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

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We have measured the ^{95}Zr β-decay branching ratios to high (<1%) precision. This serves as a proof-of-principle demonstration that the method we have developed can be used to measure the γ rays and β branching ratios of long-lived fission products. As previously reported [1-2], we collected a high-purity sample of ^{95}Zr (and its decay daughter ^{95}Nb) on a thin (40 μg/cm^2) carbon-foil backing by using a low-energy mass-separated beam of A=95 ions from CARIBU at Argonne National Lab. The subsequent decay measurements were performed at Texas A&M with the HPGe detector for γ-ray measurements and the Livermore 4π gas counter for β counting.

We performed a thorough analysis of the collected data to obtain γ-ray intensities and β branching ratios in the decay of ^{95}Zr and ^{95}Nb. The γ-ray intensity (I_γ) per β decay for a particular transition was determined from the β-γ coincidence detection rate ($R_{\beta\gamma}$) relative to the β particle detection rate from the decay of the isotope of interest (R_β), through the relation

$$I_\gamma = \frac{R_{\beta\gamma}}{R_\beta}\frac{1}{\varepsilon_\gamma}\frac{\varepsilon_\beta}{\varepsilon_{\beta\gamma}}.$$

Here ε_γ is the photopeak efficiency of the HPGe detector for detecting the γ ray of interest. These efficiencies were obtained from the CYLTRAN simulation and are reported in Table I. $\varepsilon_{\beta\gamma}$ is the efficiency for detecting the β particles emitted in coincidence with that γ ray, and ε_β is the efficiency for detecting the β particles from all decay branches of the isotope of interest in the 4π gas counter. The $\varepsilon_\beta/\varepsilon_{\beta\gamma}$ efficiency ratio of the 4π gas proportional counter deviates from unity because the β-particle energy spectra in the ratio are not identical, and a different fraction of low-energy particles are lost to self-absorption in the carbon foil or to the electronic threshold of the counter. We determined the efficiency ratio using simulations of the detector response to the β-decay spectra under consideration.

Table 1. Detector efficiencies for the γ rays in the decay of ^{95}Zr and ^{95}Nb, and the β particle efficiency ratio for the 4π gas counter.

Source	E_{γ_i}	ϵ_{γ_i} (%)	$E_{\beta_{max}}$	$\varepsilon_\beta/\varepsilon_{\beta\gamma}$
^{95}Zr	724.2 keV	0.2977(9)	399.4(18)	0.9987(1)
^{95}Zr	756.7 keV	0.2884(9)	366.9(18)	1.0011(1)
^{95}Nb	765.8 keV	0.2860(9)	159.8(5)	1.0000(1)

Before building the simulation model, we used the measured ratio of β-γ coincidences over γ singles ($R_{\beta\gamma}/R_{\gamma}$) to obtain the efficiency of the 4π gas counter ($\varepsilon_{\beta\gamma}$) for detecting the β particles emitted in coincidence with several detected γ rays. We then adjusted the β-detection threshold setting used in the simulation model to best reproduce those measured 4π gas counter efficiencies. The comparison between the experiment and the simulation of the 4π gas counter detector efficiency for the different β-decay transitions is presented in Fig. 1. The insert shows the χ^2 values obtained from the difference between the measured values and simulated values for a range of thresholds. These results indicate that the detector threshold was 1.1(3) keV. Table I shows the $\varepsilon_{\beta}/\varepsilon_{\beta\gamma}$ efficiency ratios calculated with that threshold.

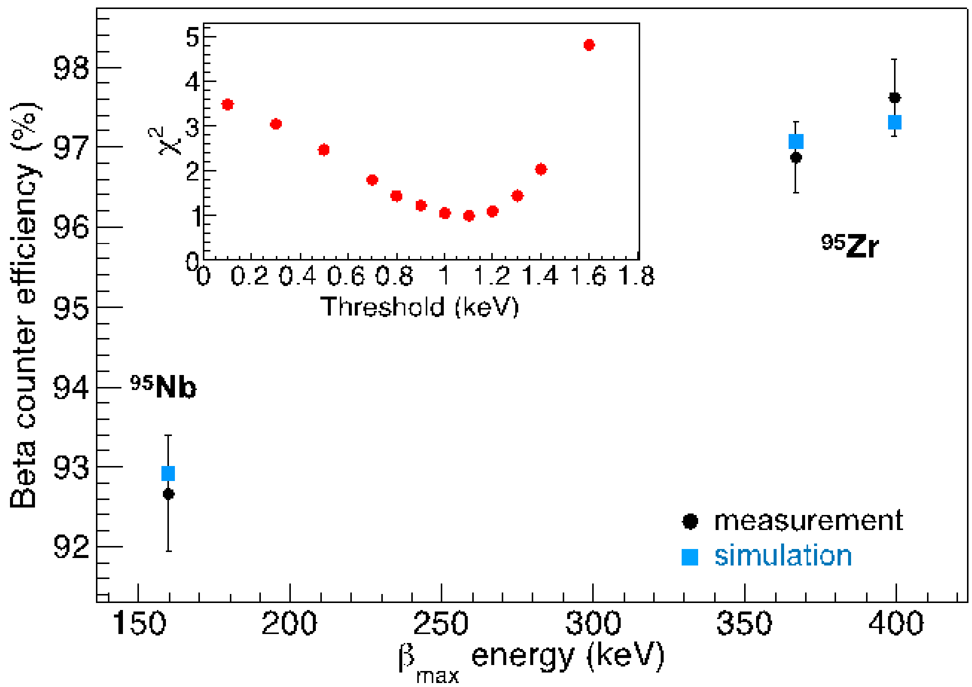

FIG. 1. Comparison of the measured gas counter efficiencies for the highest-intensity transitions in the decays of ^{95}Zr and ^{95}Nb to simulation results obtained with GEANT4 with a 1.1-keV threshold. The insert shows the χ^2 values obtained from the difference between the measured values and simulated values for a range of thresholds. These results indicated the detector threshold was 1.1(3) keV.

We calculated the β-decay branching ratios from the γ-ray intensities after including small contributions from conversion electron emission. The comparison between our results and the evaluated values [3] are presented in Table II. The complete uncertainty budget for γ-ray intensities from the decay of ^{95}Zr and ^{95}Nb is given in Table III. We are currently working on finalizing our results and uncertainties, and writing a manuscript that describes in detail our experimental approach, which will be submitted to the journal Nuclear Instruments and Methods in Physics Research A.

Table II. Preliminary results for the γ-ray intensities and β branching ratios for ^{95}Zr and ^{95}Nb obtained in this experiment in comparison with the evaluated values [3].

Isotope	E_γ	$I_{\gamma(\text{this work})}$	I_γ	$BR_{\beta(\text{this work})}$	BR_β
^{95}Zr	724.2 keV	0.4380(25)	0.4427(22)	0.4387(25)	0.4434(22)
^{95}Zr	756.7 keV	0.5441(28)	0.5438(22)	0.5449(28)	0.5446(22)
Ratio	724.2 keV/756.7 keV	0.805(6)	0.814(4)		
^{95}Nb	765.8 keV	0.9962(94)	0.9981(1)	0.9962(95)	0.9997(1)

Table III. The preliminary uncertainty budget for γ-ray intensities measured in this study.

Source	724.2 keV	756.7 keV	765.8 keV
R_{β_γ}	0.0049	0.0041	0.0078
R_β	0.0008	0.0008	0.0044
ϵ_γ	0.0030	0.0031	0.0031
$\epsilon_\beta/\epsilon_{\beta\gamma}$	0.0001	0.0001	0.0001
Total Uncertainty	0.0058	0.0052	0.0095

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

[2] K. Kolos *et. al.*, *Progress in Research*, Cyclotron Institute, Texas A&M University (2017-2018), p. I-30.

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