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Matrix effect correction in active neutron interrogation for residual fissile mass assessment in radioactive waste drums

J.P. Scotta^{a,*}, L. Tamagno^a, C. Passard^a, R. Antoni^a, O. Gueton^a, J.C. Vandamme^b,
F. Guillaumin^b, L. Villatte^b

^a CEA Cadarache, Nuclear Measurement Laboratory, Cadarache F-13108 Saint-Paul-lez-Durance, France

^b Orano Recyclage, La Hague, France

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ABSTRACT

Metallic waste in the form of shells and nozzles remains following the reprocessing of spent fuel at La Hague plant (France). Before the final disposal, drums containing the waste are transported to the compaction facility where its volume is reduced by a factor of 5. With the objective of controlling the criticality/safety levels, an active neutron interrogation system at the entrance of the facility is used to assess the residual fissile mass remaining in the waste, which relies on prompt fission neutron detection (known as the differential die-away technique, DDA). The measured signal is proportionally linked to the fissile mass by a calibration coefficient. However, two effects can produce an inaccurate prediction. The number of induced fissions for the same fissile mass varies according to the waste matrix composition, particularly the neutron slow-down and absorption ratio. Secondly, the presence of fissile clusters can impact the neutron interrogation due to an increase in self-shielding effect. This work presents a matrix effect correction method based on the use of internal flux monitors (sensitive to the neutron absorption ratio) and the transmission signal (sensitive to the slow-down ratio). It relies on a numerical model of the whole measurement cell developed with the Monte-Carlo N-Particle transport (MCNP) code. The calibration coefficient of 72 different matrices, representative of the waste produced at La Hague, were simulated and a statistical model using a multilinear regression was then established. This simulated-based approach provides a more robust and comprehensive estimation of the residual fissile mass without impacting accuracy thanks to the flux monitors and the transmission signals.

1. INTRODUCTION

The development of low-carbon energy sources is mandatory in the current context of mitigating the consequences of global warming. The nuclear industry has its own solution, i.e. the recycling of spent fuel. About 96% of a spent fuel can be potentially recovered in order to produce Mixed-Oxide fuel (MOX) [1].

The Orano La Hague reprocessing plant is a world leader in nuclear material recycling [2]. Spent fuel undergoes several stages within the reprocessing plant before it is returned to the customer. After leaving the nuclear power plant, spent fuels are stored in decaying pools for several years in order to reduce the radioactivity level and thus the heat decay. Afterwards, the fuel assemblies are cut into 1-cm pieces (shells) in the shearing and dissolution facility, while the nozzles are separated from the waste. The shells are then transferred to a nitric acid bath where the fuel (fissile material and fission products) is dissolved in order to be

separated from the structural materials. The fuel solution is then transferred to another facility where the fissile material is separated from the fission products. These fission products are vitrified and finally conditioned, while the recovered uranium and plutonium is purified to produce MOX fuel. The shells and nozzles are loaded in waste drums and then transported to the shells and nozzles compaction facility. Eight nuclear measurements stations in the shearing and dissolution facility ensure the online process monitoring and for criticality/safety control.

In the shells and nozzles compaction facility, the structural waste volume will be reduced by a factor of 5. Subsequently, the waste is then placed in special canisters and returned to customers. Within the scope of criticality/safety control, a residual fissile mass (RFM) measurement is performed before (measurement station 0) and after (measurement station 2) the compaction stage. The fissile mass remaining in the shells is due to an imperfect dissolution process. Partially dissolved shells (resulting in a non-regular shape) may also limit the contact between the

* Corresponding author.

E-mail address: juan-pablo.scotta@cea.fr (J.P. Scotta).

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fuel and the acid, encouraging the development of fissile clusters.

This work focuses on the determination of the fissile mass in the measurement station 0. Our approach is based on that used in a previous study on station 2; it uses a matrix effect correction methodology to account for the neutron absorption and the slow-down properties of the structural waste. The influence of the matrix effect on nuclear measurements was already identified and studied in previous works [3–5]. The second aspect which negatively impacts the mass prediction is the presence of fissile clusters. Depending on the size of these clusters, they can have a self-shielding effect on interrogating neutrons which, instead of producing fissions, will be absorbed by the resonant isotopes (^{238}U and ^{240}Pu) of the fuel. Thus, a smaller number of prompt fission neutrons will reach the detectors, resulting in an underestimation of the mass.

A factorial experimental design of 72 different configurations was simulated using the latest numerical model of the measurement cell that was developed with the MCNP transport code [6]. These configurations were chosen so as to represent the variation range of the waste geometry and composition.

The next section of this paper will explain the active neutron measurement principle implemented in station 0 of the shells and nozzles compaction facility. Section III describes the calculation methodology while section IV provides the results and interpretations. The conclusion and future prospects are given in the final section.

2. THE ACTIVE NEUTRON INTERROGATION SYSTEM OF THE MEASUREMENT CELL

The active neutron interrogation system implemented in the measurement station relies on the prompt fission neutrons detection using the differential die-away technique, pioneered by Los Alamos [7,8]. Two SODERN Deuterium-Tritium pulsed generators deliver $\sim 1.10^9$ n/s at 14 MeV energy and 125 Hz with a pulse duration of 200 μs .

The interrogation neutrons are slowed-down within the measurement cell by the waste matrix and the surrounding materials. Neutrons close to thermal energies levels induce fissions with the fissile isotopes of the matrix: ^{235}U , ^{239}Pu and ^{241}Pu . Prompt fission neutrons are measured by the ^3He pressurized proportional counters, surrounded by a polyethylene block designed to slow them down and increase the probability of being absorbed by the ^3He nuclei. The whole measurement cell includes 249 detectors divided into blocks of 83 detectors each, thereby covering three sides of the cell (see section 3.2). A thin layer of cadmium surrounding the detection block makes it possible to discriminate the useful fast neutrons (fissions) from the thermal interrogating neutrons (generators). The waste is surrounded by a lead shielding to reduce the gamma rate. Graphite is added as a neutron reflector to increase the number of neutrons which will interrogate the waste. Data acquisition is carried out over a specific time frame between each generator pulse to ensure time discrimination. A fission prompt neutron window typically starts from about 1 ms after the pulse and lasts about 2 ms. The active neutron measurement duration is about 15 min, which is sufficient to ensure a strong count rate and thus a very low statistical measurement uncertainty.

The useful prompt signal (S) is obtained after subtracting the background signal provided by the delayed neutrons, spontaneous fission neutrons, neutrons created by photonuclear reactions on lead, parasite emissions of the generators and electronic noise. As previously mentioned, the prompt signal is proportionally linked to the residual fissile mass (RFM) by the calibration coefficient (CC) as follows:

$$S = \text{RFM} \cdot \text{CC} \quad (1)$$

In addition, there is an Am–Be source of $\sim 1.10^5$ n/s near the drum (see Section 3.2), whose original purpose is to make sure the detection system is operating correctly. Before the waste drum enters the station, the neutron source is placed in the cavity and data acquisition is carried out for about 5 min. The reference value is then compared against the actual measurement value. As suggested by certain studies [9,10], the

transmission signal may provide a valuable information on the matrix neutron slow-down property. Since 2019, transmission measurements (15 min) are also performed on all waste drums at the station 0 in the compaction facility. Once the transmission measurement has been completed, the Am–Be source is removed from the cavity and the active neutron measurement can be performed. The net transmission signal (T_{net}) is obtained by subtracting of a ~ 30 -min passive measurement (PM) and the background noise (BN):

$$T_{\text{net}} = T_{\text{raw}} - PM - BN \quad (2)$$

Internal flux monitors (available at the station 0 since 2015) are located inside the measurement cavity opposite to the neutron generator and behind the lead shield (see Section 3.2) so as to limit the undesired gamma-dose. These monitors are ^3He proportional counters also surrounded by a cadmium layer except for a small uncovered window facing the waste drum. According to previous studies [9,10], the internal monitors are sensitive to the waste matrix neutron absorption ratio. The internal monitor signal is acquired during the active neutron interrogation measurement. Finally, there is an external flux monitor to correct any fluctuation emission from the D-T neutron generator.

The methodology used to calculate the calibration coefficient is defined in the following section; it takes into account the diverse matrix characteristics.

3. Calculation methodology

3.1. The factorial experimental design

Based on several years of operating experience (OPEX) provided by La Hague spent fuel reprocessing plant, the measured waste drums present very diverse characteristics in terms their physical properties, material nature, process handling, etc. Due to the large number of different possibilities, an experimental calibration is unfeasible. The solution is to numerically simulate a reduced group of possibilities carefully chosen through a factorial experimental design (FED). In order to create a FED the main matrix parameters need to be identified as they are representative of the typical waste geometry and composition. In statistics, a FED is used to reduce the number of simulations needed to be carried out exploiting the sparsity of effects principle and the fact that many runs in a full experimental design are redundant [11].

A total of 12 parameters with different variation levels were chosen for this study. The list of parameters and levels is provided in Table 1 and a description of each is given hereafter. The parameters which show three levels account for the minimum, the maximum and the median values expected to be found at the reprocessing plant. A high number of levels indicates a strong variability in the parameter, which is the case for those describing six levels. Each of these parameters contribute to increasing or decreasing the neutron absorption or slowing-down ratio, or the self-shielding effect in the case where fissile agglomerates are present.

Table 1

Parameters and number of levels representative of the expected waste at station 0 of the compaction facility.

Parameter	Number of levels
Neutron absorption ratio	6
Neutron slow-down ratio	6
Absorption fraction in nozzles	3
Waste density	6
Total fissile mass	6
Fuel type	3
Undissolved particle concentration	3
Drum filling height	6
Fraction of fissile mass in clusters	6
Hydraulic dumper height	3
Dripping water	3
Nozzles distribution	2

- Neutron absorption ratio (NAR) (%): ratio in mass of the structural materials which absorb neutrons (stainless steel and nickel-rich alloys) to the total structure materials (stainless steel, nickel-rich alloys and zirconium alloys).

$$NAR = \frac{m_{ss} + m_{nickel}}{m_{ss} + m_{nickel} + m_{Zr}} \quad (3)$$

- Neutron slow-down ratio (NSDR) (g/L): amount of water in all the waste drum volume.

$$NSDR = \frac{m_{H_2O}}{V_{shells/nozzles}} \quad (4)$$

- Absorption fraction in nozzles (AFN) (%): proportion of absorbing materials only present in the nozzles to the total absorbing materials. As nozzles are not exposed to the nitric acid bath in the shearing and dissolution facility, they are not contaminated with fissile material, thus the absorption of interrogating neutron is less impacted than in shells.

$$AFN = \frac{[m_{ss} + m_{nickel}]_{nozzle}}{m_{ss} + m_{nickel}} \quad (5)$$

- Waste density (WD).
- Total fissile mass (TFM) (g): amount of fissile contamination in the waste (TFM).
- Fuel type (FT): the isotopic concentration of different fuels depending on their decay time in the cooling pool and the burn-up ratio.
- Undissolved particles concentration (UPC) (kg): isotopes which remain after the dissolution process and have a significant neutron absorption cross section. Their impact compared with NAR isotopes is of second order.
- Drum filling height (H) (cm): total height of the shells and nozzles in the waste drum.
- Fraction of fissile mass in clusters (fFC) (%): fissile mass which is only present in the fissile clusters.

$$fFC = \frac{\text{fissile mass in clusters}}{TFM} \quad (6)$$

- Hydraulic dumper height (HDH) (cm): height of the water column created due to the waste drum weight once it is settled in the measurement station.
- Dripping water (DW) (L): amount of water drained at the bottom of the drum after being rinsed during the final stage of the shearing and dissolution facility.
- Nozzles distribution: possible geometrical configurations of the nozzles inside the drums. Either they can be uniformly distributed in the waste or establish a cylinder in the center of the matrix (penalizing scenario).

In a full experimental design, it would be necessary to simulate around 15 million different waste matrices given the significant number of parameters and levels (unfeasible from a calculation time viewpoint). The use of a factorial experimental design is therefore necessary. The statistical software R [12] contains a large list of ready-to-use FED for several applications. The minimum number of cases fulfilling the orthogonal condition is 72 for our research. This criterion guarantees that the effect of any case can be estimated independently from the rest of the cases because they should be uncorrelated [13].

3.2. The numerical model of the measurement cell

In order to simulate 72 measurement cases, a full 3-D numerical model was developed with the Monte Carlo transport code MCNP6.1.

The ENDF-B/VII.1 [14] nuclear data library was used for the calculations. The exact numerical model details will not be given in this article for confidentiality reasons with respect to the design of the measurement station currently in operation at La Hague. A simplified view of the model is shown in Fig. 1.

The whole waste matrix is divided into 24 angular sectors, 5 radii and 10 different heights (1200 meshes). The shells are modelled as a homogeneous material while the nozzles consist of cylinders with a fixed radius and a height dependent on the mass. The fissile clusters are represented as inner cylinders inside each mesh with a fixed radius and a height dependent on the fissile mass in the clusters. The rest of the fissile mass is homogeneously distributed in the shells. An advantage of calculating the shells and nozzles separately from the fissile clusters allows us to evaluate their single contribution to the total measured signal.

The model was qualified based on experimental measures performed in the shells and nozzles compaction facility at La Hague between 2018 and 2020. Five different standard matrices (with different amount of fissile masses) were assessed, representing the expected waste to be measured at the station. In each case, different proportions between steel and polyethylene were established to represent the variation in neutron absorption and slow-down. The qualification process consisted in adapting the MCNP model to obtain the best possible agreement between the measured and the calculated prompt signal and the active background signal (background when measuring the waste matrix without fissile mass). The model uncertainty for each observable was computed as the standard deviation of the average experimental-to-simulation gap for all 5 cases, considering an equiprobability distribution. If σ_X represents the standard deviation of X and X_{Ei}/X_{Ci} is the experimental to calculated gap of X for the waste matrix i , then it can be demonstrated as such:

$$\sigma_X = \frac{\max(X_{Ei}/X_{Ci}) - \min(X_{Ei}/X_{Ci})}{\sqrt{12}} \quad (7)$$

Finally, a model uncertainty of 7.0% (1σ) was obtained for the prompt signal, 3.1% (1σ) for the internal flux monitor signal and less than 1% (1σ) for the transmission signal.

3.3. Calculation of the global calibration coefficient

In order to calculate the calibration coefficient for each matrix configuration, a two-step simulation was carried out to first determine the fission rate, and then the prompt neutron detection efficiency. This made it possible to significantly reduce the computational cost compared with a single-step simulation. An MCNP variance reduction technique ("meshed" weight windows in time discretization) was applied to accelerate convergence and also reduce the calculation time. Around 100,000 h of MCNP calculation were needed to complete the simulation of the whole factorial experimental design.

The calibration coefficient, measured in $\text{counts} \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ is obtained as follows:

$$CC = F \cdot \bar{v} \sum_{j=1}^n (\varepsilon_j \cdot \tau_{fissj}) \quad (8)$$

where \bar{v} is the mean number of neutrons emitted per fission, F is the normalization factor which depends on the neutron generator set up parameters (frequency, neutron emission fission prompt neutron window acquisition), n is the total number of meshes in the matrix, τ_{fissj} is the total fission rate of the j mesh and ε_j is the prompt neutron detection efficiency which represents the number of counts in the detectors per prompt fission neutron. It is calculated as follows:

$$\varepsilon_j = k_{det} \cdot V_{det} \cdot RR(n, p)_j \quad (9)$$

where k_{det} is the electronic loss coefficient of the whole ${}^3\text{He}$ proportional

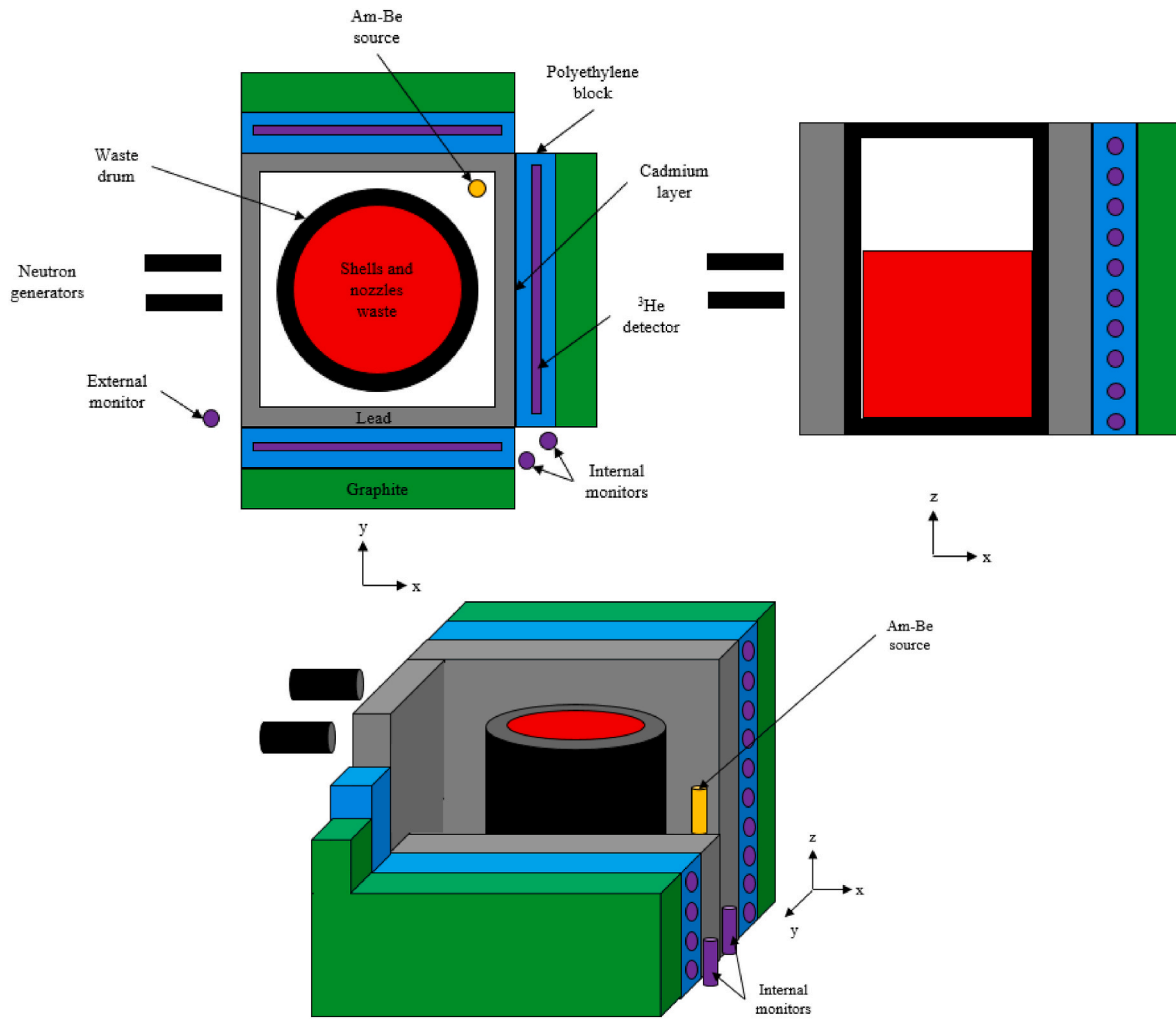


Fig. 1. Simplified numerical model of the waste drum at the measurement station 0 in the shells and nozzles compaction facility. The x-y view is given on the top left image, while the x-z view is shown on the top right image. The image below presents the overall layout.

counters, V_{det} is the ${}^3\text{He}$ proportional counters active volume and $RR(n, p)_j$ is the ${}^3\text{He}(n, p){}^3\text{H}$ reaction rate per volume unit and per fission neutron for mesh j .

Given the fissile mass fraction in the shells and nozzles, and clusters, the global calibration coefficient of the waste matrix is then obtained as the sum of each individual contribution as follows:

$$CC_{global} = CC_{S\&N}(1 - f_{FC}) + CC_{FC} \cdot f_{FC} \quad (10)$$

where $S\&N$ stands for shells and nozzles and FC stands for fissile clusters. The three fissile isotopes (${}^{235}\text{U}$, ${}^{239}\text{Pu}$ and ${}^{241}\text{Pu}$), and less significantly the fertile isotope ${}^{238}\text{U}$, contribute to the calculation of the shells/nozzles and fissile clusters calibration coefficients. On average, ${}^{235}\text{U}$ and ${}^{239}\text{Pu}$ contribute in the same proportion and represent 90% of the total prompt signal, while the fast fissions from the ${}^{238}\text{U}$ are less than 1%.

The total prompt signal is calculated as the sum of the shells and nozzles prompt signals and the fissile clusters prompt signals:

$$S = S_{S\&N} + S_{FC} \quad (11)$$

Considering (1), then (11) becomes:

$$S_X = m_X^{comb} (\eta_9 \cdot CC_X^9 + \eta_5 \cdot CC_X^5 + \eta_1 \cdot CC_X^1 + \eta_8 \cdot CC_X^8) \quad (12)$$

where the sub index X should be replaced by shells and nozzles or by fissile clusters, η is the isotopic concentration, 9, 5, 1, 8 denote the ${}^{235}\text{U}$, ${}^{239}\text{Pu}$, ${}^{241}\text{Pu}$ and ${}^{238}\text{U}$ isotopes respectively and m_X^{comb} is the total fuel

mass.

The internal flux monitor signal (IM) is obtained in a single-step calculation, according to the following equation:

$$IM = F \cdot k_{det} \cdot V_{IM} \cdot RR(n, p) \quad (13)$$

where V_{IM} defines detection active volume of the internal monitors and $RR(n, p)$ is the ${}^3\text{He}(n, p){}^3\text{H}$ reaction rate per volume unit and per D-T neutron source. The transmission signal (T) is measured in a passive mode and is then calculated similarly to (13).

The final goal is to obtain a correlation between the global calibration coefficient and the measured signals (prompt signal, internal monitor signal and transmission signal) in order to predict the total residual fissile mass in a waste drum through (1).

4. RESULTS AND DISCUSSIONS

4.1. Waste matrix effect

To understand the importance of applying a matrix effect correction method, Fig. 2 shows the residual fissile mass as a function of the measured prompt signal S . The 72 configuration matrices of the factorial experimental design are represented in this figure. The uncertainties correspond to the model uncertainty for the prompt signal (see Section 3.2). Cases with a very large fissile mass should exhibit a strong signal. Nevertheless, some configurations with a high neutron absorption

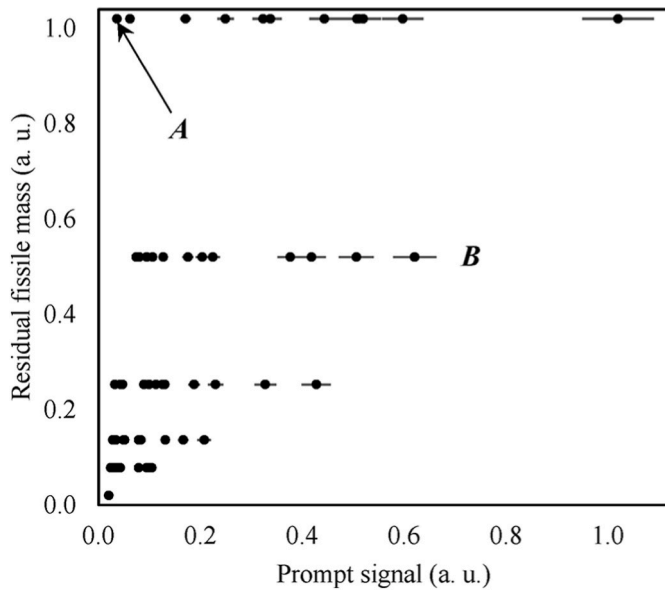


Fig. 2. Residual fissile mass as a function of the prompt signal. Each point describes a different waste matrix. The letter A represents the configuration with a very high neutron absorption ratio, maximum amount of fissile mass and weakest prompt signal. The letter B describes the waste matrices with the same amount of contamination but with a variability in the measured signal up to $\sim 50\%$.

power (indicated by the letter A) deliver a very weak signal, even though the mass (thus the number of fissions) is maximum. This is because most of the thermal interrogating neutrons are being absorbed by the waste, so very few are capable of inducing fissions. This may be a safety concern due to an underestimation of the residual fissile mass. A second feature, indicated by the letter B, concerns the variability in the prompt signal for a given mass. Here S varies up to 50% for the same amount of contamination, which is unacceptable if the main goal is to precisely predict the fissile mass in waste drums. These drawbacks need to be taken into account when developing a statistical model of the calibration coefficient, using an internal flux monitor and a transmission measurement. Using only the prompt signal is not enough to correctly determine the residual mass.

4.2. Internal flux monitor and transmission signals

Fig. 3 shows the sensitivity of the internal monitors signal to the absorption mass (mainly due to stainless steel and Inconel alloy in the shells and nozzles). The uncertainties correspond to the model uncertainty for the internal flux monitor signal (see section 3.2). The correlation coefficient between both factors is -0.75 . Waste with a low neutron absorption property will produce a large signal as many interrogating neutrons will create events in the flux monitors (no cadmium protection).

In a transmission measurement, the signal due to fissions is negligible compared with the interrogating neutrons being slowed-down. This is because the amount of fissile mass expected in typical structural waste is not large enough to produce a reckonable multiplication effect using an Am-Be source. In addition, as the source is continuously emitting neutrons, it is not possible to discriminate the fission neutrons from those produced by $^4\text{He}+^9\text{Be}$ reaction. When performing an active interrogation with a source 10,000 times more powerful in a pulsed mode, it is possible to measure fission prompt neutrons over a selected specific time-window (done between generator bursts). **Fig. 4** represents the transmission signal as a function of the amount of water in the drum (representative of the neutron slow-down ratio). The uncertainties correspond to the model uncertainty for the transmission signal (see

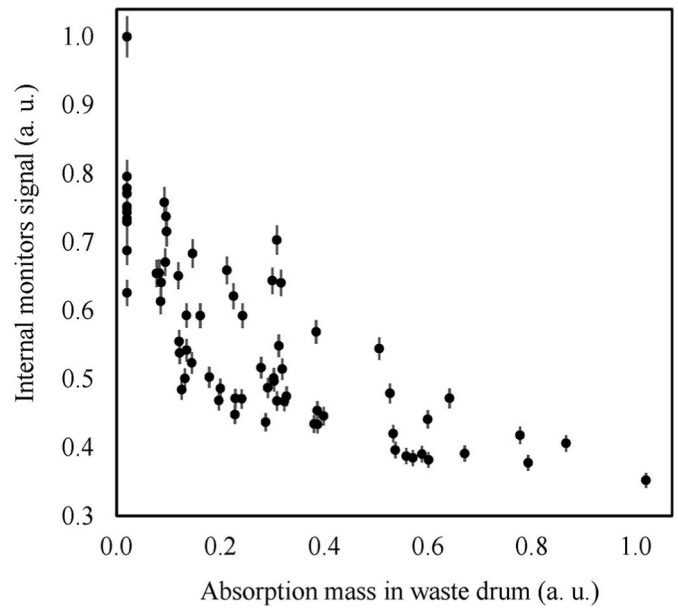


Fig. 3. Internal flux monitor signal as a function of the absorption mass for 72 different matrix compositions.

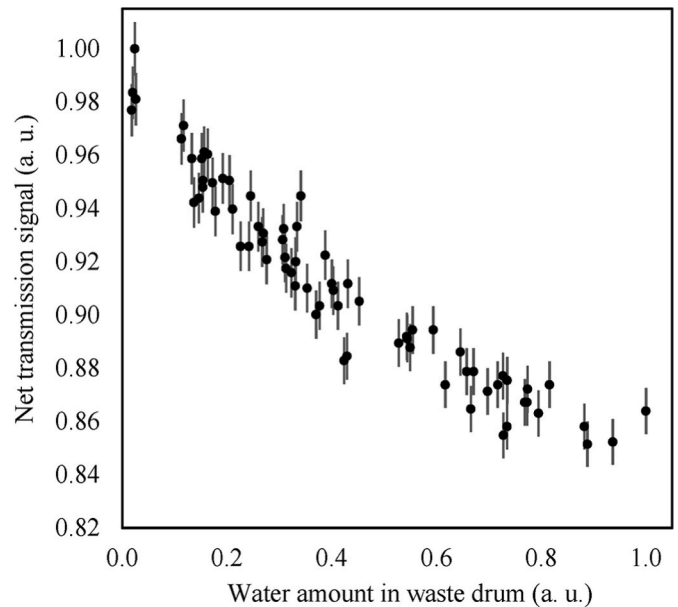


Fig. 4. Transmission signal as a function of the total water amount for 72 different matrix compositions.

Section 3.2). As neutrons scatter (mostly) with hydrogen in water, they slow down within the waste. Therefore, the probability of being captured by the cadmium layer surrounding the ^3He detectors increases, and therefore the transmission signal decreases. The transmission signal is highly correlated to the slow-down capability (-0.95).

As already suggested in previous studies [9,10], this confirms that the internal monitor and the transmission measurement signals are both responsive to the waste neutron absorption and slowing-down characteristics.

4.3. Calibration coefficient

Three models are discussed in this section. The first is a model based on the metal mapping (MM) of waste drums, which is used as reference

for comparison with the present work. It is representative of the historical approach that has been used at La Hague reprocessing plant. This model does not exploit waste observables but the neutron absorption ratio, the waste density and the absorption fraction in nozzles. These parameters are calculated by estimating the amount of Inconel, zirconium and stainless steel as well as the number and mass of nozzles in each waste drum. A second model exploiting the prompt and the internal flux monitor signals will be described later. This model will not be used in the future at the reprocessing plant; it is only given here to clarify the role of using an interrogation neutron flux monitor and a transmission signal, and to highlight their importance independently. Finally, the last model proposed will benefit from the prompt signal, the internal monitor and the transmission signals. This correlation, which is the objective of the present work, will be implemented in the next years in the measurement and interpretation algorithm of station 0 in the shells and nozzles compaction facility.

Fig. 5 shows the correlation between the global calibration coefficients simulated with the MCNP code and those predicted by the MM model. The 95% prediction bands are shown. The closer the correlation slope is to the line of $y = x$, the better the prediction. The figure of merit chosen to quantify the model quality will be the regression standard deviation relative to the mean value of the calibration coefficient CC. In this case, the relative standard deviation of the correlation is about 46% and the slope is close to 0.88, which is not satisfactory. Even though this model explicitly considers an important aspect of the waste matrix such as the neutron absorption, it still lacks of information about the neutron slow-down. Furthermore, the uncertainty of each model parameter is estimated to be as large as 5% (1σ). Thus, the performance of this model is insufficient to precisely predict the residual fissile mass even though safety margins are conveniently applied to ensure the sub-criticality condition during operation.

The following model now considers the prompt signal and the internal flux monitor signal. Fig. 6 provides the correlation between the global calibration coefficients simulated with the MCNP code and those predicted by the model. The relative standard deviation is close to 18% with a slope of 0.98, for a dynamic range variation of 3 decades. The performance prediction is more than 2.5 times better than the metal mapping model. Neither is the performance impacted by cases containing the fissile clusters in the waste matrix as they are within the 95%

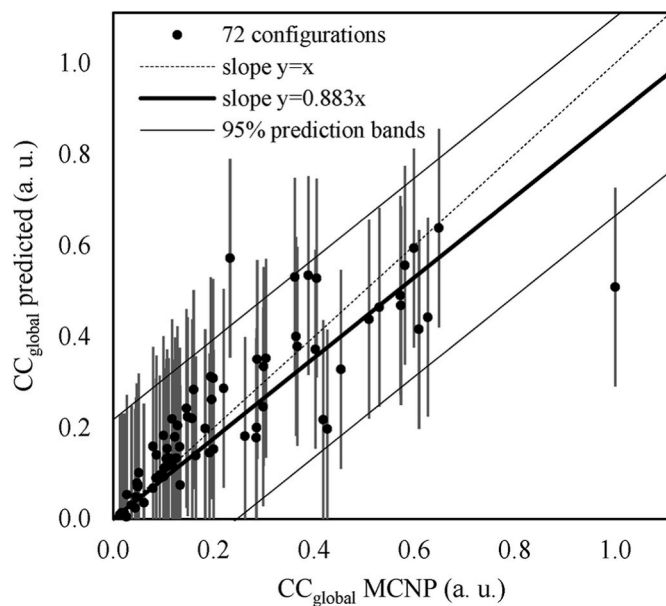


Fig. 5. Correlation between CC_{global} , predicted with the absorption ratio, absorption fraction in the nozzles and density, and the CC_{global} simulated with the MCNP code for the metal mapping model.

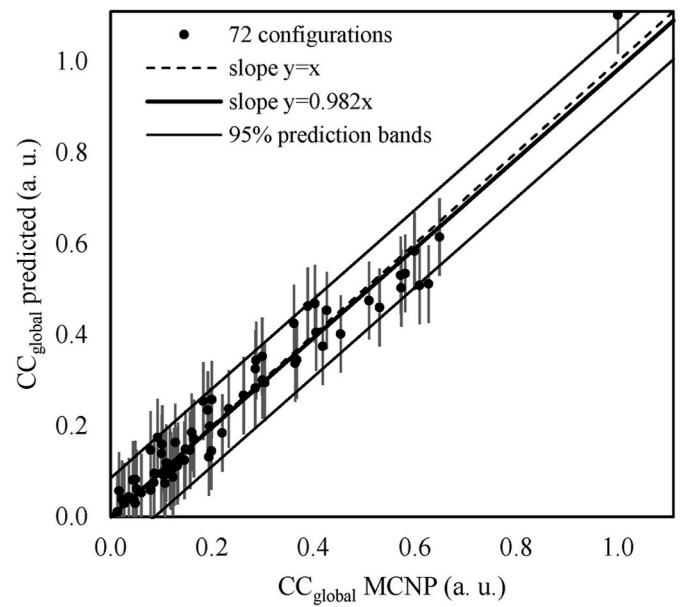


Fig. 6. Correlation between CC_{global} , predicted with prompt signal and internal monitor signal, and the CC_{global} simulated with MCNP code.

prediction bands, highlighting the robustness of the method. The gain with respect to the MM model is noticeable.

The last model taking into account the prompt signal, the internal monitor signal and the transmission signal is shown in Fig. 7. In practice, the ratio of the transmission signal to the correct functioning signal is used rather than the transmission measurement itself in order to eliminate any acquisition channel drift. As it can be seen, the points are much closer to the line of $y = x$ than in the previous model. Adding information about the neutron slowing-down process of the matrix improves the quality of the regression up to 13% of the mean calibration coefficient. This is an improvement of a factor 1.4 relative to the flux monitor/prompt signal model and by a factor of 3.5 compared with the MM model.

The last model was challenged significant amount of OPEX available at La Hague compaction facility. Considering that the transmission

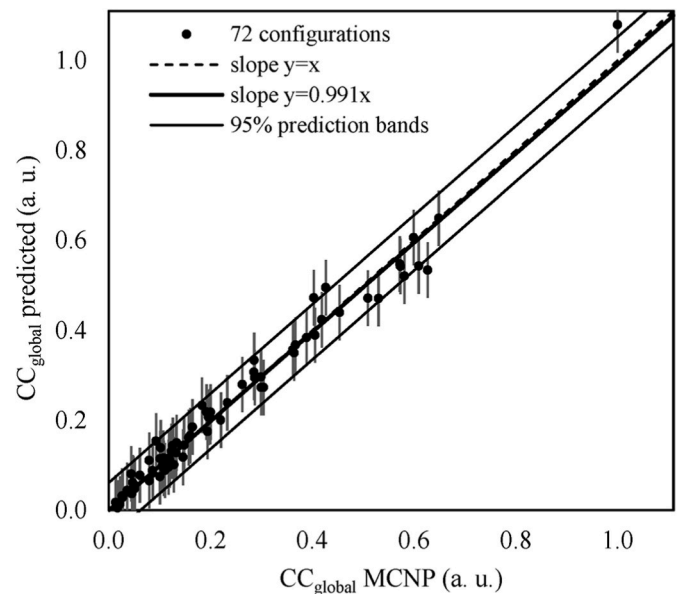


Fig. 7. Correlation between CC_{global} , predicted with S, IM and the transmission measurement signal T, and the CC_{global} simulated with MCNP code.

measurements for the residual fissile mass assessment have been used since 2019, a total of 1050 waste drums were reinterpreted and results were compared against the predictive model currently at operation. The criticality margin was not exceeded in any of the cases, even though larger safety margins were established. For instance, a larger penalty coefficient was applied to the final calculation of the fissile mass in order to consider possible spatial heterogeneities in the residual contamination. Additionally, this work provides a more robust prediction as it accounts for potential fissile agglomerates in the waste due to dissolution process defects. On average, the residual fissile mass results 10% larger (in some cases overestimated by a factor up to 2.8) compared with the current model.

5. CONCLUSIONS

Residual fissile mass measurements are performed at station 0 of the shells and nozzles compaction facility at La Hague spent fuel reprocessing plant. Due to a high variability in the composition and geometry of the metallic waste, the impact on the fissile mass prediction is significant. This paper describes a matrix effect correction approach based on internal flux monitors and transmission measurements. The internal flux monitor signal is sensitive to neutron absorption, while the transmission signal is correlated to the neutron slow-down property. A new predictive model of the global calibration coefficient (which is used to determine the residual fissile mass) has therefore been developed using a multilinear regression in terms of the waste observables: the internal monitor signal, the transmission signal and the prompt signal. The relative standard deviation of the correlation is about 13% of the mean calibration coefficient, reducing the uncertainty prediction by a factor of 3.5 considering the metal mapping model (descriptive of the historical approach used nowadays at La Hague). The model described herein boasts greater robustness as it is able to provide estimates with fissile clusters (created in the previous dissolution process) distributed homogeneously in the waste matrix. This model will be implemented in the measurement and interpretation algorithm of station 0 in the shells and nozzles compaction facility at Orano's La Hague plant in order to predict the fissile contamination of all waste drums.

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CRediT authorship contribution statement

J.P. Scotta: Writing – original draft, Investigation. **L. Tamagno:** Investigation. **C. Passard:** Validation, Investigation. **R. Antoni:** Investigation. **O. Gueton:** Project administration. **J.C. Vandamme:** Funding

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Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Data availability

The data that has been used is confidential.

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