P_n). Experimentally unknown P_n values for some of the very exotic isotopes were taken from Ref. [1] with uncertainties of 0% to 100% for caution. The detection efficiency was found to range from 40% to 80%, depending on whether the ions were implanted near the surface or the center of a silicon layer. The typical rate of β -like background events was less than ~0.5 counts/s. Systematic errors on P_n and experimental half-lives were estimated and combined with statistical uncertainties. Figures 2(a)-2(d) show

Nuclide	$T_{1/2}^{\exp}$	$T_{1/2}^{ m lit}$	Nuclide	$T_{1/2}^{\exp}$	$T_{1/2}^{\text{lit}}$
⁹⁷ Kr	60^{+6}_{-5}	63 ± 4 [13]	¹⁰⁶ Y	62^{+25}_{-14}	
⁹⁸ Kr	42 ± 4	46 ± 8 [13]	^{107}Y	41^{+15}_{-9}	
⁹⁹ Kr	13^{+34}_{-6}	40 ± 11 [13]	^{108}Y	25^{+66}_{-10}	
¹⁰⁰ Kr	7^{+11}_{-3}		¹⁰⁶ Zr	186^{+11}_{-10}	260^{+35}_{-30} [9]
⁹⁷ Rb	168_{-53}^{+144}	169.9 ± 0.7 [13]	¹⁰⁷ Zr	138 ± 4	150^{+40}_{-30} [9]
⁹⁸ Rb	102 ± 4	114 ± 5 [13]	¹⁰⁸ Zr	73 ± 4	
⁹⁸ Rb*		96 ± 3 [13]	¹⁰⁹ Zr	63^{+38}_{-17}	
⁹⁹ Rb	54.2 ± 1.3	50.3 ± 0.7 [13]	¹¹⁰ Zr	37^{+17}_{-9}	
¹⁰⁰ Rb	48 ± 3	51 ± 8 [13]	¹⁰⁹ Nb	100_{-8}^{+9}	130 ± 20 [9]
¹⁰¹ Rb	31^{+5}_{-4}	32 ± 5 [13]	¹¹⁰ Nb	86 ± 6	170 ± 20 [7]
¹⁰² Rb	35^{+15}_{-8}	37 ± 3 [13]	¹¹¹ Nb	51^{+6}_{-5}	• • •
¹⁰⁰ Sr	181^{+16}_{-13}	202 ± 3 [13]	¹¹² Nb	33^{+9}_{-6}	• • •
¹⁰¹ Sr	113 ± 2	118 ± 3 [13]	¹¹² Mo	120^{+13}_{-11}	
¹⁰² Sr	85 ± 15	69 ± 6 [13]	¹¹³ Mo	78^{+6}_{-5}	• • •
¹⁰³ Sr	68^{+48}_{-20}		¹¹⁴ Mo	60_{-9}^{+13}	
¹⁰⁴ Sr	43_{-7}^{+9}		¹¹⁵ Mo	51_{-19}^{+79}	• • •
¹⁰⁵ Sr	40^{+36}_{-13}		¹¹⁵ Tc	83_{-13}^{+20}	73^{+32}_{-22} [13]
¹⁰³ Y	234_{-15}^{+18}	230 ± 20 [7]	¹¹⁶ Tc	56^{+15}_{-10}	• • •
¹⁰⁴ Y	197 ± 4	180 ± 60 [13]	¹¹⁷ Tc	89_{-30}^{+95}	
¹⁰⁵ Y	83^{+5}_{-4}	160^{+85}_{-60} [9]		50	

TABLE I. β -decay half-lives for the nuclides identified in in-flight fission of ²³⁸U in the present work ($T_{1/2}^{exp}$). Previous experimental results ($T_{1/2}^{lit}$) are given for comparison. ⁹⁸Rb^{*} corresponds to the higher spin isomers. All times are given in milliseconds.

examples of β -decay curves for ¹¹¹Nb, ¹⁰⁸Zr, ¹⁰⁶Y, and ¹⁰³Sr, respectively, by taking the time correlation of the implanted nuclei with all associated decay events from 10 to 5000 ms after the implantation.

The results obtained in the present work, $T_{1/2}^{exp}$, are listed in the second column of Table I. The half-lives of 18 of the most neutron-rich nuclei (100 Kr, $^{103-105}$ Sr, $^{106-108}$ Y, $^{108-110}$ Zr, 111,112 Nb, $^{112-115}$ Mo, and 116,117 Tc) are reported here. In addition, significant improvements have been achieved in the accuracy of the measurements for 104,105 Y, 106,107 Zr, and 109,110 Nb. Results given for 102 Sr and 110 Nb, which differ significantly from previous measurements, include systematic uncertainties from β and β -delayed γ -ray analysis [14]. The systematic half-life data reported here should provide a stringent test for model calculations.

The results of the present work are compared to two theoretical calculations that are used to model the astrophysical *r* process [15]. The first is the finite-range droplet mass model with the deformed quasiparticle randomphase-approximation (FRDM + QRPA) [16] as well as the new version with the first-forbidden part of β decay taken [1]. The second is KTUY + GT2, i.e., the KTUY mass formula [17] combined with the second generation of the gross theory of β decay (GT2) without bottom raising [18]. The KTUY mass formula is composed of two parts, one representing the general trend of the masses and the other representing the deviations from these trends. The GT2 describes the general behavior of the β -strength functions in a statistical manner where the deformation and shell effects are taken into account through Q_{β} values which are input data for the model.

The half-lives of the Kr to Tc isotopes are displayed as a function of neutron number and compared to the predictions of the two models discussed above in Figs. 3(a)-3(h). The underestimation of $T_{1/2}$ for the N = 65 isotones by the FRDM + QRPA predictions, which have been reported as being due to the domination of low-lying Gamow-Teller strength in the case of Zr and Nb [7], is also observed here for Rb, Sr, and Y. The KTUY + GT2 model overpredicts the $T_{1/2}$ values for Mo and Tc below N = 70, but generally reproduces the experimental data more accurately than the FRDM + QRPA calculation does for the other isotopic chains.

The heavier neutron-rich ${}_{36}$ Kr to ${}_{42}$ Mo isotopes measured in this work are proposed as waiting-point nuclei by some models of the *r* process [8,9,19]. Figure 4(a) shows the ratio of $T_{1/2}$ predicted by the FRDM + QRPA calculation to those deduced in the present work, which lie at the boundary of the *r*-process path. The systematic deviation of the FRDM + QRPA model, which generally underpredicts the half-lives of the odd-*Z* nuclides and overpredicts those of the even-*Z* nuclides, seems to improve with increasing neutron richness. This is not the case for the *r*-process nuclei with $A \approx 110$, where a systematic overprediction of the half-lives is seen by factors of about 2 and 3 or more for the ${}_{40}$ Zr and ${}_{41}$ Nb isotopes, respectively. The mass number dependence of the half-lives according to the



FIG. 3 (color online). Neutron number dependence of β -decay half-lives for (top) even-Z (a) Kr, (b) Sr, (c) Zr, and (d) Mo, and (bottom) odd-Z (e) Rb, (f) Y, (g) Nb, and (h) Tc. Filled circles and open triangles represent results from the present work and previous studies, respectively. The respective solid and dotted lines are predictions from the FRDM + QRPA models, while the dashed lines are from the KTUY + GT2.

KTUY + GT2 model is shown in Fig. 4(b). It is noted that the results display very little or even no systematic dependence, and generally provide a better description of the data across this mass region than the FRDM + QRPA model does. Below A = 102, the KTUY + GT2 calculation overestimates some of the experimental results by a factor of about 2; however, it should be noted that the magnitude of the experimental uncertainties of the halflife for Kr isotopes is rather large. Figure 4(c) shows test results of the FRDM + GT2 model, rather than FRDM + ORPA, to extract differences in the treatment of the β -strength functions. Much smaller deviations, predicted by the FRDM + GT2 model, suggest that the GT2 succeeds in capturing the essence of β -strength functions. Figure 4(d) shows the difference between Q_{β}^{FRDM} and $Q_{\beta}^{\rm KTUY}$ as a function of atomic number. A suppressed odd-even staggering is clearly evident, but the FRDM model predicts a Q_{β} value of about 1 MeV less than that of the KTUY model at $A \approx 110$. A small enhancement in the FRDM + GT2 predictions, by a factor of 2 or so around A = 110, may be explained by the underestimation of Q_{β} values in the FRDM calculation. The data suggest that one of the main problems associated with β -decay half-life predictions is related to uncertainties involved with binding-energy calculations and β -strength functions.

As discussed by Möller *et al.* [1], the sum of the halflives of the *r*-process nuclei up to the midmass region, i.e., around A = 130, determines the rate of *r*-matter flow at



FIG. 4 (color online). Mass number dependence of the ratio of theoretical $T_{1/2}$ values from (a) FRDM + QRPA [16], (b) KTUY + GT2 [17,18], and (c) FRDM + GT2, to the experimental values deduced in the present work. (d) The difference between Q_{β} values predicted by the FRDM and KTUY mass formulas.

N = 82. Following this prescription, the relatively short half-lives of the Zr and Nb isotopes deduced in the present study suggest a further speeding up of the classical *r* process, and shed light on the issue concerning the low production rates of elements beyond the second *r*-process peak. The results presented here also make an impact on the abundances of nuclei at the second peak, since the peak position and shape in the solar abundances around A = 110-140 can be reproduced better by decreasing the half-life of the *r*-process nuclei by a factor of 2 to 3 [2].

In summary, the β -decay half-lives of the very neutronrich nuclides ${}^{97-100}$ Kr, ${}^{97-102}$ Rb, ${}^{100-105}$ Sr, ${}^{103-108}$ Y, ${}^{106-110}$ Zr, ${}^{109-112}$ Nb, ${}^{112-115}$ Mo, and ${}^{115-117}$ Tc, all of which lie close to the astrophysical *r*-process path, have been measured (for 18 nuclei) or their uncertainties have been reduced significantly. The results suggest a systematic enhancement of the β -decay rates of the Zr and Nb isotopes by a factor of 2 or more around A = 110 with respect to the predictions of the FRDM + QRPA model. The results also indicate a shorter time scale for matter flow from the *r*-process seeds to the heavy nuclei. More satisfactory predictions of the half-lives from the KTUY + GT2 model, which employs larger Q_{β} values, highlights the importance of measuring the half-lives and masses of very exotic nuclei, since such knowledge ultimately leads to a decrease in the uncertainty of predicted nuclear abundances around the second *r*-process peak.

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