International Conference on Nuclear Data for Science and Technology 2007
DOI: 10.1051/ndata:07175

Use of the nuclear data in the reactor dosimetry: limitations, improvements and perspectives

C. Destouches[1,a], C. Domergue[1], D. Beretz[1], G. Gregoire[1], and B. Habert[2]

[1] Commissariat à l'Énergie Atomique, Centre de Cadarache, DER/SPEx/LDCI, 13108 Saint-Paul-lez-Durance, France
[2] Commissariat à l'Énergie Atomique, Centre de Cadarache, DER/SPRC/LEPH, 13108 Saint-Paul-lez-Durance, France

Abstract. One of the main objectives of reactor dosimetry is the determination of the physical parameters characterizing the neutron field in a specific place. Reaction rates and fluence rates for example, are derived from activity measurements of irradiated samples using a neutron interpretation process, which needs the knowledge of appropriate nuclear cross sections. For reactor dosimetry, activation cross sections come from international dosimetry activation files, IRDF, EAF, themselves retrievals from international nuclear libraries as ENDF, JEFF, JENDL... These nuclear data are used not only for the activity measurement and the reaction rate derivation (mean values) but also to perform spectrum unfolding and to assess output uncertainties (covariance). Thus, their accurate knowledge is a significant stake for reactor dosimetry improvement, especially for the industrial applications as shows the high uncertainty value of 15% (k = 1) commonly encountered for the calculated neutron flux (E > 1 MeV) on the vessel and internal structures of a power reactor. After a schematic presentation of the reactor dosimetry process, each step, activity measurement, reaction rate derivation and spectrum unfolding, is analysed in order to point out where and how the nuclear data are used. A critical review of the available nuclear data drawn from the new release of the international dosimetry file IRDF2002 is made and improvements and lacks, from a reactor dosimetry point of view, are listed. Then the benchmark concept is analysed in terms of representativeness and use in the dosimetry interpretation: How to use a cross section, which has a C/E significantly different from unity? Finally, the paper concludes with a synthesis of the needs in evaluation improvements for nuclear data in terms of new cross sections estimations, covariance matrices improvements, and experimental benchmark database extension.

1 Introduction

One of the main objectives of reactor dosimetry is the determination of the physical parameters characterizing the neutronic field in a specific location. Reaction rates and fluence rates are derived from activity measurements of irradiated dosimeters using an interpretation process, which needs the prior knowledge of appropriate nuclear cross sections. For reactor dosimetry, activation cross sections are taken from international dosimetry activation files, IRDF2002 [1], EAF2005 [2], themselves extractions from international nuclear libraries ENDF [3], JEFF [4], JENDL [5]... These nuclear data are not only used for activity measurements and reaction rate derivation (mean values) but also to perform spectrum unfolding and to assess output uncertainties (covariance). Thus, their accurate knowledge is a significant stake for reactor dosimetry improvement, especially for the industrial applications as shows the high uncertainty value of 15% (k = 1) commonly allowed for calculated neutron flux (E > 1 MeV) on the vessel and internal structures. Although the reactor dosimetry process uses reputed qualified nuclear data to derive flux and spectrum, attention should be paid to possible results inconsistencies which are often due to lack in nuclear data.

After a schematic presentation of the reactor dosimetry process, each step, activity measurements, reaction rate derivation and spectrum unfolding is analysed from a dosimetry point of view in order to point out where and how the nuclear data are used. A critical review of the available nuclear data used is also performed with a particular focus on the international dosimetry file IRDF2002 [1]: improvements and lacks, from a reactor dosimetry point of view, are listed. Then, the benchmark concept is analysed in terms of representativeness and use in the dosimetry interpretation. Finally, the study concludes with a synthesis of the needs in evaluation improvements for nuclear data.

2 Reactor dosimetry

First, dosimeters are selected according to their response functions on the studied neutron spectrum (nuclear cross section) and to their characteristics with respect to activity measurement process (nature and energy of the emitted particles, decay constant). Table 1 gives a list of currently used isotopes, associated decay constants and their typical response of energy range on the neutron spectrum. Moreover, the shape and the weight of these dosimeters are optimised for the irradiation conditions. Figure 1 shows as example a set of dosimeters.

These dosimeters are arranged within the irradiation device according to required information: gradient and level of neutron flux, neutron spectrum. At the end of irradiation in a zero power reactor, a test reactor or a power reactor, dosimeters are sent for measurements and interpretation to the LDCI laboratory, part of the Reactor Study Department located at the CEA-DEN-CADARACHE centre (France).

[a] e-mail: christophe.destouches@cea.fr

Table 1. Main reaction used in reactor dosimetry.

Reaction	Typical response energy range on a ^{235}U fission spectrum [10]
^{197}Au(n,γ)^{198}Au	Thermal+epithermal
^{59}Co(n,γ)^{60}Co	Thermal+epithermal
^{55}Mn(n,γ)^{56}Mn	Thermal+epithermal
^{115}In(n,γ)^{116}In	Thermal+epithermal
^{237}Np(n,f)	0.7 MeV < E < 5.7 MeV
^{103}Rh(n,n')^{103m}Rh	0.7 MeV < E < 5.8 MeV
^{93}Nb (n,n')^{93m}Nb	1.0 MeV < E < 5.6 MeV
^{115}In(n,n')^{115m}In	1.1 MeV < E < 5.9 MeV
^{238}U(n,f)	1.4 MeV < E < 6.7 MeV
^{58}Ni(n,p)^{58}Co	2.1 MeV < E < 7.5 MeV
^{54}Fe(n,p)^{54}Mn	2.3 MeV < E < 7.5 MeV
^{46}Ti(n,p)^{46}Sc	3.7 MeV < E < 9.6 MeV
^{63}Cu(n,α)^{60}Co	4.7 MeV < E < 11.0 MeV
^{48}Ti(n,p)^{48}Sc	5.9 MeV < E < 13.0 MeV
^{56}Fe(n,p)^{56}Mn	5.4 MeV < E < 12.0 MeV
^{27}Al(n,α)^{24}Na	6.4 MeV < E < 12.0 MeV
^{24}Mg(n,p)^{24}Na	6.5 MeV < E < 12.0 MeV
^{51}V(n,α)^{48}Sc	E > 11.4 MeV

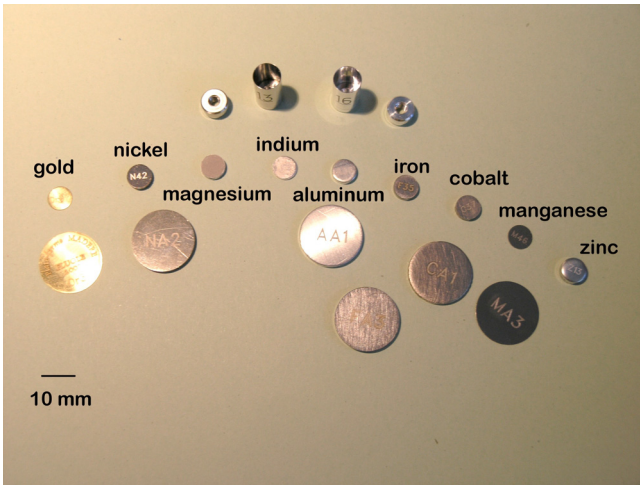

Fig. 1. Example of dosimeters shapes.

The neutron fluence of each dosimeter is evaluated in a three steps process, using calculated neutron spectra, dosimetry cross sections libraries [1] and irradiation conditions:

- Step 1) Activity measurement and estimation of the uncertainty for each dosimeter.
- Step 2) For each dosimeter, the main reaction rates and the neutron fluxes are calculated using activities and a neutron activation code which treats the activation chain for the selected isotope.
- Step 3) Determination of the neutron spectrum and neutron fluence using the whole dosimeters set results and a spectrum unfolding code such as STAY'NL [6].

3 Use of nuclear data in reactor dosimetry

3.1 Measurement process

A detection device, typically a Germanium High Purity diode, gives dosimeter counting rates of specific γ or X emission peaks. Activity is derived using the experimentally stated detector efficiency and nuclear data, mainly, decay constant, emission intensity I (Iγ or I_X) and attenuation coefficient (μ/ρ).

In order to achieve a better accuracy on the activity values, measurement process is tuned to assess and minimize corrections such as self-absorption of emitted ray (function of dosimeter's thickness) or decay constant correction during acquisition. Other experimental uncertainties are reduced to the minimum. Activities are usually given with an accuracy lower than 1% (k = 1) for γ emitters and about 4% (k = 1) for X emitters.

However, measurement results are penalized for some isotopes by strong uncertainties associated with I and μ/ρ. Indeed, one will note the case of the ^{115m}In with uncertainties on Iγ (E = 336.24 keV), which reaches 4.8% (k = 1) [10]. Situation is bad also for ^{103m}Rh which Kx peak intensity emissions present uncertainties around 6% (k = 1).

Concerning the attenuation coefficient, μ/ρ which may induce a few tens percents correction for X measurements, they are badly known and modelled. Thus, values must be experimentally stated before use for each type of dosimeters (thickness, shape) using a well known X source and an attenuation formulae. Associated uncertainties are often derived in a simplified way.

3.2 Reaction rate and flux derivation

Each dosimeter activity is analysed using an activation code treating the whole activation chain associated to the isotope of interest in order to derive reaction rates. Code entries are irradiation history, calculated neutron parameters (neutron spectrum) and activation cross sections drawn from activation dosimetry library. We will focus on the two last kinds of data. First, input neutron parameters, mainly neutron spectrum at the dosimeter location, are calculated, with attention paid to the local geometrical flux depletion and self-shielding effect, by neutron codes such as TRIPOLI [8] or MCNP [9]. They use nuclear data drawn from international standard nuclear libraries, JEFF, ENDF and JENDL. For core source spectrum calculation, codes use a mathematical function for fission neutron spectra that are usually given in the libraries in a Watt formalism [10]. The shape of this core neutron spectrum is of great importance for high-energy neutrons, because these core neutrons are those who, when slowed down, damage the reactor structural material and the reactor vessel.

Calculated neutron parameters uncertainties, when they exist, are often roughly estimated because uncertainty data are sometimes poor, especially for structure material and for the fission spectrum. Moreover, the calculation duration linked to the complexity of the modelling do not allow to perform a direct analytic propagation: uncertainties propagation is made only by stochastic simulation or sensitivity analysis. Thus,

a final uncertainty about 10% (k = 1) is typically, usually determined in a Bayesian way, associated to the output neutron spectrum.

Secondly, convoluting activation cross sections with the above calculated spectra, one obtains reaction rates for all the reaction of interest. Activation cross sections data tabulated in the energy range [1×10^{-10} MeV; 20 MeV] are drawn from the international IRDF2002 library [1]. When these cross sections do not exist in the library, trans-uranian cross sections for example, data are taken from the international libraries used for neutrons calculations quoted above. The IRDF2002 evaluation library has been performed on behalf of the IAEA and is described in detail in ref. [1]. This library is based on a compilation of the main international libraries, JEFF3.0, JENDL/D-99, RRDF-98, and ENDF/B-VI, CENDL [1] and its previous release, IRDF90V2 [1]. This library contains cross sections for 66 neutron activation reactions with associated covariance both in a multi-group (SAND) and point-wise format, total cross sections of the above isotopes completed with three cover material, radiation damage cross sections (multi-group), decay data, fission yields and isotope abundances for all reaction and isotope of interest.

Although this release of IRDF2002 has seriously improved the quantity and the quality of the nuclear data used for reactor dosimetry, its analysis made in the library report itself [1], and our feedback lead to the following remarks:

1) Only diagonal covariance matrices are available for the cross sections of the following reactions: ^{23}Na(n,γ)^{24}Na, ^{93}Nb(n,γ)^{94}Nb, ^{115}In(n,γ)^{116m}In and covariance information for the cross sections of the ^{197}Au(n,γ)^{198}Au and ^{235}U(n,f) reaction have to be improved.
2) High uncertainties values (>5%) are still present for more than 20% of the isotopes of interest such as ^{58}Ni(n,2n)^{57}Ni, ^{60}Ni(n,p)^{60}Co ...
3) The resonance integral has a large deviation from the recommended experimental values for ^{55}Mn(n,γ)^{56}Mn, ^{58}Fe(n,γ)^{59}Fe and ^{93}Nb(n,γ)^{94}Nb.
4) Deviations of C/E values from unity by more than 5% are observed for the following reactions: ^{24}Mg(n,p)^{24}Na, ^{65}Cu(n,2n)^{64}Cu, ^{103}Rh(n,n$'$)^{103m}Rh and ^{232}Th(n,f).
5) Some experimental cross section measurement values are missing (titanium for example).
6) Use of the ^{63}Cu(n,α)^{60}Co reaction cross section lead to a systematic overestimation of the neutron flux (E > 1 MeV) compare to ^{54}Fe(n,p)^{54}Mn, ^{58}Ni(n,p)^{58}Co or ^{93}Nb(n,n$'$)^{93m}Nb. One explication of this discrepancy could be the shape of the ^{235}U and/or ^{239}Pu fission spectra at high energy used in the core neutron code calculation: the threshold of this reaction is about 6 MeV.

3.3 Spectrum unfolding

Neutron spectrum and neutron fluence is derived using reaction rates of the whole dosimeters set, and a spectrum unfolding code such as STAY'NL [6].

Based on the use of the associated covariance, these codes derive the best estimate spectrum value using reaction rates deduced from measurement, a representative calculated neutron spectrum and multi-group nuclear cross sections drawn from the activation libraries (IRDF2002). For example, STAY' NL Code provides a solution by a direct least squares method for minimization of the following χ^2 function:

$$\chi^2 = {}^t\left(A^0 - A\right) \cdot (\Gamma_A + \Gamma_{A^0})^{-1} \cdot \left(A^0 - A\right) \quad (1)$$

where
A^0: matrix of reaction rates (step 2)
A: matrix of calculated (adjusted) reaction rates
$\Gamma_A 0$: variance-covariance matrix of reaction rates (step 2)
Γ_A: variance-covariance matrix of calculated reaction rates.
As one can see, cross section and calculated spectrum associated covariance matrices have a direct impact on the output values and have to be realistic and well conditioned. Too low spectrum covariance values, often encountered when only statistical dispersion in the calculated spectrum uncertainties is taken into account, would not let any unfolding possible. On the contrary, too large spectrum uncertainties make impossible the entry neutron spectrum to bring any valuable information; In this case, the unfolding process does not change input reaction rates.

Finally, when the unfolding process is correctly performed, output neutron spectrum uncertainties are rigorously derived and reduced to less than 5% (k = 1) [11], compared to the 1015% uncertainty associated to the input calculated spectrum.

4 Experimental benchmark

Integral cross section experimental benchmarks are performed in well known and calculable spectra: the Maxwellian thermal spectrum at specified neutron temperature, the 1/E slowing down spectrum in hydrogenous moderator, the spontaneous fission neutron field of ^{252}Cf fission and the mono-energetic 14-MeV neutron field from a D-T. Measured data are stored in international databases, EXFOR [12] or compilations [10, 13]. Then, they are compared to spectrum averaged cross sections calculated for the theoretical spectrum function in terms of global C/E with associated uncertainties.

Limitations of these benchmark comparisons come from their dependence on the representativeness of the neutron spectrum theoretical formulae compared to the experimental one such as, for example, the ^{235}U fission spectrum modelled by the Watt formulae [10]. Moreover, measurements are often 10 or 20 years old and nuclear data used for activity measurements have since then been improved (cf. 3.1) leading to some possible re-evaluations.

A problem arises when cross section has a C/E significantly different from unity. How to use it? In a first approach, besides increasing the results uncertainty, we correct the spectrum averaged cross section with the C/E value obtained in the reference benchmark, but this approach does not take into account the spectra difference between the benchmark and the real neutron spectrum. Thus, other cross sections, whose energy ranges partially cover the studied cross section one, are used within the spectrum unfolding.

Finally, the lack of cross section accuracy increases directly uncertainties of the outputs data. New experimental

benchmarks with new and accurate reaction rate measurements supplemented by studies on the representativeness of the theoretical fission formulae would be of great interest for improving the cross sections integral validation.

5 Synthesis

This paper has briefly presented the main steps of the reactor dosimetry process. Each step, activity measurement, reaction rate derivation and neutron spectrum unfolding process has been analysed from a dosimetry point of view and improvements are asked in the knowledge of:

1. For activity measurements: μ/ρ especially for X measurements, Iγ of the ^{115m}In, I_X for the ^{103m}Rh.
2. For cross sections: improvements are useful but before, a new in-depth analysis of the recent releases of ENDFB-VII, JEFF3, EAF2005, JENDL3.3 should be done with a focus at the covariance matrices. Results of this study should give place to a new release of the IRDF2002 evaluation.
3. For fission spectrum modelling: a study of the fission spectra modelling would be interesting to improve the accuracy of the experimental benchmarks and of the neutron core spectrum calculations by neutron codes.
4. For benchmarks: an extension of the database with new reactions and integral measurements in fission spectra (^{252}Cf, ^{235}U, or mixed ^{239}Pu-^{235}U) should be realised.

People involved in reactor dosimetry, as "neophyte" final users, hope that new libraries releases will be more user-friendly, especially the covariance matrices. Finally, an update of the synthetic but very useful *Nuclear data guide for reactor neutron metrology* compilation book [10] would be very appreciated. For example, this could be an evolution of the JANIS [14] database interface toward a sole data sheet per element giving all main isotope reactions of interest (activation schemes…) and associated nuclear data (isotope abundance, decay constants, peak emission probabilities, disintegration schemes…).

References

1. *International Reactor Dosimetry File 2002 (IRDF-2002)*, Technical Report series No. 452, Vienna, 2006.
2. R. Forrest, *The European Activation File, EAF-2005*, International Conference on Nuclear Data for Science and Technology, Vol. 769, May 2005, pp. 157–160.
3. M.B. Chadwick, P. Oblozinsky et al. (CSEWG collaboration), *ENDFB-VII: Next Generation Evaluated Nuclear data Library for Nuclear Science and Technology*, Nucl. data Sheets **107**, 2931 (2006).
4. *The JEFF-3.1 library*, NEA/OCDE, JEFF Report 20 (2006).
5. *JENDL-3.3 – The Japanese Evaluated Nuclear Data Library, version 3.3*, JAERI (2000).
6. N.P. Kocherov, *Neutron Metrology file NMF-90*, IAEA Report INDC(NDS)-347 (1996).
7. M.-M. Bé et al., *Nucléide. Table of Radionuclides Monographie*, BIPM-5, (2006) ISBN-92-822-2204.
8. TRIPOLI-4.3, *Coupled Neutron, Photon, Electron, Positron 3-D, Time Dependent Monte-Carlo, Transport Calculation*, NEA, NEA-1716/02 (02-Oct. 2004).
9. J.F. Briesmeister, *MCNP: A General Monte Carlo N-Particle Transport Code, version 4C*, Los Alamos National Laboratory report, LA-13709-M, March 2000.
10. W.L. Zijp, J.H. Baard, H.J. Nolthenius, *Nuclear data guide for reactor neutron metrology* (1989), ISBN 0-7923-0486 1.
11. C. Destouches et al., *Assessment of neutronic parameter's uncertainties obtained within the reactor dosimetry framework: development and application of the stochastic methods of analysis*, Physor 2006 (2006), Vancouver (to be published).
12. V. McLane, EXFOR Basics *A Short Guide to the Nuclear Reaction Data Exchange Format*, IAEA, IAEA-NDS-206 Rev. 2000/09, BNL-NCS-63380-2000/05 Rev (2000).
13. S.F. Mughabghab, *Atlas of neutron resonances – Resonance parameters and thermal cross sections Z = 1-100*, NNDC-BNL 5th.
14. JANIS, *A Java-based nuclear data display program*, AEN, NEA-1760/01 (2004), www.nea.fr.