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

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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 retrievals 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).

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Table 1. Main reaction used in reactor dosimetry.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Typical response energy range on a (^{235}\text{U}) fission spectrum [10]</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{197}\text{Au}(n,\gamma)^{198}\text{Au})</td>
<td>Thermal+epithermal</td>
</tr>
<tr>
<td>(^{59}\text{Co}(n,\gamma)^{60}\text{Co})</td>
<td>Thermal+epithermal</td>
</tr>
<tr>
<td>(^{55}\text{Mn}(n,\gamma)^{56}\text{Mn})</td>
<td>Thermal+epithermal</td>
</tr>
<tr>
<td>(^{115}\text{In}(n,\gamma)^{115m}\text{In})</td>
<td>Thermal+epithermal</td>
</tr>
<tr>
<td>(^{237}\text{Np}(n,\gamma)^{237m}\text{Np})</td>
<td>0.7 MeV &lt; (E) &lt; 5.7 MeV</td>
</tr>
<tr>
<td>(^{103}\text{Rh}(n,n')^{103}\text{mRh})</td>
<td>0.7 MeV &lt; (E) &lt; 5.8 MeV</td>
</tr>
<tr>
<td>(^{93}\text{Nb}(n,n')^{93}\text{mNb})</td>
<td>1.0 MeV &lt; (E) &lt; 5.6 MeV</td>
</tr>
<tr>
<td>(^{115}\text{In}(n,n')^{115}\text{mIn})</td>
<td>1.1 MeV &lt; (E) &lt; 5.9 MeV</td>
</tr>
<tr>
<td>(^{239}\text{U}(n,f)^{239m}\text{U})</td>
<td>1.4 MeV &lt; (E) &lt; 6.7 MeV</td>
</tr>
<tr>
<td>(^{58}\text{Ni}(n,p)^{58}\text{Co})</td>
<td>2.1 MeV &lt; (E) &lt; 7.5 MeV</td>
</tr>
<tr>
<td>(^{54}\text{Fe}(n,p)^{54}\text{Mn})</td>
<td>2.3 MeV &lt; (E) &lt; 7.5 MeV</td>
</tr>
<tr>
<td>(^{40}\text{Ti}(n,p)^{40}\text{Sc})</td>
<td>3.7 MeV &lt; (E) &lt; 9.6 MeV</td>
</tr>
<tr>
<td>(^{63}\text{Cu}(n,\alpha)^{63}\text{Co})</td>
<td>4.7 MeV &lt; (E) &lt; 11.0 MeV</td>
</tr>
<tr>
<td>(^{48}\text{Ti}(n,p)^{48}\text{Sc})</td>
<td>5.9 MeV &lt; (E) &lt; 13.0 MeV</td>
</tr>
<tr>
<td>(^{56}\text{Fe}(n,p)^{56}\text{Mn})</td>
<td>5.4 MeV &lt; (E) &lt; 12.0 MeV</td>
</tr>
<tr>
<td>(^{27}\text{Al}(n,\alpha)^{27}\text{Na})</td>
<td>6.4 MeV &lt; (E) &lt; 12.0 MeV</td>
</tr>
<tr>
<td>(^{24}\text{Mg}(n,p)^{24}\text{Na})</td>
<td>6.5 MeV &lt; (E) &lt; 12.0 MeV</td>
</tr>
<tr>
<td>(^{51}\text{V}(n,\alpha)^{51}\text{Sc})</td>
<td>(E &gt; 11.4) MeV</td>
</tr>
</tbody>
</table>

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 \(\gamma\) or \(X\) emission peaks. Activity is derived using the experimentally stated detector efficiency and nuclear data, mainly, decay constant, emission intensity \(I\) (\(I_{\gamma}\) or \(I_{X}\)) and attenuation coefficient \((\mu/\rho)\).

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 \(\gamma\) emitters and about 4% \((k = 1)\) for \(X\) emitters.

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

Concerning the attenuation coefficient, \(\mu/\rho\) 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 formulæ. 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 x 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 above quoted. 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 covariation 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: 23Na(n,γ)24Na,
93Nb(n,γ)94Nb, 115In(n,γ)116mIn and covariance information
for the cross sections of the 197Au(n,γ)198Au and
235U(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 58Ni(n,2n)57Ni,
56Ni(n,p)56Co . . .
3) The resonance integral has a large deviation from the
recommended experimental values for 55Mn(n,γ)56Mn,
58Fe(n,γ)59Fe and 93Nb(n,γ)94Nb.
4) Deviations of C/E values from unity for more than 5%
are observed for the following reactions: 24Mg(n,p)24Na,
65Cu(n,2n)64Cu, 103Rh(n,n)103mRh and 232Th(n,f).
5) Some experimental cross section measurement values are
missing (titanium for example).
6) Use of the 63Cu(n,α)60Co reaction cross section lead to a
systematic overestimation of the neutron flux
(E > 1 MeV) compare to 54Fe(n,p)54Mn, 58Ni(n,p)58Co
or 93Nb(n,n')93mNb. One explication of this discrepancy
could be the shape of the 235U and/or 239Pu 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 neu-
tron 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 \( \chi^2 \) function:

\[
\chi^2 = (A^0 - A) \cdot (\Gamma_A + \Gamma_{A'})^{-1} \cdot (A^0 - A) \tag{1}
\]

where
A^0: matrix of reaction rates (step 2)
A: matrix of calculated (adjusted) reaction rates
\( \Gamma_A \): variance-covariance matrix of reaction rates (step 2)
\( \Gamma_{A'} \): variance-covariance matrix of calculated reaction rates.

As one can see, cross section and calculated spectrum associ-
ated 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 per-
formed, 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 252Cf 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 235U fission spectrum modelled
by the Watt formulae [10]. Moreover, measurements are often
10 or 20 years old and nuclear data used for activity measure-
ments 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 di-
rectly 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: $\mu/\rho$ especially for $^{115m}$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 ENDF-B-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

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).