Beta-strength and anti-neutrino spectra from total absorption spectroscopy of a decay chain ^{142}Cs→^{142}Ba→^{142}La

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Abstract. Beta decays of mass A = 142 isobaric chain starting from ^{142}Cs have been investigated by means of Modular Total Absorption Spectrometer (MTAS) and on-line mass separation at Oak Ridge National Laboratory. The beta strength distribution derived for ^{142}Cs decay from MTAS spectra is showing significant differences in β-feeding pattern when compared to the values listed at nuclear databases. MTAS results are shifting the associated anti-neutrino energy spectrum towards lower energies. A decay pattern deduced for ^{142}Ba is similar to earlier reported results.

1. Introduction

The Modular Total Absorption Spectrometer (MTAS) array has been designed, constructed, characterized and applied to the decay studies of ^{238}U fission products at the Holifield Radioactive Ion Beam Facility (HRIBF) at Oak Ridge National Laboratory [1–3]. MTAS measurements yield true β-strength distribution verifying and helping to develop the microscopic description of β-decay. In particular, the decay heat release in fission products during a nuclear fuel cycle as well as respective anti-neutrino energy spectra can be determined from the complete β-decay scheme. Over seventy decay products of ^{238}U fission products measured at ORNL include 22 decays of high priority for decay heat analysis [4]. These measurements include the ^{142}Cs activity assessed recently as the third most important contributor to the high energy $\bar{\nu}$ spectra in nuclear reactors [5–7]. The significant differences between β-feeding values derived from MTAS measurement and Evaluated Nuclear Structure Data Files (ENSDF) database are substantially impacting the reference reactor anti-neutrino flux and its spectral properties, see, e.g., [8].

2. The modular total absorption spectrometer

MTAS measurement campaigns were performed in January 2012, March 2015, October-December 2015 and January 2016 at the On-Line Test Facility, namely the mass separator on-line to the ORNL Tandem accelerator. MTAS detector array consists of 19 NaI(Tl) hexagonal shape detectors. Each crystal is ∼53 cm long and about 20 cm maximum diameter. The weight of NaI(Tl) detector material in MTAS array is ∼1000 kg, covering over 99% of the solid angle around the measured activities. The entire array is surrounded by over 5000 kg of lead and SWX-227A neutron shielding foam. This shielding reduces the laboratory gamma background by a factor of 1000. MTAS auxiliary detectors include two 1-mm-thick Si β-trigger detectors (each divided into 7 strips, each ∼8.5 mm wide), placed at the center of the NaI(Tl) array around the tape transporting the activities [1]. The signals detected in these silicon counters are used as β-triggers. Applying coincidence condition between silicon detectors and MTAS suppresses typical laboratory background by at least three orders of magnitude. The γ-ray efficiency for full-energy deposition of a single γ-ray in on-line conditions is about 81% at 300 keV and about 71% at 6 MeV [3]. The MTAS thresholds for individual detector, each from 18 regular modules, is about 25 keV. For the central module it is around 30 keV.

3. MTAS measurement and ENSDF database comparison

We evaluate the energy spectra measured with MTAS to determine β-feeding intensities to known and unknown

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Table 1. The intensities of β-transitions in ^{142}Ba decay determined from MTAS measurement in comparison to the value from ENSDF and TAGS results of Ref. [14].

| level energy [keV]	I_β ENSDF	I_β TAGS [11]	I_β MTAS
225.3	3.5(7)	4.6	3.8
425.0	5.4(2)	6.0	5.8
1204.3	46(2)	46	45.8

energy levels in the daughter nuclei, including the ground state. The evaluation method is based primarily on the techniques applied in the analysis of other total absorption spectrometers spectra, see e.g., [9–11]. However, our analysis is profiting from the increased efficiency and high segmentation of MTAS [2,3,8]. The MTAS detector response function was simulated with the GEANT4 toolkit. This generated MTAS response function is verified with through measurements of single γ-ray and γ-cascades emitted in the decay of calibration sources like ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{60}Co and ^{24}Na. MTAS energy spectra are divided into two response regimes. These correspond to energy levels below threshold energy with de-excitation γ-paths known from high energy resolution gamma spectroscopy data and to unknown β-fed levels above the threshold energy grouped in 25 keV wide bins. More details of MTAS spectra analysis method are given in [1–3,8]. In this paper we present MTAS results on measured ^{142}Cs and ^{142}Ba activities compared to GEANT4 simulations using currently available ENSDF entries [12]. Beta-gated MTAS spectra are analyzed to reduce background. Also the pile-up analysis is included into our evaluation procedure [2]. The decay chain of the A = 142 includes the very short-lived ^{142}Cs (1.7 s) isotope which decays to ^{142}Ba activity having a half-life of 10.6 min, followed by the decay of ^{142}La (91.1 min) to stable ^{142}Ce.

3.1. Decay of ^{142}Ba

There were several measurements of ^{142}Ba decay, see, e.g., [13], and summarized recently in Nuclear Data Sheets [12]. These measurements were done using high-resolution Ge detectors as well as total absorption gamma spectroscopy (TAGS) [14]. Interestingly, the ^{142}Ba decay is a rare case where the high-resolution measurements point to practically the same β-decay pattern as deduced from the TAGS measurement. It could be explained by a relatively low beta decay energy value of $Q_\beta = 2212(5)$ keV. Indeed, ^{142}Ba MTAS spectrum is in general agreement with earlier data (Fig. 1). Figure 2 presents the comparison of an overall fit of the simulated decays following the ENSDF entry for the ^{142}La level scheme populated in ^{142}Ba decay (colored components) and their sum (orange), to the measured MTAS spectrum (black). The sum of the individual decay paths follows well MTAS measured data. The γ-rays with intensity much smaller then 10% of the strongest γ-ray de-exciting each level were not taken into account in this particular simulation.

The intensities of β-feeding determined from MTAS measurement were compared to the values from ENSDF entries and Greenwood's measurements [14]. Examples of the results for main levels are listed in Table 1.
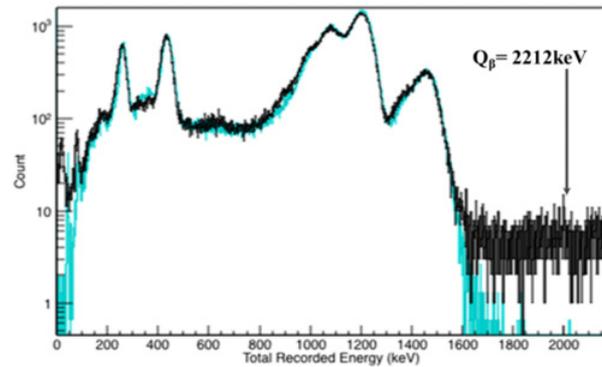
Figure 1. Background subtracted ^{142}Ba MTAS data (black) compared to the simulated MTAS response for ^{142}Ba decay events based on the ENSDF entry (cyan).
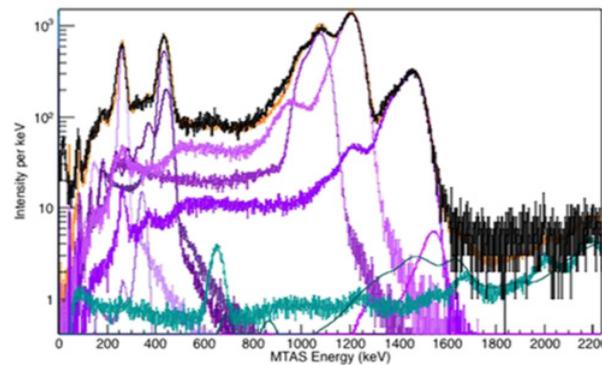

Figure 2. Fit of simulated γ-ray response functions (colored) for complex decays of individual levels to the total ^{142}Ba->142La MTAS decay data (black). The sum of all the simulated components (orange) is in good agreement with an experimental spectrum.

3.2. Decay of ^{142}Cs

The MTAS-measured energy spectrum for ^{142}Cs decay is compared to the simulated MTAS response using decay data listed in the current ENSDF entry in Fig. 3. The reduction of the ground-state β-feeding and the β-feeding to the first-excited 2^+ state at 360 keV is clearly visible, as well as the presence of new β-fed levels at higher excitation energies. The derived β-feeding intensities for ^{142}Cs decay with $Q_\beta = 7325(9)$ keV are presented in Fig. 4. The MTAS-determined ^{142}Cs β-feeding to the ^{142}Ba ground-state is 44(2)%, lower than 56% listed in ENSDF. The upper limit for beta-feeding to the first-excited 2^+ state in ^{142}Ba is 0.5% [8], at least an order of magnitude lower than reported earlier 7% [12].

## 4. High energy part of reactor anti-neutrino spectrum for ^{142}Cs

The ^{142}Cs beta decay was found to be among top 3 contributors to the high energy reactor $\bar{\nu}$ spectrum in recent evaluations [5–7]. We have re-evaluated the spectrum of anti-neutrinos emitted in this decay basing on new MTAS data. The change in the $\bar{\nu}$ energy spectrum of ^{142}Cs is shown in Fig. 5. The fraction of $\bar{\nu}$ with energy above 5 MeV changes from 20% to 14(1)% the total $\bar{\nu}$ flux from ^{142}Cs decay, while the fraction of ^{142}Cs $\bar{\nu}$ with energy below 1.8 MeV increases from 11% to 23(3)%
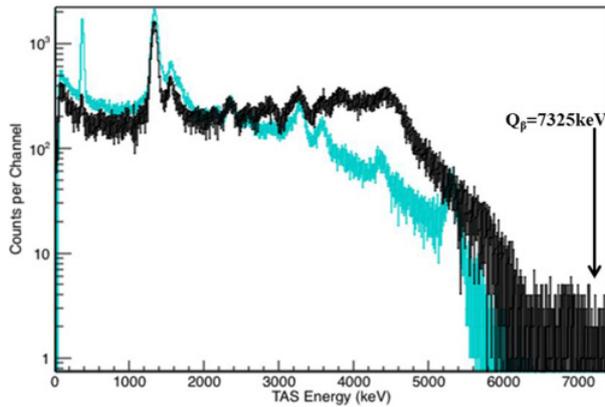
Figure 3. Background subtracted ^{142}Cs MTAS data (black) compared to the simulated MTAS response to ^{142}Cs decay events based on the ENSDF data (cyan), see [8].
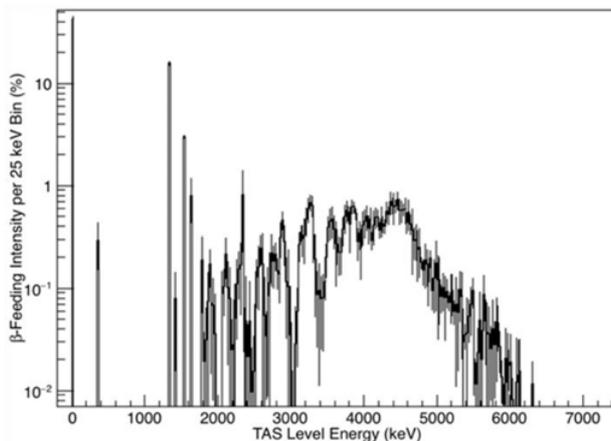
Figure 4. Average ^{142}Cs β-feeding intensity with the uncertainty based on the number of γ-rays in the de-excitation cascade from each level. The fit for the ground state β-feeding is at 44(2)%.
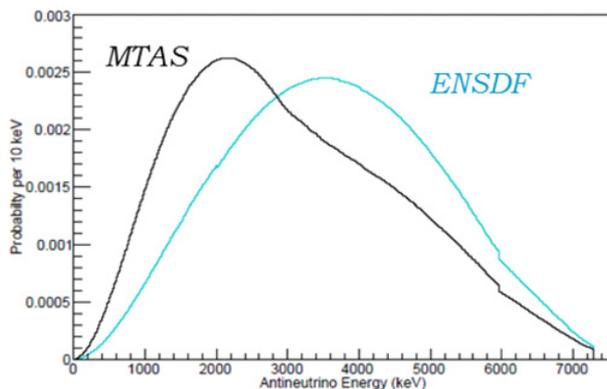
Figure 5. ^{142}Cs $\bar{v}$ energy spectrum derived from MTAS data (black) compared to the $\bar{v}$ energies obtained from the latest ENSDF entry (cyan).

[8]. Such change can be expected for complex decays of fission products, with high Q-value and influenced by the Pandemonium Effect [15] after the measurement using the total absorption technique. It shows the reduction of previously reported feeding to low-lying states and a corresponding increase of β-strength at higher excitations in the daughter nucleus. It shifts the corresponding $\bar{v}$ spectrum to lower energies.

These changes reduce the number of anti-neutrino interactions with matter. The modification of ^{142}Cs decay scheme alone reduced the reported reactor $\bar{v}$ anomaly of 94.6(22)% [16] to 95.7(22)%. Simultaneously, it increased the high-energy "shoulder" from $\sim$ 10% to $\sim$ 12% excess, compare [5–8,16].

5. Summary

The Modular Total Absorption Spectrometer (MTAS) was used to measure the beta-gamma activities in the mass A = 142 decay chain. Two isobars from this chain, ^{142}Ba and ^{142}Cs, are presented here. For both of them, the new β-feeding pattern was obtained. For ^{142}Ba decay, with low Q$_\beta$ of 2.2 MeV, MTAS spectra analysis reproduce known decay pattern obtained previously using high resolution gamma spectroscopy as well as total absorption measurements [14]. MTAS measurements of ^{142}Cs, with much higher Q$_\beta$ of 7.3 MeV, resulted in a major modification of the decay scheme. The ground-state β-feeding is reduced from 56% (ENSDF) to 44(2)% (MTAS). For the β-feeding to the first-excited 2$^+$ state at 360 keV only a limit of β-feeding $\sim$0.5% can be given, while the ENSDF value is listed as 7%. Our measurements are showing also the presence of new β-fed levels at higher excitation energies in ^{142}Ba. MTAS measurements confirmed the importance of ^{142}Cs decay as one of the top three contributors to the high energy $\bar{v}$ spectral shape in nuclear reactors, and as so far the most important activity changing the expected high-energy anti-neutrino flux. MTAS results for ^{142}Cs shift the total $\bar{v}$-spectrum toward lower energies. It results in a reduction of reactor $\bar{v}$ anomaly, roughly from 94.6(22)% to 95.7(22)% and is enhancing high-energy "bump" from $\sim$ 10% to $\sim$ 12% excess, for the reference anti-neutrino flux [8] calculated using a summation method.

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