Performance Assessment of a Correction Matrix Effect Method Based on a ^{3}He Drum Monitor on a DDT Measurement Station for the Assay of Fissile Mass in Large-Size Drums – 14026

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ABSTRACT

The fissile mass in radioactive waste drums filled with compacted metallic residues (spent fuel hulls and nozzles) produced at AREVA La Hague reprocessing plant is measured by neutron interrogation with the Differential Die-away measurement Technique (DDT), on the ACC “Atelier Compactage Coques et embouts” waste compaction facility. In the future, old hulls and nozzles mixed with Ion-Exchange Resins will be measured. The ion-exchange resins increase neutron moderation in the matrix, compare to the waste measured in the current process. In this context, the Nuclear Measurement Laboratory (NML) of CEA Cadarache is studying a matrix effect correction method, based on a drum monitor (^{3}He proportional counter) signal. A previous study [1] performed with the NML R&D measurement cell PROMETHEE 6 has shown the feasibility of the method, and the capability of MCNP simulations to reproduce experiments and to estimate the performances of the proposed correction. The next step of the study concerns the implementation of the method on the industrial station and the assessment of the final performances. This paper is more specifically focused on the establishment of the correlation between the prompt calibration coefficient of the ^{239}Pu signal and the drum monitor signal. This work was performed using MCNP simulations and a fractional factorial experimental design composed of 9 matrix parameters representative of the variation range of historical waste. Calculations demonstrate that the method allows the assay of the fissile mass roughly within a factor of 2, while the matrix effect without correction ranges on 2 decades.

INTRODUCTION

AREVA NC is preparing to process, characterize and compact old spent fuel metallic waste stored at La Hague reprocessing plant in view of their future storage. The packaging intended for a large part of these historic waste must be done in CSD-C canisters “Colis Standard de Déchets Compactés” on the ACC hulls and end pieces compaction facility [2]. However, the measurement of the residual fissile materials must take into account differences in the waste matrix between historic and currently processed metallic waste. These components are mainly made of stainless steel, nickel-based steel and zirconium. However, the presence of Ion Exchange Resins (IER), though in minor proportions, gives to the historic waste matrix a moderating capacity that is not encountered in currently processed waste.

The NML of CEA Cadarache is exploring options for implementing a matrix effect correction on the industrial neutron measurement station “P0” which is based on the Differential Die-away Technique [3] [4]. This correction is based on the use of an internal ^{3}He monitor which is sensitive to matrix materials [5]. In order to validate this method, the differences between the experimental results and those from MCNP calculations [6] have been investigated with the “PROMETHEE 6” R&D cell of CEA Cadarache [7] [8], which was modified to approach neutron moderation and absorption properties of the industrial P0 cell. This "experiment / calculation" benchmark reported in reference [1] allowed to validate the MCNP model which can now be used for P0. This paper presents the performances of the matrix effect correction estimated with MCNP calculations a fractional factorial experimental design composed of 9 matrix parameters representative of the variation of historical waste.

PROMETHHE 6 EXPERIMENT/SIMULATION BENCHMARK

The differential die-away technique [3][4] is implemented in PROMETHEE 6 (see fig. 1) using a D-T pulsed generator emitting 14 MeV neutrons with typical 125 Hz frequency and 200 µs pulse duration. Neutrons are moderated in the measurement cell and the waste drum materials. Thermal neutrons induce fissions on fissile isotopes like ^{235}U, ^{239}Pu and ^{241}Pu. Fission fast neutrons are then measured by fast neutron detection blocks made of ^{3}He counters, surrounded by polyethylene to slow down fission neutrons and increase their absorption cross section in ^{3}He, and by cadmium to discriminate fission fast neutrons from thermal interrogating neutrons flux. This measurement is performed over a specific time window (“prompt area”) starting a few hundred microseconds after the end of the pulse and lasting a few milliseconds.

The matrix correction is based on 2 internal flux monitors (25 cm long and 1 cm diameter ^{3}He proportional counters located in the drum cavity), the signal of which (S_{MI}) being also integrated on the “prompt area”.

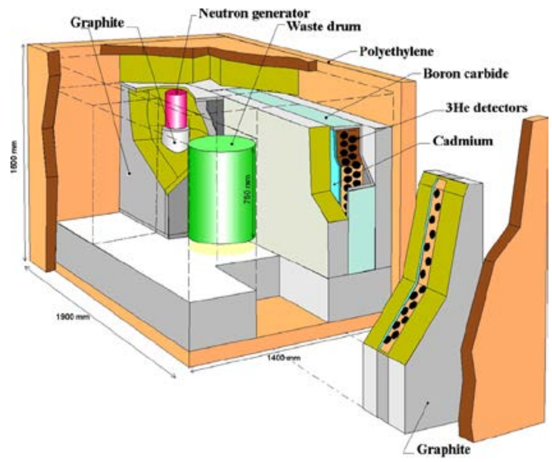


Fig. 1. Schematic view of the PROMETHEE 6 measurement cell.

A limited number of representative matrices were studied to assess the feasibility of the correction method with PROMETHEE 6, as envisaged for the experimental calibration procedure of historic waste on the industrial P0. MCNP simulation will be widely used in P0 calibration because of the high cost of experiments.

The fissile material measurements in the test matrices were performed with five ^{235}U samples fixed on an aluminium rod and inserted in aluminium tubes in known positions in the waste matrix. This device provides a nearly homogeneous vertical distribution of the fissile material in the drum. Measurements were carried out for different radius locations to assess the effect of the fissile material radial position.

To approach the measurement conditions of the industrial P0 station (drums filled with an homogeneous matrix), the discrete values of the prompt calibration coefficient CP5 (in counts per second and per gram of ^{235}U) for each radial position have been fitted to describe CP5 as a function of the radius (*r*), and this function was then integrated, to obtain the $CP5_{hom}$ coefficient representative of a homogeneous fissile material repartition (eq. 1).

$$CP5_{\mathrm{hom}} = \frac{1}{\pi R^2}\int_0^R CP5(r)\,2\pi\, r\, dr \qquad \text{(eq. 1)}$$

The correlation between $CP5_{hom}$ and the flux monitor signal S_{MI} is shown in fig. 2.

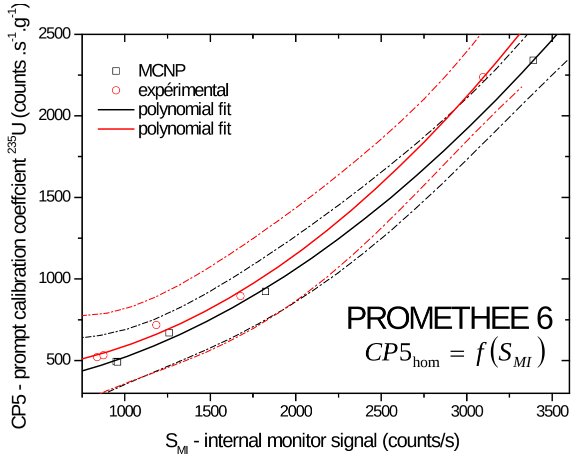


Fig. 2. Relationship between the prompt neutron calibration coefficient for an homogeneous fissile material distribution $CP5_{hom}$ and the internal monitor mean signal S_{MI} for both experiment and MCNP calculation with PROMETHEE 6, from reference [1].

Fig. 2 shows a good agreement between the experimental and calculated regression curves in the case of PROMETHEE 6. This approach allows a prediction of $CP5_{hom}$ as a function of S_{MI} using a quadratic regression (eq. 2) with a limited uncertainty, while CP5 ranges on about two decades depending on the matrix composition.

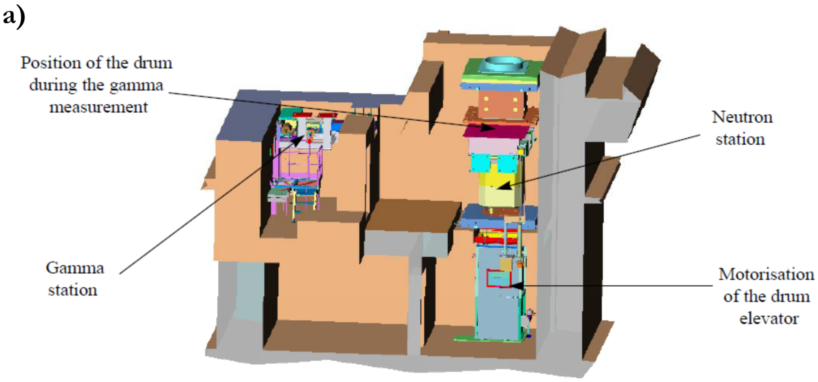
$$\left[CP5_{\mathrm{hom}}\right]_{reg} = f_{reg}\left(S_{MI}\right) = a_o + a_1\left(S_{MI} - \overline{S}_{MI}\right) + a_2\left(S_{MI} - \overline{S}_{MI}\right)^2 \qquad \text{(eq. 2)}$$

The good agreement between experiment and calculation shows that numerical simulation can be used to predict the performance of the method for the industrial station.

INDUSTRIAL MEASUREMENT CELL

The neutron measurement cell, so called P0-2 is located at the entrance of the ACC (see fig. 3a). It is mainly used to determine the fissile mass in the drums to ensure subcriticality up to the intermediate storage area at the facility exit.

a)



b)

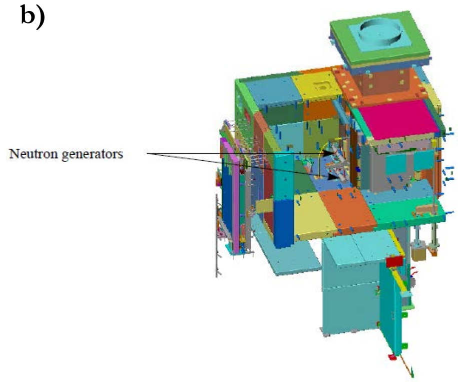


Fig. 3. a) general layout of the incoming measurement station on ACC compacting facility b) layout of the P0-2 measurement cell [10]

The P0 neutron measurement cell includes two GENIE 36 neutron generators developed by SODERN for the ACC compacting project and three blocks containing each 83 ^{3}He proportional counters with a one-meter length 2.54 cm diameter, and 4 bar filling pressure (150NH100 type manufactured by AREVA Canberra) covering three sides of the cell. The empty cell measurement efficiency is around 8%. The drums are rotated during the measurements. Fig. 3 b) shows the layout of the cell and fig. 4 is the MCNP model used for the establishment of the correlation $CP9 = f(S_{MI})$ between the prompt calibration coefficient of the ^{239}Pu signal and the internal monitor signal.

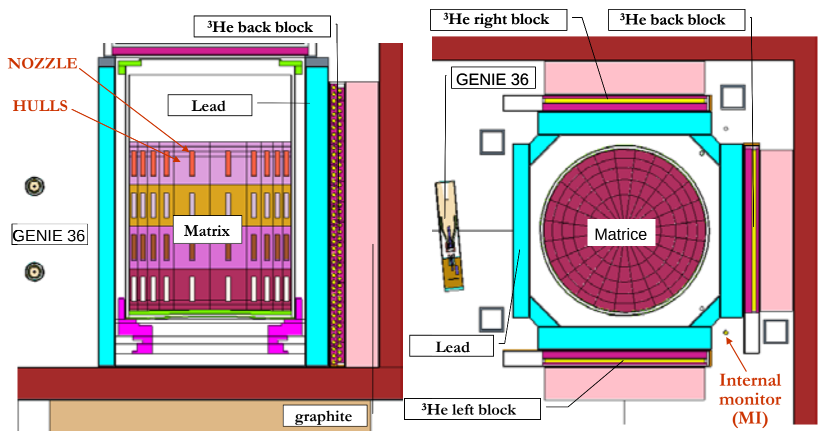


Fig. 4. MCNP plots of P0-2 measurement cell showing 800 litres drum and the internal monitor (MI)

The internal monitor is a ^{3}He detector of 1 m length, 2.5 cm diameter, located vertically behind the 13 cm thick lead shielding present in the measurement cavity to protect all the detectors from the gamma radiations of the high-level waste. The detector is surrounded by a 0.5 mm Cd layer with a slight opening toward the measured drum to be only sensitive to the thermal interrogating neutron flux within the matrix. The internal monitor signal is obtained with MCNP, in a single calculation step, according to equation 3.

$$S_{MI} = k_{\det} V \frac{\tau_{(n,p)}}{\Delta t_{zp}} \frac{E_n}{f_t} \qquad \text{(eq. 3)}$$

In which $k_{\det}$ is an electronic losses coefficient ($k_{\det}$ ~ 0.75), V is the volume of the internal monitor (490 cm^3), $\tau_{(n,p)}$ is the ^{3}He(n,p)t reaction rate per volume unit and per source neutron calculated by MCNP, Δt_{zp} is the prompt neutron area (from 1020 to 3127 µs), E_n is the 14 MeV neutron generator total emission ($\cong$ 2 x 2.10^9 n.s^{-1} in the nominal operation) and f_t is the generator pulse frequency (125 Hz).

The prompt calibration coefficient is carried out in two MCNP calculation steps, which allows a good representation of fission rate and detection efficiency heterogeneities according to the location within the matrix, as well as a large gain in computing time. A mesh grid of 1200 cells is defined to perform such calculations. It includes 24 angular sectors, 5 radii and 10 heights (see Fig 4.). Most of the cross sections are taken from the ENDF-BVI library supplied with the MCNPX 2.5.0 code [10]. The final CP9 result is provided by equation 4.

$$CP9 = k_c \frac{\overline{\nu}\, E_n}{f_t\, \Delta t_{zp}} \sum_{j=1}^{j=1200} \left(\varepsilon_{np}\right)_j \left(\tau_{fiss}\right)_j \qquad \text{(eq. 4)}$$

In which $\overline{\nu}$ is the mean number of neutrons by fission, $\left(\tau_{fiss}\right)_j$ is the fission rate for mesh j provided by MCNP, $\left(\varepsilon_{np}\right)_j$ is the detection efficiency for mesh j also provided by MCNP and k_c is a time convolution coefficient to take into account the delay between fission and the detection of prompt neutrons. It should be noted that in order to reduce the duration of simulation calculations, MCNP convergence speed-up techniques have been implemented. The “Meshed Weight Windows” with time discretization was used [11].

Matrix effects on interrogating neutrons mainly depend on 9 parameters of the characteristic expected drum content. 36 matrices have been defined using a factorial experience design for those factors, on two levels with a midpoint for 7 of them, and just two levels for the 2 others [11]. For each parameter, extreme limits (min, max) and the median value (med) are representative of the variation range of the real waste. The two main parameters are a moderating ratio (MDR) and an absorbing ratio (ABR). ABR represents the absorbing capacity due to presence of both usual 304L stainless steel (Fe 72%, Cr 18%, Ni 10%) and of nickel-rich steel (Fe 19%, Cr 19%, Ni 52,4%) in end pieces. It is defined as the mass ratio given by expression 5.

$$ABR = \frac{m_{stainless\ steel} + 1{,}4\, m_{nickel\ based\ steel}}{m_{stainless\ steel} + m_{nickel\ based\ steel} + m_{Zirconium}} \qquad \text{(eq. 5)}$$

In which the 1.4 factor reflects that for the same mass, nickel-rich steel has a 40% larger thermal neutron absorption cross section than 304L steel. Zirconium is the main element in this kind of waste in terms of mass fraction but its neutron absorption cross section is very low.

MDR represents the moderating property provided by residual water spread within the matrix and by the presence of IER. It is defined as expression 6.

$$MDR = \frac{m_{Water} + m_{IER}}{V_{Hulls\,\&\,Nozzle}} \qquad \text{(eq. 6)}$$

In which m_{Water} and m_{IER} are the water and IER masses in gram unit, $V_{Hulls\,\&\,Nozzle}$ is the hulls and nozzle filling volume of the drum in litre unit and IER mass. The seven other parameters are:

- the bulk specific density,
- the drum filling height,
- the nozzle proportion in ABR – this parameter describing the proportion of absorbing materials (stainless steel and nickel rich steel) in either nozzles or the hull matrix. In the MCNP model, the fissile mass is indeed located in the hull matrix but not in the nozzle, which has an effect on neutron absorption,
- the nozzle location distribution within the matrix - Fig. 5 give the MCNP model for the two level (centred or spread) of this parameter,
- the residual water after the drying process - because the waste drums are filled with water for the storage and transport,
- the concentration of difficult-to-dissolve particles in the spent fuel reprocessing (such as Nb, Ru, Mo …), which have a significant neutron absorption cross section,
- self-shielding in fissile material.

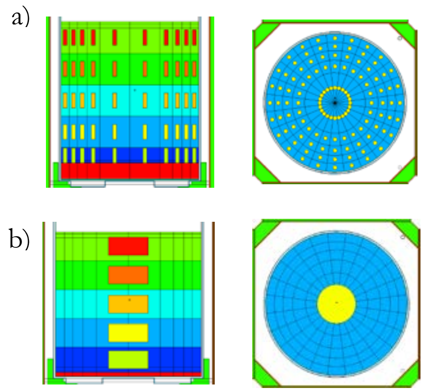


Fig. 5. MCNP plot for a) spread level nozzle distribution
b) centred level nozzle distribution

Beyond the 36 matrices defined with the factorial experimental design, 20 “Extra-cases” were modelled and included in the correlation law performance assessment in order to take into account fissile mass location and matrix material heterogeneity.

Fig. 6 shows the CP9 versus S_{MI} correction established from the 36+20 matrices modelled with MCNP. The quadratic fit used in fig. 2 would allow a prediction of CP9 as a function of S_{MI} with a limited uncertainty. However, it is required to decrease the residual standard deviation for low CP9 values to avoid a large overestimation of the fissile mass. For this purpose, a quadratic regression is used for S_{MI} values located below a given S_{MI} threshold (so called "low MI area” in fig. 6) and another quadratic fit, independent of the previous one is calculated above this threshold (so called "high MI area" in fig. 6). For the reliability of this approach, a sufficiently large and quite similar number of cases is present in the 2 regions. The cut-off value was determined to optimize the residual standard deviation in the low MI area.

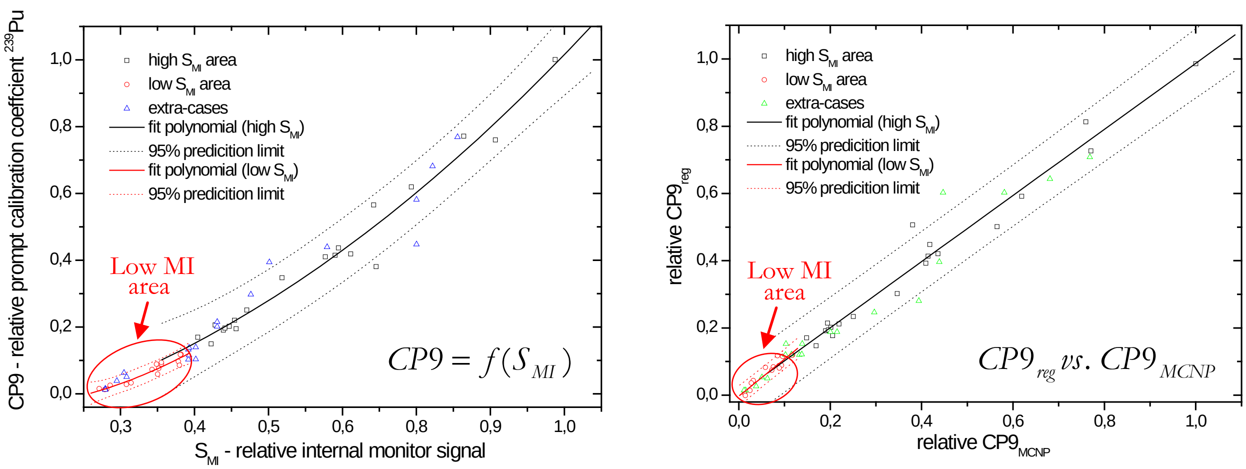


Fig. 6. a) Correlation between the prompt neutron calibration coefficient and the internal monitor signal S_{MI} b) Relationship between the predicted CP9 (i.e. $CP9_{reg}$) and the one calculated, with MCNP (the objective being closer to $CP9_{reg}=CP9_{MCNP}$). Both S_{MI} and CP9 are normalized to their maximal values.

The standard deviation obtained with this approach in the low MI area allows a prediction of CP9 as a function of S_{MI} with a limited overestimation of roughly 100% (2 standard deviations uncertainty), while CP9 ranges on about two decades as a function of the matrix composition, which is a dramatic reduction of the matrix effect uncertainty.

The final assessment of the maximum fissile mass, requested for criticality purposes, involves taking into account other effects related to matrix heterogeneities, self-shielding in fissile material, and background noise calibration. Regarding the significant decrease of the matrix effect uncertainty owing to the proposed method, the total standard deviation on the ^{239}Pu prompt calibration coefficient including the other effects leads to a maximum overestimation of the residual fissile mass that is consistent with the criticality measurement objective for the future recovery of historic waste.

CONCLUSION AND PERSPECTIVE

The matrix effect correction method based on internal flux monitors has been proposed to reduce the uncertainty on the fissile mass measured in the P0 industrial system at AREVA NC La Hague reprocessing plant, for the characterisation of historic radioactive waste drums. A preliminary study performed with the PROMETHEE 6 measurement cell of the NML of CEA Cadarache has shown that experiment and MCNP calculations are in good agreement for a large set of test matrices and fissile material positions, proving that numerical simulation could be used to predict the performance of the method for the industrial station [1].

The assessment of the performances of the method on the industrial station was thus performed by modelling 56 matrices with MCNP to establish the correlation between the prompt calibration coefficient of the ^{239}Pu signal and the drum monitor signal. The prediction law of the ^{239}Pu prompt calibration coefficient leads to an overestimation of the fissile mass roughly a factor 2 taking into account a 2 standard deviation uncertainty, while the uncorrected signal ranges on 2 decades for the studied matrices. This allows the total standard deviation on the residual fissile mass to be consistent with the criticality objectives for the future recovery of historic waste.

Next step will consist in confirming the reliability of this method with an experimental benchmark on the industrial P0 system, by performing dedicated calibration measurements with a few matrices representative of historic waste.

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