

TOTAL ABSORPTION SPECTROSCOPY OF FISSION FRAGMENTS RELEVANT FOR REACTOR ANTINEUTRINO SPECTRA DETERMINATION*

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The contribution of each fission fragment to the reactor antineutrino spectra was determined using the summation method based on the existing information on fission yields and decay data contained in nuclear databases and the reactor evolution code MURE. The beta decay of some of the main contributors has been studied using the Total Absorption Spectroscopy (TAS) technique during two experimental campaigns at the IGISOL facility, in Jyväskylä (Finland). Results on the decay of ^{92}Rb , the most important contributor in the 4–8 MeV energy region are reported. The status of the analysis of the second experiment is presented as well.

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1. Introduction

Nuclear reactor cores are intense sources of antineutrinos, for instance, in standard 900 MWe Pressurized Water Reactors (PWR) there are produced around 10^{21} antineutrinos per second. This huge flux allows for studying neutrino physics despite the low interaction cross section that those elusive particles offer to the experimentalists. The antineutrino detection is usually done through the inverse beta decay process in which the incoming antineutrino interacts with a proton producing a neutron and a positron

$$\bar{\nu}_e + p \rightarrow n + e^+. \quad (1)$$

This mechanism is used in reactor antineutrino experiments such as Double Chooz (France) [1], Daya Bay (China) [2] and RENO (South Korea) [3]. Two detectors are used in those experiments, one near the reactor to measure the emitted antineutrino spectra and one at $\approx 1\text{--}2$ km to measure the oscillated antineutrino spectra. In this way, the magnitude of the oscillation parameter θ_{13} is studied. However, the Double Chooz experiment took data only with the far detector from 2011 to December 2014 when the near detector was finally installed and operative. For this reason, the data analysis to estimate the oscillation parameter θ_{13} demanded an alternative determination of the emitted antineutrino spectra.

A smaller rate of emitted antineutrinos than predicted was found for different experiments located at short distances from the reactor core, where no neutrino oscillation is expected to occur. This is the so-called “reactor anomaly” [4] and motivated the search for a new type of neutrinos, the *sterile* neutrinos.

The results of the three neutrino experiments [1–3] presented an unexplained distortion in the positron energy spectra in the 4–8 MeV region [5]. It is evident when plotting the measured *versus* expected ratio for the near detectors (except for Double Chooz as the near detector was not available yet) where no oscillation is supposed to occur. The origin of this distortion remains to be clarified. For these two main reasons, an alternative determination of the emitted antineutrino spectra from a nuclear reactor is needed.

Usually, there are two ways to accomplish such task, first, the *conversion method* [6], in which the measured integral beta spectra are converted into antineutrino spectra under certain assumptions. Second, the *summation method* [7], in which one adds the individual contributions from each decay branch of the fission fragments. The latter method allows for calculating reactor antineutrino spectra when no integral beta spectrum has been measured and it is the only method useful as a predictive tool for future nuclear reactors. For these reasons, our group decided to use the summation method as a tool for calculating antineutrino spectra. However, the summation method requires reliable information on the beta decay of, at least, the most relevant contributors. This means that the information on the nuclear databases should be reliable in terms of Q values, beta feeding distributions, types of beta transition (allowed, 1st forbidden, 2nd forbidden, etc.) and fission yields for each fissionable isotope.

1.1. Pandemonium effect and total absorption spectroscopy technique

High resolution spectroscopy based on HPGe detectors is the main technique used for studying beta decay properties of nuclei through the measurement of the gamma rays from the de-excitation of the daughter nucleus. The low efficiency of these detectors for the detection of high-energy gamma-rays and the high fragmentation of the de-excitation paths from levels located at high excitation energies make difficult the measurement of beta feeding at high excitation energies. This experimental difficulty was first underlined by Hardy *et al.* [8] and it is usually referred to as *Pandemonium* effect. It causes an underestimation of feeding for levels at high excitation energies and an overestimation for levels at low excitation energies. As a consequence, the average gamma energies are underestimated and the average beta and neutrino energies are overestimated in the nuclear databases for nuclei which were studied using High Resolution Spectroscopy techniques. The usual way to overcome this effect is the Total Absorption Spectroscopy (TAS) technique. Using a large scintillation crystal of high intrinsic efficiency and ideally covering 4π solid angle, one is sensitive to the full de-excitation cascade and, therefore, the beta feeding rather than the individual gamma rays.

The impact of this effect on the calculation of the reactor decay heat due to the gamma and beta radiation emitted at the reactor cores was presented in Ref. [9]. Those TAS results on the decay of $^{102,104-107}\text{Tc}$, ^{105}Mo , and ^{101}Nb nuclei [9, 10] were found remarkably relevant in the determination of the reactor antineutrino spectra as well as it was presented in Ref. [11]. Figure 2 of that article shows the ratio of the antineutrino spectra calculated with and without the new TAS data for the four main fissionable isotopes.

The impact was up to 8% for $^{239,241}\text{Pu}$, around 3.5% for ^{238}U and 1.5% for ^{235}U in the energy range from 0 to 6 MeV. One observes as the new TAS results displace some statistics in the energy spectra from high neutrino energies ($2 < E < 6$ MeV) to lower energies ($E < 2$ MeV).

2. Experiments

An evaluation of the most relevant contributors to the reactor antineutrino spectra was done using the summation method [7], the nuclear data considered in Ref. [11] and the MCNP Utility for Reactor Evolution (MURE) [12]. In the simulations, it was assumed a 450 days irradiation period and a standard fuel composition for PWR reactors of 52% ^{235}U , 33% ^{239}Pu , 6% ^{241}Pu and 8.7% ^{238}U . The results are shown in Table I and were already published in Ref. [13]. They indicated ^{92}Rb as the most relevant case in the energy region of 4–8 MeV with special importance at 6–8 MeV.

The most feasible cases from the experimental point of view among those shown in Table I were selected under the criteria of production yields and decay properties. Two experiments were carried out devoted to study them together with other cases of relevance for reactor decay heat calculations.

TABLE I

Main contributors to a standard PWR antineutrino energy spectrum computed with the MURE code [12], coupled with the list of nuclear data given in Ref. [11], assuming a fuel composition of 52% ^{235}U , 33% ^{239}Pu , 6% ^{241}Pu and 8.7% ^{238}U , for a 450 day irradiation period and using the summation method described in [11]. Values are given in percentage (table from Ref. [13]).

	4–5 MeV	5–6 MeV	6–7 MeV	7–8 MeV
^{92}Rb	4.74	11.49	24.27	37.98
^{96}Y	5.56	10.75	14.10	
^{142}Cs	3.35	6.02	7.93	3.52
^{100}Nb	5.52	6.03		
^{93}Rb	2.34	4.17	6.78	4.21
^{98m}Y	2.43	3.16	4.57	4.95
^{135}Te	4.01	3.58		
^{104m}Nb	0.72	1.82	4.15	7.76
^{90}Rb	1.90	2.59	1.40	
^{95}Sr	2.65	2.96		
^{94}Rb	1.32	2.06	2.84	3.96

2.1. Jyväskylä 2009 experiment

Several rubidium and bromine isotopes were studied in this experiment performed at the IGISOL facility [14] in the University of Jyväskylä in 2009. Proton-induced fission on an uranium target was the production mechanism to obtain the radioactive isotopes. The JYFLTRAP double Penning trap was used to extract a very clean beam of ^{92}Rb using the mass-selective buffer gas cooling technique [15]. The purity of the beam is a key issue for TAS measurements since the removal of contaminations in the analysis induces difficult-to-estimate systematic uncertainties to the beta-feeding distribution. The *Rocinante* TAS spectrometer, composed of 12 BaF_2 crystals arranged in a cylindrical shape of diameter 25 cm and height 25 cm, was used. A silicon detector was placed close to the measurement point to tag on the beta particles and remove contributions to the TAS spectra from the room background. Figure 1 (left) shows schematically the experimental setup and a picture of the TAS detector used.

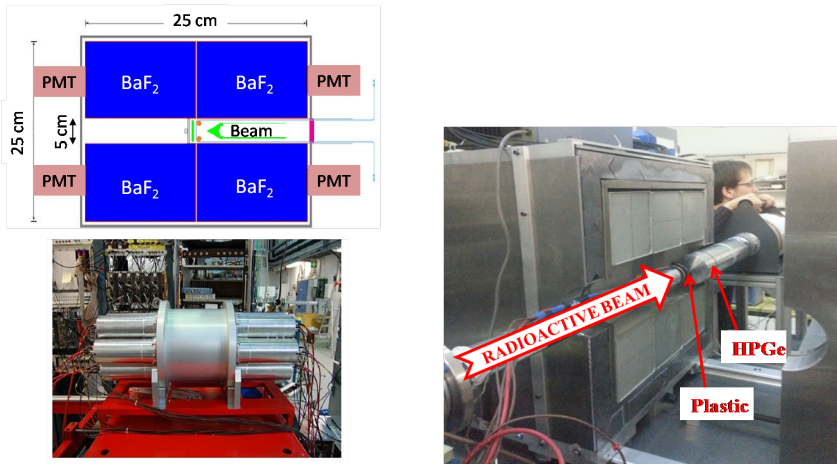


Fig. 1. (Left) The *Rocinante* Total Absorption Spectrometer used in the 2009 experiment at IGISOL facility at the University of Jyväskylä. (Right) DTAS detector used in the 2014 experiment.

2.1.1. Results on ^{92}Rb decay

The data analysis was performed using TAS β -tagged spectra to suppress the room background contamination. The analysis was performed following the TAS data analysis described in Ref. [16]. The results obtained for ^{92}Rb decay have been recently published in Ref. [13]. The feeding distribution from this analysis has been convoluted with the response of our detector to this particular decay in order to obtain the recalculated spectrum that is shown in comparison with the experimental one in Fig. 2 (a).

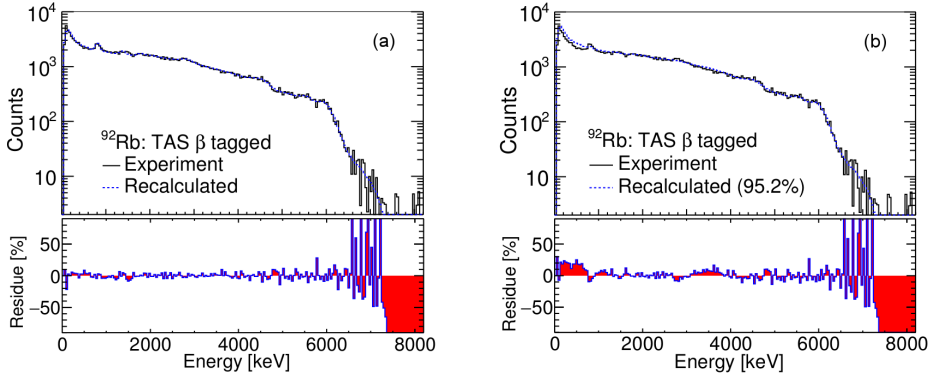


Fig. 2. (a) Comparison of experimental and reconstructed spectra found in our TAS analysis of ^{92}Rb decay (figure from Ref. [13]). (b) The same as (a) when fixing the direct ground state feeding to the value of 95.2% reported in Ref. [17].

A relevant result is the direct ground state feeding. We obtain a 87.5(25)% feeding in contrast with the previously reported value of 95.2(7)% in ENSDF database [17]. Our value provides the best reproduction of the experimental spectrum, $\chi^2 = 630$, and if one fixes the ground state feeding in our analysis to the 95.2% reported in Ref. [17], the value of the χ^2 grows to 2048 and the reproduction of the experimental spectrum is much worse, specially in the low-energy region, as it can be seen in Fig. 2 (b). The resulting feeding distribution is shown in Fig. 3 (left) in comparison with the previous knowledge on this decay from high resolution studies as registered in ENSDF database [17]. We can observe that the feeding placed at states between 800 and 2000 keV were previously underestimated.

The impact of our results on the calculation of the antineutrino spectra is presented in Fig. 3 (right). The plot shows the ratio between the antineutrino spectra calculated with and without our results on ^{92}Rb for the four main fissionable isotopes, $^{235,238}\text{U}$ and $^{239,241}\text{Pu}$. Three different sets of nuclear data were evaluated and the results are shown in Fig. 3(b). The curve showing values larger than 1 for the ratio with/without the new TAS data in the 5-8 MeV energy region (black curve in the online version) is the one where more impact is found, as expected, since an old version of the ENSDF database was used in Ref. [19]. It reaches an impact of up to 27% correction for the case of ^{235}U . The other two datasets explored show very similar results and give values smaller than 1 in the 5-8 MeV energy region. Those curves correspond to the data set used in Ref. [11] (red line in the online version) and that from Ref. [18] (green line in the online version) which is basically the most updated ENSDF database [17]. For those two datasets, corrections of 4.5% for ^{235}U , 3.5% for ^{239}Pu , 2% for ^{241}Pu , and 1.5% for ^{238}U are found.

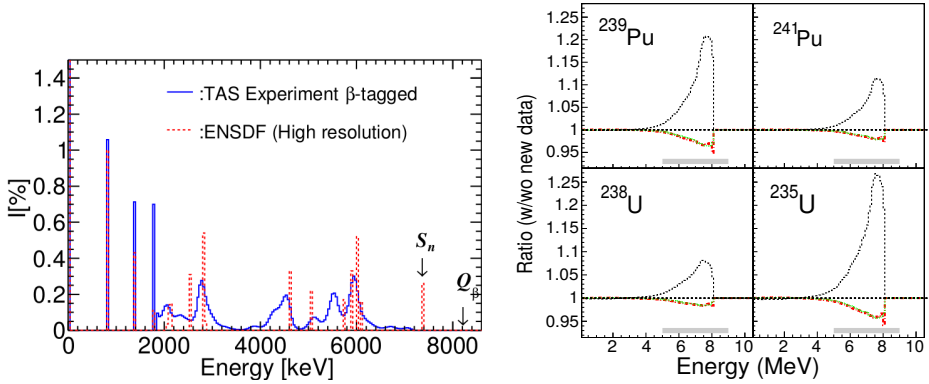


Fig. 3. (Left) Comparison of beta feeding distribution of ^{92}Rb decay found in our work [13] with the information in the ENSDF database [17]. (Right) Ratio of the antineutrino spectra calculated with and without the new TAS information on ^{92}Rb decay for the four main fissionable isotopes. Three sets of nuclear data are evaluated: Ref. [11] in red, Ref. [18] in green and Ref. [19] in black (figures from Ref. [13]).

2.2. Jyväskylä 2014 experiment

The second experiment was carried out in 2014 at the IGISOL facility as well [20], after the upgrade of the facility to the new IGISOL-4 [21]. The new DTAS detector [22], developed for the future DESPEC experiment at FAIR Facility in GSI, Darmstadt (Germany), was first used in this experiment. It is composed of 18+1 NaI(Tl) crystals of dimensions $15 \times 15 \times 25 \text{ cm}^3$. A picture of the experimental setup can be seen in Fig. 1 (right). The measurement of 12 nuclei interesting from the point of view of antineutrino spectra determination was done together with 11 cases interesting for reactor decay heat calculations. The data analysis is currently on-going including calibrations and test of the *Geant4* simulations. These simulations describe the experimental setup in a detailed way with the aim of obtaining the response of the TAS detector to the radiation involved in the decays to study. The validation of these simulations is done by the comparison of experimental and simulated spectra of calibration sources.

3. Summary and conclusions

An alternative determination of the reactor antineutrino spectra is required by reactor neutrino experiments. The summation method needs reliable information from nuclear databases. However, the beta decay of some fission fragments studied using the high resolution spectroscopy suffers from the *Pandemonium* effect. The use of the TAS technique provides a way to determine feeding distributions in a more reliable manner.

The most relevant contributors to the reactor antineutrino spectra were determined using the summation method and the reactor evolution code MURE. Two experiments to study the most feasible cases have been performed at the IGISOL facility in Jyväskylä (Finland).

Results obtained for the most relevant contributor in the 4–8 MeV range, ^{92}Rb , as well as the status of the data analysis of the second experiment are presented.

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