Needs for accurate measurements of spectrum and multiplicity of prompt γ emitted in Fission

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1. Introduction

The γ energy accounts for about 10% of the total energy released in the core of a thermal or fast reactor. The γ-energy release is much greater in the core of the reactor than in its structural sub-assemblies (such as reflector, control rod followers, dummy sub assemblies). However, because of the propagation of γ from the core regions to the neighboring fuel-free assemblies, the contribution of γ energy to the total heating can be dominant. The γ–heating in a center of a typical fast reactor core [Lut01] comes from several components, roughly:

- 20% from the γ produced in radiative capture
- 40% from the prompt γ emitted by fission fragments
- 30% from the delayed γ produced by fission products
- 10% from the inelastic scattering reactions.

The prompt γ from the fission of ^{235}U, ^{239}Pu, and to a lesser extent ^{241}Pu contribute predominantly to the total γ-energy release in thermal or fast cores fueled with either uranium oxide (UOx) or mixed oxide (MOx). Since the characteristics of γ (multiplicity, total energy and spectrum shape) produced by radiative capture and inelastic scattering is considered to be fairly known, the γ from the fission process becomes the major source of uncertainty in the prediction of γ-heating.

2. Required uncertainties on γ-heating in reactors

Four of the six nuclear systems identified by the Gen-IV international forum are fast reactors (FR). The high performance required for these future FR's calls for very innovative core characteristics compared with conventional fast-reactor designs, which in turns, give rise to new challenges in neutronics methods and data. Presently, the criteria for designing Gen-IV reactors lead to cores using steel or ceramic reflectors without UO_2 blankets. Therefore, it becomes important to calculate precisely the gamma-heat deposition in these materials as well as in control-rod followers or dummy sub-assemblies. For instance, a 7.5% (1σ) uncertainty is required for the energy deposition in non-fuelled zones of Gen-IV fast reactors [Rim05].

The experimental Reactor Jules Horowitz (JHR), expected to start in 2012 at Cadarache, will feature experimental devices in its central part. The γ produced in the surrounding fuel elements account for more than 90% of the total heating of the experimental devices [Bla05]. γ heating needs to be known with a good accuracy and according to the designers, a 7.5% (1σ) uncertainty is required.

3. *Discrepancies between integral measurements and calculations of γ-heating*

In order to verify the adequacy of the calculation of γ heating in fast reactors, the CIRANO and the BALZAC experimental programs in the MASURCA facility have been performed. TLD and γ-fission chambers were used to derive the experimental heating values with rather low uncertainties of about 6%. The comparison of calculated and measured γ-heating values has shown a significant underestimation by about 10% in the PuO2/UO2 core region, by 16% in the reflector and by 11% in the diluent zone [Lut01]. Because of its importance, the γ-production data from Pu239 in current nuclear-data libraries is suspected.

For the JHR project, γ-heating measurements of the experimental programme ADAPh have been performed both in the UO_2 core of the MINERVE reactor and in the MOX core of the EOLE reactor; both reactors being located at Cadarache. The comparison between calculated and measured γ-ray absorbed doses in TLD shows a large underestimation in both cores MINERVE UO2: C/E=0.72 ± 7.5% (1σ) and EOLE MOX: C/E=0.75 ± 7.5% (1σ) [Bla05]. The systematic bias observed is attributed to deficiencies in γ-production data in the evaluated file. As U235 is the most important contributors in prompt γ production in the MINERVE UO2 core, a deep analysis of this isotope data is required. Similarly to fast systems, the EOLE MOX core is indicating that γ-production data from Pu239 in current nuclear-data libraries is highly suspicious.

4. *Prompt fission γ-production data in nuclear-data libraries*

In the most recent nuclear-data libraries, γ-production data for the prompt γ emitted in fission are described the file MF12 (γ multiplicity) along with the file MF15 (pointwise description of the spectrum). Redundant information can be found in file MF1 - MT458 (Components of energy release due to Fission). A quick inspection of the recent libraries, JEFF3.1, ENDF/B-VI.8 and JENDL3.3, shows that evaluated data are based on rather old measurements of the photon spectrum of U235 [Ver72], [Pee71]. In general, the same spectrum shape is chosen for every fissile isotope. The total energy Eγ and multiplicity Nγ are also based on measurements or simple systematics (see next section). No incident-neutron energy dependence is described in any existing nuclear-data libraries.

5. *Overview of existing differential measurements*

The prompt γ-spectra have been measured for the main fissile isotopes in the early 70's. Table 1 summarizes the measurements that can be found in the open literature.

Isotope	Total energy MeV	Total Multiplicity MeV	Average Energy MeV	Energy Range MeV	Reference
U233	6.69 ± 0.3	6.31 ± 0.3	1.06 ± 0.07	*0.09-10.0*	*[Ple72]*
U235	6.43 ± 0.3	6.51 ± 0.3	0.99 ± 0.09	*0.09-10.0*	*[Ple72]*
	6.51 ± 0.4	6.70 ± 0.3	0.97 ± 0.05	*0.14-10.0*	*[Ver72]*
	7.18 ± 0.3	7.45 ± 0.3	0.96 ± 0.05	*0.14-10.0*	*[Pee71]*
	7.25 ± 0.3	8.13 ± 0.3	0.89 ± 0.05	*0.01-10.5*	*[Pee71]*
PU239	6.73 ± 0.35	6.88 ± 0.4	0.98 ± 0.07	*0.09-10.0*	*[Ple72]*
	6.81 ± 0.3	7.23 ± 0.3	0.94 ± 0.05	*0.14-10.0*	*[Ver72]*
Cf252	7.06 ± 0.3	8.32 ± 0.4	0.85 ± 0.06	*0.09-10.0*	*[Ple72]*
	6.84 ± 0.3	7.80 ± 0.3	0.88 ± 0.04	*0.14-10.0*	*[Ver72]*
	8.6	*10*	*0.90* ± 0.06		*[Bow58]*
	6.7 ± 0.4				*[Nar73]*

Based on the existing measurements, some systematics have been derived by Hoffman et al [Hof74] and Frehaut et al. [Fre88] to predict the total γ-energy and multiplicity as a function of fissioning-nuclei mass.

Table 1 illustrates the significant dispersion of the experimental values and demonstrates the high uncertainty (~15%) that should be assigned to the total prompt γ-energy. More recent measurements have been performed on ^{252}Cf [Ham95], [Gla89] to get the spectrum shape versus fission-fragments mass, unfortunately, the total γ-energy or the number of γ was not reported. Nevertheless, these measurements are of great importance to check the modeling of the fission process and fragment decay.

6. *Modeling issues*

Over the past 20 years, an important effort has been done to predict the γ-production data for radiative capture and inelastic scattering process and is summarized for instance in the CRP report of IAEA [CRP99]. However, very little work has been devoted to the prompt γ produced by fission. So far, a tractable model (such as the Madland-Nix model for prompt neutrons) is not available for evaluation purposes and evaluators have to rely on the old measurements described in the previous section.

The main issue is to be able to predict the excitation-energy and spin distribution of the fission fragments. Even with the most sophisticated fission theories, these quantities are not accurately predicted as a function of fragment mass and charge. Furthermore, a rigorous modeling requires to treat the decay of the fission fragments and the neutron-γ emission by the Hauser-Feschbach model. This cumbersome approach (see [Gru01] for instance) requires the knowledge of a great number of nuclear parameters of highly-deformed nuclei (level density, transmission coefficient for gamma and neutron and so on.). These issues emphasize the needs for accurate measurements.

7. Conclusion

As illustrated by the generation-IV design objectives and the JHR project, a better accuracy on γ-heating prediction is required by nuclear-reactor analysts. Integral measurements performed in thermal and fast mock-up have demonstrated a significant underestimation of the total γ-heating that deserves a deeper understanding.

One of the major source of uncertainty comes from prompt γ emitted in fission. The data found in modern nuclear-data libraries are still based on experiments performed in the early 70's and a high uncertainty is expected on quantities like total γ energy or multiplicity.

To reduce the present uncertainties, the total energy, multiplicity and the spectrum shape of prompt photons emitted in fission needs to be accurately measured. The priority is to measure ^{235}U and ^{239}Pu at thermal and fast incident-neutron energies. The required accuracy is 7.5% for the γ energy and the multiplicity. A more important effort should be also devoted to the modeling of these quantities.

References

[Lut01] A. Lüthi, R. Chawla, and G. Rimpault, *Nucl. Sci. Eng.* 138, ·3 ·(2001). ·

[Rim05] G. Rimpault, *Proceedings of the Workshop On Nuclear Data Needs for Generation IV*, April 2005, Antwerp, Belgium.

[Bla05] D. Blanchet, *Proc. of M&C 2005 International Topical Meeting on Mathematics and Computation, Supercomputing, Reactor Physics and Nuclear and Biological Applications* **,** September 12–15, 2005, Palais Des Papes, Avignon, France.

[Pee71] R.W.Pelle and F.C. Maienschein, *Phys. Rev.* **3**, 1, 373 (1971).

[Ple72] F. Pleasonton et al. Phys. Rev. C6, 1023 (1972)

[Ver72] V. V. Verbinski et al., Phys Rev C7, 1173 (1973).

[Bow58] H. R. Bowman and S.G.thomson, *Proc. 2nd International Conference on Peaceful uses of Atomic Energy*, Vol. 15, 212 (1958).

[NaR73] E. Nardi, A. Gavron and Z. Faenkel , Phys. Rev. C8, 2293 (1973).

[Val69] Val'skii et al. , Sov. Jour. Of Nucl. Phys. 10, 134 (1969).

[Hof74] Hoffman and Hoffman, Ann. Rev. Nucl. Sci. 24 (1974).

[Fre88] J. Frehaut. *I.A.E.A consultants meeting physics of neutron emission in fission* Mito, Japan (1988).

[Ham95] F.J. Hambsh, J. van Aarle and R. Vogt, *XIII Meeting on physics of Nuclear. Fiss*ion Obninsk (1995).

[Gla98] P. Glassel et al., Nucl. Phys. A502 (1989).

[CRP99] INDC(NDS)-412 Dec.199.[Gru01] O. T. Grudzevitch, Phys. At. Nucl., Vol. 64, No.1 (2001).