Decays of the Three Top Contributors to the Reactor $\bar{\nu}_e$ High-Energy Spectrum, ⁹²Rb, ^{96gs}Y, and ¹⁴²Cs, Studied with Total Absorption Spectroscopy

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We report total absorption spectroscopy measurements of 92 Rb, 96gs Y, and 142 Cs β decays, which are the most important contributors to the high energy $\bar{\nu}_{\rho}$ spectral shape in nuclear reactors. These three β decays contribute 43% of the $\bar{\nu}_e$ flux near 5.5 MeV emitted by nuclear reactors. This $\bar{\nu}_e$ energy is particularly interesting due to spectral features recently observed in several experiments including the Daya Bay, Double Chooz, and RENO Collaborations. Measurements were conducted at Oak Ridge National Laboratory by means of proton-induced fission of ²³⁸U with on-line mass separation of fission fragments and the Modular Total Absorption Spectrometer. We observe a β -decay pattern that is similar to recent measurements of 92 Rb, with a ground-state to ground-state β feeding of 91(3)%. We verify the 96gs Y ground-state to ground-state β feeding of 95.5(20)%. Our measurements substantially modify the β -decay feedings of ¹⁴²Cs, reducing the β feeding to ¹⁴²Ba states below 2 MeV by 32% when compared with the latest evaluations. Our results increase the discrepancy between the observed and the expected reactor $\bar{\nu}_e$ flux between 5 and 7 MeV, the maximum excess increases from $\sim 10\%$ to $\sim 12\%$.

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Nuclear reactors generate most of their energy by the fission of isotopes of uranium and plutonium creating two radioactive neutron-rich nuclei. These fission products decay towards stable nuclei, emitting β particles and $\bar{\nu}_{e}$ s, as well as γ rays and neutrons in their decay chains. Recently, there is great interest in the β decays of fission products motivated by the direct $\bar{\nu}_e$ measurements that use nuclear reactors as intense $\bar{\nu}_{\rho}$ sources [1–6]. The number of measured reactor $\bar{\nu}_{e}$ interactions with detector matter is 0.95(2) of the expected number of events and is often referred to as the "reactor $\bar{\nu}_e$ anomaly" [3–5]. There is also up to a 10% excess of high-energy $\bar{\nu}_e$ events in the 5 to 7 MeV $\bar{\nu}_e$ energy range that is referred to as the "shoulder" [1-3,7]. These results might constitute a hint of new physics in the neutrino sector, including the possible existence of sterile neutrinos [4,5]. However, in order to fully analyze the unexpected features of the measured $\bar{\nu}_{e}$ energy spectrum, the associated $\bar{\nu}_{e}$ spectrum must be understood to better than a few percent. Earlier studies [7-12] and the present measurements demonstrate that this is not yet the case.

There is other interesting physics that can be explored with β -delayed decay products from nuclear reactors. The energy released by the nuclear fuel through the β -delayed decay of fission products is known as decay heat. Decay heat accounts for approximately 8% of the total energy from nuclear fission, and it is the only source of heating of nuclear fuel rods after stopping the controlled chain reaction. Understanding β -decay features during decay heat release in nuclear fuel contributes to the optimization of energy production and most importantly to the analysis of reactor safety [13]. Reliable measurements of β -strength patterns also point to the structure of energy levels involved in β decay and the related neutron and proton singleparticle state evolution in neutron-rich nuclei.

One approach toward obtaining a more reliable prediction of the $\bar{\nu}_e$ flux from nuclear reactors is to accurately measure individual β decays of the most important fission products. One method to improve β decay measurements is to use total absorption spectroscopy [14]. Performing total absorption β -decay studies of several hundred radioactive nuclei is a major experimental undertaking requiring advanced experimental and data analysis techniques. However, it has been assessed that for $\bar{\nu}_e$ with energy between 5 and 7 MeV, there are a few known nuclei that are abundantly produced in the reactor that contribute substantially to the $\bar{\nu}_e$ flux in this energy region [8,11]. The three largest contributors, ⁹²Rb, ^{96gs}Y, and ¹⁴²Cs, are characterized by β transitions to the ground state or to lowenergy excited states that dominate the decay pattern and are expected to create 43% of the $\bar{\nu}_e$ flux near 5.5 MeV [11]. In the present study we report new decay measurements obtained using total absorption spectroscopy for these three fission products.

The decays of ⁹²Rb, ^{96gs}Y, and ¹⁴²Cs were measured at Oak Ridge National Laboratory. These three activities were produced by inducing fission in a UC_x target with a 40 MeV, 50 pnA proton beam. The isotopes were extracted, ionized with a surface-ionization source, accelerated to 40 keV, and analyzed by means of an on-line separator with a mass resolution of $M/\Delta M = 600$ [15]. Radioactive beams were collected on a transport tape, which was then moved into the Modular Total Absorption Spectrometer (MTAS), measured, and then transported away afterwards to prevent buildup of the associated long lived activities.

MTAS consists of 19 hexagonal NaI(Tl) crystals forming a 4π array [16–18]. There is over 1000 kg of NaI(Tl) detector material in MTAS, covering over 99% of the solid angle around the measured activities. The γ -ray efficiency for full γ -ray energy absorption in on-line conditions is a flat 81% from 300 to 800 keV, and then drops smoothly to 72% at 5 MeV [18]. At the center of MTAS there are two 1 mm thick silicon detectors that are used as β triggers. The silicon detectors suppress laboratory background by at least 3 orders of magnitude. In addition to the active background suppression, there are over 5000 kg of lead shielding around MTAS; see Refs. [16–19] for further details.

We evaluate the MTAS measured energy spectrum to determine β -feeding intensities to known and unknown energy levels in the daughter nuclei, including the ground state. The evaluation technique is based primarily on techniques demonstrated and applied to other total absorption spectrometers, but the analysis is enhanced because of the increased efficiency and segmentation of MTAS [17,19–22]. Only a brief overview of our analysis is presented.

The MTAS response to γ rays and electrons is simulated with the GEANT4 toolkit [23]. We have verified the generated MTAS response functions to single γ -ray and two- γ cascades [16–18]. MTAS energy spectra are divided into two response regimes: energy levels below a threshold energy with deexcitation γ paths known from highresolution data and unknown β -fed levels above the threshold energy modeled in 25 keV bins. The higher energy regime has high level density [21]. These simulated response functions are then fit to the measured MTAS data. For β decay to states at higher excitation energy, there is an uncertainty associated with the number of γ rays involved in the deexcitation path, but this is mitigated by the high efficiency of MTAS. The very high full γ -ray energy efficiency is important to our analysis, since it implies a change in MTAS peak γ -ray efficiency of about 0.5 when detecting four γ rays of total energy E compared to detecting a single γ ray of the same total energy. This property is true over the energies of interest for our β -decay studies and demonstrates how an immediate qualitative evaluation of raw MTAS γ spectra is possible; see Fig. 1. The γ multiplicity can be identified by comparing the spectra in different crystals of MTAS [19]. The MTAS efficiency can be calculated from the γ multiplicity for a given energy level deexcitation pattern.

In Fig. 1 we compare the measured energy spectrum for 142 Cs decay, to the simulated MTAS response using decay data in the current Evaluated Nuclear Structure Data File (ENSDF) database [24,25]. The reduction of both the ground-state β feeding and the large reduction of the β feeding to the first-excited 2⁺ state at 360 keV can be seen, as well as the presence of new β -fed levels at higher excitation energies. When coupled with a deconvolution method, such as fitting routines based on an iterative technique described in previous work [20], our analysis allows for a quantitative estimate of β -feeding intensities. The numerical deconvolution and evaluations in this study are based on the approach in Ref. [20], but we have



FIG. 1. Background subtracted ¹⁴²Cs MTAS energy spectrum (black) compared to the simulated MTAS response to 1.9×10^{6} ¹⁴²Cs decay events based on the ENSDF data (cyan). We observe β feeding to low-lying states (below 2500 keV) to be more weakly populated than previous measurements, while β feeding to higher lying levels are more strongly fed. The peak around 6850 keV is a sum of the ¹²⁷I and ²³Na neutron capture peaks. The number of counts in the neutron capture peak is consistent with the simulations of 0.09% β -neutron branching fraction in Ref. [24].

modified it to include experimental information on γ multiplicities and decay paths measured with MTAS.

An uncertainty in the response function arises due to the detector geometry in the center of MTAS in our GEANT4 simulation, such as the nonactive volume around the silicon detectors and signal cables. This impacts the simulation of β particles and the simulated ground-state response most, but becomes less influential for higher energy levels fed by β decay. We have a simulation model of the interior of MTAS that reproduces large ground-state feedings and compares well with off-line measurements of ⁹⁰Sr activity. Based on simulating then deconvolving ENSDF decay patterns we add a systematic 2% relative uncertainty to the β -response error budget. When simulating the β and $\bar{\nu}_e$ spectra we assume that the 0^- to 0^+ ground-state to groundstate β -decay transitions for the three nuclei have β transitions of an allowed shape, as is expected based on previous measurements and calculations [8,11,26,27].

An example of an overall fit of the simulated response functions with a single γ transition deexciting 25 keV bins and their sum compared to the measured MTAS spectrum of ⁹²Rb is shown in Fig. 2. The sum of the individual decay paths is indistinguishable from the measured data, which is true for almost all possible sets of decay paths involving up to four γ rays. Hence, a good fit to the full MTAS data is required, but it is not sufficient for a deconvolution to be accurate. The number of γ rays involved in the decay must also be measured experimentally to determine the error bar on fitted intensities. Using the modular construction of MTAS to fit the number of γ rays for each decay is



FIG. 2. Fit of simulated single γ -ray response functions (colored) to the total ⁹²Rb MTAS data (black). The sum of all the simulated components is indistinguishable from the data. The ground-state β feeding (broad mauve curve) is the dominant contribution to all channels except at the highest energies. Each of the lower fed levels have large β components and the β contribution diminishes as the level energy increases because the lower energy β particles associated with these higher levels do not deposit as much energy in MTAS. We account for pile up and random coincidence events contributions in our analysis; these smaller effects are not shown in the picture to preserve clarity.

the basis of our analysis of β -strength distributions and their uncertainties.

The derived β -feeding intensity for ${}^{92}\text{Rb}$ [$T_{1/2} =$ 4.48(3) s, $Q_{\beta} = 8095(6)$ keV] is shown in Fig. 3. Earlier reported decay schemes of ⁹²Rb used to derive $\bar{\nu}_{\rho}$ spectra have varied substantially in the ground-state to ground-state intensity, from 51(18)% [8,28] to 95.2(7)% [11,29]. The latter uncertainty is entirely based on the uncertainty of the absolute 815 keV γ intensity of 3.2(4)% [30], which means that the ground state β -feeding error is underestimated in Ref. [29]. In addition, the evaluation [29] does not rely on any total absorption measurements. Therefore, the possible influence of the pandemonium effect can not be excluded [31], again making the quoted small error likely unreliable. The main backgrounds for the ⁹²Rb decay are its daughter activity, ⁹²Sr, and less than 1% contamination of 91 Sr. There are 2.4×10^7 events in the β -gated MTAS spectrum. Our result for the ground-state feeding of 91(3)% is consistent with the most recent total absorption measurement of 87.5(25)% [27].

The β decay of 96gs Y $[T_{1/2} = 5.34(5) \text{ s}, Q_{\beta} = 7103(6) \text{ keV}]$ was measured following the decay of implanted 96 Rb and 96 Sr ions. The isomer 96m Y $(T_{1/2} = 9.6 \text{ s})$ is not extracted from the ion source, nor is it produced in the decay chain of 96 Rb and 96 Sr. The analysis of the 96gs Y data is more complex than the 92 Rb analysis. 96 Rb decay has both β - γ and β -neutron- γ branches so that 96 Sr $[T_{1/2} = 1.07(1) \text{ s}]$ and 95 Sr $[T_{1/2} = 23.90(14) \text{ s}]$ activities are present. These associated decays have half-lives that bracket the 96gs Y half-life, but these contaminants can be separated by a deconvolution method of the energy spectra as a function of half-life. Another difficulty with the 96gs Y analysis is the existence of $0^+ \rightarrow 0^+ E0$ decays. MTAS is relatively insensitive to E0 decays with energies below 1.6 MeV, and, in the case of 96gs Y, the weak E0 signal is overwhelmed by



FIG. 3. Average ⁹²Rb β -feeding intensity with the uncertainty based on the number of γ rays in the deexcitation cascade from each level. The fit for the ground state β feeding is off scale at 91(3)%.



FIG. 4. Average ¹⁴²Cs β -feeding intensity with the uncertainty based on the number of γ rays in the deexcitation cascade from each level. The fit for the ground state β feeding is off scale at 44(2)%.

the ground-state feeding. We assume the β measurements reported for the 1581 keV *E*0 decay are correct [32,33]. Based on the previous measurements, we also take into account a small feeding through this 1581 keV state, which does not affect our estimate of the ground-state β -feeding intensity. Our result for the β -strength ground-state feeding, 95.5(20)%, verifies the previous data 95.5(5)% adopted in ENSDF [34].

The derived β -feeding intensity for ¹⁴²Cs $[T_{1/2} = 1.684(14) \text{ s}$, $Q_{\beta} = 7325(9) \text{ keV}]$ is shown in Fig. 4. The details of the A = 142 isobar analysis are given in Refs. [16,35]. We determine the ¹⁴²Cs β feeding to the ¹⁴²Ba ground state to be 44(2)%, as compared to 56% in ENSDF [24]. We reduce the ¹⁴²Cs β feeding to the first-excited 2⁺ state in ¹⁴²Ba from 7% to < 0.5%.

We use the new MTAS data to evaluate $\bar{\nu}_e$ production in nuclear reactors. The change to the emitted $\bar{\nu}_e$ energy spectrum of ¹⁴²Cs is shown in Fig. 5. The fraction of $\bar{\nu}_e$ s with energy above 5 MeV changes from 20% to 14(1)% of the total $\bar{\nu}_e$ flux from ¹⁴²Cs β decay. The fraction of ¹⁴²Cs $\bar{\nu}_e$



FIG. 5. Calculated ¹⁴²Cs $\bar{\nu}_e$ energy spectrum from the data in the present study (black) compared with the expected $\bar{\nu}_e$ energy for the latest ENSDF data (cyan).

with energy below 1.8 MeV increases from 11% to 23(3)%. This reduces the fraction of expected $\bar{\nu}_e$ events detected in a typical $\bar{\nu}_e$ experiment with a threshold of 1.8 MeV. This change is rather typical for complex decays of fission products influenced by the pandemonium effect [31] and corrected by the total absorption technique [36]. It shows the reduction of previously reported β feeding to low-lying states and a corresponding increase of β strength at higher excitations in the daughter nucleus, which shifts the $\bar{\nu}_e$ spectrum to lower energies, thereby reducing the number of $\bar{\nu}_e$ that interact with matter.

The effect on the emitted $\bar{\nu}_e$ energy spectra shown by nuclear fuel component due to the three new MTAS measurements of ⁹²Rb, ^{96gs}Y, and ¹⁴²Cs activities are shown in Fig. 6. The fission fraction yields used to calculate the spectra in Fig. 6 are taken from the Evaluated Nuclear Data File ENDF/B-VII.1 [28] and the decay data is taken from ENSDF. In order to calculate the measured $\bar{\nu}_{e}$ spectrum, the emitted reactor $\bar{\nu}_e$ flux should be weighted by the $\bar{\nu}_e + p \rightarrow e^+ + n$ cross section. This cross section increases quadratically with the $\bar{\nu}_e$ energy [37], so that higher energy $\bar{\nu}_e s$ contribute more substantially to the overall number of measured $\bar{\nu}_e$ interactions. We calculate that there is a reduction in detected $\bar{\nu}_e$ from a typical nuclear reactor of 1.1%. This will be discussed further in Ref. [38]. For a highly enriched nuclear fuel (practically 100% ²³⁵U fissions) used in research reactors, the measured $\bar{\nu}_{e}$ flux from 5 to 7 MeV is reduced by 0.976(+9, -8). For a lowenriched nuclear fuel in a commercial nuclear reactor with fuel fractions of 0.584 ²³⁵U, 0.076 ²³⁸U, 0.29 ²³⁹Pu, and 0.05 ²⁴¹Pu, as adopted in Ref. [8], the measured $\bar{\nu}_{e}$ flux over the 5 to 7 MeV energy range is reduced to 0.977(8) of the ENSDF evaluated $\bar{\nu}_e$ flux. We have performed the same calculations using the Joint Evaluated Fission and Fusion File (JEFF-3.1) [38] and calculate flux reductions from 5 to 7 MeV that are within one σ of the ENDF/B-VII.1 results for all fuel types. This $\sim 2\%$ decrease in reference flux



FIG. 6. Ratio of the emitted $\bar{\nu}_e$ energy spectrum based on the measurements presented in this study to the current ENDF/B-VII.1 by nuclear fuel type, ²³⁵U (black), ²³⁸U (red), ²³⁹Pu (green), and ²⁴¹Pu (blue).

increases the measured high-energy $\bar{\nu}_e$ shoulder by ~2% for energies between 5 and 7 MeV for either nuclear fuel source considered here.

In summary, we have measured the decays of fission products, ⁹²Rb, ^{96gs}Y, and ¹⁴²Cs that are top contributors to the high-energy component of the reactor $\bar{\nu}_e$ spectrum using the Modular Total Absorption Spectrometer. For ⁹²Rb decay, we obtain results close to a recent total absorption measurement [27], with more statistics and a more model independent analysis. We verify previous ground-state to ground-state measurements of $^{96gs}Y \beta$ decay adopted in ENSDF. However, our measurement of 142 Cs β decay leads to a major revision of the decay scheme and the emitted $\bar{\nu}_e$ energy spectrum. There is a reduction to the ¹⁴²Cs contribution to the reactor $\bar{\nu}_e$ should r from 5 to 7 MeV, increasing the reported excess of detected high energy $\bar{\nu}_e$ s. One should note that the ¹⁴²Cs case may be considered as typical among fission products with large β -decay energies that are located in the region of deformed nuclei, which are likely to have high level densities at high excitation energies and a widely distributed β -strength pattern. Such decays have to be measured using the total absorption technique to get reliable β -strength distributions and the corresponding $\bar{\nu}_{e}$ spectra. When compared with the current ENSDF data, for a typical low-enriched uranium fuel mixture used in commercial reactors we see a reduction of 0.977(8) of the expected measured reactor $\bar{\nu}_e$ flux over the 5 to 7 MeV energy range. For a highly enriched uranium fuel used in a typical research reactor we see a decrease in the expected measured $\bar{\nu}_e$ flux of 0.976(+9, -8) over the 5 to 7 MeV energy range. The present findings increase the reported reactor $\bar{\nu}_e$ anomaly ratio by 0.011(1) to 0.96(2), and will be discussed in further detail in Ref. [38]. In addition, the findings enhance the maximum excess of measured high-energy reactor $\bar{\nu}_e$ s in the 5 to 7 MeV range from about 10% to 12%. While the evaluation of new data is likely to reduce or even remove the reactor $\bar{\nu}_e$ anomaly, the 5 to 7 MeV shoulder in the detected reactor $\bar{\nu}_e$ spectra might be further enhanced.

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