Total Absorption Spectroscopy of Fission Fragments Relevant for Reactor Physics and Nuclear Structure and Astrophysics

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Abstract. The accurate determination of reactor antineutrino spectra remains a very hot research topic, where new questions have emerged in recent years. Indeed, after the "reactor anomaly" – a deficit of measured antineutrinos at short baseline reactor experiments with respect to spectral predictions – the three international reactor neutrino experiments Double Chooz, Daya Bay and Reno have evidenced spectral distortions in their measurements with respect to the same spectral predictions. This puzzle is called the "shape anomaly". Recently summation calculations of reactor antineutrino spectra based on the use of nuclear data have obtained the best agreement to date with the reactor neutrino flux measurements at the level of 2% thanks to a decade of Total Absorption Gamma-ray Spectroscopy (TAGS) measurements at the radioactive beam facility of the University of Jyväskylä in two experimental campaigns. A selection of the results obtained so far is presented.

1 Motivation of β decay studies

The elementary radioactive processes of α, β and γ emissions are nowadays quite well understood and experimentally constrained. However, in the case of β decay, finding a faithful description of its properties has been a long and arduous task. In fact, even today, the effort to describe Weak interactions is still ongoing. Although β rays were observed and identified as electrons at the end of the nineteenth century, it took roughly another 30 years to solve the puzzle of the continuous β spectrum shape thanks to the hypothesis of the existence of a new particle, the neutrino [1]. It then took another 30 or more years to characterise the weak interaction Hamiltonian as being of V-A type. Nevertheless, the theory built by Fermi in the thirties allowed him to determine a β decay probability and from there predict the β energy spectrum [2]. It opened the way to compute the half-life of a nucleus and quantities such as the β strength which allows one to constrain nuclear models. Indeed, our understanding of the β decay process is of fundamental importance in many domains of physics, including *inter alia* reactor physics, fundamental neutrino physics, nuclear structure and astrophysics.

Nuclear reactors are the most intense and pure controlled sources of low-energy electron antineutrinos. In the case of the Pressurised Water Reactor, they derive their power from the fission of the four isotopes ^{238}U, ^{235}U and ^{239}Pu and ^{241}Pu. In the fission process, two fission products (FP) are created which are neutron-rich nuclei. They undergo β or β-n decays at the origin of the production of a large amount of electron antineutrinos, i.e. $\sim 10^{20}$ $\bar{\nu}$ in a 1GW power reactor, but also of electrons and γ-rays from the de-excitation of the daughter nuclei produced. Several consequences follow that require a knowledge of β-decay if they are to be understood and handled properly. Firstly

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the average energy released in the β and γ emissions is the source of a large part of reactor decay heat (DH). It is essential for safe control of nuclear reactors and for reactor waste management that this is understood. Secondly in a β decay process, when the daughter nucleus is produced in an excited state above the neutron emission threshold, a β-delayed neutron might be produced before γ de-excitation occurs. This is important for the operation and control of the chain reaction in reactors. It is also true that this process plays a role in the astrophysical r process of nucleosynthesis. Thirdly the extremely large number of antineutrinos produced close to a reactor could be useful for reactor monitoring and non proliferation studies and it is essential for fundamental neutrino physics.

The measurement of the decay products of well identified fission products in the laboratory, offers a unique tool to constrain the various physics domains mentioned above. The measurement of γ or β emission is useful for decay heat calculations, antineutrino energy spectra predictions and delayed neutron fraction determination.

Moreover, from the determination of the β feeding intensity[1], I_β, achievable via γ-ray measurements, one can determine the β strength to individual levels in the daughter nucleus that is of interest for nuclear physics and nuclear astrophysics. For a given level energy in the daughter nucleus, the β strength is inversely related to its partial half-life and thus gives access to the transition probability to this level. The β strength distribution comparison, as a function of the excitation energy in the daughter nucleus, is a very sensitive test of theoretical models, which could improve our understanding of the residual interactions between nucleons as has been shown already [3].

In addition, an accurate determination of the β strength places constraints on the theoretical models that are complementary to integrated observables such as half-lives and P_n values. They are very important ingredients in r-process calculations [4].

2 Solving the Pandemonium effect

2.1 Pandemonium effect in Nuclear Databases

A contribution to the observables listed above arises from fission fragments that are short-lived nuclei and have large Q_β. Such cases are usually associated with complex decay patterns involving high γ-multiplicity cascades in the de-excitation of the numerous levels fed in β-decay. Before the 1990s, the conventional detection techniques used for such measurements were germanium detectors with excellent resolution but highly inefficient at high energy and to measure the full cascade. This leads to the failure to detect part of the γ-cascade or some of the high energy γ-rays, thus generating an overestimate of the high-energy part of the fission product β spectra. This was called the Pandemonium effect which is responsible for a strong bias present in nuclear databases (NDB) and de facto in any physics domain which uses NDB [5]. It implies, for example, the propagation of large uncertainties in any calculations using NDB involving nuclei with large Q values, such as summation calculations [6]. In the 1970s, summation calculations for decay heat, based on various databases assembled world-wide, were generally trusted. However important discrepancies were observed comparing the DH calculations and benchmark experiments mainly because of the Pandemonium effect. These differences were partially compensated by the inclusion of average β and γ energies derived from the Gross Theory of β decay which was able to compensate for the missing β-strengths or the information [7, 8] on certain nuclei. Since then this temporary solution was replaced step-by-step with the use of measured data with a new detection technique: total absorption γ-ray spectroscopy TAGS (section 2.2). Several collaborations and some world-wide efforts based on TAGS were born with the support of international agencies as the International Atomic Energy Agency (IAEA) and the Nuclear Energy Agency. Figure 2 in [9] shows how the inclusion of 7 nuclei ($^{102;104;105;106;107}$Tc, ^{105}Mo, and ^{101}Nb) measured by the TAGS collaboration in 2007 using the JYFLTRAP at Jyväskylä, and thus corrected for the Pandemonium effect, has improved the computation of the γ component of the decay heat for a fission burst of ^{239}Pu in the 4–3000 s range and solved the long-standing discrepancy between integral data [10] and predictions.

2.2 Total Absorption Gamma-ray Spectroscopy

In order to overcome the acceptance and intrinsic efficiency issues of HPGe detectors for γ-ray detection, one can use a calorimeter to cover as large a volume as possible. This is the purpose of a Total Absorption Spectrometer (TAS) containing large crystals providing as close to 4π coverage as possible. In contrast with a HPGe which detects precisely the individual γ-rays in a cascade, the crystals absorb the full γ energy released by the γ cascades in the β-decay process and the total energy of the cascade is obtained directly. Ideally, a TAS would give access to the β intensity, I_β, which can then be linked to the β strength. In the actual experiment, one has to consider the detector response which modifies the physical signal. In order to extract the true level energy feeding one has to solve the inverse problem [11, 12]. The measured spectrum is first cleared of pile-up and contaminants, while the response function of the total spectrometer is accurately simulated and used to deconvolve the measured spectrum to obtain the beta feeding probability. It depends on the branching ratios for the different de-excitation paths of the states populated in the decay and on the detector response itself.

Two TAGS experimental campaigns have been carried-out in 2009 and 2014 at the IGISOL in Jyväskylä with the main motivation of studying decay heat and the reactor antineutrino spectrum. For the first time, a TAS has been coupled to a double Penning trap (JYFLTRAP) in order to obtain sources of very high isobaric purity. Two different TAS detectors, both segmented, were used for those campaigns: Rocinante (in 2009) made of 12 BaF_2 crystals of very good efficiency coupled with a silicon detector for β coincidences [13] and DTAS (in 2014) composed of 16

[1] The probability to feed a given level energy in the daughter nucleus

to 18 NaI crystals coupled to a plastic detector for β coincidences [14]. In total, roughly 30 nuclei, of top priority for reactor and neutrino physics, were measured.

2.3 Impact on the antineutrino reactor anomaly

In the fundamental neutrino physics sector, in the last 15 years much effort has been dedicated to the measurement of the θ_{13} oscillation parameter [15–17]. Besides its measurement, which has been a great success, it has revealed two questions as yet unanswered, referred to as "reactor anomalies"[18], in comparing the detected antineutrino energy spectra with the predictions of the Huber-Mueller (H-M) conversion model [19, 20].

Firstly there is a 6% deviation in the computed flux compared with the measured one that was found in 2011. This has led to extensive experimental and theoretical researchs to bring to light the existence of sterile neutrinos [18]. In 2017, the Daya Bay collaboration showed that the 6% discrepancy could come from a potential bias in the ^{235}U spectrum measured in the 1980s by Schreckenbach *et al.*, and converted in the H-M model [21]. To show it, they measured the inverse β decay yield (IBD) as a function of the fission fraction coming from ^{235}U and ^{239}Pu and compared it to the H-M model. The conclusion was that a 5% deviation in the flux mainly came from the ^{235}U [22].

The second reactor anomaly is due to the shape of the spectra between 5 and 7 MeV which presents a bump not explained so far by nuclear model predictions [23].

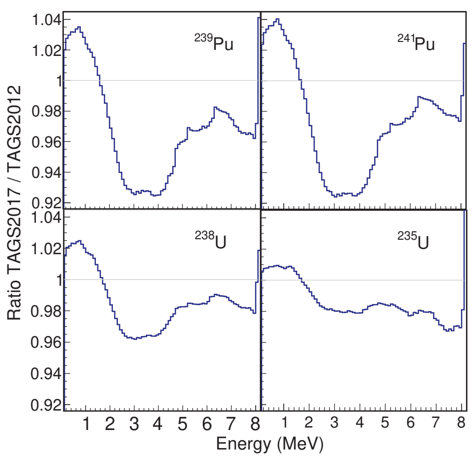

Figure 1. Accumulated impact of the TAGS data of the ^{86,87,88}Br and ^{91,92,94}Rb decays measured with Rocinante on the antineutrino spectra with respect to that published in 2012 [3, 24].

In 2012, we published an updated version of our summation calculations[2] for antineutrino energy spectra prediction in which we quantified the relative impact of the seven Pandemonium-free measurements of nuclei, measured in 2007 on the spectra [24] and quoted in section 2.1. A description of the cocktail of NDB and models used for the decay data is given in [24]. The ratios of the antineutrino spectra computed with our model including the seven Pandemonium-free cases over the same calculation performed using older NDB for those nuclei for the four fissile nuclei present in the reactor core exhibited a noticeable deviation from unity [24]. It was later shown that this behaviour is systematic, when correcting for the Pandemonium effect in data. The effect is to increase the spectra below 2-3 MeV and decrease it in the energy region above which dominates the flux. The limits of this systematic behaviour depend on the Q values of the nuclei involved. With respect to our predictions in 2012, we have since quantified the cumulative impact of the TAGS beta intensities of the other nuclei measured with Rocinante in 2009 on the antineutrino spectra generated after the thermal fission of ^{235}U, ^{239}Pu and ^{241}Pu, and fast fission of ^{238}U [3]. The results are presented in Fig 1 in comparison with the spectra built with the most recent NDB for the same nuclei and containing only TAGS data from [24]. The decrease of the 2 plutonium spectra above 1.5 MeV is remarkable, reaching 8%. The impact on the two uranium isotopes amounts to about 2% and 3.8% in the 3 to 4 MeV range in ^{235}U and ^{238}U respectively.

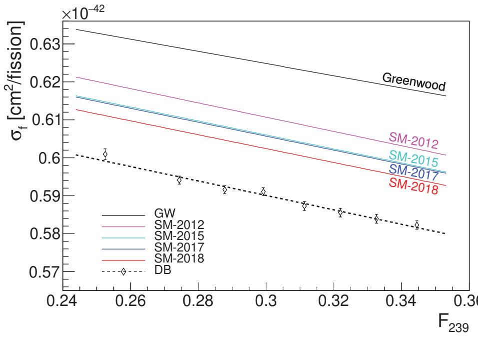

Figure 2. Comparison of the IBD yield computed with the summation antineutrino spectrum obtained using the fission fractions published in [22] for ^{239}Pu and a decade of TAGS data.

The cumulative impact of the nuclei measured in 2014 in Jyväskylä with the DTAS detector has been studied as well and presented elsewhere [3]. It has shown an important deviation from unity in the energy range of interest for the shape anomaly. The consideration in the calculation of the ^{100,102}Nb isotopes in particular and their isomers corrected for Pandemonium bias implied a strong decrease of the spectra peaked at 4.5 MeV and a strong increase at 6.5 MeV, in the region of the shape distortion. However, it was not enough to fully explain the observed bump.

In order to highlight and quantify the systematic impact of the TAGS data on the detected antineutrino flux, we have studied the absolute impact of a decade of TAGS measurements on the calculated antineutrino energy spectra by updating our summation model [25]. Beside other modifications presented in [25], but not commented on here, we have step by step included in the calculations the results analysed over 10 years. The total antineutrino IBD yields have then been computed for the different summation models called SM2012, SM2015, SM2017 and SM2018 (which stands for "Summation Model" followed

[2]NB: the summation method is the only alternative we have so far to the H-M model.

by the year of publication of the TAGS decay data added in the calculations) as a function of the ^{239}Pu fraction of fission. The result is presented in Fig 2. A systematic reduction of the detected flux is observed when correcting for the Pandemonium effect in data as well as a systematic reduction of the discrepancy with the Daya Bay results shown in the figure with the small diamonds. Considering this systematic and understood trend, we expect the discrepancy to reduce further with the inclusion of future TAGS data leaving less room for the reactor anomaly in flux.

3 Recent TAGS results for $^{96gs/m}$Y

Recently the collaboration has published new results associated with the decay of ^{96}Y and its isomer [26] measured in 2014 at the IGISOL IV facility. For several reasons the measurement of the decay of both the ^{96}Y ground state (gs,GS) and isomer (m) is interesting. i) For the decay heat, the decays of both nuclei produce almost 5% of the DH around 10s after the thermal fission of ^{235}U. The ground state being of first priority for the IAEA experts in the case of U/Pu and Th/U fuels and the isomer being of priority 1 for Th/U fuel [27]. ii) Concerning reactor antineutrino spectra, the ^{96gs}Y is the second most important contributor to the spectra in the region of the bump. iii) Eventually, it has some interest for the study of the structure of the daughter ^{96}Zr which lies in a region of phase transition and emergence of shape-coexisting states[28]. The β decay of these nuclei has been measured before but either the decays of the gs and the isomer were mixed or the isomeric 8+ state was not produced. Therefore, the analysis of their decay patterns had to rely to some extent on previous high-resolution spectroscopy measurements. This motivated the necessity to operate at IGISOL as the facility allows one to separate the two states, which have an energy difference of 1540 keV between the ground state (7103keV) and the isomeric one, using a buffer-gas cooling technique.

The gs ^{96}Y GS 0- β minus decays to the ^{96}Zr gs with ~95% probability. The isomeric state 8+ also decays by β^- to ^{96}Zr following another decay scheme with a 1.54 MeV energy difference. In addition, the ^{96}Y gs decays 1.26% of the time to the excited level at 1581.34 keV which de-excites via an E0 decay to the ^{96}Zr GS. The purified β-gated spectra have been extracted offline. In case of ^{96gs}Y, only the summing-pile-up has been subtracted whereas in the case of the isomer, both contamination coming from the ^{96gs}Y and the ^{96}Sr have been subtracted due to some purification issues in JYFLTRAP. For the resolution of the inverse problem, the known part taken from ENSDF [29] has been considered up to 4389.5keV with some assumptions made for some levels. Above this energy, a statistical model has been used. Then the response function has been obtained from Monte Carlo (MC) simulation. For the specific case of the β decay of the ground state, the "traditional" procedure [12] has been slightly modified to take into account the E0 de-excitation of the 0+ level at 1581.6keV instead of the usual γ response. This de-excitation occurs by means of conversion electrons in competition with pair production due to the fact that the energy involved is larger than the pair production threshold. It has been simulated in the MC response taking into account a certain probability of pair production found in the literature. This has, of course, modified the response function of the spectrometer as well as the efficiency of the plastic β detector [26].

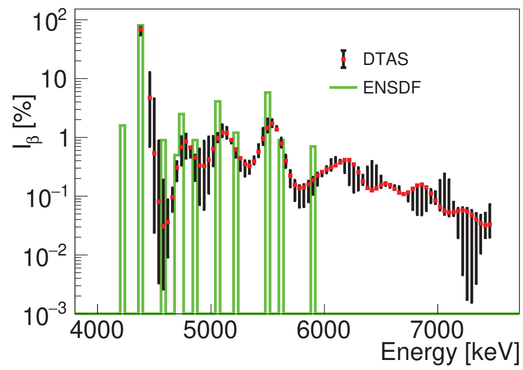

Figure 3. β intensity of the ^{96m}Y obtained as described in the text (red dots with error bars) and compared with high-resolution γ-spectroscopy data from ENSDF (green line).

The β intensity distribution obtained for the GS did not present Pandemonium pattern. The distribution obtained for the first time for the isomer is presented in Fig 3 and compared with the high-resolution spectroscopy values from ENSDF. Even if this nucleus did present a clear Pandemonium effect, its impact is small on the antineutrino energy spectrum calculation since it is a minor contributor in the 5-7MeV range. For the DH, we obtain a clear change in the average γ and β energies due to its Pandemonium nature but as for the antineutrino, the cumulated impact of both nuclei in decay heat summation calculations was found to be very small both for the electromagnetic and the light particle components. Nevertheless, those results are of extreme importance as they contribute to reduce the uncertainties quoted in NDB for ^{96}Y.

4 Conclusion and outlook

In summary, the TAGS data measured during the last decade at the IGISOL facility of the University of Jyväskylä have impressively improved the agreement between the summation calculations built with the nuclear data for reactor antineutrino spectra and decay heat with respectively reactor neutrino experiments and integral decay heat measurements. A few highlights and recent results have been presented in these proceedings to illustrate these achievements. Nevertheless further new TAGS measurements are needed to help us to understand the shape anomaly in reactor neutrino physics, persistent discrepancies between integral measurements of decay heat and summation calculations and provide reliable new predictions of these observables for innovative fuels and reactor designs.

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