Monte Carlo simulations of neutron coincidence counting systems in passive mode
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1 Abstract

Monte Carlo modelling studies of three different passive neutron-counting chambers: JCC51, JCC62 and FUNE are presented in this paper. The qualification process of the models consists of performing qualitative and quantitative comparisons between simulation and measurement for a $^{252}\text{Cf}$ calibrated source. Parametric sensitivity studies allow an evaluation of some of the uncertainties due to modelling hypotheses. The simulation of the response of the coincidence counting systems is done using the codes MCNP and SOURCES4C, and the “two-parameters method”. The consistency of the calculation scheme is here evaluated with five plutonium oxide samples, perfectly characterized concerning their mass and isotopic composition.

The first results obtained with those characterized or calibrated materials show that MCNP can provide a total count with a deviation of about 3% compared to the measured value, and the combined MCNP / two-parameters method can provide total counts with about 5% and real counts with about 15% of deviation compared to the measured value.

2 Introduction

In application of the 25 July 1980 Act that provided for nuclear materials to be nationally controlled in France, the Institute for Radiation-protection and Nuclear Safety (IRSN) carries out inspections covering the physical protection of nuclear materials, physical follow-up and accounting within French facilities. The inspectors visit the nuclear fuel cycle facilities with their own transportable “non destructive assay” devices to characterise the nuclear materials held by operators.

Simulation of measurement devices can be used as a complementary tool to optimise their performance and to provide them with an additional calibration and assessment method. For this purpose, the three IRSN passive neutron counters: JCC51, JCC62 and FUNE are modelled with MCNP.

We present in this paper the qualification process of our simulations. The quality of the three models is first assessed by performing qualitative and quantitative comparisons between simulation and measurement for a $^{252}\text{Cf}$ calibrated source. For this purpose, we work on the detection efficiency and the die-away time, that characterize such a neutron system. Parametric sensitivity studies of nuclear, physical, chemical, geometrical and environmental data allow an evaluation of some of the uncertainties due to modelling hypotheses. The simulation of the response of the coincidence counting systems is then performed using the codes MCNP and SOURCES4C, and the “two-parameters method” [1]. The consistency of the calculation scheme is evaluated for five plutonium oxide samples, perfectly characterized concerning their mass and isotopic composition.

3 Description of the neutron coincidence counting systems

IRSN inspectors use three transportable passive neutron counters: JCC51 (see figure 1), JCC62 (see figure 2) and FUNE (see figure 3), designed to quantify the plutonium present in
different container configurations. The measurement of neutron counts (singles and doubles) is performed using neutron multiplicity decoders such as Canberra JSR12 or AMSR. The devices are thermal neutron well counters, composed of $^3$He detectors surrounded by high-density polyethylene (HDPE), moderator and reflector material. The sample cavity is shrouded with a cadmium liner designed to prevent re-entry of thermal neutrons.

The JCC51 neutron-counting device, manufactured by CANBERRA, is used in passive mode to measure the plutonium held in small dimension containers (typical $H \times \Omega = 30 \times 16$ cm), with a mass range of 50 g to 10 kg. The neutron-measuring chamber is a thermal neutron well counter using forty-two $^3$He detectors, arranged in two concentric rings, to reach detection efficiency of about 31%.

The JCC62 passive neutron-counting device, manufactured by CANBERRA, is used to measure the plutonium held in large dimension containers (typical $H \times \Omega = 75 \times 16$ cm), with a mass range from about 50 g to about 10 kg. The neutron-measuring chamber is a thermal neutron well counter using twelve $^3$He detectors, arranged in a concentric ring. Each detector is surrounded by polyethylene and cadmium sleeve in the central region to flatten the axial response and decrease counter die-away time. This device is under-moderated, thus providing detection efficiency of about 7%.

The FUNE passive neutron-counting device, developed within the IRSN [2], is used to measure the plutonium contained in waste drums (100 and 200 litres), with a mass range of about 10 mg to several hundred grams. This thermal neutron well counter comprises fourteen trapezoidal blocks of high-density polyethylene, each containing two $^3$He detectors. The $^3$He detectors are arranged in a concentric ring in the system and the detection efficiency provided is about 14%.

The plutonium quantity, held in bulk plutonium samples, can be estimated from the experimental neutron counts (singles and doubles) obtained with each of the devices, using a method called “two-parameters method”. This method, based on the model developed by Hage and Cifarelli [3] for a known efficiency system, can be used to quantify plutonium in known chemical form and isotopic composition samples.

4 Simulation with a calibration source

4.1 Modelling hypothesis

The code MCNP (Monte-Carlo N-Particles) [4] is used to determine $^3$He detector neutron detection efficiency of the devices. It calculates the number of neutron captures (n,p) occurring in the active parts of the $^3$He counters, for a neutron emitted by a source. The 3D

![Image: JCC51, JCC62, FUNE]
The description of the measuring system includes the physical (density), chemical (composition) and nuclear (cross sections) characteristics of the materials, and also a geometric description of the neutron source.

Figures 4, 5 and 6 present axial and radial views of the three devices, modelled using MCNP4C.

A model is built as realistically as possible, but it is very hard to accurately describe all the components of such a device. It is important to have thorough knowledge of the materials that form the neutron moderator and absorber: hypothesis and verifications are thus performed to arrive at a better description. In the case of detector tubes, the manufacturers give some parameters such as the active length or fill pressure, but wall thickness and material, end caps and added gases are harder to approach. As simulation quality is a function of the approximations made, parametric studies enable us to quantify the influence of data known to be inaccurate and its uncertainty on the result. A compromise has to be found between the detail of the description and precision.

4.2 Comparison between simulation and measurement

The quality of the model is first assessed by comparing the experimental and simulated shape of detection efficiency by moving a calibration source inside the sample cavity. We studied the axial and radial response of a $^{252}$Cf calibration source in the sample cavity to check the...
geometry of the model. Figures 7, 8 and 9 show only the axial response of detection efficiency, obtained experimentally and by MCNP simulation.

The axial and radial profiles of the JCC51 device present maximum efficiency in the middle of the sample cavity. The detection efficiency varies by around 20% between the centre and the top (or the bottom) of the device. The location of nuclear material inside the volume of detection is a cause of uncertainty taken into account when inspectors use experimental neutron counts to quantify plutonium held in a container centred in the sample cavity, by the two-parameters method.

The efficiency axial profile of the JCC62 device presents a constant efficiency zone over a height of 75 cm. This enables the plutonium present in elongated containers to be quantified without any position uncertainty. The efficiency is standardised in the central zone because a sheet of cadmium (see figure 5) surrounds the moderator-counter sets. The end of the cadmium sheet and the presence of polyethylene reflectors (see figure 5) are at the origin of the profile contour surrounding the constant zone.

The efficiency profile obtained for the FUNE device is not symmetrical about the middle of the measurement cavity. This is due to the rotating drum rack support hole at the bottom of the system (see figure 3). The detection efficiency varies by around 20% between the centre and the top (or the bottom) of the device.

4.3 Evaluation of the characteristic parameters

We compare here the characteristic parameters measured by the devices against the simulated parameters: detection efficiency and neutron die-away time in the system.

Table 1 presents both the measured and simulated values of the characteristic parameters (detection efficiency and die-away time) obtained with the three devices for a $^{252}$Cf source centred in the cavity. The experimental uncertainties include source emission uncertainty and counting statistical uncertainty, which are negligible given the fluence of the source specified on the certificate. The statistical uncertainties associated with the simulated values come from calculations running at least a million or more particles. The uncertainties are given at 2σ.
As regards experimental uncertainties, the simulations are consistent with the experiment for the JCC51 and JCC62 systems. In the case of FUNE, the simulations are 8% lower than the measurements. As the parametric study (see § 4.3) does not explain this deviation, a second analysis, in progress, consists of comparing the experimental detection efficiency given by each detection block.

The die-away time is calculated using a weighted least squares with a single exponential decay function to fit the evolution of efficiency with time, calculated by MCNP. At a moment, the number of neutrons disappearing by unit of time is proportional to the number of neutrons present in the device. Thus the number of neutrons present in the device drops exponentially over time with a mean die-away time. The results of the simulation are consistent with the experimental values for the three systems, showing that the quantity of moderator is accurately modelled.

### 4.4 Modelling uncertainties

Some simulation uncertainties can be assessed by parametric sensitivity studies of the nuclear (spectra, cross sections), geometric (³He detectors position, tubes thickness), physical (density), chemical (composition) and environmental data (repository premises). The influence of some parameters is studied hereafter, considering a $^{252}$Cf source inside the measurement cavity. The results are reported in table 2.

#### Table 2: detection efficiency ($\varepsilon$), die-way time ($\lambda$)

<table>
<thead>
<tr>
<th></th>
<th>JCC51</th>
<th>JCC62 [5]</th>
<th>FUNE</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{252}$Cf</td>
<td>EXP</td>
<td>SIMU</td>
<td>EXP</td>
</tr>
<tr>
<td>$\varepsilon$ ($\sigma_e$)</td>
<td>0.3080 (0.0040)</td>
<td>0.3001 (0.0020)</td>
<td>0.0682 (0.0012)</td>
</tr>
<tr>
<td>$\lambda$ ($\mu$s)</td>
<td>50.5</td>
<td>50</td>
<td>26.5</td>
</tr>
</tbody>
</table>

| Table 1: detection efficiency ($\varepsilon$), die-way time ($\lambda$) |

**geometrical data**: the position of $^3$He detectors embedded in HDPE could have a significant influence on the result if the moderator thickness was not accurately known. Cadmium thickness and density has no effect, due to fact that a cadmium sheet of 3 times the mean free path of thermal neutrons in cadmium (~ 0.004 cm) is sufficient to stop those neutrons. In this study, only detectors tube thickness influence is analysed, with variations of $\pm$ 2 mm.

**physical and chemical data**: HDPE density can vary from 0.90 to 0.97 g/cm$^3$. Regarding detector filling gas, the fill pressure around 4 atm. has little effect on efficiency (the effect become significant for 4 ± 0.5 atm.: 2.9%, 4.5% and 3.9%), similarly the gas added to $^3$He to reduce wall effect losses (1 or 5% CO$_2$).

<table>
<thead>
<tr>
<th></th>
<th>JCC51</th>
<th>JCC62</th>
<th>FUNE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tube thickness</td>
<td>1.6%</td>
<td>1.5%</td>
<td>1.7%</td>
</tr>
<tr>
<td>HDPE density</td>
<td>1.9%</td>
<td>3.7%</td>
<td>1.0%</td>
</tr>
<tr>
<td>Detection gas composition</td>
<td>1.1%</td>
<td>1.3%</td>
<td>1.5%</td>
</tr>
<tr>
<td>Spectrum</td>
<td>2.2%</td>
<td>2.8%</td>
<td>2.6%</td>
</tr>
<tr>
<td>Cross section library</td>
<td>0.1%</td>
<td>0.1%</td>
<td>0.2%</td>
</tr>
<tr>
<td>Measurement premises</td>
<td>0.2%</td>
<td>0.8%</td>
<td>1.1%</td>
</tr>
</tbody>
</table>

Table 2: parametric studies
**nuclear data:** two sets of cross section data, evaluated at the room temperature, are studied. The ENDF-BVI library is the most recent, but ENDF-BV is more convenient when considering natural elements: all the isotopes of an element have to be defined using the ENDF-BVI library; this is not the case for ENDF-BV and subsequently ENDF-BV is easier to use. In our case for a $^{252}$Cf source placed inside these devices, the influence of the library is small. Regarding spectrum data, the use of two Watt spontaneous fission spectra, reference [4] and [6], shows a slightly influence on the simulation result.

**environment data:** the simulation of air inside and around the device has a negligible influence on detection efficiency, compared to void. However the room scatter effect, which may be negligible for a source centred in the sample cavity, should be taken into account for a source located at the bottom of the JCC62 and FUNE devices.

The total uncertainty assessed by this sensitivity study reach 3.5 % for JCC51 and FUNE, and 5 % for JCC62 (more sensitive since it has been developed to measure fuel assemblies). The position of $^3$He detectors embedded in HDPE could have a significant influence on the result, but it is accurately known. Other parameters like source description, $^3$He detector modelling, experimental counting loss (depending on high voltage and discrimination threshold adjustments),… are harder to evaluate accurately.

5 Simulation of plutonium oxide containers

The simulation of the response of the coincidence counting systems is evaluated for five plutonium oxide powder samples, perfectly characterized regarding their mass and isotopic composition.

5.1 Calculation scheme

The code MCNP only allows to estimate the total neutron count rate, starting from a 3D description of the nuclear material and the definition of its emission spectrum. Neutron emission (spectrum and strength), coming both spontaneous fission and ($\alpha$,n) reactions, is evaluated from a SOURCES4C [6] calculation.

Experimentally, the quantity of plutonium held in bulk plutonium samples, can be estimated from the neutron counts (singles and doubles) obtained for each of the devices, using the two-parameters method. The experimental parameters, total counting rate $T$ and real counting rate $R$ (neutron coming from spontaneous fissions) are expressed as a function of:

- the device measurement characteristics (detection efficiency, die-away time, predelay, and gate width) and the known nuclear data. All these data are included in the K term,
- three physical characteristics of the sample: $m^{240\text{Pu}}_{\text{eff}}$ (effective mass of $^{240}$Pu), $\alpha$ (ratio between ($\alpha$,n) reaction and neutron from spontaneous fission) and $M$ (multiplication factor).

\[
T = f_1(K, m^{240\text{Pu}}_{\text{eff}}, \alpha, M) \\
R = f_2(K, m^{240\text{Pu}}_{\text{eff}}, \alpha, M)
\]

This model can be used, combined with MCNP and SOURCES4C, to estimate the single and real rates. MCNP is used here to estimate first the detection efficiency and the die-away time of the system with a calibrated $^{252}$Cf source, and then to estimate the multiplication factor of the sample.
Comparison of the measurements and simulations of five samples of PuO₂ powder of low burn-up type isotopic composition is performed for the three devices, covering a mass range of 50 - 1000 g. Plutonium oxide samples are perfectly characterized concerning their mass and isotopic composition. The measurements were done in the PERLA (PERformance Laboratory) reference laboratory at JRC/Ispra in 1997 with JCC51 and FUNE [1], and in 2001 with JCC62 [5].

The PuO₂ samples are modelled with as much detail as possible, taking the containers into account. Since the masses and isotopic compositions of the nuclear materials are certified, the single unknown quantity is powder density. Gamma scanning measurements were performed to evaluate the height of the powder in the internal containers; the associated uncertainty has also an influence on the powder density and the multiplication factor.

Table 3 indicates, for each sample and device, the Pu mass, the neutron emission \( S_{\text{neutron}} \) calculated with SOURCES4C, the experimental counts (total neutron count rate \( T_{\text{exp}} \) and real neutron count rate \( R_{\text{exp}} \)), the total counts \( T_{\text{MCNP}} \) and multiplication factor \( M_{\text{MCNP}} \) simulated with SOURCES4C and MCNP, and the simulated counts (total neutron count rate \( T_{\text{sim}} \) and real neutron count rate \( R_{\text{sim}} \)), obtained with the two-parameters method.

As a general rule simulations are consistent with the experiment. The deviations observed on total count rates are less than 5 % with the three devices, when \( T \) is calculated in one step by MCNP. Those ones are slightly higher, up to 11 % when \( T \) is calculated in two steps (MCNP + two-parameters method) because of the combination of two sources of error (simulation uncertainties and model assumptions). The reals are estimated with an error less than 15 %, that is interesting, considering that the experimental uncertainty associated to the Pu mass, resulting from the two-parameters method, is about 15% for FUNE and JCC51 because of the nuclear material position effect, and 5 % for JCC62. The emission spectrum of the neutron source (PuO₂) still stay the most critical parameter.
6 Conclusions

The results of the model validation process using the typical parameters of a neutron device (efficiency and die-away time) reveal that the JCC 51 and JCC62 devices match well, the deviations are less than 3% with a modelling uncertainty estimated of about 5%. Regarding the FUNE device, the sensitivity study cannot explain the -8% deviation between simulation and measurement observed on detection efficiency. As the device is formed by the association of 14 trapezoidal blocks, the individual block-by-block “simulation/measurement” study currently performed should help us to understand and to correct the bias.

The calculation neutron counts scheme, using the two-parameter method hypothesis, applied to five bulk plutonium oxide samples with the three devices, allow to reach a first approximation of the doubles expected, with an accuracy of 15% here. The total count rates calculated directly by MCNP are more consistent than those issued from the two-parameters method, because of the model’s assumptions. We should now progress to predicting coincidence and multiplicity simulation directly by Monte Carlo calculations.

7 Acknowledgements

We would like to thank Paolo Peerani and Marc Looman (JCR/Ispra) for performing the characterisation of the JCC62 device, at Ispra in 2001.

8 References

[1]: T. Lambert. Application of passive neutron coincidence counting for the control of plutonium waste container: method with two or three parameters. INMM Proceedings 40, 1999
[7]: W.B. Wilson, R.T. Perry et al… SOURCES4C: a code for calculating (α, n), spontaneous fission, and delayed neutron sources and spectra. Los Alamos National Laboratory report LA-UR-02-1839, April 2002.