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Nuclear data production, calculation and measurement: a global overview of the gamma heating issue

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Abstract. The gamma heating evaluation in different materials found in current and future generations of nuclear reactor (EPR^{TM}, GENIV, MTR-JHR), is becoming an important issue especially for the design of many devices (control rod, heavy reflector, in-core & out-core experiments…). This paper deals with the works started since 2009 in the Reactor Studies Department of CEA Cadarache in ordre to answer to several problematic which have been identified as well for nuclear data production and calculation as for experimental measurement methods. The selected subjects are:

- Development of a Monte Carlo code (FIFRELIN) to simulate the prompt fission gamma emission which represents the major part of the gamma heating production inside the core,
- Production and qualification of new evaluations of nuclear data especially for radiative capture and inelastic neutron scattering which are the main sources of gamma heating out-core
- Development and qualification of a recommended method for the total gamma heating calculation using the Monte Carlo simulation code TRIPOLI-4©
- Development, test and qualification of new devices dedicated to the in-core gamma heating measurement as well in MTR-JHR as in zero power facilities (EOLE-MINERVE) of CEA, Cadarache to increase the experimental measurement accuracy.

1 Context

Nuclear heating in power reactor is coming from the energy deposition of charged particles produced by neutron and photon interactions, and the radioactive decay of fission products. Inside the core, 10% of the deposited energy is due to the gamma when it represents almost the entire heating in the reflector (Figure 1).The main source of gamma heating in the core is the fission reaction which represents about 60% (one third from the delayed photons) of the total amount followed by the radiative capture (about 20%) and the inelastic scattering. This distribution may be different from one to another reactor depending on its specific material balance.

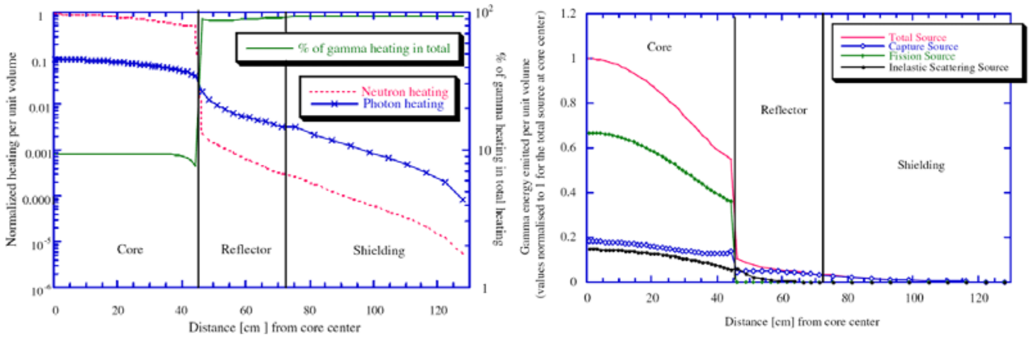

Fig. 1. Energy deposition in a Fast Reactor & Gamma heating relative contribution sources [1]

In the next few years, the cooling system in power reactors for fast as well as for thermal designs will be a major subject. The new concepts involved for the construction of this future reactors (Pressurized Water Reactor, GENIV as Fast Reactor, Material Testing Reactor-Jules Horowitz Reactor) are based on the addition of devices that will be exposed to high levels of nuclear heating inside or close to the core. In PWR, the addition of new features in view to increase safety (ex: burnable poisons such as UO_2-$\mathbf{Gd}_2O_3$ pins for PWR…), performance (**steel** control rod followers and diluents for FR) and lifetime (stainless **steel** heavy reflector) is directly affecting the primary coolant sizing. Similar issues are encountered in the JHR conception with **Hf** control rods, **Al** structures and a **Be** reflector. The designs of the experimental setups that will be introduced in the core or in the reflector imply also a good evaluation of the nuclear heating level in both locations. A well detailed description of the gamma heating determination stake is given in reference [2]. Improving nuclear data evaluations, reducing experimental measurement uncertainties and with the help of dedicated experimental programs, a better description and simulation of the phenomena can be reached. This principal was at the origin of the motivation to start several PhD works at CEA. At the same time, the Gamma Heating Working Group was created to coordinate the different works (meeting twice a year). A compilation of these works is presented in the following parts.

2 NUCLEAR DATA PRODUCTION

The nuclear data library JEFF3.1.1 contains most of the photon production data. Nevertheless, some data were missing (no photon production for all neutron interaction for Gd, Cd, Ag, In, which are nuclei of particular interest for JHR and PWR, no spectrum of the prompt gamma emitted by fission of $^{239,\ 241}$Pu) or inconsistent (^{54,56,57,58}Fe, ^{62}Ni) [3], or based on quite old measurements (prompt fission gamma emission of ^{235}U). A new evaluation for radiative capture and inelastic scattering is proposed. These data are performed with the nuclear reaction code TALYS using a level density model and a strength function model to describe the transition probabilities. A nuclear structure library is also needed (RIPL2.0 by default or the compilation of latest measurements performed at the Budapest facility EGAF and the ENSDF library). For example, the ^{54}Fe analysis shows that the total gamma emission energy after a radiative capture is underestimated for more than 50%. At the end, 16 nuclei have been checked and new prompt gamma spectra from neutron radiative capture are proposed for the next beta version of JEFF (MF6-MT102 standards) [4].

As it has been shown on figure 1, the main contribution to the inner core gamma heating is coming from the fission reaction. It is also well known that even in the most recent nuclear data libraries (JEFF3.1.1, ENDF/B-VI.8 and JENDL3.3) gamma production data for the prompt gamma emitted in fission are lacking or based on quite old measurements. For the main fissile isotopes (^{235}U, ^{239}Pu), the prompt gamma spectra were measured in the early 70’s and great discrepancies are observed between the obtained measurements. Since that time, very little work has been devoted to the prompt gamma production from fission even if new measurements have been or will be done. Moreover, the fission process is highly complex to be described by theoretical models (spin distribution and excitation energy) and even today, no qualified model is available for evaluation purpose. Nevertheless, a Monte Carlo code (FIFRELIN) dedicated to the simulation of the fission fragment de-excitation is being developed at CEA-Cadarache [5]. A specific module based on the de-excitation gamma cascade simulation which needs a density level distribution model associated to a strength function, has been developed for the prompt fission gamma emission simulation [6]. Preliminary results have been obtained for a one nucleus de-excitation (^{55}Mn radiative capture, de-excitation of ^{144}Ba) and ^{252}Cf spontaneous fission. These first results indicate that the choice of the model describing the density level is the main source of discrepancy before the strength function. A good agreement has been reached for these tested cases even if a systematic overestimation is observed for ^{55}Mn radiative capture and ^{252}Cf spontaneous fission.

3 SIMULATION CODES

The Monte Carlo simulation code TRIPOLI-4© associated to the JEFF3.1.1 library and up to date evaluations, is used as a reference for the validation of gamma heating calculation. The last version (TRIPOLI4.8) allows coupled neutron-photon-electron-positron calculations. A methodology will be investigated to reach reasonable duration which will be very useful for the evaluation of the prompt gamma contribution. The delayed gamma part will be obtained combining PEPIN code results for the generation of the input gamma spectra and coupled photon-electron-positron TRIPOLI calculation. The final objective is the qualification of the coupled neutron-photon deterministic calculations with the APOLLO2 code using the Method Of Characteristic.

4 INTEGRAL EXPERIMENTS AND INSTRUMENTATION

4.1 Introduction to gamma heating formalism

Nuclear heating can be shared into a neutron and a photon contribution. The neutron or photon heating is due to the kinetic energy released by the secondary charged products induced by the uncharged particles interactions with the traversed material. The heating rate $H(E)$ generated by a neutral particle of incident energy E in a given volume of material made of i elements, is commonly expressed in terms of KERMA (Kinetic Energy Release in MAterials) factors $k_{i,j}$ [7]:

$$H(E) = \sum_i \sum_j \rho_i k_{i,j}(E)\phi(E) \quad [\text{eV.s}^{-1}] \tag{1}$$

Here, ρ_i is the number density of material i [cm^{-3}], $\phi(E)$ is the particle volume integrated flux [cm.s^{-1}] at E and $k_{i,j}(E)$ is the kerma factor for element i and reaction j at energy E:

$$k_{i,j}(E) = \overline{E}_{i,j}(E)\sigma_{i,j}(E) \tag{2}$$

The kerma factor represents the total kinetic energy released $\overline{E}_{i,j}(E)$ [eV] in element i by the charged reaction products and recoil nucleus produced during the reaction j at incident energy E and with the corresponding microscopic cross section $\sigma_{i,j}(E)$ [cm^2]. As the gamma heating is concerned, the main contributions are coming from the photoelectric effect, the Compton scattering and the pair production. The useful experimental KERMA unit [Gy] is related to the deposited energy in a material integrated over time (i.e. irradiation duration) and generally corrected with a normalization factor (i.e. source intensity or power reactor correction factor).

4.2 Experimental results

Zero power facilities called EOLE and MINERVE dedicated to experimental neutron physics purposes are located at CEA Cadarache. The experimental programs carried out are devoted to both measurement technique development and simulation code experimental validation. In such reactors, the gamma heating measurements are done with Thermo-Luminescent Detectors (TLD) or Optically Stimulated Luminescent Detectors (OSLD).

Recently, the ADAPH$^+$ program has been carried out in the UO_2 core of the MINERVE research reactor (R1-UO2 configuration to be representative of a PWR standard spectrum) with calcium fluoride TLD-400 (CaF_2:Mn) and Al_2O_3:C OSLD. The objective was to reduce the different sources of uncertainties by improving the gamma heating calibration procedure and the interpretation of the experimental results [8]. The first step consisted in irradiating the detectors with a standard ^{60}Co gamma-ray source to define the calibration curves. Each TLD-400 is characterized with its own calibration curve because previous works have demonstrate that within the same manufacturing batch a 15% standard deviation could be reached when it is only 2% for OSLD. The detectors were set in aluminium pillboxes which had sufficient thickness to ensure electronic equilibrium in the detector [9] for this experimental set-up. Moreover, one particular advantage of the OSLD is that they can be read repeatedly (only once for the TLD). The uncertainties on the calibration factors remained close to 2% even for the TLD. The evaluation of uncertainties associated to the absorbed dose measurements in the core is calculated by combining the uncertainties on the calibration factors and of the signal response repeatability. The results have been compared to a two step Monte Carlo simulation needed to estimate the cavity correction factor associated to the in-core irradiation (respectively 1.07 and 1.05 for the TLD and OSLD). The simulated results have also been corrected with the delayed gamma ray contribution which is not evaluated in the calculation and represents 32% of total gamma dose. At last, the experimental measurements must be corrected of the neutron contribution due to the sensitivity of the detectors to neutrons with different energies. This contribution of neutron dose to the total dose integrated has been evaluated to respectively 8.6% and 5.9% for TLD and OSLD. Finally, the C/E remains to 1.05±5.3% for TLD and 0.96±7.0% for OSLD (k=2). Further work is planed to be done to explain the discrepancy between the two types of detector and to define a procedure for the delayed gamma ray and neutron contributions calculation.

The integral experiment called PERLE has been performed in EOLE facility. It is a regular PWR type core, with 3.7% enriched UO_2 pins surrounded by a 22 cm thick stainless steal reflector. Some TLD have been positioned inside the core and the reflector to measure the gamma KERMA. One of the purposes of PERLE program is to estimate the gamma heating in a heavy reflector for EPR™. The stainless steal is a Fe, Ni, Cr alloy. The nuclear data library fails in giving accurate evaluation for photon production of Fe isotopes and ^{62}Ni (see chapter 2). The PERLE experiment will allow qualifying new data evaluations for these isotopes [3]. Moreover, a recommended method for the total gamma heating one step calculation will be developed with The Monte Carlo simulation code TRIPOLI-4© with the objective to evaluate directly the deposited energy within the whole geometry.

The AMMON program is currently performed in the EOLE facility. This program is dedicated to the JHR neutron parameters validation. It is made of 7 JHR cylindrical assemblies inserted in an aluminium alloy rack surrounded by a driver zone consisting in 3.7% enriched UO_2 fuel pins. Different configurations will be tested (inserting Hf rod in the central JHR assembly, replacing a JHR assembly by a Be cylinder…). These configurations will be instrumented with TLD and OSLD detectors to evaluate the gamma KERMA in aluminium, hafnium and beryllium which are problematic element especially for the design of future experimental devices (performance, safety), and the aluminium structure of the beryllium reflector (sizing the coolant system). The first results are waited for the end of 2012.

A specific experiment called CARMEN-1, has been recently tested in the 70 MW power reactor OSIRIS (CEA-Saclay). The objective of this program is to combine the analysis of neutron and photon flux measurements for the JHR core mapping [10]. The instrumentation is composed of three different kinds of sensors for the neutron flux measurement (U and Pu fission chambers, Self Powered Neutron Detector), two different kinds of sensors for the photon flux (Ionization chamber, Self Powered Gamma Detector) and two other different ones are dedicated to the nuclear heating (Differential calorimeter and Gamma Thermometer commonly used in MTR). Three different methods have been chosen to estimate the nuclear heating from temperature measurements (by the mean of a preliminary calibration, the zero reading method and the current addition method). The results obtained will be compared with the goals to reduce uncertainties of the nuclear heating and to identify the more appropriate method. They will allow correlating the measurements between the two sensors and the two materials in which the nuclear heating is measured (graphite in calorimeter and stainless steel in GT). Even if the nuclear heating measured is mainly due to photon, the neutron contribution must be corrected (from few % in stainless steal up to 10% for graphite) combining neutron and photon measurements. This device will be use in the JHR core. One of the objective will be the measurement of the nuclear heating which is a crucial parameter because it drives the temperature of the future experimental devices and their samples.

5 Conclusion

During the last few years many programs have been supported especially for new generation reactor: EPR™ (GENIII$^+$) and MTR-JHR. One of the subjects of interest was the measurement of gamma heating for checking nuclear data evaluations which are obviously failing for some isotopes of very high importance (Fe, Hf, Al, Be). Different programs dedicated to improve the measurement methods (ADAPH$^+$ in MINERVE, CARMEN-1 in OSIRIS) and the nuclear data for photon production (PERLE and AMMON in EOLE) have been planed. All these works are linked and have to be developed using the progress of each others. They are still under study by the way of PhD works started since 2009 which are presented in this paper. The role of the Gamma Heating Working Group is to make exchange easier between students and to enlarge the public to others research group (CEA,DEN,DTN, Mc Master University from Canada, University of Marseille-Provence). In the end, better nuclear data, validated calculation methods and tools are waited for an accurate estimation of gamma heating and flux inside reactors

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