LAWRENCE LIVERMORE NATIONAL LABORATORY

LLNL-TR-740533

Precision Gamma-Ray Branching Ratios for Long-Lived Radioactive Nuclei

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October 24, 2017

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

FINAL REPORT

Precision Gamma-Ray Branching Ratios for Long-Lived Radioactive Nuclei

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Abstract

Many properties of the high-energy-density environments in nuclear weapons tests, advanced laser-fusion experiments, the interior of stars, and other astrophysical bodies must be inferred from the resulting long-lived radioactive nuclei that are produced. These radioactive nuclei are most easily and sensitively identified by studying the characteristic gamma rays emitted during decay. Measuring a number of decays via detection of the characteristic gamma-rays emitted during the gamma-decay (the gamma-ray branching ratio) of the long-lived fission products is one of the most straightforward and reliable ways to determine the number of fissions that occurred in a nuclear weapon test. The fission products ^{147}Nd, ^{144}Ce, ^{156}Eu, and certain other long-lived isotopes play a crucial role in science-based stockpile stewardship, however, the large uncertainties (about 8%) on the branching ratios measured for these isotopes are currently limiting the usefulness of the existing data [1,2]. We performed highly accurate gamma-ray branching-ratio measurements for a group of high-atomic-number rare earth isotopes to greatly improve the precision and reliability with which the fission yield and reaction products in high-energy-density environments can be determined. We have developed techniques that take advantage of new radioactive-beam facilities, such as DOE's CARIBU located at Argonne National Laboratory, to produce radioactive samples and perform decay spectroscopy measurements. The absolute gamma-ray branching ratios for ^{147}Nd and ^{144}Ce are reduced <2% precision. In addition, high-energy monoenergetic neutron beams from the FN Tandem accelerator in TUNL at Duke University was used to produce ^{167}Tm using the ^{169}Tm(n,3n) reaction. Four-time improved branching ratio of ^{167}Tm is used now to measure reaction-in-flight (RIF) neutrons from a burning DT capsule at NIF [10]. This represents the first measurement of RIF neutrons in any laboratory fusion system, and the magnitude of the signal has important implications for fundamental plasma science and for weapons physics.

Background and Research Objectives

Many properties of the HED environments in nuclear weapons tests, National Ignition Facility (NIF) shots, and the interior of stars and other astrophysical bodies must be inferred from the resulting long-lived radioactive nuclei that are produced. These radioactive nuclei are most easily and sensitively identified by studying the characteristic gamma-rays emitted during decay. For example, one of the most straightforward and reliable ways to determine the number of fissions that occurred in a nuclear weapon test is to detect the characteristic gamma-ray radiation emitted from the long-lived fission products (^{147}Nd, ^{144}Ce, and others) that were produced. However, these gamma-rays are emitted in only a fraction of the decays, and this fraction (known as the gamma-ray branching ratio) must be known accurately to determine the total number of fissions. In addition, the flux of reaction-in-flight (RIF) neutrons at NIF has recently been successfully measured by a LLNL-LANL team from a burning DT capsule using the production of ^{167}Tm via (n,3n) reactions on a ^{169}Tm puck [10]. The ratio of RIF neutrons to primary 14-MeV neutrons was determined by measuring the ratio of gamma-rays from the decay of ^{167}Tm and ^{168}Tm atoms produced in the puck – the ^{167}Tm atoms can only be produced by RIF neutrons with energies above 15 MeV via the ^{169}Tm(n,3n) reaction, while the ^{168}Tm atoms are dominantly produced by the 14-MeV neutrons. However, the branching ratio of gamma-ray transition energy of 207.8 keV is known to 19% (I_{gamma}=42±8%) [9].

Scientific Approach and Accomplishments

Thought, the level schemes of the isotopes of interest are well known, there are certain challenges in measuring these gamma-ray branching ratios, mainly, due to impurities, self-attenuation in the sample, etc. These challenges can be largely avoided by producing ultra-thin samples nearly free of radioactive contaminants at the CAlifornium Rare Isotope Breeder Upgrade (CARIBU) facility, located in Argonne National Laboratory and measuring the absolute number of decays utilizing betta-gamma counter, which is essentially close to 100% [3, 4] efficient, in conjunction with precision gamma-ray spectroscopy. The experimental approach to determine the gamma-ray branching ratios consists of the following steps: (1) producing ultrapure sources of ^{147}Nd, ^{144}Ce, and ^{156}Eu using CARIBU beams, (2) performing gamma-spectroscopy using a custom-made 4pi gas proportional counter in conjunction with gamma-ray spectroscopy using the precisely-calibrated HPGe detector at Texas A&M University [5,6].

(1) Source Production: We produced high-purity sources on thin (40 microgram/cm^2) carbon foil backings by harvesting isotopes at CARIBU. The CARIBU mass separator has a mass resolution of ΔM/M ~ 10E-4. Consider the example of decay chain of mass 147 shown in Figure 1. Within hours after implantation of the mass 147 isotopes, all the shorter-lived species decay to ^{147}Nd. CARIBU can currently deliver ~10E5 ions/s of mass 147, so nearly 10E10 ions were collected in 1 day. A large volume of HPGe detector was used to monitor the implantation rate and to provide additional information on the purity of the ion beam by looking for characteristic gamma-rays from other fission products. Beta-delayed neutron emission will produce isotopes along the A=146 chain, but the longest-lived isotope is ^{146}Pr with a half-life of 24.2 min and therefore all of this radioactivity will decay away in less than a day. The A=144 is the nearest mass chain with an isotope that has a half-life longer than 1 day (^{144}Ce) and the presence of this isotope was monitored by measuring the 133.5-keV gamma ray that is emitted in 11% of the decays. The quantity of the ^{147}Nd atoms collected using this technique in 1 day of CARIBU beam time were 1x10E10. These samples had enough intensity and few other radioactive contaminants. The produced source was shipped overnight to Texas A&M for the beta-gamma coincidence, and X-ray spectroscopy measurements.

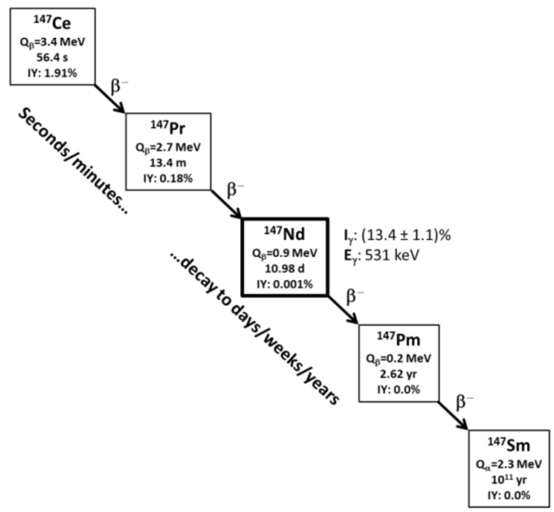

Figure 1. For the mass 147 chain, the independent yield (IY) from fission initially generates mostly ^{147}Ce (and nuclei that decay to ^{147}Ce and some ^{147}Pr nuclei. These nuclei all have half-lives of seconds to minutes and they betta decay within ~2 hours to create a pure sample of ^{147}Nd. The only remaining radioactive contaminant is ^{147}Pm from the decay of ^{147}Nd, which is a small background that can be precisely taken into account.

(2) Beta Spectroscopy: The collected carbon foil, was positioned in the center of the windowless 4pi gas proportional counter for beta detection. This type of detector has a low-energy threshold of <2 keV and yields essentially 100% detection efficiency for charged particles. The low-energy threshold is crucial for decays such as ^{147}Nd beta decay, which has a Q value of only 0.9 MeV and is very challenging to study by other techniques because conversion electrons are emitted in addition to beta particles in over 50% of the decays.
(3) Gamma-ray Spectroscopy: The collected sources at CARIBU were gamma-ray counted using large volume HPGe detector that has been painstakingly calibrated at Texas A&M and has an efficiency that is known to about 0.2% over the energy range of 50 keV to 2 MeV. The measurements, performed both with and without coincident beta particle, was performed in the standard geometry.

The ^{169}Tm(n,3n)^{167}Tm reaction cross section and the branching ratio of the 207.8-keV gamma ray following the EC decay of ^{167}Tm were measured at the FN Tandem accelerator in TUNL at Duke University. The branching ratio was measured using the facilities at UC Berkeley and the 88-Inch Cyclotron at LBNL. A thin holmium foil was irradiated using a 29-MeV alpha-particle beam inducing the ^{165}Ho(α,2n)^{167}Tm reaction. The foil was counted using well-calibrated coaxial and planar HPGe detectors. The branching ratios for the emission of 207.8-keV and 531.5-keV gamma rays were determined with fractional uncertainties of approximately 3.8%. These uncertainties are five times smaller than the previous measured results [9]. Using this measured branching ratio for the 207.8-keV transition, a subsequent experiment was conducted to measure the ^{169}Tm(n,3n)^{167}Tm cross section, shown on Figure 2. The cross section was measured with a precision of 5%–8% in the 17–22 MeV energy range that is important for determining the RIF-neutron fluence at NIF. In addition, the cross sections for ^{58}Ni(n,2n) and ^{94}Zr(n,α) were measured over the same energy range, also shown on Figure 2.

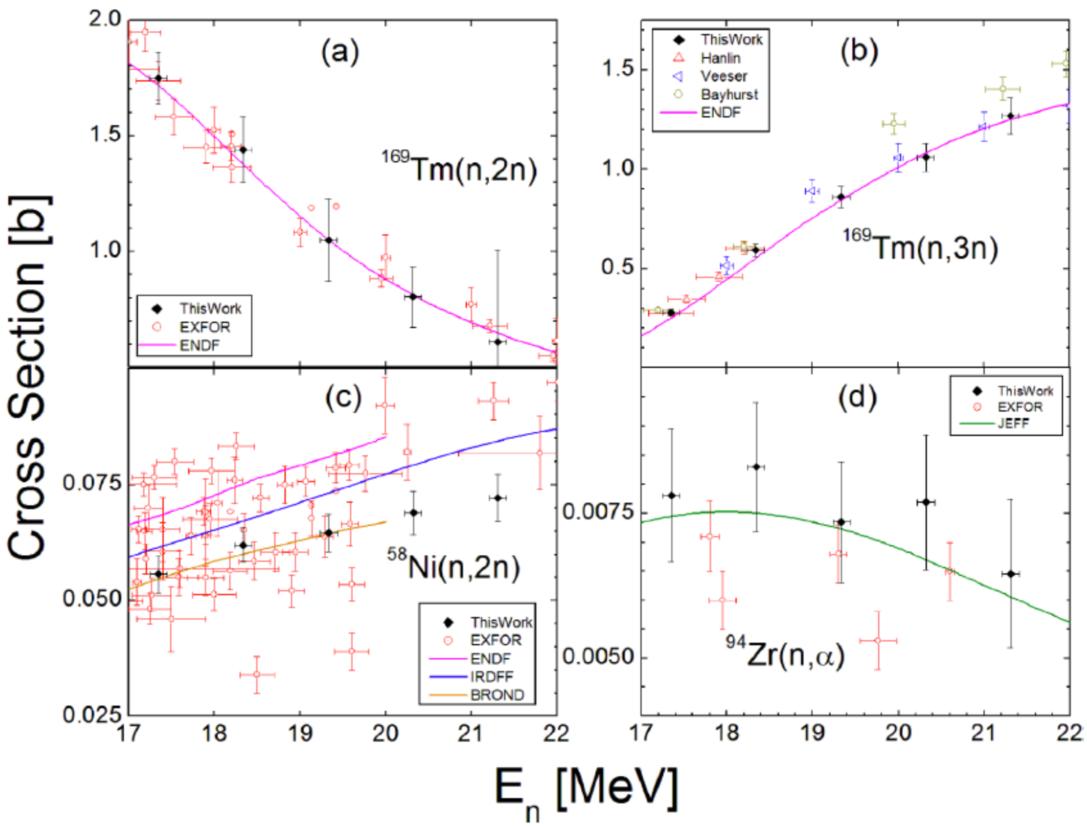

Figure 2. Experimental results from different neutron induced cross sections measured in this project [7] compared with the previous data summarized in EXFOR and evaluations [8,9].

Impact on Mission

We addressed important nuclear data needs for stockpile stewardship science by exploiting new opportunities to harvest fission products and developing new techniques for precision decay measurements. This effort also aligns with the Laboratory core competencies of nuclear, chemical, and isotopic science and technology and high-energy-density science. For several isotopes, nuclear data limits the precision with which conclusions can be drawn about high-energy-density environments of interest to the weapons complex (absolute fission-product yield studies) and ignition fusion efforts (the flux of reaction-in-flight neutrons). Having a precisely-calibrated source and HPGe detector available at LLNL will be enormously beneficial to the Livermore counting capability and can impact many strategically important programmatic missions like stockpile stewardship and nuclear forensics by providing overall better nuclear data for fission. This additional capability can open up new opportunities to attract additional funding from WCI, GS, NIF, and the DOE Office of Science. The results of this LDRD are important for WCI planning of their nuclear data program by demonstrating the capability to produce 1% measurements of relevant branching fractions.

The ^{169}Tm(n,3n)^{167}Tm cross-section provide reliable nuclear data which reduce the overall uncertainties on the measured reaction-in-flight yield at NIF to about 5%–8% in an important energy region between 17 and 22 MeV. In addition, the absolute mamma-ray branching ratios following the electron-capture decay of ^{167}Tm were improved by factor of five.

Conclusion

We have measured the gamma-ray branching ratios of four high-atomic-number rare earth isotopes (neodymium-147, cerium-144, and thulium-167), with 1 to 2% precision which greatly improve the nuclear data used to understand fission yield and reaction-in-flight neutrons. We have developed an isotope-harvesting technique that can be used to make ultrapure radioactive sources at CARIBU and other radioactive-beam facilities such as the Facility for Rare Isotope Beams at Michigan State University. In addition, we develop a 4-pi gas proportional detector that can be used to precisely measure branching ratios of any fission product or reaction product with a half-life over one hour. Finally, we attracted a postdoctoral researcher (Kay Kolos) and two graduate students (Brian Champine from UC Berkeley and Amber Hennessy from UC Irvine) to LLNL to assist with the challenge of developing new precision measurements of radioactive decay.

Using the ^{169}Tm(n,3n) reaction at National Ignition Facility represent the first measurement of reaction-in-flight neutrons in any laboratory fusion system, and the magnitude of the signal has important implications for fundamental plasma science and for weapons physics [10]. The branching ratio and the ^{169}Tm(n,3n) cross-section results provide more reliable nuclear data for an important diagnostic that is used at the National Ignition Facility to estimate the yield of reaction-in-flight neutrons produced via the inertial-confinement-fusion plasma in deuterium-tritium capsules.

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B. Champine: APS -2015 Fall Meeting of the APS Division of Nuclear Physics, Santa Fe, October 28 - 31, 2015. LLNL-PRES-678879

B. Champine: PhD dissertation: "Experiments to Improve Nuclear Data for High Energy Density Environments". UC Berkeley 2016.

K. Kolos: Invited talk on "Precision beta-decay studies for fundamental science and applications" at the Gordon Research Conference for Nuclear Chemistry, June 22nd 2017. LLNL-PRES-733511

K. Kolos: PLS Post-Doc seminar on "Measurements of g-ray branching ratios for fission products", July 15th 2017.

K. Kolos: Poster on "Improving beta-decay studies for fundamental science and applications" with N. Scielzo at the ASCAC/LDRD Review, January 5th 2017. LLNL-POST-716118

K. Kolos: Poster at the postdoc symposium LLNL "Measurements of gamma-ray branching ratios for fission products", June 14th 2017. LLNL-POST-732912