Precision γ-ray branching-ratio measurements for long-lived fission products of importance to stockpile stewardship

K. Kolos,[1] A.M. Hennessy,[2] J.A. Clark,[3] J.C. Hardy,[4] V.E. Iacob,[4] G.E. Miller,[2] E. Norman,[5] G. Savard,[3] N.D. Scielzo,[1] A.J. Shaka,[2] M.A. Stoyer,[1] and A.P. Tonchev[1]
[1]*Lawrence Livermore National Laboratory,* [2]*University of California at Irvine,* [3]*Argonne National Laboratory,* [4]*Texas A&M University,* [5]*University of California at Berkeley*

Introduction

One of the most straightforward and reliable ways to determine the number of fissions that occurred in a chain reaction is done via detection of the characteristic γ rays emitted during the β-decay of the fission product. These γ rays are emitted in only a fraction of the decays, and this fraction (the γ-ray branching ratio) must be known accurately to determine the total number of fissions. The fission products ^{95}Zr, ^{144}Ce, and ^{147}Nd (along with other long-lived isotopes) play a crucial role in science-based stockpile stewardship. The γ-ray branching ratio of ^{147}Nd is known to only 8% uncertainty [1], hence leading to an 8% contribution to the uncertainty in fission-chain yield. The ^{144}Ce γ-ray branching-ratio values [2] are dominated by the results of a single measurement [3] performed about 40 years ago and have yet to be confirmed at that precision. Precise values of these absolute γ-ray branching ratios are desired for national-security applications and to greatly improve the precision and reliability with which the number of fissions can be determined.

Although the level schemes of the isotopes of interest are well known, there are several challenges in precisely measuring these γ-ray branching ratios. These isotopes have Q values of less than 1.2 MeV and β-energy spectra that peak near zero energy. The efficiencies of the β detector and γ-ray detector have to be well characterized and impurities and self-attenuation of the low-energy β particle in the sample must be minimized. The approach that has been developed in the past year to determine the γ-ray branching ratios consists of producing radiopure sources using low-energy ion beams from the CARIBU facility at Argonne National Laboratory (ANL) and performing β counting using a custom-made 4π gas proportional counter in coincidence with γ spectroscopy using the precisely-calibrated HPGe detector [4,5] at Texas A&M University.

Previous Work

High-purity samples of ^{95}Zr (50 Bq), ^{144}Ce (360 Bq), and ^{147}Nd (1500 Bq) were collected on thin (40 μg/cm^2) carbon foil backings using low-energy mass-separated beam of A=95, 144, and 147 fission products from CARIBU over several 2-day collection periods. Within a few hours after implantation on the collection foil, all the shorter-lived species β decay to the long-lived species of interest as illustrated for the case of ^{147}Nd in Fig. 1. During collection, a HPGe detector was used to monitor the implantation rate and to provide additional information on the purity of the ion beam by detecting the characteristic γ-rays from the shorter-lived fission products that make up the beam. The produced sources, which each have half-lives of 11 days or longer, were then shipped to Texas A&M for the branching-ratio measurements. Subsequent γ-ray spectroscopy measurements at TAMU revealed small levels of ^{131}I and ^{103}Ru in the ^{147}Nd sample and ^{103}Ru in the ^{144}Ce sample, most likely from molecules containing isotopes from these lighter mass chains. These additional isotopes will necessitate corrections between 0.3-2.5% for the branching-ratio determination.

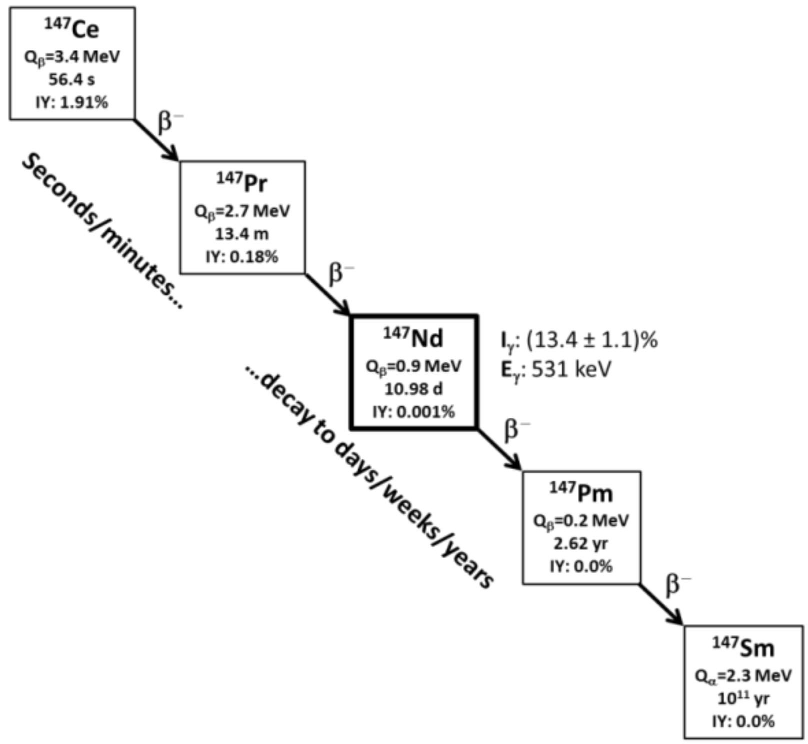

FIG. 1. CARIBU delivered intense, mass-separated beams consisting of isotopes from a single mass chain. For the mass-147 chain, the independent yield (IY) from fission initially generates mostly ^{147}Ce (and nuclei that decay to ^{147}Ce) and some ^{147}Pr nuclei. These nuclides both have half-lives of the order of minutes, and β^- decay within ~2 hours to create a pure sample of ^{147}Nd. Over time, the daughter nucleus ^{147}Pm grows in from the decay of ^{147}Nd. However, this is a small background that can be precisely tken into account.

At Texas A&M, the samples were placed in the center of a 4π gas proportional counter used for β detection. This newly-built detector has no internal windows, and therefore the β particles that emerge from the foil immediately enter the active volume of the detector. The performance of the counter was investigated using ^{95}Zr samples produced using a nuclear reactor and using CARIBU beams. The detector response agreed well with GEANT4 simulations and this work was summarized in the PhD thesis [6] of Brian Champine of the University of California at Berkeley.

The γ-ray measurements, performed both with and without coincident β-particle detection, were performed in the standard measurement geometry used by the Texas A&M group for many precise γ-ray branching-ratio measurements [7-9] and for detailed studies of internal conversion [10-12]. In this geometry, the efficiency of the detector is known to about 0.2% over the energy range of 50 keV to 2 MeV [4,5].

The decays of the ^{95}Zr, ^{144}Ce, and ^{147}Nd samples were studied over multiple weeks in 2017 and the analysis is currently underway. The γ-ray spectra for the β-γ coincidence results obtained are shown in Fig. 2. In each case, the daughter product is also radioactive and the contribution to the β emission rate from the sample must be taken into account. For the ^{144}Ce and ^{147}Nd samples, additional corrections to the β emission rate of 0.3% and ~2.5%, respectively, must be made to account for other long-lived fission products found in the sample.

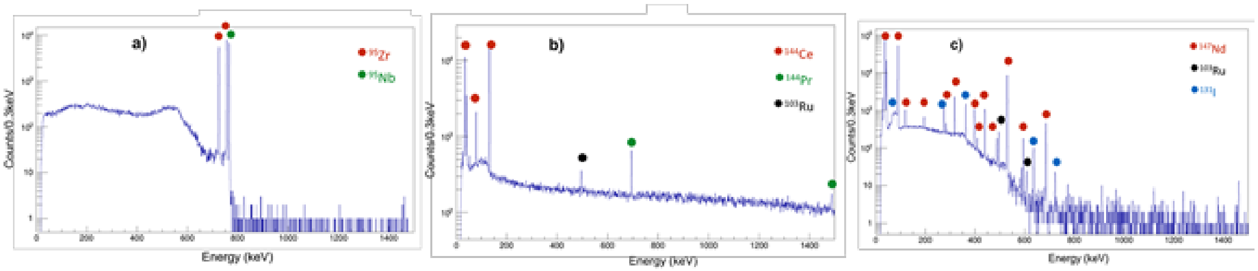

FIG. 2. The γ-ray energy spectra for β-γ coincidences for the a) ^{95}Zr, b) ^{144}Ce, and c) ^{147}Nd samples. In the ^{95}Zr sample, the only observed γ-ray lines are from the decays of ^{95}Zr and the daughter ^{95}Nb, which at the time of the measurement accounted for about 30% of the total activity of the sample. For the ^{144}Ce sample, several γ rays from the daughter ^{144}Pr ($t_{1/2}$=17.3 min), which is in equilibrium with ^{144}Ce, and a small (0.3%) contribution from the decay of ^{103}Ru can also been seen. For the ^{147}Nd sample, in addition to the many lines from the decay of ^{147}Nd, several lines from ^{131}I and ^{103}Ru indicate 2% and 0.3% contributions, respectively, to the total β-decay rate of the sample.

The γ-ray branching ratios are being determined from the ratio of the β-γ coincidence detection rates to the β particle detection rates, after correcting for detector efficiencies and the contribution to the β-decay rate from the daughter isotopes and any observed contaminants. The efficiency of the β counter largely divides out in the measurement and can be determined by comparing the number of γ-rays detected both with and without the coincidence detection of β particles. It is anticipated that with the data collected with these foils, the branching ratios can be determined to 1-2% precision. The γ-ray branching ratios in the decay of ^{95}Zr are already well established at 0.5% precision [13] and serve here as a confirmation of the accuracy of the measurement approach. These results will be a major part of the PhD thesis of Amber Hennessy, a graduate student at UC Irvine.

This work was supported in part by LLNL LDRD under project 15-ERD-015.

[1] N. Nica, Nucl. Data Sheets **110**, 749 (2009).

[2] J.K. Tuli, Nucl. Data Sheets **56**, 607 (1989).

[3] K. Debertin, U. Schotzig, K.F. Walz, and H.M. Weiss, Ann. Nucl. Energy **2**, 37 (1975).

[4] J.C. Hardy, V.E. Iacob, M. Sanchez-Vega, R.T. Effinger, P. Lipnik, V.E. Mayes, D.K. Willis, and R.G. Helmer, Applied Radiation and Isotopes **56**, 65 (2002).

[5] R.G. Helmer, J.C. Hardy, V.E. Iacob, M. Sanchez-Vega, R.G. Neilson, and J. Nelson, Nucl. Instrum. Methods Phys. Res. **A511**, 360 (2003).

[6] B. Champine, Ph.D. Thesis, University of California at Berkeley (2016).

[7] J.C. Hardy *et al.*, Phys. Rev. Lett. **91**, 082501 (2003).

[8] V.E. Iacob *et al.*, Phys. Rev. C **74**, 015501 (2006).

[9] D. Melconian *et al.*, Phys. Rev. C **85**, 025501 (2012).

[10] N. Nica *et al.*, Phys. Rev. C **71**, 054320 (2005).

[11] N. Nica *et al.*, Phys. Rev. C **77**, 034306 (2008).

[12] N. Nica *et al.*, Phys. Rev. C **80**, 064314 (2009).

[13] S.K. Basu, Nucl. Data Sheets **111**, 2555 (2010).