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

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This report describes the progress of our experimental program to precisely measure the β-decay branching ratios of ^{95}Zr, ^{144}Ce, and ^{147}Nd, which began in 2016. More information about the motivation and experimental approach can be found in our previous report [1]. The measurement described in [1] suffered from a β-detector efficiency that was ~15% lower than anticipated due to a high electronic threshold that was discovered after data collection. Therefore, we have repeated these measurements.

As in the previous experiment, high-purity samples of ^{95}Zr (150 Bq) and ^{147}Nd (1600 Bq) were collected on thin (40 μg/cm^2) carbon-foil backings using low-energy mass-separated beams of A=95 and 147 fission products from CARIBU at Argonne National Laboratory. The implanted samples were then shipped to Texas A&M where the decay-counting measurement took place. Because of its long half-life (284.91 d), we were able to reuse a previously-made ^{144}Ce (160 Bq) sample. The γ-ray and β-γ measurements were performed in the same geometry as our previous experiment: The sample was inserted in the middle of a 4π gas proportional counter for β-particle detection and was positioned 15.1 cm from the HPGe detector. We also improved the signal-to-background ratio in the γ-ray spectrum by a factor of 3 by adding a lead-plastic-copper layered shield around the HPGe detector. We performed multiple-day-long measurements with each sample, interleaved with background measurements. The β-γ coincidence spectra are shown on Fig.1.

A thorough analysis of the data is underway. We were able to collect sufficient statistics for a sub-percent uncertainty on all the measured sources: We have collected about 170k, 120k, and 40k β-γ coincidence counts for the most-intense γ-ray peaks in the decays of ^{147}Nd, ^{95}Zr and ^{144}Ce, respectively. We are investigating the systematic uncertainties associated with the measurement. The main challenges are establishing the β-detector and γ-ray detection efficiencies and determining the purity of the samples. The preliminary analysis shows the β detector performed as expected with an efficiency of 96-98% for β transitions with energies in the range, 100-800 keV. The experimental and simulated (GEANT4) efficiencies are in good agreement, as can be seen in Fig. 2. To determine the fraction of observed β-singles counts from the isotope of interest, the contributions from the decay of the daughter isotope and any contaminants must be taken into account. For the ^{95}Zr sample, the grow-in of the daughter ^{95}Nb accounted for about 20% of the activity; for the ^{144}Ce sample, the daughter ^{144}Pr has only a 17.28 minute half-life, so its contribution was 50%; For ^{147}Nd, the daughter ^{147}Pm has a 2.6 year half-life, and so it

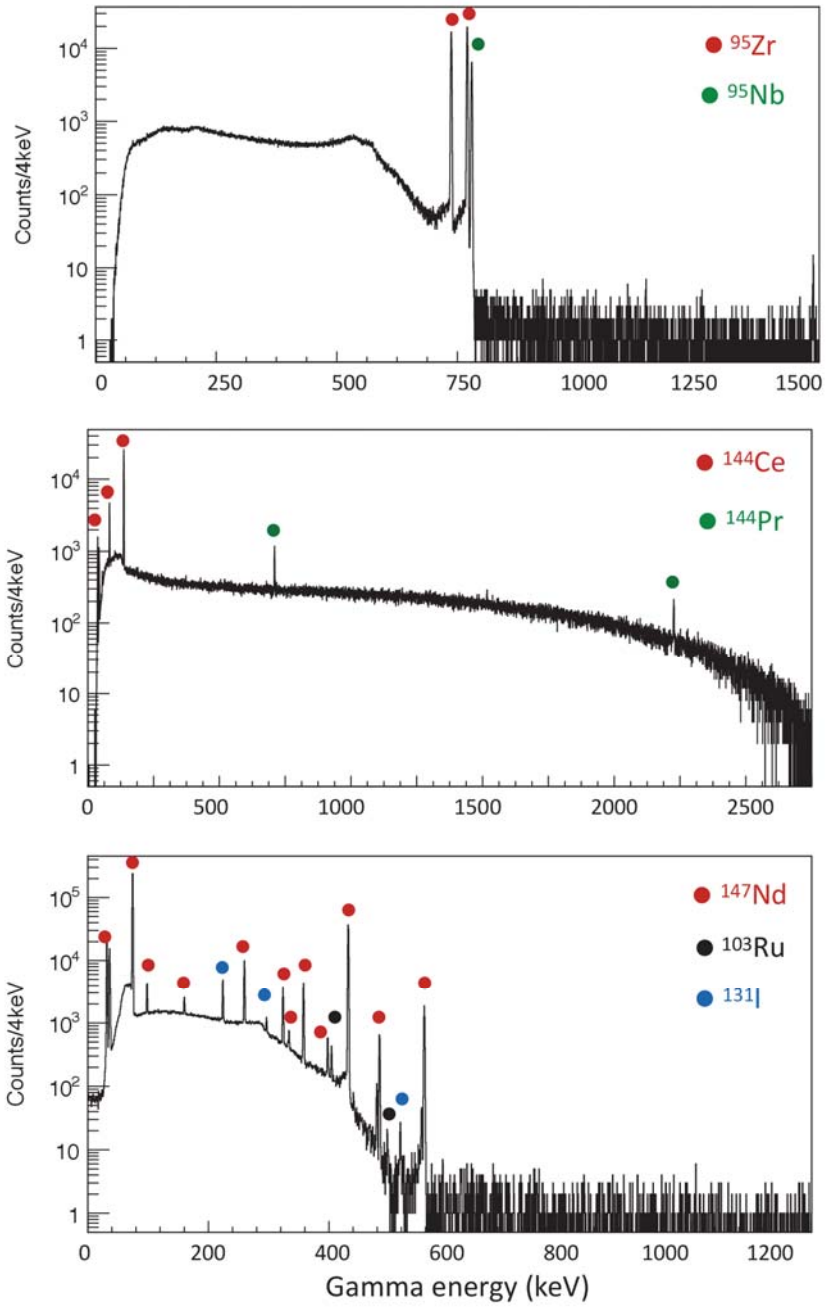

FIG. 1. The γ-ray energy spectra for β-γ coincidences for the ^{95}Zr (top), ^{144}Ce (middle), and ^{147}Nd (bottom) samples.

contributed only about 0.5%. The only additional contaminants observed in any of the samples were in the case of ^{147}Nd, where ~0.3% of the activity was from ^{131}I and ^{103}Ru.

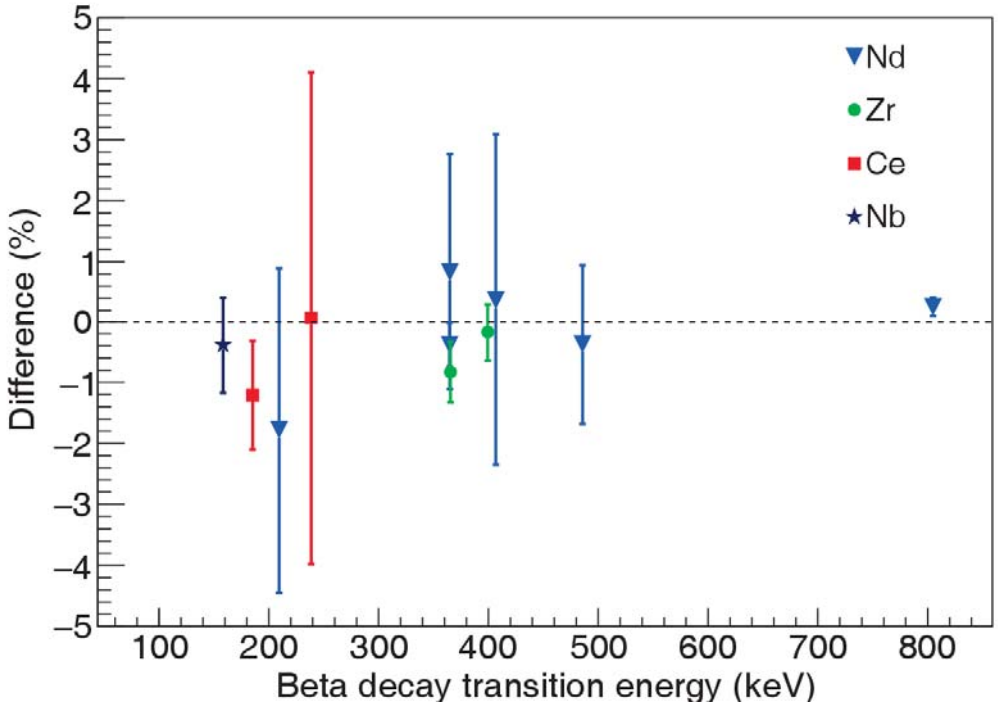

FIG. 2. The efficiency of the 4π gas proportional counter used for β detection compared with GEANT4 simulations. Figure shows the difference between the simulated efficiency and the experimental efficiency for several transitions in ^{95}Zr, ^{144}Ce, and ^{147}Nd.

With this data set we should be able to determine the branching ratios of ^{95}Zr, ^{144}Ce, and ^{147}Nd to 1-2% precision. We intend to complete the analysis by the end of the summer.

[1] K. Kolos *et. al., Progress in Research*, Cyclotron Institute, Texas A&M University (2016-2017), p. I-31.