Identification of Four New Neutron-Rich Rare-Earth Isotopes

H. Mach
Brookhaven National Laboratory, Upton, New York 11973, and Clark University, Worcester, Massachusetts 01610, and University of Maryland, College Park, Maryland 20742

and

A. Piotrowski, R. L. Gill, R. F. Casten, and D. D. Warner
Brookhaven National Laboratory, Upton, New York 11973
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Four new neutron-rich, fission-product nuclei have been identified at the on-line mass separator TRISTAN at Brookhaven National Laboratory. Their half-lives have been measured to be, for ^{156}Pm, $T_{1/2} = 28.2 \pm 1.1$ sec; for ^{159}Sm, $T_{1/2} = 15 \pm 2$ sec; for ^{160}Sm, $T_{1/2} = 8.7 \pm 1.4$ sec; and for ^{161}Eu, $T_{1/2} = 27 \pm 3$ sec. These nuclei are of particular importance in nucleosynthesis calculations, since they lie in the region of the rare-earth maximum in *r*-process abundances. A comparison of these and other new half-lives with recent calculations discloses a characteristic pattern of discrepancies.

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Information on nuclear-decay half-lives has important applications to astrophysical problems as well as to the field of nuclear structure and technology. Recently Mathews[1] has reviewed the critical data needs for the calculation of nucleosynthesis in astrophysical environments. He has singled out neutron binding energies and nuclear half-lives as the most important parameters to be determined experimentally. The situation is particularly critical in the case of rapid neutron capture in explosive stellar environments, where even the site and general scenarios involved remain uncertain. The most crucial nuclei are those near the abundance peaks in the *r* process at mass numbers 80, 130, 165, and 195.

There exist only two sets of half-life estimates for a large number of neutron-rich nuclei: the work of Takahashi, Yamada, and Kondoch[2] using a simple model and the more accurate calculations by Klapdor, Metzinger, and Oda,[3] which incorporate a detailed treatment of β strength functions. In the few years since the calculations of Klapdor, Metzinger, and Oda,[3] a number of new neutron-rich nuclei have been found and a systematic pattern of discrepancies between the experimental and predicted values appears to be emerging. In general, it has been shown[4] that the calculations tend to overestimate the experimental half-lives in the $A \approx 70$ region and underestimate them for $A \approx 190$. Since deviations from the calculated half-lives range over more than an order of magnitude, there remain large uncertainties in the input data for different astrophysical models. In order to gain a better understanding of the systematics of these discrepancies, and to provide new data for nucleosynthesis calculations, we have identified a number of new neutron-rich nuclei in the $A \approx 160$ mass region: ^{156}Pm, ^{159}Sm, ^{160}Sm, and ^{161}Eu.

The neutron-rich rare-earth nuclei are difficult to study by on-line mass-separation techniques because of the low fission yield and relatively long diffusion times from the target material. Consequently, fast chemical-separation techniques or other exotic techniques are normally applied, which are limited to the close vicinity of the stability line. As demonstrated by Piotrowski, Gill, and MacDonald,[5] a thermal ion source recently developed at TRISTAN has achieved very high temperatures ($\approx 2500\,°\mathrm{C}$) and faster fission-product diffusion, making it possible to extract on-line short-lived rare-earth nuclei with useful yields. The results presented here were obtained in a survey of the mass range $156 \leq A \leq 164$ by use of a modified version of this ion source which included several improvements to reduce further the fission-product diffusion time.

The measurements were performed at the on-line mass separator TRISTAN at Brookhaven National Laboratory.[6] The rare-earth activity was produced by thermal-neutron fission of about 5 g of enriched ^{235}U impregnated on graphite cloth disks. The radioactive elements were ionized by the thermal ion source and extracted to form an ion beam. After mass separation by a 90° magnet, the radioactive ion beam of the selected isobar was deposited onto a movable aluminized Mylar tape. After a predetermined period of time the accumulated source was transported within ≈ 0.3 sec to a counting station, which was comprised of a small planar Ge, a large Ge, and a large Ge(Li) detector. Gamma-ray singles, γ-multispectra scaling (GMS), and γ-γ-t coincidences involving any two of the detectors were simultaneously accumulated. The multiscaling cycle was started with the arrival of the source at the counting station and lasted over a period of time ΔT selected for a particular isobar. During this time 32 consecutive spectra were collected. The decay curves required no dead-time corrections except for

TABLE I. Observed and predicted half-lives for the new neutron-rich rare-earth isotopes.

Nuclide and Decay	Experimental: This Work (sec)	Experimental: Previous Measurements (sec)	Theoretical: Takahashi et al.[2] (sec)	Theoretical: Klapdor et al.[3] (sec)	$R = \frac{T_{1/2}(\exp)}{T_{1/2}(\mathrm{calc})}$ [a]
^{152}Pr→Nd		3.2 ± .2 (Ref. 7)	14	3.93	.81 ± .05
^{155}Pm→Sm		48 ± 4 (Ref. 8)	56	21.2	2.26 ± .19
^{156}Pm→Sm	28.2 ± 1.1		27	8.39	3.36 ± .13
^{157}Sm→Eu	402 ± 24	480 ± 30 (Ref. 9) 480 ± 60 (Ref. 10)			
^{158}Sm→Eu	312 ± 12	331 ± 5 (Ref. 11)			
^{159}Sm→Eu	15 ± 2		50	10.1	1.49 ± .20
^{160}Sm→Eu	8.7 ± 1.4		83	6.06	1.44 ± .23
^{160}Eu→Gd	31 ± 4	50 ± 10 (Ref. 10) 53 ± 10 (Ref. 12) 41 ± 4 (Ref. 13)			
^{161}Eu→Gd	27 ± 3		56	24.8	1.09 ± .12
^{162}Eu→Gd	≈6 [b]		20	9.35	
^{163}Gd→Tb		68 ± 3 (Ref. 14)	126	66.3	1.02 ± .05
^{165}Tb→Dy		127 ± 6 (Ref. 15)	158	146	.87 ± .04
^{168}Dy→Ho		510 ± 30 (Ref. 16)	2238	132	3.86 ± .23

[a]Calculations of Klapdor, Metzinger, and Oda (Ref. 3) used in the ratio.
[b]Tentative assignment.

the case of ^{157,158}Sm isotopes, for which the γ rays from long-lived isotopes were used to extract the normalization parameters.

All of the observed transitions in the energy range 20–1500 keV were carefully analyzed and successfully identified from a combination of E_γ, I_γ, GMS-decay, and coincidence relationships. All short-lived impurity lines were exclusively those of barium and their decay products, which were 17 and 19 mass units lighter than the particular rare-earth isobar under study. These ions were part of exotic molecular beams formed at high temperature in the ion source. The decay of Ba isotopes and their decay chains are well known, and have provided in-beam energy and intensity calibrations.

A characteristic feature of the rare-earth decay spectra is the presence of strong and highly converted low-energy transitions. Consequently, the new isotopes were positively identified by the presence of a characteristic set of $K\alpha_2$, $K\alpha_1$, $K\beta_1'$, and $K\beta_2'$ x rays. The associated γ lines were identified on the basis of GMS decay curves identical to those of the x rays and on the basis of x-γ and γ-γ coincidences. The half-lives measured for the new isotopes are listed in Table I. These were estimated by an averaging of the values obtained for strong, single transitions in each decay. The half-lives of the x rays were not included in the average because of the possibility of small systematic errors inherent in the analysis of the complex x-ray region. Figure 1 illustrates the GMS data for one transition representative of each new isotope. Each of the nuclei under study presented special problems, which are discussed in the following paragraphs.

^{156}Pm.—Eleven transitions were found associated with the decay of ^{156}Pm on the basis of strong x-γ and γ-γ coincidences. The half-life measurement represents the average of six results involving the 75.7-, 117.8-, and 174.1-keV transitions and two sets of GMS measurements with GMS cycle times of $\Delta T = 48$ and 96 sec.

^{159}Sm.—The decay of ^{159}Sm was initially missed in the survey of the $A = 159$ isobar with the GMS cycle time of $\Delta T = 640$ sec. The off-line analysis of the first bins of the GMS spectra revealed clearly the presence of Eu x rays with a $T_{1/2}$ for $K\alpha_1 + K\alpha_2$ of 19 ± 5 sec and for $K\beta_1'$ of 17 ± 6 sec. Two γ lines have been

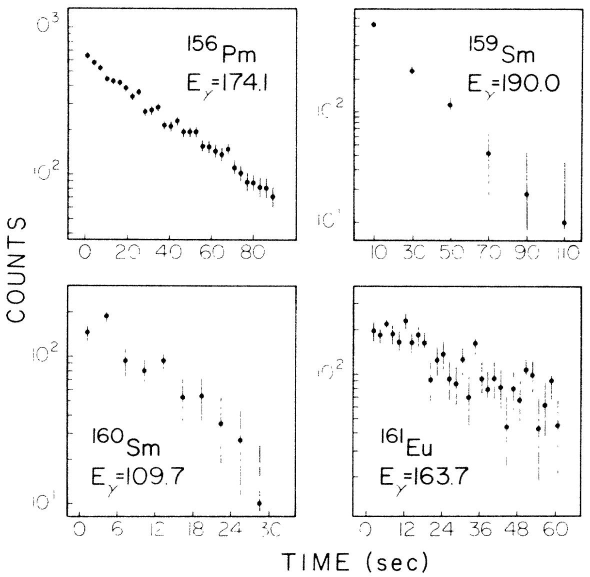

FIG. 1. Example of data representative of each new isotope. E_γ represents the energy in kiloelectronvolts of a transition in the daughter nucleus.

found associated with this decay, 190 ± 0.3 keV ($T_{1/2} = 15 \pm 2$ sec) and a weak transition at 114.3 ± 0.4 keV ($T_{1/2} = 16 \pm 4$ sec), on the basis that these transitions could not be identified with any other decay consistent with their E_γ and $T_{1/2}$ values.[17]

^{160}Sm.—The decay of ^{160}Sm has also been confirmed in the off-line analysis of the first bins of the GMS spectra measured with the cycle time of 96 sec. The half-lives of the Eu x rays were found to be $T_{1/2}(K\alpha_1 + K\alpha_2) = 14 \pm 6$ sec and $T_{1/2}(K\beta_1') = 11 \pm 4$ sec and are probably affected by the close-lying intense Gd x rays. Only one transition at 109.7 ± 0.3 keV ($T_{1/2} = 8.7 \pm 1.4$ sec) was found with a half-life comparable to those of the x rays and in weak coincidence with Eu x rays in the gates involving various detector pairs. Furthermore, no other association could be made for the 109.7-keV transition consistent with its E_γ and $T_{1/2}$ values.[17]

^{161}Eu.—Five transitions of energy 71.9 ± 0.2, 91.9 ± 0.2, 163.7 ± 0.2, 293.9 ± 0.3, and 314.3 ± 0.3 keV were assigned to the decay of ^{161}Eu on the basis of γ-γ and x-γ coincidences and the measured half-lives. Four of them fit immediately into the level scheme of ^{161}Gd as revealed in the ^{160}Gd(d,p) reaction.[18] The $T_{1/2}$ value is the average of the measurements for the 71.9-, 91.9-, and 163.7-keV transitions.

^{162}Eu.—The barium impurity lines and contaminant transitions from a long-lived ^{157}Eu$\rightarrow$^{157}Gd decay made the search difficult. Weak evidence was found for a short-lived component in the europium x rays with a half-life of ≈ 6 sec. The GMS spectrum was measured with $\Delta T = 32$ sec.

No results can be reported for the $A = 163$ and 164 isobars as a result of the presence of contaminants.

We have measured half-lives of three other nuclei, which have been previously investigated by other techniques. Inasmuch as the on-line mass separator provides a different way of accessing these nuclei, it is interesting to note that our results are consistently lower and, in the case of ^{157}Sm and ^{160}Eu, substantially lower, than those reported in the literature (see Table I).

The new half-life measurements in the $A \approx 160$ region are summarized in Table I. A comparison to the theoretical predictions discloses that the model of Klapdor, Metzinger, and Oda[3] generally underestimates, while that of Takahashi, Yamada, and Kondoch[2] systematically overestimates, the measured half-lives. Similar observations have been made[4] for Yb, Lu, Ra, and Ac nuclei.

The model of Klapdor, Metzinger, and Oda was tested[3] on a large number of known nuclei. For those with half-lives less than 1000 sec, the calculations reproduced 58% of the results within a factor of 2, which is similar to the value of 67% for the nuclei of Table I. Thus in the case of new isotopes in the $A \approx 160$ region, the theoretical estimates are found reliable within the expected limits. This is in contrast to the results for a number of new isotopes with $A < 80$ where only 28% of them are reproduced within a factor of 2. Moreover, since the average half-life for the lighter-mass nuclei is only 3 sec, which is much less than for the nuclei of Table I, one would have expected,[3] *a priori,* that the calculations of Ref. 3 would have been more, not less, accurate for them.

In order to assess the calculations[3] in more detail, it is convenient to use ratios of experimental to calculated half-lives. These ratios, denoted R, are listed in column 6 of Table I and disclose that, between ^{156}Pm and ^{165}Tb, R decreases monotonically. Moreover, there are strong and similar discontinuities between ^{152}Pr and ^{155}Pm and between ^{165}Tb and ^{168}Dy. Apparently, there is a sawtooth pattern where R decreases monotonically between turning points ($Z = 59$ and 65) and then jumps suddenly. It would be of interest to search for this pattern in other mass regions.

Since the publication of Ref. 3 about fifty new isotopes have been discovered. These nuclei are scattered unevenly throughout the neutron-rich region from $Z = 6$ to $Z = 89$. However, those near $Z = 28$ provide a particularly extensive group and offer an interesting comparison with the present results. A compilation of the new half-life measurements in the $Z = 28$ region was recently made by Bosch *et al.*,[4] and Fig. 1 of their paper provides a visual presentation of the ratios of experimental to theoretical results.

The sawtooth pattern observed at $A \approx 160$ appears also in the vicinity of the closed shell at $Z = 28$, but it takes even more extreme values. From Cr nuclei, the

R values decrease almost monotonically to reach $R \approx 0.1$ for Co nuclei and then jump at $Z = 28$ to $R \approx 2.5$ for Cu isotopes. These data suggest that the turning points of the sawtooth pattern may be related to shell closures. One may also note that the very large discrepancy between experiment and theory, $R = 0.03$, observed for ^{64}Co ($Z = 27$) was considered[3] as an isolated spurious effect. Yet this result and recent results for other new Co isotopes would be consistent with a deep minimum in the sawtooth pattern observed just below $Z = 28$.

In this context, the turning points for the $A = 160$ region remain unexplained. For spherical nuclei, there is a small subshell gap due to the $g_{7/2}$ shell closure at $Z = 58$ and a larger one due to the $d_{5/2}$ shell closure at $Z = 64$. Although the turning points at $Z = 59$ and 65 appear close to the values of $Z = 58$ and 64 mentioned above, the deformation typical for the $A \approx 160$ nuclei would be expected to wash out any shell effects.

In conclusion, the new half-life measurements for the $A \approx 160$ region are in rather good agreement with the calculations of Ref. 3. Nevertheless, the discrepancies that do exist display an interesting pattern. If investigated further, the systematics of the discrepancies could lead to the nuclear processes missed in the model calculations and indirectly could contribute to an improvement in the half-life estimates. There is a pressing need to provide a more realistic assessment of the calculations, particularly in the vicinity of closed-shell nuclei. Already for the $A < 80$ nuclei the discrepancies are higher than expected. Moreover, there seems to be a suggestion that the discrepancies take extremal values in the vicinity of closed-shell nuclei. At present it is difficult to assess the impact of those observations on the calculation of nucleosynthesis. However, the above results and discussions strengthen Mathews's call[1] for more experimental work in the vicinity of closed-shell nuclei.

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