Total absorption spectroscopy of fission fragments relevant for reactor antineutrino spectra

M. Fallot[1,a], A. Porta[1], L. Le Meur[1], J.A. Briz[1], A.-A. Zakari-Issoufou[1], V. Guadilla[2], A. Algora[2,13], J.-L. Taìn[2], E. Valencia[2], S. Rice[3], V.M. Bui[1], S. Cormon[1], M. Estienne[1], J. Agramunt[2], J. Äystö[4], L. Batist[14], M. Bowry[3], R. Caballero-Folch[5], D. Cano-Ott[6], A. Cucoanes[1], V.-V. Elomaa[7], T. Eronen[7], E. Estévez[2], G.F. Farrelly[3], L.M. Fraile[15], M. Fleming[16], E. Ganogliu[17], A.R. Garcia[6], W. Gelletly[2,3], M.B. Gomez-Hornillos[5], D. Gorelov[7], V. Gorlychev[5], J. Hakala[7], A. Jokinen[7], M.D. Jordan[2], A. Kankainen[7], P. Karvonen[7], V.S. Kolhinen[7], F.G. Kondev[8], J. Koponen[7], M. Lebois[18], T. Martinez[6], P. Mason[3], E. Mendoza[6], F. Molina[2], M. Monserrate[2], A. Montaner-Pizá[2], I. Moore[7], E. Nácher[19], S.E.A. Orrigo[2], H. Penttilä[7], A. Perez[2], Zs. Podolyák[3], I. Pohjalainen[7], P.H. Regan[3,9], J. Reinikainen[7], M. Reponen[7,10], S. Rinta-Antila[7], J. Rissanen[7], B. Rubio[2], T. Shiba[1], V. Sonnenschein[7], A.A. Sonzogni[11], J.-C. Sublet[16], V. Vedia[15], A. Voss[7], C. Weber[7,12], and J.N. Wilson[18]

1 SUBATECH, CNRS/IN2P3, Université de Nantes, Ecole des Mines de Nantes, 44307 Nantes, France
2 IFIC (CSIC-Univ. Valencia), Valencia, Spain
3 Department of Physics, University of Surrey, Guildford GUZ7XH, UK
4 Helsinki Institute of Physics, 00014 University of Helsinki, Finland
5 Universitat Politécnica de Catalunya (UPC), 08034 Barcelona, Spain
6 CIEMAT, Madrid, Spain
7 Department of Physics, PO Box 35, 40014 University of Jyväskylä, Finland
8 Argonne National Laboratory, Argonne, USA
9 National Physical Laboratory, Teddington, Middlesex TW11 0LW, UK
10 RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan
11 National Nuclear Data Center, Brookhaven National Laboratory, Upton NY 11973-5000, USA
12 Faculty of Physics, Ludwig-Maximilians University Munich, Am Coulombwall 1, 85748 Garching, Germany
13 MTA ATOMKI, Hungary
14 Petersburg Nuclear Physics Institute, 188300 Gatchina, Russia
15 Universidad Complutense, Grupo de Física Nuclear, CEI Moncloa, 28040 Madrid, Spain
16 United Kingdom Atomic Energy Authority, Culham Science Centre, Abingdon OX14 3DB, UK
17 Department of Physics, Istanbul University, 34134 Istanbul, Turkey
18 Institut de Physique Nucléaire d'Orsay, CNRS/IN2P3, 91406 Orsay, France
19 Instituto de Estructura de la Materia, CSIC, 28006 Madrid, Spain

Abstract. The accurate determination of reactor antineutrino spectra remains a very active research topic for which new methods of study have emerged in recent years. Indeed, following the long-recognized reactor anomaly (measured antineutrino deficit in short baseline reactor experiments when compared with spectral predictions), the three international reactor neutrino experiments Double Chooz, Daya Bay and Reno have recently demonstrated the existence of spectral distortions in their measurements with respect to the same predictions. These spectral predictions were obtained through the conversion of integral beta-energy spectra obtained at the ILL research reactor. Several studies have shown that the underlying nuclear physics required for the conversion of these spectra into antineutrino spectra is not totally understood. An alternative to such converted spectra is a complementary approach that consists of determining the antineutrino spectrum by means of the measurement and processing of nuclear data. The beta properties of some key fission products suffer from the pandemonium effect which can be circumvented by the use of the Total Absorption Gamma-ray Spectroscopy technique (TAGS). The two main contributors to the Pressurized Water Reactor antineutrino spectrum in the region where the spectral distortion has been observed are ^{92}Rb and ^{142}Cs, which have been measured at the radioactive beam facility of the University of Jyväskylä in two TAGS experiments. We present the results of the analysis of the TAGS measurements of the β-decay properties of ^{92}Rb along with preliminary results on ^{142}Cs and report on the measurements already performed.

1. Introduction

The fission process is at the origin of thermal energy widely used worldwide for electricity production. In general in this process two fission products, that are beta minus emitters, are produced together with a few neutrons. In the beta minus decay process of these nuclei, an electron and an electron antineutrino are emitted, often accompagnied by de-excitation gamma rays from

[a] e-mail: fallot@subatech.in2p3.fr

DOI: 10.1051/epjconf/201714610002

the daughter nuclei. Sometimes a delayed neutron is emitted, if the daughter nucleus is populated above its neutron emission threshold. The delayed neutrons allow the control of the criticality of a reactor core, while the electrons and gamma-rays are at the origin of the decay heat emitted even after the reactor shuts down. The antineutrinos escape and can be detected with ton-scale detectors to solve basic science, or can be used to image the reactor fuel, as a potential reactor monitoring tool [1]. Antineutrinos pass through large quantities of matter without interacting, and it is impossible to shield against them. This property combined with the experience and technical know-how in reactor antineutrino detection developed internationally since their discovery in 1956, attracted the interest of the International Atomic Energy Agency which has asked its member states to perform sensitivity studies. The required R&D for new detector designs suited for reactor monitoring and new reactor simulations to predict the antineutrino characteristics associated with various reactor designs are still ongoing worldwide [2].

In parallel, efforts have been made to study the beta decay properties of the fission products. Indeed, antineutrino energy spectra associated with new nuclear fuels are not known, and they can be calculated by means of the measurement and processing of nuclear data. It was shown that the data associated with important contributors to the antineutrino spectra could be biased by the Pandemonium effect [3] and require new measurements with the TAGS technique [4]. Yet another method to compute antineutrino spectra from thermal fission of ^{235}U, $^{239;241}$Pu was reinvestigated recently, based on the conversion of integral beta spectra measured at the ILL research reactor of Grenoble three decades ago. The obtained converted spectra were found to be about 3% lower than the previous ones [5,6] used as references by reactor neutrino experiments, which led to the “Reactor Anomaly” [7], triggering a huge experimental effort in the search for sterile neutrinos at short distances from small reactor cores. Note that in the text below, we employ the generic term neutrinos meaning electron antineutrinos when appropriate. More recently, the three large reactor neutrino experiments Double Chooz, Daya Bay and Reno have shown evidence of shape distortion when their measurements are compared with the antineutrino spectra built with the conversion method, especially in the region 4 to 8 MeV [8]. This distortion reaches a 10% deviation with respect to the measured spectra. These results attracted even more attention to the modelling of reactor antineutrino spectra and the underlying ingredients in each calculation method. Hayes et al. [9] showed that forbidden non-unique transitions could play an important role in the normalization of antineutrino spectra and distort potentially the converted spectra. Recently Huber et al. [10] have carried out statistical studies which conclude that there may be a problem with the shape of the measured ILL beta spectrum arising from ^{235}U thermal fission [11]. The construction of reactor antineutrino spectra with nuclear data is the only alternative to the converted spectra, in the absence of a new precise measurement of integral beta spectra from actinide thermal fissions. It is thus timely to try to improve the predictions of the summation method by reducing the errors associated with the nuclear data.

2. Can nuclear data help to solve the reactor antineutrino puzzle?

Mueller et al. [5] have reinvestigated the summation methods as applied to reactor antineutrino spectra. The need to identify potential biases in nuclear data has already been stressed. This issue can be resolved experimentally by the use of Total Absorption Gamma-ray Spectroscopy (TAGS). A TAGS is a calorimeter made of large crystals of high intrinsic efficiency, ideally covering a 4π solid angle. In Fallot et al. [4] spectra created with the summation method using, whenever possible, data free of the Pandemonium problem were presented, including in particuler recently measured TAGS data at the IGISOL facility at Jyväskylä using for the first time a TAGS combined with the available Penning trap JYFLTRAP [12]. Such measurements showed that five amongst the seven measured nuclei suffered strongly from the Pandemonium effect [13,14]. A TAGS experimental campaign, based on candidates that suffered from the Pandemonium problem and made an important contribution to the antineutrino spectra, was started then and new measurements were performed at Jyväskylä [15]. A few recent results of these experiments will be presented in section III. The second important ingredient entering in the summation spectra is the fission yields. Sonzogni et al. [16] have shown that fission yield nuclear data should be chosen with great care as data which have not been updated can bias the shape of spectra built with the summation method. For instance the usage of the uncorrected $ENDF/B-VII$ fission yields produces summation spectra with a shape which recalls the distortion observed in the three reactor neutrino experiments [17], while JEFF3.1 yields, which do not suffer from the inconsistancies found in $ENDF/B-VII$, do not reproduce such a pattern. Sonzogni et al. also show in their paper that a potential contribution from fast fissions in a PWR would not reproduce the distortion observed in the three reactor neutrino experiments with respect to converted spectra [18].

In table I, the list of the main nuclei contributing to a typical PWR antineutrino spectrum, in the bins above 4 MeV, is reproduced from [15]. This list has been established with the summation method using the ingredients presented in [4,19]. It has been cross-checked by participants attending a TAGS consultants' meeting organized by the Nuclear Data Section of the International Atomic Energy Agency, to which Sonzogni et al. have presented similar results [20]. A more extensive table was extracted from our summation calculations, in agreement with Sonzogni et al.'s results, and can be found in [21]. Overall, one can see that the number of nuclei contributing to these bins is small enough to give the hope to produce summation calculations with reduced systematic errors due to decay data on a relatively short time scale, i.e. comparable to the time scale of short baseline reactor neutrino experiments chasing sterile neutrinos.

3. Results from experiments at Jyväskylä

In late 2009, the decays of seven isotopes of Br and Rb were studied at the JYFL facility of Jyväskylä (Finland). The motivations were several. Some of these nuclei are

Table 1. Main contributors to a standard PWR antineutrino energy spectrum computed with the MURE code coupled with the list of nuclear data given in [4] and assuming that they have been emitted by ^{235}U (52%), ^{239}Pu (33%), ^{241}Pu (6%)and ^{238}U (8.7%) for a 450 day irradiation time and using the summation method described in [4]. Reproduced from [15].

	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%

delayed neutron emitters. Indeed, a topic of interest for nuclear astrophysics and the r-process is the determination of gamma-neutron competition above the neutron emission threshold [22,23]. Some of these nuclei are of first priority for decay heat calculations (U/Pu and Th/U cycles) [24]. ^{92}Rb and ^{93}Rb were measured as main reactor antineutrino contributors, with ^{92}Rb assessed to be an important second priority contributor to decay heat [25].

^{92}Rb alone exhausts 16% of the antineutrino energy spectrum emitted by PWRs in the region of energy 5 to 8 MeV. It is also a candidate Pandemonium nucleus, and its ground state to ground state beta branch is a first forbidden non unique transition with a large branching ratio. The reconstructed spectrum obtained after cleaning of the data from daughter contamination and pile-up events, and so solving the inverse problem with the method developed in [26] is shown in the upper panel of Fig. 1 in blue, superposed on the data (black line). One can see the very good agreement between the two. Our analysis gives a ground state to ground state branch of 87.5%±3.0%. The error has been obtained by variation of the input parameters of the analysis i.e. small variations in the thicknesses of materials in the detector simulation, level densities, spin-parities of ill-defined levels in the daughter nucleus, etc. It is thus an envelope obtained by summing quadratically all the studied effects. More details of the analysis can be found in [15]. Figure 2 displays the ratios between the antineutrino spectra calculated using the results presented in [15], with respect to the data for ^{92}Rb decay used in [4] (thick red dashed-dotted line), [20] (green dotted line) and [17] (black dashed line), assuming they would all use the same fission yields and the same beta decay data apart from the nucleus of interest. In this way, we compare only the beta decay properties of the nucleus involved. While [4] used the data from [20,27] used the latest version of the $ENDF/B-VII$ library. The two sets of data give very similar antineutrino spectra, which is why the impact of the new TAGS data on these two sets is almost the same (red and green lines superposed on the plots). Note that the sharp drop in one single bin located at the Q value comes from the fact that we use the most recent adopted Q value of 8095 keV for ^{92}Rb which is different from the one measured by the authors of [27] at the time. In their calculation, the authors of [17] used

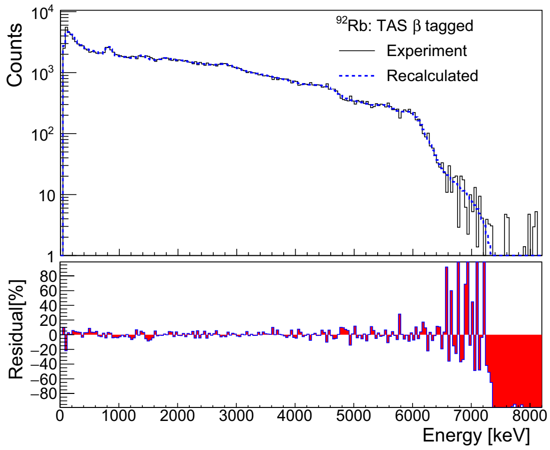

Figure 1. Upper panel: comparison between measured spectrum (black continuous line) and reconstructed one (blue dashed line) with the feeding obtained from the TAGS data analysis. Lower panel: residues between the two curves reported in the upper panel.

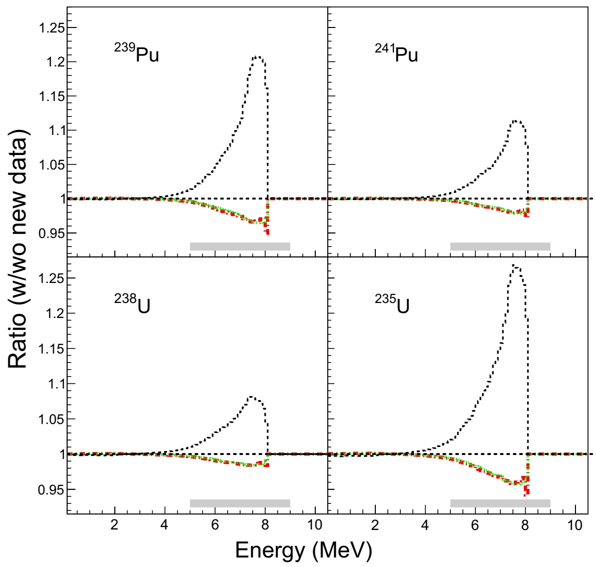

Figure 2. Ratio between the antineutrino spectra calculated using the latest TAGS results with respect to the data on ^{92}Rb decay used in [4] (thick red dashed-dotted line), in [20] (green dotted line) and in [17] (black dashed line). The sharp drop in the ratio, in one single bin located at the Q value of the ^{92}Rb, is due to the different values in Q given in [27] and $ENSDF$, that were used to reconstruct the antineutrino spectrum.

the $ENDFBVI.8$ database without any update, in which the ground state to ground state beta branch of the ^{92}Rb amounts only to 50%, a value which is well below the other experimental results. ^{92}Rb being dominant in the 5 to 8 MeV region, the impact on the computed antineutrino spectra is enormous. This shows that a careful choice of data is mandatory for building summation method spectra. In Fig. 2 the grey horizontal bar indicates the region of the distortion observed by reactor antineutrino experiments with respect to converted spectra, which coincides with the region dominated by the contribution of ^{92}Rb.

In 2014, a second experimental campaign was carried out at the Jyväskylä facility, using for the first time the DTAS detector, made of 18 NaI crystals developed by the IFIC of Valencia (Spain) [28], coupled for the first time

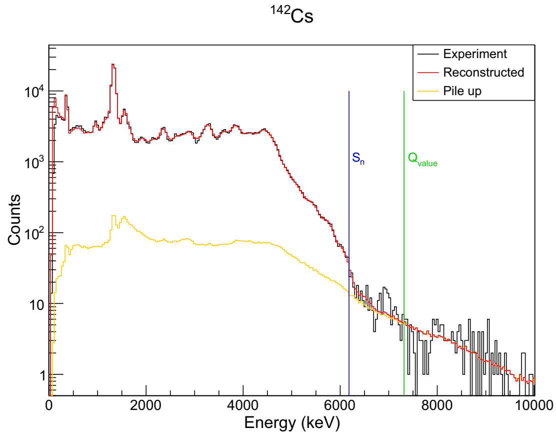

Figure 3. ^{142}Cs: comparison between measured spectrum (black continuous line) and reconstructed one (red continuous line) with the feeding obtained from the TAGS data analysis. The contribution of pile-up signals is shown (yellow line).

to the new IGISOL-4 facility. The experiment provided also the opportunity of to use for the first time the precision Penning trap with IGISOL-4. This experiment was very successful as twelve nuclei of importance for the antineutrino spectra and eleven of importance for reactor decay heat were measured. The analyses were undertaken by the Subatech and IFIC teams. We show in Fig. 3 preliminary results for the ^{142}Cs nucleus, the third main contributor to the spectrum (see Table 1). In the figure, the locations of the Q-value and the neutron emission threshold are indicated by vertical lines. The yellow line represents the contribution of the pile-up signals to the data. The red line is the result of the reconstruction of the TAGS data with the feeding found with the Bayes method [26]. It is already in very good agreement with the data shown in black.

Overall, TAGS data have now been obtained for eight of the top eleven nuclei presented in Table 1. The reader can find the other new results showing the impact of ^{87,88}Br and ^{94}Rb on antineutrino spectra in [22] and [29]. The results for ^{140}Cs and ^{100,100m}Nb from the same experimental campaign are presented in [30]. Previous TAGS measurements on $^{102;104-107}$Tc, ^{105}Mo, and ^{101}Nb isotopes were also found to have a non-negligible impact on the antineutrino spectrum in this energy region [13]. All of this progress gives confidence in our ability to predict the antineutrino energy spectra from the main uranium and plutonium isotopes with reduced errors in this energy region where only a few nuclei contribute thanks to nuclear data to be acquired in the near future. Beyond the experimental challenge, the other challenge is to propagate the errors of the decay and fission yield data on the summation calculations.

4. Summary and outlooks

In summary, already a substantial number of the nuclei that are major contributors to the PWR antineutrino spectrum have been measured with the TAGS technique. The nuclear data community has also reached an agreement on the nuclear data sets to be used for the summation calculations following agreements at the TAGS consultant meetings organized to discuss and review TAGS by the IAEA Nuclear Data Section and dedicated workshops organized in Valencia (Spain), Seattle (USA) and Nantes (France). Overall, a lot of progress has been made since this work was initiated. Recent papers have shown that unknowns in nuclear physics have a significant impact on the estimates of reactor antineutrino spectra (for instance, forbidden non-unique transitions), but they also show that an alternative method of predicting these spectra with smaller errors may soon come from nuclear data.

The authors acknowledge the nuclear data section of the IAEA and especially Paraskevi (Vivian) Dimitriou for fostering research in the field through the TAGS consultant meetings and the CRP about beta-delayed neutron emission. This work was supported by the CHANDA European project, the In2p3 institute of CNRS, and the NEEDS challenge through the NACRE project. This work was supported by the Academy of Finland under Project No. 213503, Nuclear and Accelerator-Based Physics Research at JYFL. This work was supported by Spanish Ministerio de Economía y Competitividad under grants FPA2008- 06419, FPA2010-17142 and FPA2011-24553 and FPA2014-52823-C2-1-P, CPAN CSD-2007-00042 (Ingenio2010), and the program Severo Ochoa (SEV- 2014-0398) and by EPSRC and STFC (UK). Work at ANL was supported by the U.S Department of Energy under contract DE-AC02-06CH11357.

References

[1] M. Fallot, Nuclear Data Sheets **120**, 137 (2014)
[2] L. Giot, M. Fallot, contribution to these proceedings
[3] J.C. Hardy et al., Phys. Lett. B **71**, 307 (1977)
[4] M. Fallot et al., Phys. Rev. Lett. **109**, 202504 (2012)
[5] Th. Mueller et al., Phys. Rev. C **83**, 054615 (2011)
[6] P. Huber, Phys. Rev. C **84**, 024617 (2011)
[7] G. Mention et al., Phys. Rev. D **83**, 073006 (2011)
[8] Double Chooz and Reno Collaborations in Proceedings of the Neutrino 2014 Conference, `http://neutrino2014.bu.edu/`; Daya Bay Collaboration in Proceedings of the ICHEP 2014 Conference, `http://ichep2014.es/`
[9] A.C. Hayes et al., Phys. Rev. Lett. **112**, 202501 (2014)
[10] P. Huber, arXiv:1609.03910
[11] A.A. Hahn et al., Phys. Lett. B **218**, 365 (1989) and references therein
[12] T. Eronen et al., Eur. Phys. J. A **48**, 46 (2012)
[13] A. Algora et al., Phys. Rev. Lett. **105**, 202501 (2010)
[14] D. Jordan et al., Phys. Rev. C **87**, 044318 (2013)
[15] A.-A. Zakari-Issoufou, Phys. Rev. Lett. **115**, 102503 (2015)
[16] A.A. Sonzogni, E.A. McCutchan, T.D. Johnson, and P. Dimitriou, Phys. Rev. Lett. **116**, 132502 (2016)
[17] D.A. Dwyer, T.J. Langford, Phys. Rev. Lett. **114**, 012502 (2015)
[18] A.A. Sonzogni et al., contribution to these proceedings
[19] M. Estienne et al., Nucl. Data Sheets **120**, 149 (2014)
[20] A.A. Sonzogni et al. , Phys. Rev. C **91**, 011301 (R) (2015)
[21] P. Dimitriou, A.L. Nichols, IAEA report INDC(NDS)-0676, Feb. 2015, IAEA, Vienna, Austria

[22] J.L. Taìn et al., contribution to these proceedings
[23] J.L. Tain et al. Phys. Rev. Lett. **115**, 062502 (2015)
[24] A. Algora et al., contribution to these proceedings R471
[25] T. Yoshida, A.L. Nichols, Nuclear Energy Agency report NEA/WPEC-25 (2007), OECD-NEA, Paris, France, ISBN 978-92-64-99034-0
[26] J.L. Taìn et al., Nucl. Instr. Methods Phys. Res. A **571**, 728 (2012)
[27] G. Rudstam et al., At. Data Nucl. Data Tables **45**, 239 (1990)
[28] V. Guadilla et al., Nucl. Instr. Methods Phys. Res. B **376**, 334 (2016)
[29] E. Valencia et al., Phys. Rev. C **95**, 024320 (2017)
[30] V. Guadilla et al., contribution to these proceedings