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Identification of new neutron-rich rare-earth isotopes produced in ^{252}Cf fission

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New neutron-rich isotopes of several rare-earth elements have been identified using on-line mass separation. The observed decay rates of K x rays in the mass separated fractions for which $153 \leq A \leq 162$, together with the associated γ-decay spectra, provided direct isotopic identifications. New isotopes observed, with values of their half-lives given in parentheses, are ^{153}Pr (4.3 ± 0.2 s), ^{155}Nd (8.9 ± 0.2 s), ^{156}Nd (5.47 ± 0.11 s), ^{157}Pm (10.90 ± 0.20 s), ^{158}Pm (4.8 ± 0.5 s), and ^{162}Eu (10.6 ± 1.0 s). In addition, we have confirmed recent reports of the observation of ^{153}Nd (28.9 ± 0.4 s), ^{154}Nd (25.9 ± 0.2 s), ^{156}Pm (26.70 ± 0.10 s), ^{159}Sm (11.37 ± 0.15 s), and ^{160}Sm (9.6 ± 0.3 s). These half-life values are compared to the results of theoretical predictions.

In this Rapid Communication, we report on the identification of several new neutron-rich rare-earth isotopes in the mass region between $A = 153$ and 162. At the present time, information on the decay properties of neutron-rich rare-earth nuclides, more than two to three units of Z off the line of beta stability (along lines of constant A) is sparse. This results from the fact that such nuclides cannot be produced in conventional nuclear reactions [e.g., (n,p), (n,α), (n,2p), (p,3p), etc.] and that their yields in thermal neutron fission of ^{235}U are low. Furthermore, until the recent development of high-temperature thermal ion sources, coupled on-line to mass separators, suitably rapid separation techniques were not available. (The lower limit on detectable half-lives achieved using a fast radiochemical separation technique employing high performance liquid chromatography was > 30 s, e.g., see Refs. 1 and 2.)

A unique feature of the isotope separation on line (ISOL) system at the Idaho National Engineering Laboratory (INEL) is its use of spontaneous fission of ^{252}Cf as the source of fission fragments. In spontaneous fission of ^{252}Cf, the fission yields of nuclides with $A > 150$ is significantly enhanced over the corresponding yields from thermal-neutron fission of ^{235}U (e.g., see Refs. 1 and 2). Thus, the INEL ISOL system is particularly well suited for studies of neutron-rich rare-earth nuclides. The results reported in this paper represent the initial ones from a program to systematically study the nuclear properties of such nuclides.

Half-lives, together with the beta-decay energies and the distribution of Gamow-Teller strength, in neutron-rich nuclides such as those reported here are of importance in astrophysical calculations involving the rapid neutron-capture process (r process) (e.g., see Ref. 3). In calculations involving the r process, because of lack of experimental data, it has been necessary to utilize predicted values of the half-lives of the relevant nuclides. Thus, as new data become available, it is desirable to check the validity of these predictions. At the present time, there exist two sets of half-life predictions, an earlier set by Takahashi, Yamada, and Kondoh[4] based on the gross theory of beta decay and a recent set by Klapdor, Metzinger, and Oda[5] based on a microscopic model employing a description of the distribution of beta strength.

In a recent publication by Mach *et al.*,[6] in which four new neutron-rich rare-earth isotopes were reported, the level of agreement between recent data in the mass region from $A = 152$ to $A = 168$ was compared with these model predictions. Their overall conclusion was that the model of Takahashi *et al.*[4] systematically overestimates and the model of Klapdor *et al.*[5] generally underestimates the measured half-lives. Furthermore with respect to the model of Klapdor *et al.*[5] they suggested that the ratio R (experimental-to-predicted half-life) follows a sawtooth pattern where R decreases monotonically between turning points (of the sawteeth) at $Z = 59$ and 65.

The new neutron-rich rare-earth isotopes identified to date using the INEL ISOL system include ^{153}Pr, ^{155,156}Nd, ^{157,158}Pm, and ^{162}Eu, with improved half-life values also being obtained for the recently reported iso-

topes, ^{153,154}Nd, ^{156}Pm, ^{159}Sm, and ^{160}Sm. These new isotopes extend the region of experimentally measured half-lives by roughly one unit of Z (along lines of constant A) and, therefore, present an opportunity to further expand the range over which experiment and model predictions can be compared. In particular, it is of interest to check how well these new isotopes fit into the pattern of the systematics of the discrepancies, between experiment and model predictions, suggested by Mach *et al.*[6]

The source of fission products used for the INEL ISOL system is two $\sim$200-μg electrodeposits of the spontaneously fissioning isotope ^{252}Cf (Ref. 7). These sources, which were electrodeposited at Oak Ridge National Laboratory, are located in a specially designed hot cell, and are mounted inside a pressurized chamber which forms an integral part of the gas-jet transport arrangement coupling the ^{252}Cf sources on line to the ion source of the mass separator. With this arrangement, the ^{252}Cf fission products are transported with high efficiency ($>50\%$) using a NaCl-aerosol-loaded He-gas mixture into the electron-bombardment heated hollow-cathode ion source used in the present series of measurements. A more detailed description of the INEL ISOL system and its operation is given in Ref. 8.

Following mass separation, the selected mass fraction was collected on a movable tape which, after a predetermined collection time, was moved to a detector station where time-multiscaled x ray and γ spectra were recorded. For each isotope studied, the collection-counting cycle was typically chosen to be approximately three times the estimated half-life with three to six time-multiscaled x- and γ-ray spectra being recorded within this cycle time. For each isotope studied, some 140 to 1700 cycles were combined to obtain the final sets of spectra. Also, for each isotope studied, spectra were accumulated using longer cycle times, characteristic of the daughter activities expected in that mass fraction.

Two detectors, located opposite each other, were used for the measurements: a 200-mm^2×10-mm intrinsic planar Ge detector having a 50-μm Be window and energy resolution of 0.55 keV at 40 keV for the x ray and low-energy γ rays, and a 50-cm^3 Ge(Li) detector for the higher-energy γ rays. The former detector viewed the collection tape through a 127-μm Mylar window while the latter detector viewed the collection tape through a 0.67-mm Al window and a 0.48-g/cm^2 Cu plus 0.63-g/cm^2 Sn absorber which was used to reduce the γ-ray summing effects.

With the excellent energy resolution of the x-ray detector, x-ray peaks for adjacent rare-earth elemental fractions were fully resolved. Thus, with this arrangement, unique isotopic identification was obtained from the mass separation and the x-ray identification combined. Furthermore, γ rays having comparable decay characteristics to those of the observed x rays could then generally be given a unique isotopic assignment.

Detailed examination of spectra measured for specific mass fractions clearly indicated that cross contamination from neighboring masses was a negligible problem. We did, however, observe some level of contamination in the higher mass factions (with $A>157$) from molecular beams of LaO and CeO. The identification and extent of the molecular contaminants could, however, be clearly identified from the presence of the Ce and Pr x rays and their associated γ rays.

The adopted half-life values for the new isotopes and those other isotopes measured in this work are given in Table I. These values generally represent an average of the individual values measured for the K x rays and the prominent γ rays which could be associated with each isotope. Specific comments on the data obtained for individual isotopes listed in Table I are as follows:

^{153}Pr. Identification of the ^{153}Pr isotope was accomplished in two separate experiments, with collection-counting cycle times of 12 and 16 s each. The half-life value was obtained from an average of individual values involving the Nd K x rays and the 50.0-, 141.8-, and 191.8-keV γ rays. Separate half-life values from the x

TABLE I. Half-life values measured for the new neutron-rich rare-earth isotopes and their comparison with the results of two theoretical predictions.

	Experimental		Theoretical			
	Present work	Previous work	Takahashi *et al.* (Ref. 4)	Klapdor *et al.* (Ref. 5)	R = (observed/predicted) Takahashi *et al.* (Ref. 4)	Klapdor *et al.* (Ref. 5)
Isotope	(sec)	(sec)	(sec)	(sec)		
^{153}Pr	4.3 ± 0.2		14	2.81	0.31	1.53
^{153}Nd	28.9 ± 0.4	32 ± 4 (Ref. 9)	58		0.50	
^{154}Nd	25.9 ± 0.2	26 ± 2 (Ref. 10)	77		0.34	
^{155}Nd	8.9 ± 0.2		18	15.3	0.49	0.58
^{156}Nd	5.47 ± 0.11		24	8.36	0.23	0.65
^{156}Pm	26.70 ± 0.10	28.2 ± 1.1 (Ref. 6)	25	8.39	1.07	3.18
^{157}Pm	10.90 ± 0.20		22	4.68	0.50	2.33
^{158}Pm	4.8 ± 0.5		10	2.31	0.48	2.08
^{159}Sm	11.37 ± 0.15	15 ± 2 (Ref. 6)	31	10.1	0.37	1.13
^{160}Sm	9.6 ± 0.3	8.7 ± 1.4 (Ref. 6)	67	6.06	0.14	1.58
^{162}Eu	10.6 ± 1.0		18	9.35	0.59	1.13

rays and the γ rays are in excellent agreement, being 4.4 and 4.2 s, respectively.

^{153}Nd. Identification of the ^{153}Nd isotope was first reported by Pinston *et al.*[9] In the present work, however, a total of 48 γ-ray transitions could be associated with this decay, compared with the eight transitions reported earlier. The half-life value was obtained from an average of individual values involving the Pm K x rays and the 32.2-, 105.5-, 345.0-, 418.3-, and 967.1-keV γ rays.

^{154}Nd. In the earliest report of ^{154}Nd, by Buchtela,[10] a half-life value of (40 ± 10) s was proposed with the principal γ-ray components being at 0.4 and 0.7 MeV. However, a recent report by Karlewski *et al.*[11] is in conflict with this, in that they find a half-life value of (26 ± 2) s with no evidence for strong γ-ray peaks near 0.4 and 0.7 MeV. In the present work, we confirm the assignment of Karlewski *et al.*,[11] with some 35 γ rays being assigned to the ^{154}Nd decay. The present half-life value was obtained from an average of individual values involving the Pm K x rays and the 50.7-, 68.0-, 72.3-, 79.4-, 83.7-, 101.0-, 117.5-, 130.0-, 151.7-, 180.7-, 799.5-, 850.2-, and 908.8-keV γ rays. Based on a reexamination of the details of the experimental separation and measurements performed by Buchtela,[10] we conclude that what they observed was actually the decay of ^{155}Pm (see Ref. 1 to compare decay parameters), present as a daughter activity from ^{155}Nd decay in the original Nd chemical fraction.

^{155}Nd. A total of 16 γ rays could be definitely associated with the decay of ^{155}Nd in the present work. The half-life value was obtained as an average of individual values involving the Pm K x rays and the 67.4-, 113.1-, 180.6-, 253.1-, 419.0-, and 955.1-keV γ rays. Separate half-life values for the x rays and γ rays were in excellent agreement, being 8.7 and 9.0 s, respectively.

^{156}Nd. The half-life value for ^{156}Nd was obtained as an average of individual measurements involving the Pm K x rays and the 84.8- and 150.7-keV γ rays, which are the only γ rays which we can definitely associate with this decay at this time. Separate half-life values, however, for the x rays and γ rays are in excellent agreement, being 5.5 and 5.3 s, respectively.

^{156}Pm. In a recent paper, Mach *et al.*[6] reported the identification of ^{156}Pm, with 11 γ-ray transitions being associated with its decay. In the present work, the ^{156}Pm γ-ray spectrum that was obtained was of high statistical quality and proved to be somewhat more complex. The half-life value was obtained as an average of individual values involving the Sm K x rays and the 75.9-, 117.8-, 174.0-, 267.8-, 880.9-, 894.8-, 934.6-, 1148.5-, and 1260.1-keV γ rays.

^{157}Pm. Some 17 γ rays could be associated with the decay of ^{157}Pm in the present work. The half-life value was obtained as an average of individual values involving the Sm K x rays and the 52.6-, 108.2-, 160.5-, and 187.9-keV γ rays. Separate half-life values for the x rays and γ rays were identical.

^{158}Pm. Identification of the ^{158}Pm decay was accomplished in two separate experiments, with collection-counting cycle times of 12 and 16 s, respectively. One γ ray, at 72.7 keV, together with the Sm K x rays could be associated with this activity. The half-life values determined for the x rays and 72.7-keV γ rays were in reasonable agreement, being (4.5 ± 0.5) s and (5.5 ± 0.8) s, respectively.

^{159}Sm. Mach *et al.*[6] reported the identification of ^{159}Sm, with γ rays at 114.3 and 190 keV being associated with its decay. In the present work, some 14 γ rays were definitely associated with the decay of this activity. The half-life value was obtained as an average of individual values involving the Eu K x rays and the 75.4-, 114.4-, 179.2- 190.0-, 254.7-, and 862.0-keV γ rays.

^{160}Sm. Mach *et al.*[6] reported the identification of the ^{160}Sm decay based on their observation of the associated Eu K x rays and a single γ-ray transition at 109.7 keV. In the present work, we were able to associate only five γ-ray transitions with this decay, even though the time-multiscaled γ-ray spectra were obtained with rather good statistics, with in excess of 5000 total counts being accumulated in the dominant Eu K_α and 109.8-keV peaks.

^{162}Eu. Identification of the ^{162}Eu decay was accomplished in two separate experiments, with collection-counting cycle times of 24 and 28 s. For each experiment, the resulting γ-ray spectra were observed to contain a strong contamination from ^{146}La decay, resulting from LaO molecular beam formation in the ion source. Since the ^{146}La component was dominant, and since it had a half-life value comparable to that of ^{162}Eu, only the decay of the Gd K x rays could be used to characterize the ^{162}Eu decay. In their paper, Mach *et al.*[6] had reported only weak evidence for the observation of ^{162}Eu.

Shown in Table I, for comparison with the half-life values measured in the present work are the theoretical predictions of Takahashi *et al.*[4] and Klapdor *et al.*[5] These new half-life values are representative, as a group, of nuclides somewhat further from the line of beta stability than those discussed by Mach *et al.*,[6] but still within the same mass region. It is thus of interest to see whether the observations they made regarding the agreement between observed and predicted values still appear valid. In this regard, their overall conclusion that the model of Takahashi *et al.*[4] systematically overestimates and the model of Klapdor *et al.*[5] generally underestimates the measured half-lives still appears valid. On the average, for the data shown in Table I, the ratios R, between the observed and predicted values, are 0.46 and 1.6 for Takahashi *et al.*[4] and Klapdor *et al.*,[5] respectively.

Although no overall pattern can be discerned in the ratio R calculated using the values of Takahashi *et al.*,[4] Mach *et al.*[6] have suggested that a sawtooth pattern exists in the R values obtained using the Klapdor *et al.*[5] half-life values, with a lower turning point (of the sawtooth pattern) at $Z = 59$. Inspection of the data in Table I for those more neutron-rich nuclides shows less support for this sawtooth pattern, however. First, the new data on ^{153}Pr, with $R = 1.53$, does not support the postulation of a turning point at $Z = 59$. Rather, if such a turning point can be defined, the new Nd data would suggest that it is at $Z = 60$. Second, the concept of monotonically declining R values between turning points is not well supported by the new data. Rather, we note that it is in fact only the Pm ($Z = 61$) data which appear to have abnormally large values of R, with the R values for the $Z = 62$ and 63 nu-

clides being much closer to unity. Further data, especially in the $Z=62$ through 65 region, would help to elucidate whether such a pattern of systematic discrepancies, as proposed by Mach *et al.*,[6] actually does exist in this mass region.

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