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TAGS measurements of ^{100}Nb ground and isomeric states and ^{140}Cs for neutrino physics with the new DTAS detector

V. Guadilla[1a], A. Algora[1,2], J.L. Tain[1], J. Agramunt[1], J. Äystö[3], J.A. Briz[4], A. Cucoanes[4], T. Eronen[3], M. Estienne[4], M. Fallot[4], L.M. Fraile[5], E. Ganioğlu[6], W. Gelletly[1,7], D. Gorelov[3], J. Hakala[3], A. Jokinen[3], D. Jordan[1], A. Kankainen[3], V. Kolhinen[3], J. Koponen[3], M. Lebois[8], T. Martinez[9], M. Monserrate[1], A. Montaner-Pizá[1], I. Moore[3], E. Nácher[10], S.E.A. Orrigo[1], H. Penttilä[3], I. Pohjalainen[3], A. Porta[4], J. Reinikainen[3], M. Reponen[3], S. Rinta-Antila[3], B. Rubio[1], K. Rytkönen[3], T. Shiba[4], V. Sonnenschein[3], A.A. Sonzogni[11], E. Valencia[1], V. Vedia[5], A. Voss[3], J.N. Wilson[8], and A.-A. Zakari-Issoufou[4]

[1] Instituto de Física Corpuscular CSIC-Universidad de Valencia, 46071 Valencia, Spain
[2] Institute of Nuclear Research of the Hungarian Academy of Sciences, 4026 Debrecen, Hungary
[3] University of Jyvaskyla, Department of Physics, PO Box 35, 40014, University of Jyvaskyla, Finland
[4] Subatech, CNRS/IN2P3, Nantes, EMN, 44307 Nantes, France
[5] Universidad Complutense, Grupo de Física Nuclear, CEI Moncloa, 28040 Madrid, Spain
[6] Department of Physics, Istanbul University, 34134 Istanbul, Turkey
[7] Department of Physics, University of Surrey, GU2 7XH Guildford, UK
[8] Institut de Physique Nucléaire d'Orsay, 91406 Orsay, France
[9] Centro de Investigaciones Energéticas Medioambientales y Tecnológicas, 28040 Madrid, Spain
[10] Instituto de Estructura de la Materia, CSIC, 28006 Madrid, Spain
[11] NNDC, Brookhaven National Laboratory, Upton, NY 11973-5000, USA

Abstract. In this work we report on total absorption γ-ray spectroscopy measurements of the β decay of fission products that are important contributors to the antineutrino spectrum. The experiment was performed at IGISOL as a part of a campaign of measurements with the new DTAS spectrometer. Preliminary results of the analysis of the β decay of ^{100}Nb,^{100m}Nb and ^{140}Cs are presented.

1. Introduction

Nuclear power plants produce on average 6 antineutrinos per fission due to the β decay of fission products, which means a flux of $\sim 10^{20}$ $\overline{\nu}$/s from a 1 GW thermal reactor. For this reason, nuclear reactors are used in neutrino oscillation experiments, such as Double Chooz [1], Daya Bay [2], and Reno [3], that aim to improve the value of the neutrino oscillation mixing angle θ_{13}. In a recent study, thanks to an improvement in antineutrino spectrum calculations, a global deficit of the neutrino flux at close baseline experiments has been found [4]. This shows the necessity of a good understanding of the antineutrino spectrum.

One possible way to calculate the reactor antineutrino spectrum is the summation approach, which uses information from nuclear databases. The total spectrum is calculated as the sum of the spectra associated to the decay of each fission product weighted by the corresponding fission yield, as presented in Eq. (1).

$$S(E_{\overline{\nu}_e}) = \sum_i \left(Y_i \times \sum_j I_{ij} S_{ij}(E_{\overline{\nu}_e}) \right) \tag{1}$$

where Y_i is the cumulative fission yield of the fission product i, I_{ij} is the decay intensity to the daughter level j, and $S_{ij}(E_{\overline{\nu}})$ is the antineutrino spectrum for the transition with endpoint energy $Q_\beta - E_j$.

One of the main advantages of this method, is the possibility to identify the most important contributors to the spectrum in any energy range. In particular, it has been pointed that ^{100}Nb is one of the most relevant decays, with a contribution of 5.52% to the total antineutrino spectrum in the energy range 4–5 MeV according to [5]. Likewise, the other decay presented here, ^{140}Cs, contributes a 3.4% to the antineutrino spectrum from ^{235}U around 4 MeV [6].

On the other hand, the main disadvantage of this method is related both to the inaccuracy of the fission yields and to the incompleteness of the decay schemes. Related to the latter, it is known that the *Pandemonium* systematic error [7] may affect the I_β data from high resolution experiments with germanium detectors. This effect is due to the modest efficiency of these detectors, and it implies an underestimation of the feeding to high energy levels. A method to avoid this error is the Total Absorption γ-Ray Spectroscopy (TAGS) technique, that uses large scintillator crystals covering a solid angle close to 4π to absorb the full γ-cascades of the de-excitation of the daughter nucleus after β-decay. The TAGS technique has already shown its potential to improve decay data

[a] e-mail: `guadilla@ific.uv.es`

of importance for the reactor antineutrino spectrum calculation [5,8,9], and priority TAGS measurements have been defined by the IAEA [10], with ^{100}Nb and ^{140}Cs among the high priority cases. They are also on NEA/IAEA lists of important contributors to reactor decay heat [10,11].

In order to determine the I_β distribution with this technique, the inverse problem represented by Eq. (2) has to be solved [12].

$$d_i = \sum_{j}^{levels} R_{ij}(B) f_j \qquad (2)$$

where d_i is the number of counts in the experimental channel i, f_j represents the number of events that feed level j in the daughter nucleus, and R_{ij} is the response function of the detector, that depends on the branching ratio matrix of the decay (B) and is calculated by means of Monte Carlo (MC) simulations [13].

2. Experiment

Measurements of fission products of interest were performed with beams provided by the mass separator of the upgraded IGISOL IV facility [14] (Jyväskylä, Finland). Nuclear species were produced by proton-induced fission on a natural uranium target. One of the main advantages of this facility is the possibility of precision trap-assisted separation with the JYFLTRAP double Penning trap [15]. Beams were purified in this system before being implanted onto a tape placed at the centre of our spectrometer and in front of a plastic β detector. The cycles of the tape transport system were optimized for the half-life of each decay. Finally, in our set-up we also placed a HPGe detector behind the β detector in order to check for possible contaminants. In these measurements, we used the new Decay Total Absorption γ-ray Spectrometer (DTAS) [16], that was commissioned with radioactive beams in this experiment [17]. This is a 18-fold segmented NaI(Tl) spectrometer with rectangular crystals of 150 mm $\times$ 150 mm $\times$ 250 mm that has been designed and constructed for the DEcay SPEctroscopy (DESPEC) [18] experiment at FAIR. The total efficiency of this detector for detecting a single γ-ray is larger that 80%.

Among all the cases measured in this experimental campaign, we focus here on the decay of ^{140}Cs and ^{100}Nb. In the measurement of ^{100}Nb, special care was taken in order to distinguish experimentally between the decay of both isomers (see Fig. 1). The low spin isomer was populated through the decay of the parent, by selecting ^{100}Zr in JYFLTRAP, since it is a $0^+ \rightarrow 1^+$ transition, whereas the high spin isomer, assumed to be a 5^+ based on [19], was obtained by using the high precision Ramsey cleaning purification technique [20]. Moreover, a combined measurement with both isomers mixed was also performed.

3. First results

The TAGS analysis of the β-γ coincidences of these measurements will be described in this section. The total energy sum of the 18 crystals was reconstructed off-line. In order to calculate the response function for each

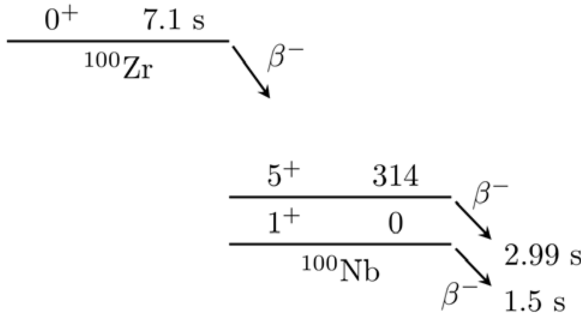

Figure 1. Scheme of the two isomers in ^{100}Nb. Spin-parity and energy (in keV) of both isomeric states, as well as the half-life of their β decay are presented. The decay of the ground state of the grandparent, ^{100}Zr, is also depicted.

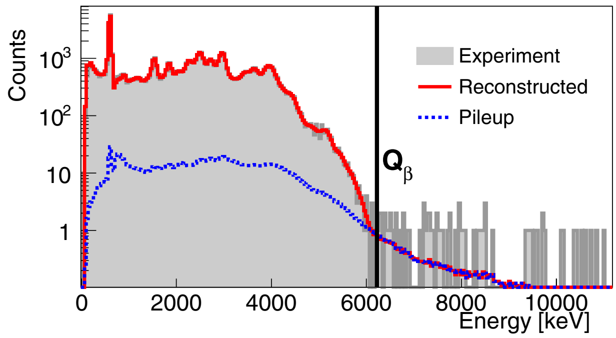

Figure 2. Relevant histograms for the analysis of ^{140}Cs: parent decay (gray filled), summing-pileup contribution (dotted blue) and reconstructed spectrum (red).

decay, the DTAS spectrometer was characterized with Geant4 MC simulations of several calibration sources. The coincidence between γs and βs guarantees a spectrum free from environmental background, however, all other possible sources of contamination must be taken into account in the analysis. One contamination present in all our measurements is the summing-pileup contribution. We calculate it with a MC procedure based on the random superposition of two stored events within the ADC gate, and a theoretical expression based on [21] allows us to normalize it.

^{140}Cs decay

The β^- decay ^{140}Cs$\rightarrow$^{140}Ba, with $Q_{\beta^-} = 6.22$ MeV and $T_{1/2} = 63.7$ s, was measured with an implantation rate of 30 ions/s [22]. The β-gated spectrum of DTAS can be seen in Fig. 2. Since the half-life of the daughter is 12.75 d, its decay does not contribute to the spectrum. The evaluation of the summing-pileup, the only contamination in this measurement, is shown in Fig. 2.

The detector response to the decay was calculated following the Valencia method [13], i.e., with information of the decay scheme at low excitation energies [23], and the nuclear statistical model at high excitation energies. The expectation maximization (EM) algorithm was applied to extract the I_β distribution [12]. In Fig. 2 the quality of the reproduction of the measured spectrum with this response is shown.

The study of this decay, apart from being relevant from the point of view of the antineutrino spectrum calculation, as mentioned in the introduction, represents

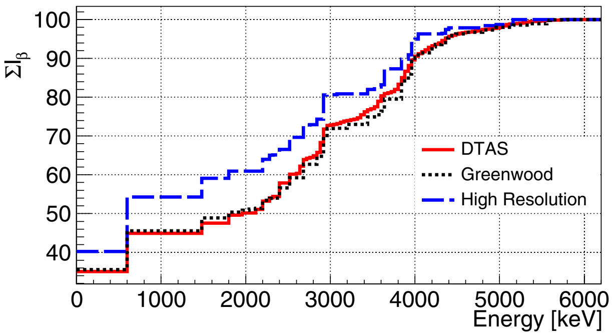

Figure 3. Comparison of the accumulated β intensity distribution for the decay of ^{140}Cs: present preliminary analysis (solid red line), previous TAGS data [24,25] (dotted black line), and high resolution data [26] (dashed blue line) are shown.

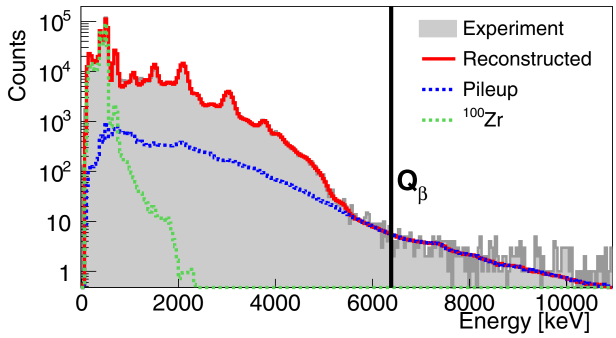

Figure 4. Relevant histograms for the analysis of ^{100}Nb ground state (g.s.): parent decay (gray filled), summing-pileup contribution (blue), grandparent decay (green) and reconstructed spectrum (red).

a cross-check for TAGS analysis techniques because it was already measured with this technique at the INEL ISOL facility by Greenwood et al. [24,25]. In Fig. 3 we show a comparison of the preliminary accumulated I_β distribution obtained in our analysis, the one previously obtained in [25] by using another method of analysis and a different set-up, and the distribution from high resolution measurements [26]. It can be seen how *Pandemonium* affected high resolution data [27,28], overestimating the β intensity at low energies. Moreover, it also shows that our preliminary data and data from Greenwood are in really good agreement.

^{100}Nb decay

The β^- decay of the low spin isomer of ^{100}Nb to ^{100}Mo, with $Q_{\beta^-} = 6.386$ MeV, was measured through the decay of the parent, as mentioned in Sect. 2. Thus, the decay of ^{100}Zr was treated as a contamination, and this contribution was calculated with MC simulations, by using the DECAYGEN event generator [29] with the information available for this decay as input [30]. The normalization of this contribution was done with the γ-ray at 400 keV coming from the decay of ^{100}Zr. Apart from that, the summing-pileup contamination was also taken into account. Again, the response function to the decay was calculated with the known information at low excitation energies [31], and the nuclear statistical model for the rest of the Q_β window.

The fit of the measured spectrum with the I_β that we obtained from the analysis is shown in Fig. 4. New β intensity is extracted from this preliminary analysis. In particular, around 4.5% of the total intensity is found above 3129.7 keV, the highest level that was seen in high resolution measurements [32].

^{100m}Nb decay

The case of the β^- decay of the high spin isomer of ^{100}Nb, with $Q_{\beta^-} = 7$ MeV, is more difficult to analyse, because it was impossible to measure it without the low spin contribution. Two different strategies were adopted to study this branch, as described in Sect. 2.

The first strategy consisted in using a high precision purification technique in the JYFLTRAP system, based on the use of a first purification trap for isobaric cleaning, followed by a second precision trap for isomeric cleaning [20]. As a result, our preliminary normalization

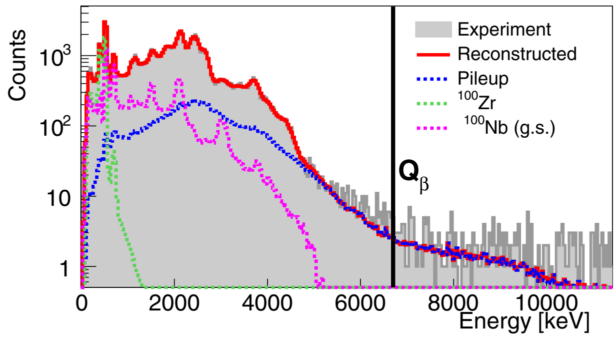

Figure 5. Relevant histograms for the analysis of ^{100m}Nb (Ramsey cleaning): parent decay (gray filled), summing-pileup contribution (blue), grandparent decay (green), low spin isomer decay (pink), and reconstructed spectrum (red).

gives a spectrum with approximately 67% high spin isomer and 23% low spin isomer. A 10% contribution from the decay of ^{100}Zr is also found, maybe due to an accidental overlap between the frequency selected in the purification process for ^{100m}Nb and the repeating frequency corresponding to ^{100}Zr, as reported in [33]. The contribution of the low spin isomeric branch was normalized by checking the peak at 695.2 keV associated to a level populated only in the decay of ^{100}Nb low spin, as well as adjusting the β penetration in the spectrometer, mainly due to the ground state to ground state transition that is only allowed for the decay of the low spin isomer. The contribution of the decay of ^{100}Zr was normalized as mentioned before.

The response function of the decay was calculated analogously to the case of the low spin isomer, but taking into account that allowed transitions in this case correspond to 4^+, 5^+ and 6^+ states in ^{100}Mo. The quality of the fit with the I_β obtained from the analysis can be seen in Fig. 5. From this preliminary results we extract 11% β intensity above 3647.2 keV, the last known populated level so far [19,32,33].

The second strategy to measure the high spin branch, was to measure both isomers together by selecting the frequency of JYFLTRAP associated to ^{100}Nb. In this measurement, the high spin component was favoured in the proton-induced fission process, and the low spin component was treated as a contaminant, in the same way as in the previous case. With our preliminary normalization

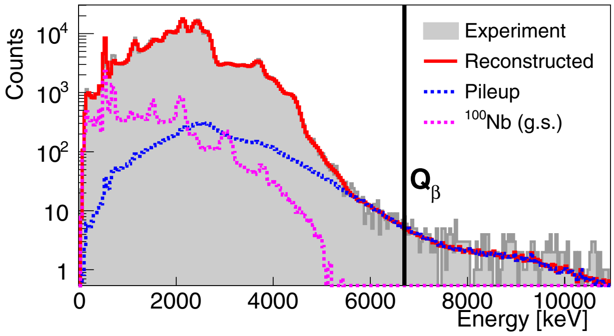

Figure 6. Relevant histograms for the analysis of ^{100m}Nb: parent decay (gray filled), summing-pileup contribution (blue), low spin isomer decay (pink), and reconstructed spectrum (red).

factor for the low spin component, it represents just a 6% of the total spectrum. The analysis was performed with the same response function as before, taking into account the summing-pileup contamination. The resulting fit for the I_β obtained is shown in Fig. 6. In this case we see a preliminary 12% β intensity above the last known populated level [19,32,33].

4. Conclusions

Measurements of the β decay of fission products that are important contributors to the antineutrino spectrum of a nuclear reactor have been carried out with the new DTAS detector at IGISOL, following the line of previous TAGS experiments that had an impact on antineutrino spectrum calculations [5,8,9]. The preliminary analysis of the decay of ^{140}Cs confirms previous TAGS results from Greenwood et al. that point to a remarkable *Pandemonium* in data obtained with germanium detectors. Moreover, the decay of the two isomers of ^{100}Nb has been measured with TAGS technique for the first time, thanks to the purification of the JYFLTRAP system. The preliminary results of this challenging measurement shows new β intensity that was not detected before in high resolution experiments. The final analysis, as well as the evaluation of the impact of these data on the antineutrino spectrum calculation with the summation method are ongoing.

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