Impact of the Total Absorption Gamma-Ray Spectroscopy on FP Decay Heat Calculations

Tadashi YOSHIDA[1], Takahiro TACHIBANA[2], Jun-ichi KATAKURA[3]

[1]*Musashi Institute of Technology, Tamazutsumi 1-28-1, Setagaya-ku, Tokyo 158-8557, Japan*
e-mail: yos@ph.ns.musashi-tech.ac.jp
[2]*Senior High School of Waseda University, Kamishakujii 3-31-1, Nerima-ku, Tokyo 177-0044, Japan*
[3]*Nuclear Data Center, Japan Atomic Energy Research Institute Tokai-mura, Ibaraki 319-1195, Japan*

We calculated the average β- and γ-ray energies, E_β and E_γ, for 44 short-lived isotopes of Rb, Sr, Y, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm and Eu from the data by Greenwood et al, who measured the β-feed in the decay of these nuclides using the total absorption γ-ray spectrometer. These E_β and E_γ were incorporated into the decay data files from JENDL, JEF2.2 and ENDF-B/VI, and the decay heats were calculated. The results were compared with the integral measurements by the University of Tokyo, ORNL and Lowell. In the case of JENDL, where the correction for the so-called Pandemonium effect is applied on the basis of the gross theory, the very good agreement is no longer maintained. The γ-ray component is overestimated in the cooling time range from 3 to 300 seconds, suggesting a kind of an over-correction as for the Pandemonium effect. We have to evaluate both the applicability of the TAGS results and the correction method itself in order to generate a more consistent data basis for decay heat summation calculations.

1. Introduction

Summation calculation is a powerful and versatile method, which is widely used to predict the various aggregate behaviors of fission products (FPs) including the FP decay heat. In doing summation calculation of the decay heat, attention has to be paid on the so-called *Pandemonium problem*[1],[2] or missing of the β-strengths in the high-energy region of the daughter nucleus in the published decay schemes of high Q-valued short-lived isotopes. Calculations based on the decay data suffering from this problem tend to underestimate the γ-ray component of the decay heat and to overestimate the β-ray component. In order to compensate these lost strengths, nuclear-model calculation based on the gross theory of β-decay[3] were performed and the calculated results were adopted in the JENDL FP Decay Data File 2000[4], the preceding version of which is known as the JNDC FP Decay Data Library[5]. The American libraries, ENDF/B-V and VI[6], adopt the same kind of correction to get rid of the Pandemonium problem. By virtue of this recipe the calculated decay heat became highly consistent with the measured one. There still remains, however, small but non-negligible discrepancy in the γ-ray component in the cooling-time range from 300 to 3000s[7]. In order to remedy this persistent disagreement, an European group (Algora and Tain as spokespersons) started the measurement of the β-feeding in the decay of some important FPs including the neutron-rich Tc isotopes[8] by using the total absorption γ-ray spectrometer (TAGS). At least in principle TAGS is free from the β-feed missing, which, for example, leads to the Pandemonium problem. In 1990s there had been an extensive program[9] in which the β-feeds in Rb, Sr, Y, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm and Eu isotopes were measured by this technique.

2. Decay Heat and TAGS

A typical TAGS system consists of a large NaI(Tl) scintillation detector having a deep axial well with the radioactive source in it[8],[9]. In principle all of the γ-rays emitted in a cascade accompanied by the de-excitation of a certain level deposit their energy in the scintilltor giving the level energy to which the β-transition takes place. Therefore the TAGS gives the level energy as the pulse energy and the β-feeding rate as the pulse height simultaneously. These are the basic information required to calculate the average β- and γ-ray energies per one β-decay of the parent nucleus. If the TAGS measurement is carried out ideally, the average β- and γ-ray energies (E_β and E_γ) obtained in this way are of the best quality and free from the Pandemonium problem. In reality, however, there exist several difficulties which might makes the TAGS away from the ideal. These are the photon loss, the β-particle contamination, the finite energy resolution and so on. Partly because of this, JENDL did not adopt TAGS data as the basis of the E_β and E_γ calculation. It is certain, however, that TAGS data are appropriate to yield the E_β and E_γ values in its intrinsic nature.

3. Calculation of the Average Energies

We calculated the average β- and γ-ray energies for 44 short-lived isotopes of Rb, Sr, Y, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm and Eu from the TAGS data obtained by Greenwood et al.[9]. The average β-ray energy release per one β-decay E_β is calculated by using the β-feeding intensities I_β as,

$$E_\beta = \sum_i R(E_i) \cdot [f_\beta(-E_i) / f_0(-E_i)] \ , \tag{1}$$

with

$$R(E_i) = \frac{I_\beta(E_i)}{\sum_i I_\beta(E_i)} \ , \tag{2}$$

$$f_\beta(-E) = \int_1^{-E/mc^2+1} mc^2 (E_0 - 1) p E_0 (-E/mc^2 + 1 - E_0)^2 F(Z_d, E_0) dE_0 \ , \tag{3}$$

where

$$f_0(-E) = \text{The integrated Fermi function.} \tag{4}$$

Here m is the electron rest mass, Z_d the proton number of the daughter nucleus of the β-decay, E_i the transition energy measured from the parent state to the i-th energy level in the daughter nucleus. The function F is the Fermi function. For simplicity, we adopted the integrated Fermi function f_0 of allowed transition for all transitions in the calculation. The average γ-ray energy release per one β-decay E_γ is also expressed as

$$E_\gamma = \sum_i R(E_i) \cdot (Q + E_i) \ , \tag{5}$$

The measured β-feeding intensities of 44 FP isotopes mentioned above are given in Ref.[9]. These FP nuclei have Q_β-values ranging from 1MeV to 6MeV. The values of E_{max}/Q_β are between 69% and 97%, where E_{max} is the maximum excitation energy in the daughter nucleus for which the β-feeding intensity is measured. We adopted these β-feeding intensities for the calculation of E_β and E_γ with the use of the above equations. However, sometimes only ambiguous data are given in the table of Ref. [9]. In such cases, we treated them in the following manner in the calculation;

(1) When only the maximum or minimum values are given, we adopted these values as the β-feeding intensities of the energy levels.

(2) When only total b-feedind intensity of more than two energy levels is given, we share out the total intensity equally among these energy levels.

The vales of E_β thus obtained are compared with those values adopted in JENDL FP Decay Data File[4]. We found that the newly calculated values are, on an average, 0.9 times smaller than those values in JENDL. As for the E_γ values, they are 1.3 times larger than the values in JENDL on an average. The reason of the decrease of E_β and the increase of E_γ is that many new β-feeding intensities are found in the region of the high excitation-energy in the TAGS measurement. The vales of E_γ are affected more directly by these high excitation-energy strength than the values of E_β as can be seen in equations (1) and (5).

4. Decay Heat Calculations

The E_β and E_γ values calculated in Sect. 3 were incorporated into the decay data files from JENDL, JEF2.2[10] and ENDF/B-VI. JENDL and ENDF/B-VI are corrected for the Pandemonium effect and JEF2.2 is not. Using these original and modified libraries, the decay heats after a burst fission were calculated with the summation method and the results were compared with the integral measurement from the University of Tokyo[11], Oak Ridge National Laboratory[12] and University of Massachusetts Lowell[13]. In Figs. 1 and 2 the results for the γ-ray component of Pu-239 are displayed for the original and the TAGS modified libraries, respectively, along with the integral measurements. In the case of JEF2.2, where any theoretical correction is not made for the missing of the β-strengths, improvement is remarkable (dotted curves). This implies that the TAGS detects the high-energy β-strengths as is expected and that the E_β and E_γ values derived therefrom correctly reflects the contribution from the high-energy β-strengths. On the other hand, in the case of JENDL (solid curves), where the correction is applied on the basis of the gross theory, the very good agreement is no longer maintained. The γ-ray component is overestimated in the cooling time range from 3 to 300 seconds suggesting a kind of an over-correction. In this cooling time range, the dominant nuclides which increase the γ-ray component (and correspondingly decreases the β-ray component) from the JENDL original are Cs-141 and La-144 as is seen from Fig. 3, which shows the effect of the nuclide-wise replacement of JENDL E_γ by TAGS energy E_γ. These 11 isotopes shown in Fig. 3 are those which have relative contributions more than 0.5 % in the cooling time range less than 100s.

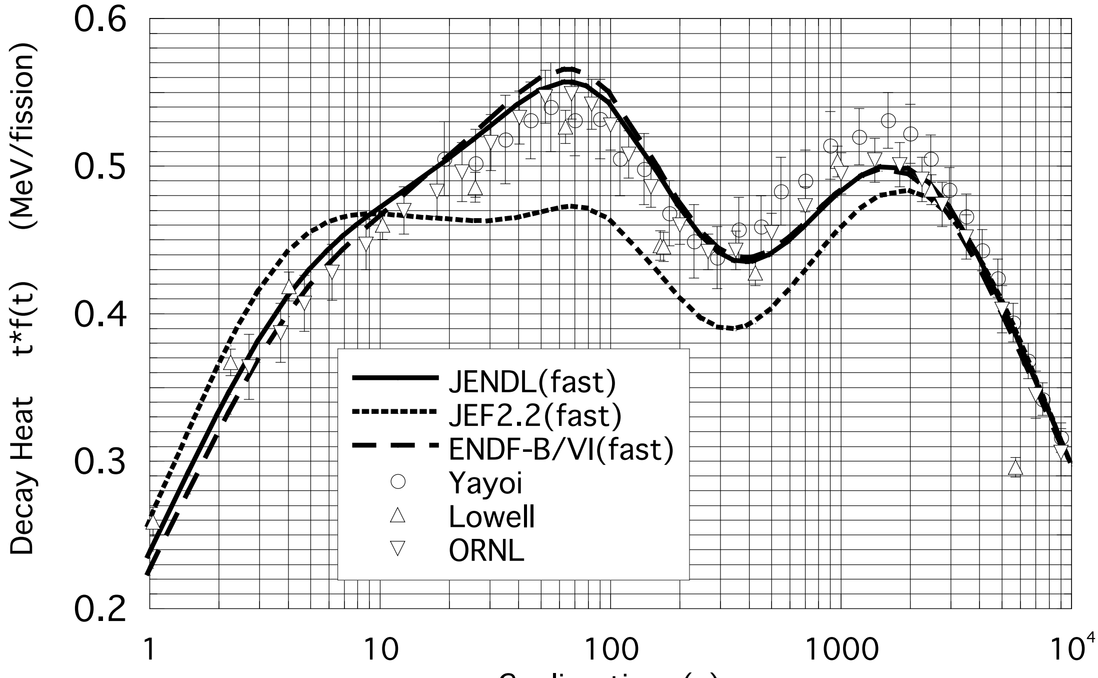

Fig. 1 Decay heat after a burst fission in Pu-239 before the TAGS correction (γ-ray component)

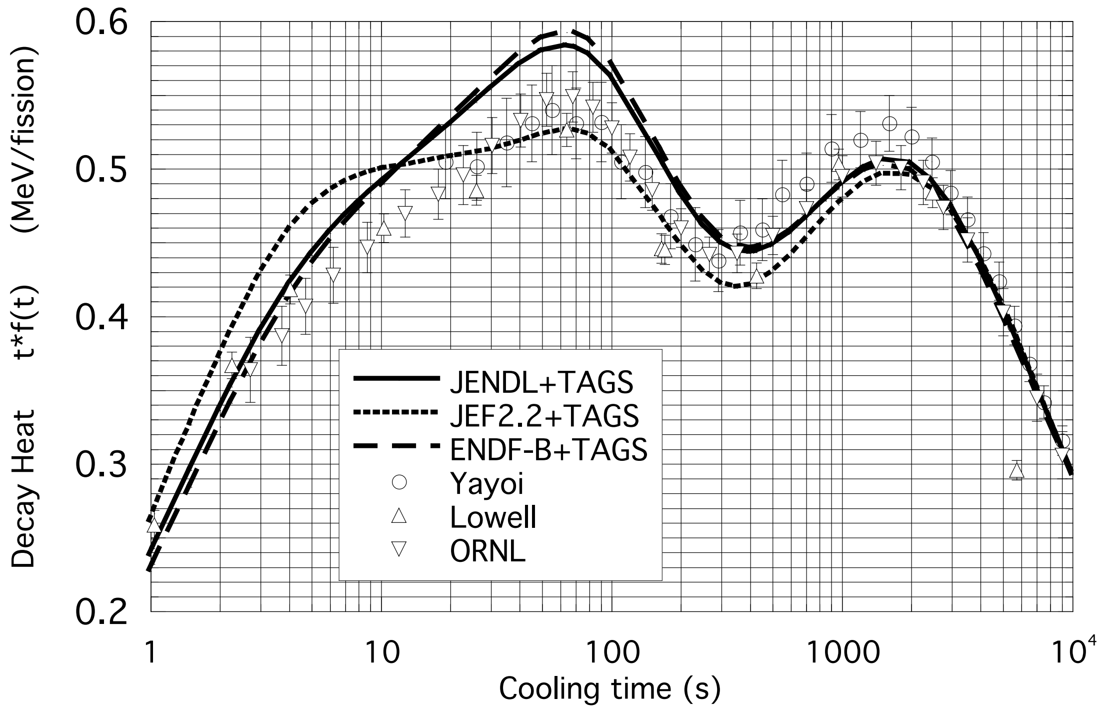

Fig. 2 Decay heat after a burst fission in Pu-239 after the TAGS correction
(γ-ray component)

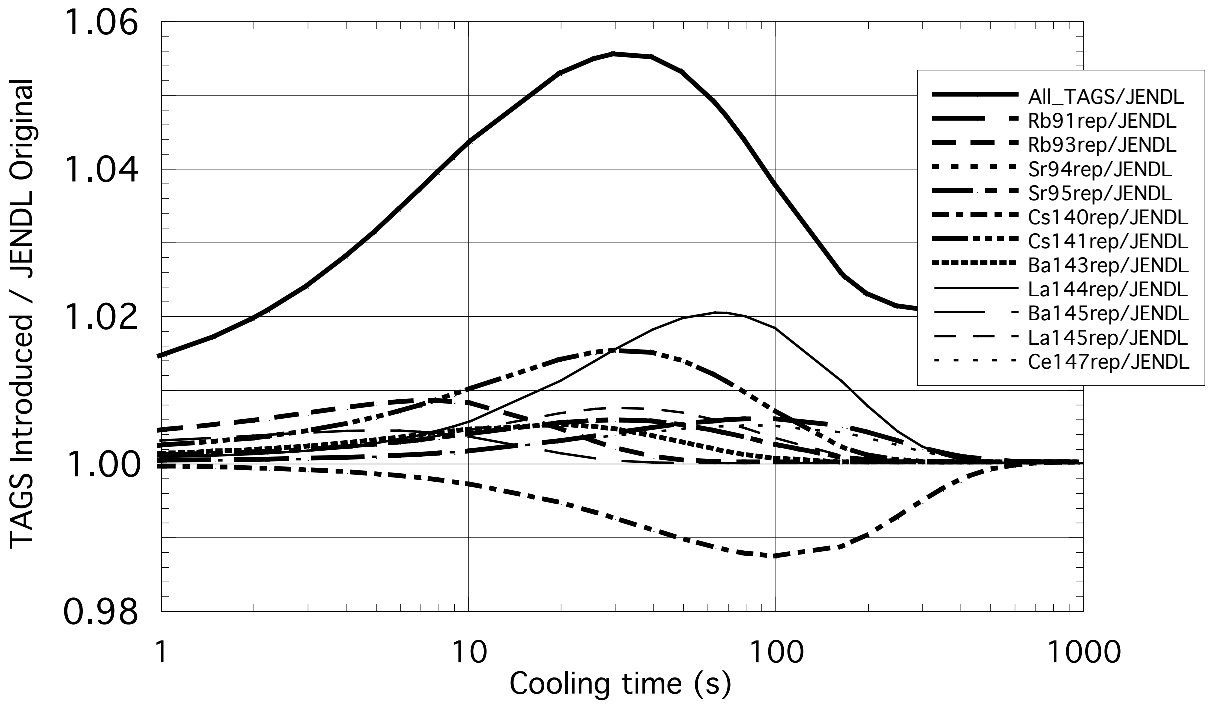

Fig. 3 Effect of introduction of TAGS energies into JENDL summation calculation
(fast fission in Pu-239, γ-ray component)

5. Discussion

As can be seen in Fig. 2, the γ-ray component of the decay heats around 50s cooling for Pu-239 becomes overestimate by adopting the newly calculated E_γ values. These overestimations, which are probably found in other fissioning systems like U-235, Pu-241 and so on too, are mainly due to the increase of E_γ values of many nuclides such as shown in Fig. 3. As for La-144, one of the largest contributors, we should point out that (1.2 ± 0.1) % is given for the β-feed intensity to the ground state of Ce-144 in Ref.[9]. However, this value is very suspicious because this transition is the third-forbidden transition (3^-) -> 0^+. This reduces the reliability of their values for La-144. This value 1.2 ± 0.1 is not adopted in the latest Nuclear Data Sheets[14]. In this way we have to evaluate all of the TAGS data nuclide by nuclide before drawing a definite conclusion.

6. Concluding Remarks

The fact that the JENDL calculation is deteriorated by the introduction of the TAGS energies is an unexpected result. It is, however, still true that JENDL reproduces the short-cooling FP decay heat extremely well[4] for almost all the fissioning systems such as Th-232, U-233, U-235, U-238, Pu-239 and Pu-241, measured at Tokyo, Oak Ridge and Lowell. Taking into the account the recent progress in the TAGS measurements, we have to reconsider the correction method and to try to generate a consistent data basis for decay heat summation calculations.

References

[1] Hardy J.C., Carrez L. C., Jonson B., Hansen P.G.: Phys. Lett., **71B**, 307 (1977)

[2] Yoshida T, Nakasima R.: *J. Nucl. Sci. Technol.*, **18**, 393 (1981)

[3] Takahashi K., Yamada M., Kondoh T.; *Atom. Data and Nucl. Data Tables*, **12**, 101 (1973) and the references therein

[4] Katakura J., Yoshida T., Oyamatsu K., Tachibana T., JENDL FP Decay Data File 2000, JAERI 1343 (2001)

[5] Tasaka K., et al.: JNDC Nuclear Data Library of Fission Products -second Version-, JAERI-1320 (1990)

[6] Katakura J., England T.R.: Augmentation of ENDF/B Fission Product Gamma-Ray Spectra by calculated Spectra, LA-12125-MS (1991) Los Alamos National Laboratory

[7] Yoshida T., Tachibana T., Storrer F., Oyamatsu K., Katakura J.: *J. Nucl. Sci. Technol.*, **36**, 135 (1999)

[8] Algora A., Tain J.L., Private communication, (2002)

[9] Greenwood R.C., Helmer R.G., Putnam M.H., Watts K.D.: *Nucl. Instr. and Meth.* **A390** (1997) 95.

[10] JEF-2.2 Radioactive decay Data, JEF Report 13 (1994) OECD Nuclear Energy Agency

[11] Akiyama, M., An,S.: *Proc. Int. Conf. on Nuclear Data for Science and Technology*, Antwerp, p.237 (1982) and references therein.

[12] Dickens J.K., Love T.A., McConnell J.W., Peelle R.W.: *Nucl. Sci. Eng.*, **78**, 126 (1981)

[13] Nguyen H.V.: *Proc. Int. Conf. on Nuclear Data for Science and Technology*, Trieste, p.835 (1997)

[14] Sonzogni A.A.: *Nuclear Data Sheets*, **93**, 599 (2001)