Decays of the Three Top Contributors to the Reactor ν[over ¯]_{e} High-Energy Spectrum, ^{92}Rb, ^{96gs}Y, and ^{142}Cs, Studied with Total Absorption Spectroscopy

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Phys. Rev. Lett. **117**, 092501 — Published 22 August 2016

DOI: 10.1103/PhysRevLett.117.092501

The decays of three top contributors to the reactor $\overline{\nu}_e$ high-energy spectrum, ^{92}Rb, ^{96gs}Y, and ^{142}Cs, 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 $\overline{\nu}_e$ spectral shape in nuclear reactors. These three β decays contribute 43% of the $\overline{\nu}_e$ flux near 5.5 MeV emitted by nuclear reactors. This $\overline{\nu}_e$ 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 ^{238}U 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 ^{142}Cs, reducing the β feeding to ^{142}Ba states below 2 MeV by 32% when compared with the latest evaluations. Our results increase the discrepancy between the observed and the expected reactor $\overline{\nu}_e$ flux between 5 and 7 MeV, the maximum excess increases from $\sim$10% to $\sim$12%.

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 $\overline{\nu}_e$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 $\overline{\nu}_e$ measurements that use nuclear reactors as intense $\overline{\nu}_e$ sources [1–6]. The number of measured reactor $\overline{\nu}_e$ interactions with detector matter is 0.95(2) of the expected number of events and is often referred to as the "reactor $\overline{\nu}_e$ anomaly" [3–5]. There is also up to a 10% excess of high-energy $\overline{\nu}_e$ events in the 5 to 7 MeV $\overline{\nu}_e$ 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 $\overline{\nu}_e$ energy spectrum, the associated $\overline{\nu}_e$ 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 single-particle state evolution in neutron-rich nuclei.

One approach toward obtaining a more reliable prediction of the $\overline{\nu}_e$ flux from nuclear reactors is to accurately measure individual β decays of the most important fission products. One way 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 $\overline{\nu}_e$ 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 $\overline{\nu}_e$ flux in this energy region [8, 11]. The three largest contributors, ^{92}Rb, ^{96gs}Y, and ^{142}Cs, are characterized by β transitions to the ground state or to low-energy excited states that dominate the decay pattern and are expected to create 43% of the $\overline{\nu}_e$ 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 ^{92}Rb, ^{96gs}Y, and ^{142}Cs 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 keV 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 three orders of magnitude. In addition to the active background suppression, there are over 5000 kg of lead shielding around MTAS, see [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 de-excitation γ paths known from high-resolution 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 de-excitation 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 de-excitation pattern.

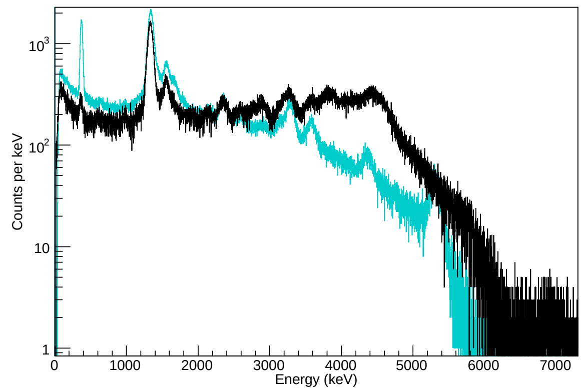

FIG. 1: (Color online) Background subtracted ^{142}Cs MTAS energy spectrum (black) compared to the simulated MTAS response to 1.9×10^6 ^{142}Cs decay events based on the ENSDF data (cyan). We observe β feeding to low lying states (below 2500 keV) to be more weakly populated than previous measurements,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

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 [20], but we have 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 non-active 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

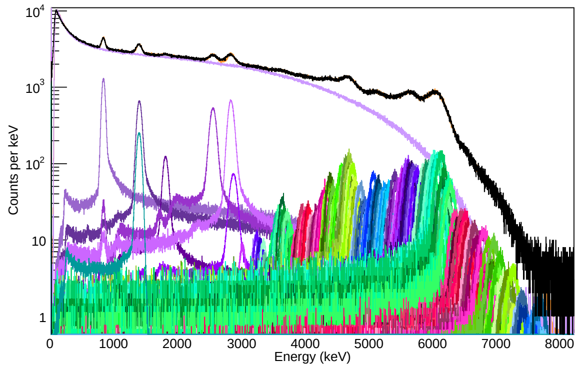

FIG. 2: (Color online) Fit of simulated single γ-ray response functions (colored) to the total ^{92}Rb MTAS data (black). The sum of all the simulated components is indistinguishable from the data. The ground-state β feeding (broad mauve curve) is the dominant contribution to all channels except at the 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.

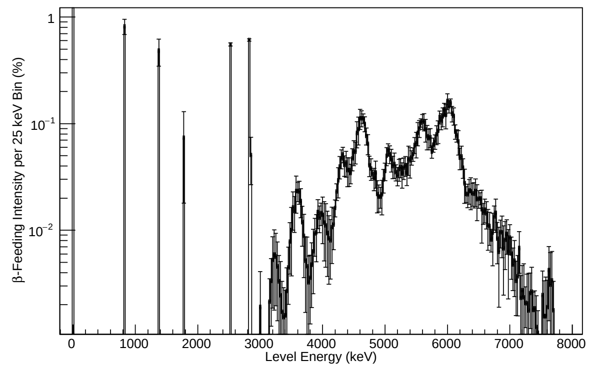

FIG. 3: Average ^{92}Rb β-feeding intensity with the uncertainty based on the number of γ rays in the de-excitation cascade from each level. The fit for the ground state β-feeding is off scale at 91(3)%.

a simulation model of the interior of MTAS that reproduces large ground-state feedings and compares well with off-line measurements of ^{90}Sr 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 $\overline{\nu}_e$ spectra we assume that the 0^- to 0^+ ground-state to ground-state 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 de-exciting 25 keV bins and their sum compared to the measured MTAS spectrum of ^{92}Rb 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 the basis of our analysis of β-strength distributions and their uncertainties.

The derived β-feeding intensity for ^{92}Rb ($T_{1/2}$=4.48(3) s, Q_β=8095(6) keV) is shown in Fig. 3. Earlier reported decay schemes of ^{92}Rb used to derive $\overline{\nu}_e$ 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 [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 ^{92}Rb decay are its daughter activity, ^{92}Sr, 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 $^{96\mathrm{gs}}$Y ($T_{1/2}$=5.34(5) s, Q_β=7103(6) keV) was measured following the decay of implanted ^{96}Rb and ^{96}Sr ions. The isomer $^{96\mathrm{m}}$Y ($T_{1/2}$=9.6 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 $^{96\mathrm{gs}}$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) s) and ^{95}Sr ($T_{1/2}$=23.90(14) s) activities are present. These associated decays have half-lives that bracket $^{96\mathrm{gs}}$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 $^{96\mathrm{gs}}$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 $^{96\mathrm{gs}}$Y, the weak E0 signal is overwhelmed by the ground-state feeding. We assume the β measurements reported for the 1581 keV E0 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].

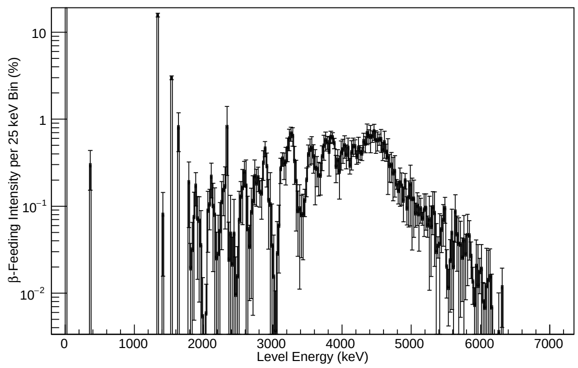

FIG. 4: Average ^{142}Cs β-feeding intensity with the uncertainty based on the number of γ rays in the de-excitation cascade from each level. The fit for the ground state β-feeding is off scale at 44(2)%.

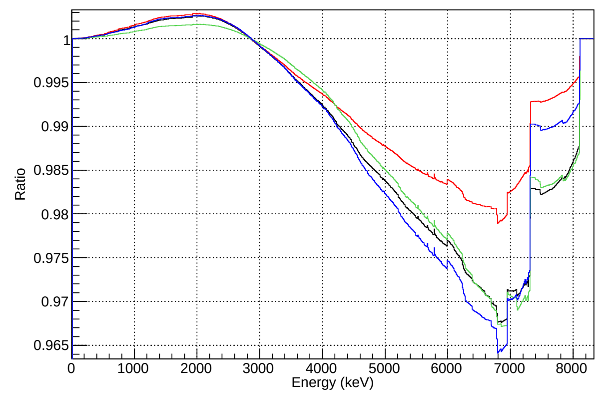

FIG. 6: Ratio of the emitted $\overline{\nu}_e$ energy spectrum based on the measurements presented in this study to the current ENDF/B-VII.1 by nuclear fuel type, ^{235}U (black), ^{238}U (red), ^{239}Pu (green), and ^{241}Pu (blue).

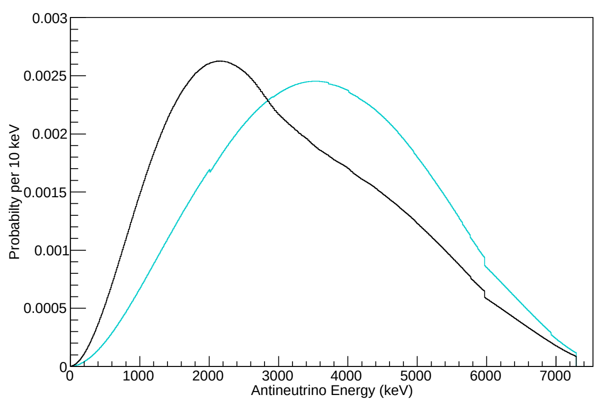

FIG. 5: Calculated ^{142}Cs $\overline{\nu}_e$ energy spectrum from the data in the present study (black) compared with the expected $\overline{\nu}_e$ energy for the latest ENSDF data (cyan).

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

We use the new MTAS data to evaluate $\overline{\nu}_e$ production in nuclear reactors. The change to the emitted $\overline{\nu}_e$ energy spectrum of ^{142}Cs is shown in Fig. 5. The fraction of $\overline{\nu}_e$s with energy above 5 MeV changes from 20% to 14(1)% of the total $\overline{\nu}_e$ flux from ^{142}Cs β decay. The fraction of ^{142}Cs $\overline{\nu}_e$ with energy below 1.8 MeV increases from 11% to 23(3)%. This reduces the fraction of expected $\overline{\nu}_e$ events detected in a typical $\overline{\nu}_e$ 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 $\overline{\nu}_e$ spectrum to lower energies thereby reducing the number of $\overline{\nu}_e$ that interact with matter.

The effect on the emitted $\overline{\nu}_e$ energy spectra by nuclear fuel component due to the three new MTAS measurements of ^{92}Rb, ^{96gs}Y, and ^{142}Cs 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 $\overline{\nu}_e$ spectrum, the emitted reactor $\overline{\nu}_e$ flux should be weighted by the $\overline{\nu}_e$ + p $\rightarrow$ e$^+$ + n cross section. This cross section is proportional to the square of the $\overline{\nu}_e$ energy [37], so that higher energy $\overline{\nu}_e$s contribute more substantially to the overall number of measured $\overline{\nu}_e$ interactions. We calculate that there is a reduction in detected $\overline{\nu}_e$ from a typical nuclear reactor of 1.1%. This will be discussed further in [38]. For a highly-enriched nuclear fuel (practically 100% ^{235}U fissions) used in research reactors, the measured $\overline{\nu}_e$ flux from 5 to 7 MeV is reduced by 0.976(+9,-8). For a low-enriched nuclear fuel in a commercial nuclear reactor with fuel fractions of 0.584 ^{235}U, 0.076 ^{238}U, 0.29 ^{239}Pu, and 0.05 ^{241}Pu, as adopted in [8], the measured $\overline{\nu}_e$ flux over the 5 to 7 MeV energy range is reduced to 0.977(8) of the ENSDF evaluated $\overline{\nu}_e$ 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 increases the measured high-energy $\overline{\nu}_e$ shoulder by $\sim$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, ^{92}Rb, ^{96gs}Y, and ^{142}Cs that are top contributors to the high-energy component of the reactor $\overline{\nu}_e$ spectrum using the Modular Total Absorption Spectrometer. For ^{92}Rb 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 β decay adopted in ENSDF. However, our measurement of ^{142}Cs β decay leads to a major revision of the decay scheme and the emitted $\overline{\nu}_e$ energy spectrum. There is a reduction to the ^{142}Cs contribution to the reactor $\overline{\nu}_e$ shoulder from 5 to 7 MeV, increasing the reported excess of detected high energy $\overline{\nu}_e$s. One should note that the ^{142}Cs 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 $\overline{\nu}_e$ 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 $\overline{\nu}_e$ 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 $\overline{\nu}_e$ flux of 0.976(+9,-8) over the 5 to 7 MeV energy range. The present findings increase the reported reactor $\overline{\nu}_e$ anomaly ratio by 0.011(1) to 0.96(2), and will be discussed in further detail in [38]. In addition, the findings enhance the maximum excess of measured high-energy reactor $\overline{\nu}_e$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 $\overline{\nu}_e$ anomaly, the 5 to 7 MeV shoulder in the detected reactor $\overline{\nu}_e$ spectra might be further enhanced.

We would like to thank the ORNL Tandem operations staff for providing the excellent quality proton beams necessary for this work. This research was also sponsored by the Office of Nuclear Physics, U. S. Department of Energy under contracts DE-AC05-00OR22725 (ORNL), DE-FG02-96ER40983 (UTK), DE-FG02-96ER40978 (LSU), DE-FG02-96ER41006 (MSU), DE-FG-05-88ER40407 (VU), and by the Polish National Science Center under contracts UMO-2015/18/E/ST2/00217 and UMO-2013/08/T/ST2/00624.

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