Precision γ-ray branching ratio measurements for long-lived fission products of importance to nuclear-security

M. Bencomo,[1] K. Kolos,[1] J.A. Clark,[2] V.E. Iacob, D. Melconian, E. Norman,[3] H.I. Park, G. Savard,[2] N.D. Scielzo,[1] and M.A. Stoyer[1]

[1]*Lawrence Livermore National Laboratory, Livermore, California,*

[2]*Argonne National Laboratory, Lemont, Illinois*

[3]*University of California Berkeley, Berkeley, California*

Continuing with our effort of precisely measuring the branching ratios for long-lived fission products we have collected and measured two radiopure ^{156}Eu samples. The samples were collected on thin (40 μg/cm^2) carbon-foil backings using a low-energy mass-separated beam of A = 156 fission products from CARIBU at Argonne National Laboratory. During collection, a HPGe detector was used to continuously monitor the implantation rate by detecting the characteristic γ rays emitted following the β decay of the shorter-lived fission products (Fig. 1). The first sample had measured activity of 375 Bq while the second one had an activity of 700 Bq. The implanted samples were then shipped to Texas A&M University for measurement of the subsequent decay.

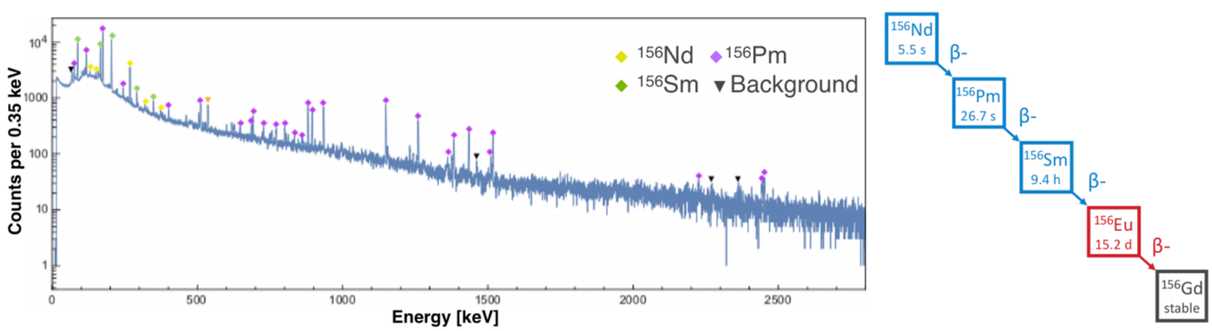

Fig. 1. γ spectrum from CARIBU during sample collection (January 2020). Most of the γ rays seen come from the A=156 decay chain.

As with previous experiments, the samples were placed in the center of a 4π proportional gas counter, with no internal windows, for β detection [1-3]. The γ rays were measured with a 0.2% absolute efficiency calibrated HPGe both with and without a coincident β particle at the nominal distance of 151 mm. This geometry has been discussed in previous reports and experiments [1, 5]. The measurement of both samples was done in 14 runs over the course of 2 weeks, where we collected over 120 hours of data for sample 1 and 220 hours for sample 2. These collection times yielded 2,091,588 and 8,096,150 coincidence events, respectively.

The analysis is currently underway. We have filtered the data to remove any noisy or faulty "cycles" encountered during the measurement. This filtering procedure reduces the total number of β singles by 3.5% (sample 1) and 5.3% (sample 2). This filter will also remove coincident events related to noisy/faulty runs by 3.5% and 18% (the large deficit in sample 2 is due to two runs where 43% and 25% of the data was noise). Further filtering of coincident events missing timing or energy information has also been done with the number of coincident events lost due to this being 0.34% for both samples. For the highest-intensity γ ray, which has an energy of 811 keV, there are over 30,000 counts for sample 1 and over 100,000 for sample 2 (Fig. 2). We have also identified a contaminant, ^{112}Ag, with a contribution of <0.3% to our total number of β particles detected.

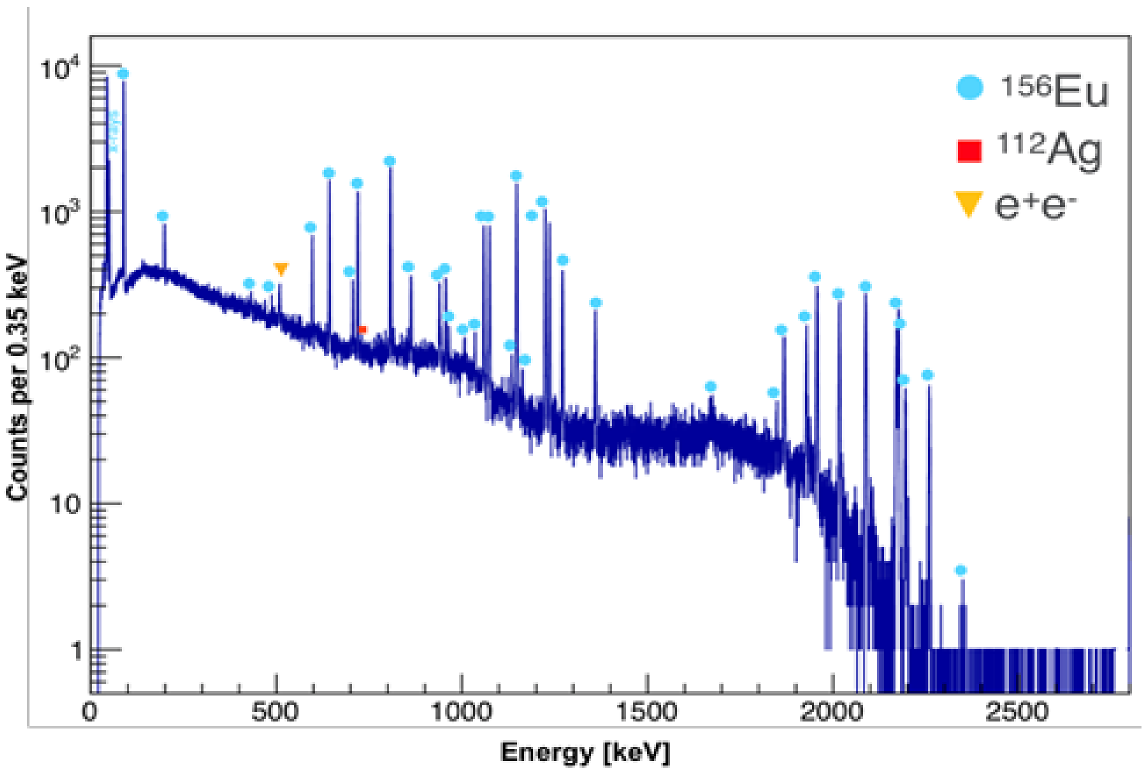

Fig. 2. Coincident γ-ray spectrum from the TAMU measurement (February 2020). The contaminant contribution is <0.3%.

Additionally, the efficiencies for all γ-ray energies, ranging from 89 to 2361 keV, have been calculated using the Cyltran Monte Carlo transport code [4] and the HPGe geometry [5]. This particular decay has close to 100 γ rays associated with it, most of which are not very intense. However, 85% of those γ rays are clearly identifiable in the measured γ spectrum, while for the rest we can set upper limits. Currently, there is work being done with the experimental data and GEANT4 simulations to obtain the efficiencies of the β detector, another crucial component to determine the branching ratios to high precision. We anticipate reaching a fractional precision of 1% for the highest-intensity transition in the decay of ^{156}Eu.

Furthermore, a proposal for "Improving decay data of the long-lived fission products ^{161}Tb and ^{111}Ag for nuclear medicine and national-security applications" has been approved by the ANL PAC for 6 days of beam collection. The scheduling for this experiment is pending, while we coordinate based on the availability of all collaborators involved.

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