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Review of nuclear data improvement needs for nuclear radiation measurement techniques used at the CEA experimental reactor facilities

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Abstract. The constant improvement of the neutron and gamma calculation codes used in experimental nuclear reactors goes hand in hand with that of the associated nuclear data libraries. The validation of these calculation schemes always requires the confrontation with integral experiments performed in experimental reactors to be completed. Nuclear data of interest, straight as cross sections, or elaborated ones such as reactivity, are always derived from a reaction rate measurement which is the only measurable parameter in a nuclear sensor. So, in order to derive physical parameters from the electric signal of the sensor, one needs specific nuclear data libraries. This paper presents successively the main features of the measurement techniques used in the CEA experimental reactor facilities for the on-line and off-line neutron/gamma flux characterizations: reactor dosimetry, neutron flux measurements with miniature fission chambers and Self Power Neutron Detector (SPND) and gamma flux measurements with chamber ionization and TLD. For each technique, the nuclear data necessary for their interpretation will be presented, the main identified needs for improvement identified and an analysis of their impact on the quality of the measurement. Finally, a synthesis of the study will be done.

1 Introduction

The constant improvement of the neutron and gamma calculation codes used in the experimental nuclear reactors goes hand in hand with that of the associated nuclear data libraries such as JEFF-3.1.1 [1], ENDF/B-VII.1 [2], JENDL4.0 [3]…The validation of these calculation schemes always requires the confrontation with integral experiments performed in experimental reactors to be completed. Nuclear data of interest, straight as cross sections, or elaborated ones such as reactivity, are always derived from a reaction rate measurement which is the only measurable parameter in a nuclear sensor. So, in order to derive physical parameters from the rough electric signal of the sensor, one needs specific nuclear data libraries. These nuclear data libraries are either directly extracted from the general libraries when the data exist, or re-evaluated in dedicated libraries such as IRDFF. However, the reactions put in games being generally of not much interest for neutron calculation codes, these data are often old and associated to high level of uncertainty when existing. As efforts needed to proceed to new evaluation or new measurements are rather consequents, a motivation for each ones

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should be established. This paper is thus going to present successively the main features of the measurement techniques used in the CEA experimental reactor facilities for the on-line and off-line neutron/gamma flux characterizations: reactor dosimetry, neutron flux measurements with miniature fission chambers and Self Power Neutron Detector (SPND) and gamma flux measurements with ionization chamber and TLD. For each technique, the nuclear data necessary for their interpretation is presented, the main needs for improvement identified as well as an analysis of their impact on the quality of the measurement. Finally, a synthesis of the study will be done.

2 Nuclear data used in the main measurement techniques

2.1 Activation measurement techniques

2.1.1 Neutron measurements

In the reactor dosimetry process applied at the CEA Cadarache [4, 5], reaction rates and neutron fluences are derived from activity measurements of irradiated dosimeters using the inversion of the Bateman equation. These dosimeters (figure 1) are selected according to their response functions on the studied neutron spectrum (neutron cross section) and to their characteristics with respect to activity measurement process (nature and energy of the emitted particles, decay constant). Typical precision estimates for these fluence monitors are estimated to range from 3 to 10%.

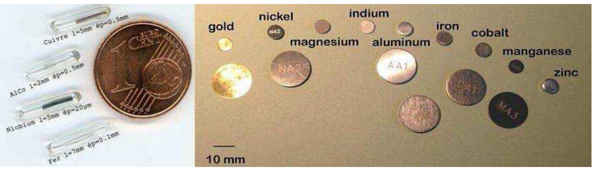

Figure 1. Typical set of dosimeters

Decay data used to derive the dosimeter's activity from the rough experimental counting spectra are extracted from specific libraries, for instance, Nucleides [6] or NUDAT [7]. For deriving neutron fluences from these activity measurements activation cross sections are taken from international dosimetry activation files, IRDFF [8], themselves extractions from the main international nuclear libraries ENDF-B/VII , JEFF3.1.1, JENDL4.0, or EAF2010 [9].

These nuclear data are not only used for activity measurements and reaction rate derivation (mean values) but also to assess output uncertainties (covariance). In addition the latter could also be used in an unfolding spectrum analysis combing several dosimeters results, each one covering different ranges of the studied neutron spectrum.

Thus, nuclear data accuracies impact both measurement and interpretation processes. Although the reactor dosimetry process is used as a reference for absolute calibration of other methods and nuclear sensors, dosimetry nuclear reactions are not of much interest for neutron calculation codes; they could present poor or discrepant evaluations in general purpose data library due mainly to a lack of experimental evaluations [4]. We shall note here the constant effort realized by the nuclear data service NDS of the IAEA to propose and maintain compilations as up to date as possible of these data with covariance matrix evaluations when possible, for example, the IRDFF validation project CRP [8]. Focusing on dosimetry measurements performed at the CEA Cadarache, particular nuclear data improvement needs could be identified. The $^{117}Sn(n,n')^{117}Sn^{m}$ reaction is an interesting estimator for upper part of the epithermal and for the fast neutron flux evaluations in short duration (minutes up to few days) and low to intermediate level power irradiations because of its unique characteristics combination: $E_{Threshold} = 0.314$ MeV, $T_{1/2} = 14$ d,
$E_{g1} = 158$ keV $I_{\gamma 1} = 86\%$.

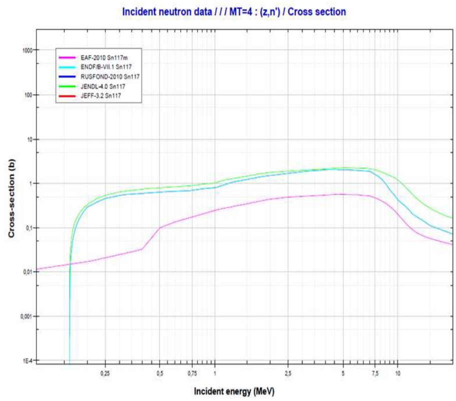

Figure 2. Comparison of the ^{117}Sn(n,n')^{117}Sn^{m} cross section evaluations.

Enriched Tin dosimeters (93% at. ^{117}Sn) have been tested in different spectra from Mockup Reactor (EOLE) to MTR facilities (OSIRIS). The associated measurement activity method has been upgraded at MADERE facility (CEA/Cadarache) up to reach 5% uncertainty (1σ). However, figure 2 extracted from JANIS 4.0 [10], shows that nuclear data still need upgrades to allow this reaction to be used in absolute way: discrepancies between the different library evaluations and lack of uncertainties.

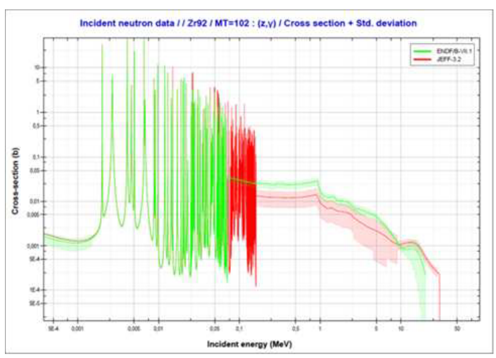

Figure 3. Comparison of the ^{92}Zr(n,γ)^{93}Zr cross section.

Continuing the investigation for reactions able to give information between 10keV and 1MeV, the ^{92}Zr(n,γ) reaction has been identified as very promising [11]. ^{92}Zr enriched Zirconium dosimeters have been irradiated under BN cover at OSIRIS giving promising results. Here again nuclear data need to be upgraded due to large discrepancies between evaluations (figure 3).
In addition, the status of the following reactions needs to be revised:

- ^{93m}Nb(n,γ)^{94}Nb cross section is unknown leading to possible errors in ^{93m}Nb activity analysis in the MTR high level thermal flux conditions. The ^{93}Nb(n,n')^{93m}Nb reaction is one of the most important estimator for fast neutron fluence in MTR and Reactor Vessel Survey.
- ^{103}Rh(n,n')^{103}Rh^{m} reaction, used in many benchmarks used for ZPR neutron calculation scheme validation, presents C/M discrepancies over 10%. Mass attenuation coefficient μ/ρ, I_x^{103}Rh^{m} and cross section should to be reevaluated.
- Calculation/Measurement discrepancies (around 10%) are still observed for ^{55}Mn(n,γ)^{56}Mn reaction [12].
- ^{237}Np(n,f) cross section used for interpretation of the Vessel Surveillance capsules shows high level of uncertainties and evaluation discrepancies in the 1keV – 100keV range.

More generally speaking, experimental benchmarks for integral cross sections should be revisited in order to take into account the effect of the decay data evolution since the measurements were made.

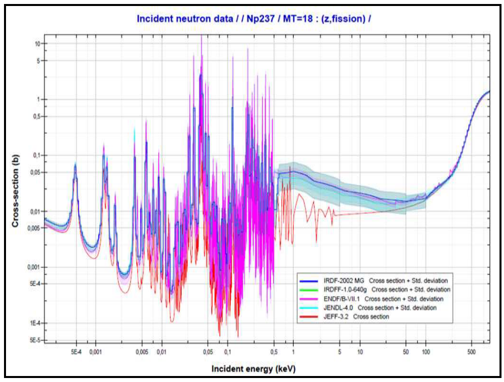

Figure 4. Comparison of the ^{237}Np(n,f) cross section.

2.1.2 Gamma measurements

Two types of dosimeters are commonly used for the photon heating measurements in the CEA mockup reactors where gamma flux level: TLD (CaF2:Mn) and OSLD (Al2O3:C). The dosimeters are made of ionic crystal containing point defects generated by the addition of impurities (Mn or C) which creates localized energy states in the band gap. If an external radiation deposits energy with the slowing down of a charged particle in the crystal lattice, an electron can go up from the valence band to the conduction band. This electron which moves in the conduction band tends to go down to the valence band and can be trapped (energy level in the band gap - Figure 4 a). The energy stored in the traps is proportional to the deposited dose in the crystal. For measurement, dosimeters are thermally (TLD) or optically (OSLD) stimulated. Trapped electrons break free from their traps and move freely in the conduction band until their recombination with a hole. The latter, is associated with a photon emission of a specific energy if the center of the hole is a luminescent center (case of a radiative recombination). The measured light intensity is detected by a photomultiplier tube in the reader and is directly proportional to the energy deposited by electrons/positrons in the crystal. Measurement Method is described in detail in [13, 14].

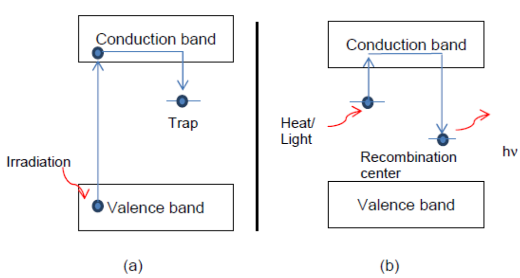

Figure 5. TLD and OSL schematic process during the irradiation (a) and the measurement step (b) after irradiation.

The measurement analysis is based on Monte Carlo TRIPOLI4© [15] calculations modelling the core exact three-dimensional geometry. The JEFF nuclear data library is used for the calculation of the neutron transport and the photon emission. The photon transport is made on the basis of the EPDL97 photo-atomic library [16]. The prompt and delayed doses deposited in dosimeters are estimated separately. The transport of 4 (neutron, photon, electron and positron) or 3 particles (photon, electron and positron) is simulated in the calculations depending whether the prompt or delayed dose is calculated. The TRIPOLI4© calculations enable to model the electromagnetic cascade shower with both electrons and positrons using EEDL (Evaluated Electron Data Library) [17] and the atomic relaxation with the EADL (Evaluated Atomic Data Library) library [17].

The evaluation of the contributions of the gamma radiation to the total and local peak power is an important stake for nuclear core physic of NPPs but also for designing irradiation devices in MTRs. Since the end of the 90s, CEA has upgraded this method for in EOLE and MINERVE facilities [18] for experimental validation of neutron/gamma calculation schemes used for reactor core modelling.

Latest experimental CEA results show a delayed dose of about 25% of the total photon energy deposition in the dosimeters. A systematic underestimation of the calculated global photon energy deposition is still remaining but has been reduced drastically from about -20% ± 13 % to -8 % ± 4.5 % (1σ) [18]. This underestimation could be partially explained by an underestimation in the JEFF3 prompt photon emission ($^{235}U(n,f)$) and for ^{27}Al neutron interactions in the sensor surrounding [18].

However, concerning measurement process itself, low level of covariance data information and lack of recent reevaluated data in EEDL, EPDL, EADL and EXDL limits uncertainty derivation. However, these files have been recently updated by D.E Cullen (August 2015) and renamed EPICS2014 [19]. Improvements brought by this version will be evaluated on existing measurements data.

2.2 Online neutron measurement techniques

2.2.1 Self Power Neutron Detector (SPND)

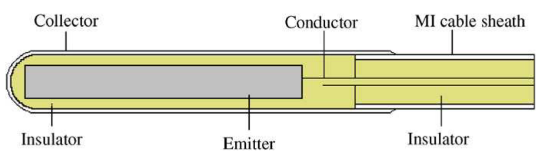

Figure 6. Schematic diagram of a SPND.

Since the 60s SPNDs are widely used in MTRs and NPPS to monitor thermal neutron flux. The SPND (Figure 6) consists of three main parts: the emitter (V, Rh, Co, Pt), the insulator (Al_2O_3 or MgO) and the collector (Inconel). Electrons are emitted when exposed to radiation then they penetrate the thin insulation around the emitter and reach the outer sheath. Some electrons are also emitted from the insulator and sheath. The net flow of electrons from the emitter gives rise to a DC signal between the emitter and sheath, which is proportional to the incident neutron flux. Rh and V SPNDs work on the basis of (n, β) reactions and are used for flux mapping, while Co and Pt SPNDs work on the basis of a (n, γ-e) prompt reactions and are used for reactor control and safety. Specific SPND with Bismuth emitter (SPGD) have also been developed to be exclusively sensible to gamma rays measurement via (γ, e) interactions. Due to different constant time in concerned reaction, SPND total time response can be generalized as follow:

$$R_{total} = R_{n_{(prompt)}} + R_{n_{(delayed)}} + R_{\gamma_{(prompt)}} + R_{\gamma_{(rdelayed)}}$$

Those reaction contributions are evaluated through the MATISSe code [20] which is a multistep Monte Carlo calculations using the JEFF3.1 neutron data library, the ENDF/B-VII.1 gamma data library and libraries associated to charged particles (EEDL,EADL). Knowledge of the shapes of the neutron and gamma spectra (figure 7) is needed for this SPND modelling. However, to give a precise absolute value of the measured thermal neutron (<5%, k=1), SPND have to be calibrated in situ with dosimetry. As the Nuclear data libraries associated to gamma and charged particles are the same as those used for TLDs, analysis performed in the previous paragraph is valid, in particular concerning covariance matrices.

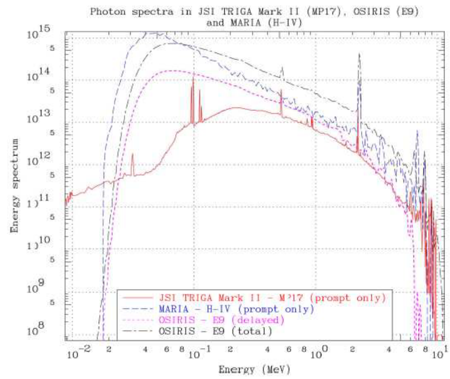

Figure 7. Typical MTR gamma spectra [20].

2.2.2 Fission chambers

Fission chambers are used for real time and online flux monitoring in experimental devices in MTRs and for spectral indexes measurements in mock-up reactor facilities. The neutron energy range of interest drives the choice of the fissile deposit (^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{242}PU, ^{237}Np, ^{241}Am, ^{243}Am). One notes, that the CEA Cadarache LDCI is one of the last supplier for subminiature fission chambers with these deposits.

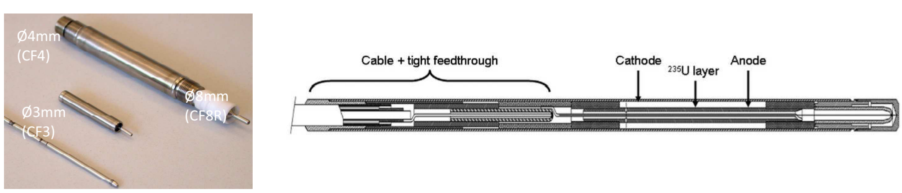

Figure 8. Fission chamber manufactured at CEA-Cadarache.

A typical fission chamber is composed of two coaxial cylindrical electrodes, one of which is covered with fissile material (Figure 8). The inter-electrode gap is filled with gas, often argon. After a neutron-induced fission in the deposit, one of the fission products is ejected into the gas, creating a large number of charge pairs. These charges are collected by a polarization voltage applied between the electrodes, leading to a current pulse. A system dedicated to fast neutron measurement, FNDS [21], has been developed at the CEA in collaboration with SCK.CEN. This device is at the moment the only system available for online fast neutron flux measurement in MTRs with an acceptable uncertainty (>10%). FNDS uses a ^{242}Pu deposit fission chamber for fast neutron flux measurement because ^{242}Pu has been shown to be the best choice for measuring the fast component in the case of a high flux with a significant thermal component [21]. Unfortunately, fission and capture cross sections of ^{242}Pu and moreover ^{243}Pu are badly known [22]. A new evaluation of these data is strongly welcomed, especially for the future use in the JHR facility.

2.2.3 Miniature Ionization Chamber

The miniature ionisation is a fission chamber without fissile coating. Recently this device has shown that the current level is sufficient to be measured and that the model used to analyse the signal has a good accordance with independent nuclear heating measurements. In particular, MCNP modelling has shown a good selectivity to the photon flux in MTR conditions with a calculated neutron contribution to the signal of the ionization chamber of less than 3% [FOURMENTEL].These innovative experimental results highlight the interest to measure the photon flux to improve the assessment of the nuclear heating in MTRs. But, nuclear data associated to gamma and charged particles need to be re-evaluated as in the case of TLD and SPND at least in terms of covariance matrices.

3 Conclusions

This paper has focused on the nuclear data aspects of the main features of the measurement techniques used in the CEA experimental reactor facilities for the on-line and off-line neutron/gamma flux characterizations. For each technique, the nuclear data necessary for their interpretation have been presented, the main needs for improvement listed and an analysis of their impact on the quality of the measurement. Beside demands for specific isotopes and reactions identified for neutron and gamma measurements, a generic one exists for having a coherent set of data with improved uncertainties. One should note here the constant effort made by the IAEA – Nuclear Data Service and by the NEA. In addition, nuclear data for charged particles improvements become necessary to allow instrumentation modelling to give results applicable for instrumentation calibration. Indeed, measurement processes developed for nuclear measurements take into account the lack of knowledge of some nuclear data into the calibration process such as for example dosimetry. Progress in nuclear data dedicated to instrumentation is propagated through more accurate measurements, leading to a better validation of the calculation schemes used for physic phenomena modelling. Finally, having a better experimentation validation, these codes will increase safety and operational margins for industrial using this instrumentation.

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