

# Aggregate Decay Behavior of Fission Products in Nuclear Reactors

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## Abstract

There exist a lot of important quantities which have their origin in aggregate behavior of unstable fission products (FP) in nuclear reactors. Decay heat is a typical example of importance. These quantities are calculated by summing up all the contributions coming from each unstable nucleus accumulated during reactor operation by using of FP decay data libraries. In their infancy, however, they were suffered from so-called the *pandemonium* problem. For decades, the difficulty caused by pandemonium problem has been circumvented by introduction of a  $\beta$ -decay theory. In recent years, the total absorption gamma-ray spectroscopy (TAGS) is saving the situation remarkably even though leaving some open problems.

## 1 Introduction

In nuclear engineering, there exist a lot of quantities of crucial importance which have their origin in aggregate behavior of fission products (FP) in reactor cores. They are the reactor decay heat, the delayed  $\gamma$ -ray spectrum, the delayed neutron fractions and their spectra among others. The flux and the spectrum of the reactor antineutrino also belong to this category which are attracting interest from neutrino physicists recent years. As these quantities generally depends on the details of the reactor operation history and the time lapse after the reactor shutdown, one has to calculate them using these conditions as inputs. The most typical way of calculation is the so-called summation method which sums up all the contributions from decaying FP nuclides existing in the reactor core.

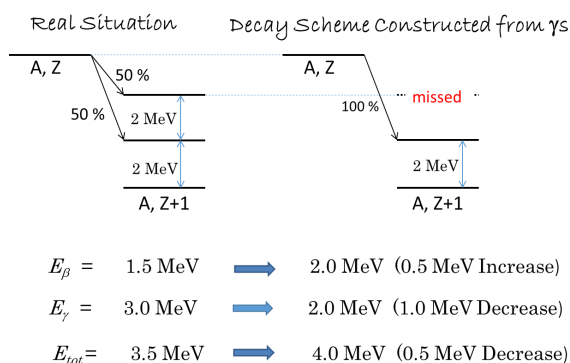
Summation calculations require a data library on the decay properties of almost a thousand of FPs which consist of the average  $\beta$ - and  $\gamma$ -ray energies, their energy spectra, the  $\beta$ -delayed particle ( $n, e, \bar{\nu}_e$ ) emission probabilities and their spectra *et al.* depending on what quantity one wants to calculate, along with exact information on each  $\beta$ -decay chain in common. Most of these nuclide-wise quantities, however, are known to be suffering from the *pandemonium* problem [1], and the history and the future of overcoming this problem will be detailed in the following sections.

## 2 Pandemonium problem in nuclear decay schemes

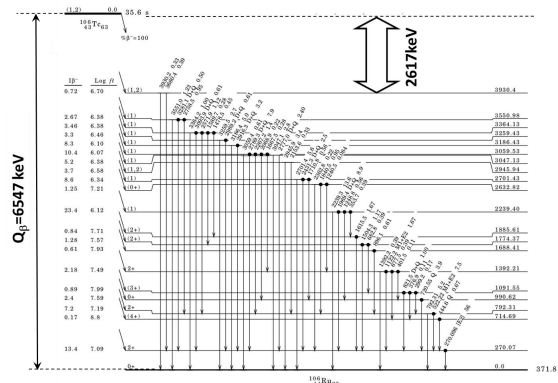
The effect of the pandemonium problem on the reactor decay heat is illustrated in Fig. 1. This is an highly simplified toy-model of  $\beta$ -decay schemes. Real decay schemes are widely used and available in the form of books/CD-ROM [2], journals [3] or internet data-basis [4]. Assume that a parent nucleus having  $Q_\beta$ -value of 6 MeV feeds the two excited levels at 2 and 4 MeV by 50% each in its daughter which will be de-excited emitting  $\gamma$ -rays. It may happen that the feed to 4 MeV level is missed in constructing the decay scheme as in the right-hand side of Fig. 1. This missing leads to remarkable overestimation of the average  $\beta$ -ray energy  $E_\beta$  and underestimation of  $E_\gamma$  both by 33% in this case.

By using a computer simulation, Hardy *et al.* warned this kind of missing is inevitable in decay schemes of highly  $Q_\beta$ -valued unstable nuclides which are constructed from hundreds of discrete high-resolution  $\gamma$ -ray energy and intensity data from experiments [1]. Reconstruction of a complex decay scheme can be a very difficult task like a extremely complicated jigsaw puzzle. In addition, completeness of the  $\gamma$ -ray data is not expected especially when the parent is a short-lived, high  $Q_\beta$ -valued nuclide. Figure2 shows an example of a decay scheme of such a nuclide,  $^{106}\text{Tc}$  [5]. In this case, the  $\beta$ -feeding

to levels above 3930 keV seems to be missed up to 6549 keV( $=Q_{\beta}$ ). Being calculated as a weighted sum of  $E_{\beta}$  and  $E_{\gamma}$ , this kind of feed (or level) missing results in over- and underestimation of  $\beta$ - and  $\gamma$ -ray components of reactor decay heat, respectively. This was revealed when extensive FP decay data libraries for decay-heat calculation were completed and tested in Japan, Europe and the US in the end of 1970's.



**Fig. 1:** Extremely simplified decay-scheme toy model with  $\beta$ -feedings followed by  $\gamma$ -transitions



**Fig. 2:** An example of complex decay scheme of a fission product nuclide with a high  $Q_{\beta}$ -value

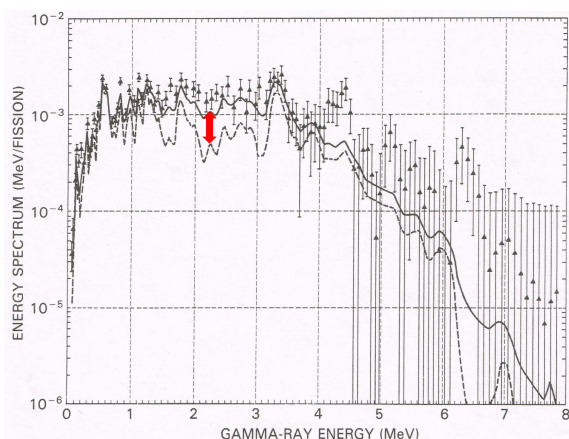
### 3 Brief history of pandemonium problem

Comparing their decay-heat calculations with sample irradiation experiments, they found big discrepancies, a serious overestimation in the  $\beta$ -ray and underestimation in the  $\gamma$ -ray component, contrary to their optimistic expectations to their new libraries in the late 1970's. After a year of discussion they came to a conclusion that this disagreement comes from the pandemonium problem. In parallel at the same period, one of the present authors demonstrated that the gross theory of  $\beta$ -decay [6–8] works remarkably well against the pandemonium problem [9]. Here in addition, let us check the pandemonium effect on the delayed  $\gamma$ -ray spectrum, or the  $\gamma$ -ray component of the decay heat. Figure 3 shows the  $\gamma$ -ray spectrum 2.7 sec after a fission burst in  $^{235}\text{U}$ . The dashed curve is calculated from high-resolution  $\gamma$ -ray based decay schemes and the solid from the gross theory [10]. Comparison with the measured spectrum by Dickens *et al.* [11] indicates the pandemonium effect (red arrow) reaches more than a factor 2 at most and that the gross-theory result is essentially pandemonium free.

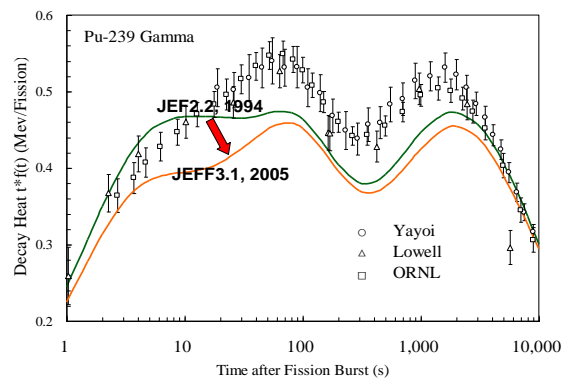
The mean  $\beta$ - and the  $\gamma$ -ray energies had been adopted for most of short-lived FPs in the Japanese evaluated nuclear data library JENDL and in its US counterpart ENDF/B-IV for decades of years until recently when they are gradually being replaced by data from the total absorption gamma-ray spectroscopy (TAGS) on which we will describe hereafter especially in the next section. Anyway, adoption of the gross-theory mean energies is the reason why both JENDL and ENDF/B-IV reproduced the direct decay-heat measurements much better than the European evaluated nuclear data data files, JEF and JEFF, which intentionally exclude theoretical predictions. Because of this policy, however, the European files indicate us the seriousness and persistence of the pandemonium effect in conventional decay schemes. As we see in Fig.4 the underestimation caused by the pandemonium effect is becoming more and more along with the passage of years. This seems to come from the increase of available high resolution  $\gamma$ -based decay schemes toward more neutron rich exotic FPs.

Even though we could circumvent the pandemonium problem by introduction of the gross theory especially for the decay heat and related quantities, experimental decay data for FPs is indispensable for wide range of applications. Construction of decay schemes has its own limitation especially for highly  $Q_{\beta}$ -valued, short-lived nuclides. The best and only way available now seems to be TAGS. The

first extensive TAGS measurements were performed by Idaho group in 1990's [12]. From the viewpoint of usefulness of the TAGS data, Working Party on International Nuclear Data Evaluation Cooperation of OECD/Nuclear Energy Agency started a research coordination of TAGS physicists and data users in 2006 [13] and this activity was then handed over to a series of Consultants' Meeting of IAEA/Nuclear Data Section. Discussions there led to first important results for technetium and molybdenum isotopes by Valencia group [14], which remarkably improved the summation calculation of plutonium decay heat. The IAEA meeting extended its scope to reactor neutrino spectra [15] and Valencia, Oak Ridge [16], Nantes [17] groups responded to this with fruitful TAGS data.



**Fig. 3:** Measured and calculated delayed  $\gamma$ -ray energy spectra

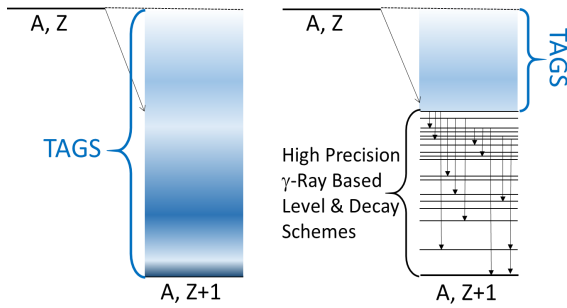


**Fig. 4:** Effect of the pandemonium problem on the decay heat after a burst fission in  $^{235}\text{U}$  sample. The vertical axis is multiplied by the cooling just for concise display

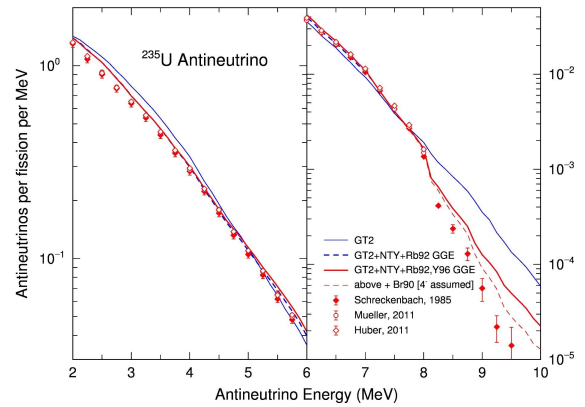
#### 4 Total absorption gamma-ray spectroscopy

The idea of TAGS is to detect the total energy of the  $\gamma$ -ray emitted just after a single  $\beta$ -transition to a certain excited level. In the case of the simplified decay model of Fig.1, a transition from the parent (A,Z) to the 4 MeV level in its daughter (A,Z+1) results in a single 4 MeV photon or two 2 MeV photons. In both cases, the total energy released in the form of  $\gamma$ -ray is 4 MeV. On the contrary, a transition to the 2 MeV level results in an energy release of 2MeV. TAGS uses a large scintillation detector within which all the photons deposit their energy and the data acquisition system gets the signal proportional to the total energy released as photons which is equal, ideally at least, to the excitation energy of the fed level. Though the principle is rather simple but its real execution is quite difficult and requires a complicated data analysis.

As is schematically shown on the left-hand side of Fig.5 TAGS gives us the  $\beta$ -strength function or the  $\beta$ -transition rate per small energy-bin almost up to  $Q_\beta$  above the ground state, the energetic ceiling. It does not, however, provide any information about the level structure or the  $\gamma$ -branching ratios which are very important components of the conventional decay schemes. In order to constitute the best possible  $\beta$ -decay diagram, the TAGS data (pandemonium free) and the current high-resolution scheme, have to be combined at a certain appropriate energy above which level missing becomes sizable as is illustrated on the left-hand side of Fig.5. Then the continuum-to-continuum (bin-to-bin) and the continuum-to-discrete (bin-to-level)  $\gamma$ -transition rates must be calculated and be added into the diagram. It may needs a reliable Hauser-Feshbach type calculation.



**Fig. 5:** TAGS data, the conventional decay schemes obtained from high resolution  $\gamma$ -ray data and their integration



**Fig. 6:** Antineutrino energy spectrum  $^{235}\text{U}$  sample under neutron irradiation at ILL high flux reactor

## 5 Antineutrino as another example

The gross theory of  $\beta$ -decay played an important role to overcome the pandemonium problem. In practice it is widely used to predict the decay behavior of nuclides far off the stability line even now. As an example we introduce here the case of reactor antineutrino  $\bar{\nu}_e$ . The curves shown in Fig.6 were obtained from the  $\bar{\nu}_e$  spectrum of each contributing FP nuclide with the summation method. All the spectra of about 500 FPs were calculated based on the improved version of the gross theory [18, 19], GT2. The NTY treatment applied here was introduced by Nakata *et al.* [20] for odd-odd decaying nuclides. GGE stands for the fact that the grand-to-grand transition rate was enhanced in order to reflect a peculiar behavior of  $^{92}\text{Rb}$  and  $^{96}\text{Y}$ . The three series of experimental data here are all based on the same electron spectrum measured by Schreckenbach *et al.* [21] by three different authors including themselves. The overall agreement between fully theoretical calculation with the experimental data is fairly good.

## 6 Concluding remarks

For decades, the difficulty caused by pandemonium problem has been circumvented by introduction of a  $\beta$ -decay theory. In recent years, the TAGS data is becoming available year by year saving the situation remarkably. The TAGS data (pandemonium free) and the current high-resolution decay-scheme should be integrated into a better description of  $\beta$ - and  $\gamma$ -decay property of FPs for application purposes with the help of some reliable statistical  $\gamma$ -decay theory.

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