First Industrial Tests of a Matrix Monitor Correction for the Differential Die-away Technique of Historic Waste Drums

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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 NC La Hague reprocessing plant is measured by neutron interrogation with the Differential Die-away measurement Technique (DDT). In the next years, old hulls and nozzles mixed with Ion-Exchange Resins will be measured. The ion-exchange resins increase neutron moderation in the matrix, compared to the waste measured in the current process. In this context, the Nuclear Measurement Laboratory (LMN) of CEA Cadarache has studied a matrix effect correction method, based on a drum monitor, namely a ^{3}He proportional counter located inside the measurement cavity. After feasibility studies performed with LMN's PROMETHEE 6 laboratory measurement cell and with MCNPX simulations, this paper presents first experimental tests performed on the industrial ACC (hulls and nozzles compaction facility) measurement system. A calculation vs. experiment benchmark has been achieved by performing dedicated calibration measurements with a representative drum and ^{235}U samples. The comparison between calculation and experiment shows a satisfactory agreement for the drum monitor. The final objective of this work is to confirm the reliability of the modeling approach and the industrial feasibility of the method, which will be implemented on the industrial station for the measurement of historical wastes.**

***Index Terms* — Differential Die-away Technique (DDT), matrix effect correction, numerical simulations, Drum monitor, radioactive waste.**

I. 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 wastes must be done in CSD-C canisters on the ACC hulls and end pieces compaction facility [1]. 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) in historic waste, though in minor proportions, gives the matrix a moderating capacity that is not encountered in the current process.

The Nuclear Measurement Laboratory (LMN) 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 [2] [3].

This matrix effect correction method is based on the use of a drum ^{3}He monitor signal sensitive to matrix materials [4]. In order to validate this method, the differences between experimental results and MCNPX calculations [5] have been first investigated with the "PROMETHEE 6" R&D cell of CEA Cadarache, which was modified to approach the neutron moderation and absorption properties of the industrial P0 cell. It has allowed an "experiment / calculation" benchmark to validate the MCNPX model used for this work about P0 [6].

A next step of the study concerned the assessment of the performances of the method in case of the industrial station, 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 factorial experimental design composed of matrix parameters representative of the variation range of historical waste [7].

Hereafter, are presented and discussed first experimental tests performed on the industrial ACC measurement system. A calculation vs. experiment benchmark has been achieved by performing dedicated calibration measurement with a representative drum and ^{235}U samples.

II. DIFFERENTIAL DIE-AWAY TECHNIQUE

A Deuterium Tritium pulsed generator emits 14 MeV neutrons with a typical frequency close to 100 Hz and with a pulse duration of a few hundred microseconds. Neutrons are moderated in the measurement cell and waste drum materials. Thermal neutrons induce fissions on isotopes like ^{235}U, ^{239}Pu and ^{241}Pu, also referred as "fissile materials" in this paper. The fast neutrons emitted during fissions are then measured by fast neutron detection blocks made of ^{3}He counters, surrounded by

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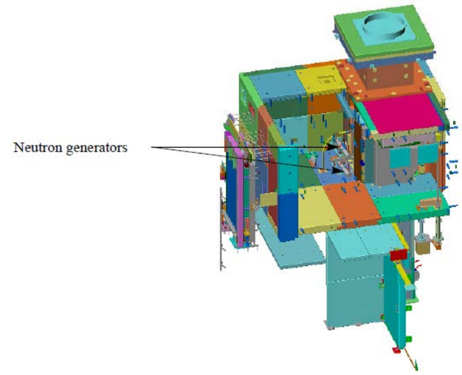
polyethylene to slow down fission neutrons and increase the (n,p) reaction in ^{3}He, and by cadmium to discriminate fast fission neutrons from thermal interrogating neutrons. This measurement is performed over a specific time window ("prompt neutron area") starting a few hundred microseconds after the end of the pulse and lasting a few milliseconds. The net prompt signal $(S_p)_{net}$ in this area is obtained by subtracting the background noise including ^{244}Cm and ^{240}Pu spontaneous fission neutrons, neutrons from photonuclear reactions on lead and, for a slight part, fission delayed neutrons. For a given matrix, $(S_p)_{net}$ is proportional to the fissile material mass uniformly distributed in the drum, which is obtained thanks to a "prompt calibration coefficient" (e.g. CP5 in counts per second and per gram of ^{235}U). Additional information about fission delayed neutrons can be obtained in another time window, a few milliseconds after the pulse, which can be used to complete the validation of the numerical model. The net delayed neutron signal $(S_r)_{net}$ is then due to delayed neutrons emitted by fission fragments and proportional to fissile and fertile mass with specific "delayed calibration coefficients" (e.g. CR5 and CR8 in counts per second and per gram of ^{235}U and ^{238}U, respectively).

III. INDUSTRIAL MEASUREMENT CELL

The industrial neutron measurement cell, called P0-2 station (P0-1 being a gamma spectroscopy station), is located at the entrance of the ACC compaction facility, see Fig. 1. It is mainly used to determine the fissile mass in the drums, in view to ensure sub-criticality during the compaction process. The P0-2 neutron measurement cell includes two high flux GENIE 36 neutron generators manufactured by SODERN, with a nominal emission of more than $2 \cdot 10^{9}$ n.s^{-1} each, and three fast neutron detection blocks containing each 83 ^{3}He 150NH100 proportional counters manufactured by AREVA Canberra, with a 1 m length, 2.5 cm diameter, and 4 bar filling pressure, covering three walls of the cell. The empty cell measurement efficiency is around 11%. The drums are rotated during the measurements.

The drum monitor is also a 150NH100 ^{3}He detector with a 1 m length, 2.5 cm diameter, and 4 bar pressure. It is located vertically behind the 10 cm thick lead shielding present in the measurement cavity, see further Fig. 2, to protect all the detectors from gamma radiations emitted by the high-level waste. The detector is partially surrounded by a 0.5 mm Cd sheet used as a collimator, to focus on thermal neutrons stemming from the drum.



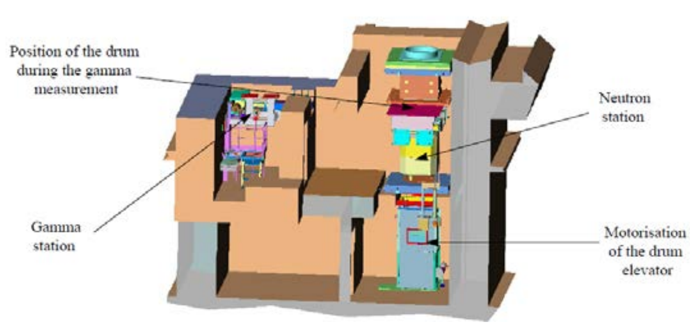


Fig. 1. Above: general layout of the incoming measurement station on ACC compacting facility; below: layout of the P0-2 measurement cell [8].

IV. EXPERIMENTAL SET-UP AND MEASUREMENT

The 800 l test drum is filled with a transport basket. Both the drum and basket are made of stainless steel with a total mass of 890 kg. In addition, a hydraulic damper provides a water volume of ~15 l located at the bottom, between the transport basket and the drum. A "water box" is also located on the bottom of the transport basket. This device, used for a previous calibration test, is composed of an empty Al cylinder filled with Zircaloy (zirconium alloy), stainless steel and water. It provides an additional amount of 9 litres of water and ~ 4 kg of steel.

In order to assess the prompt neutron calibration coefficient and the corresponding drum monitor response, a set of uranium pellets have been fixed on an aluminium guide and placed inside a vertical tube located on the drum axis, see Fig. 2. The pellets are spaced to cover the transport basket height. The total fissile mass of ^{235}U is close to 10 g and the ^{238}U mass is larger than 200 g. Self-shielding in the pellets has been estimated by MCNP calculation to be lower than 20%.

The two GENIE 36 pulsed neutron generators from EADS SODERN are operated at an average total emission of 5.5×10^{9} n/s [4π], with a 140 kV high voltage, ~100 Hz pulse frequency, and 200 µs pulse duration. The signal from ^{3}He tube is processed by current amplifiers and a 64 channels MEDAS dating electronic card. Acquisition windows after the start of the pulse are (1) a prompt neutron area from 1020 to 3127 µs, and (2) a delayed neutron area from 5570 to 8000 µs. Real counting time is typically 15 min. The drum is uniformly rotated during acquisition at a speed of about 3 rounds per min.

V. NUMERICAL SIMULATION

The MCNPX [5] numerical model of the industrial station with the ^{235}U samples is shown in Fig. 2. The source holder was described in the model but not the rest of the aluminium support of the uranium pellets due to the low capture cross section of aluminium for thermal neutrons.

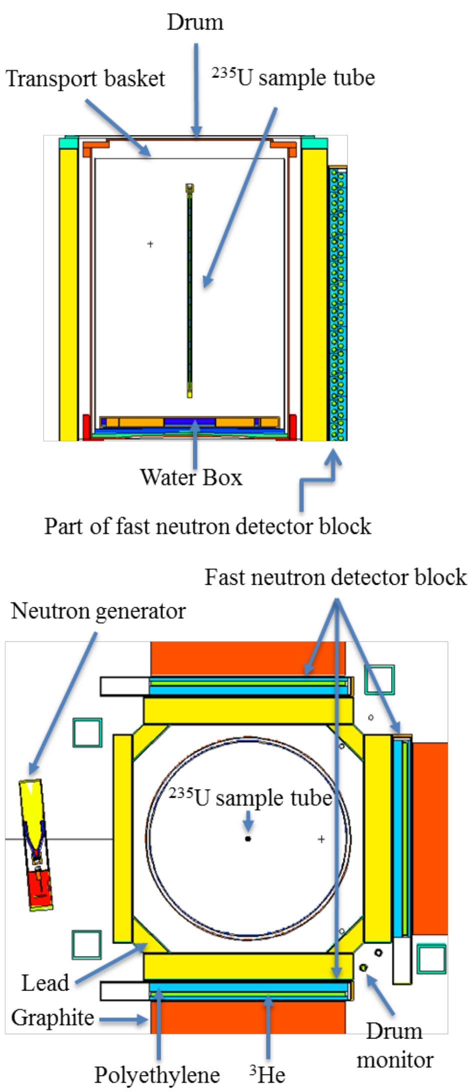


Fig. 2. MCNP plots of industrial station P0-2 showing the test drum, the tube containing the uranium samples, and the drum monitor. Above, a vertical cross section; below, a horizontal cross section.

VI. COMPARISON BETWEEN CALCULATION AND EXPERIMENT

First, electronics losses coefficients have been assessed by measuring, in passive mode, a 3.7×10^4 n/s ^{252}Cf source at the centre of the empty cavity, during a 15 min acquisition, and by comparing experiment and calculation. These coefficients principally account for count losses due to the electronic discriminator threshold of the ^{3}He detectors. The calculated detection efficiency of the fast neutron detection blocks must be multiplied by a factor $k_S = 0.807 \pm 0.025$ [1σ] to obtain the experimental detection efficiency. Standard deviation on k_s is dominated by the ^{252}Cf source relative uncertainty, which is 3% [1σ].

Matrix effects on interrogating neutrons mainly depend on two main parameters, which are a moderating ratio (MDR) and an absorbing ratio (ABR) as defined in [7]. In historical wastes, ABR mainly represents the absorbing capacity due to the presence of stainless steel (Fe 72%, Cr 18%, Ni 10%) and nickel-rich steel (Fe 19%, Cr 19%, Ni 52%) in end-pieces. MDR represents the moderating property provided by residual drip water spread within the matrix and by the presence of ion exchanging resins (IER). It was shown that S_{MI} is not sensitive to fissile materials but only to matrix characteristics [6] [7].

In order to assess the S_{MI} shift brought by the amount of steel and water contained in the test drum, an active measurement was carried out with and without the drum inside P0-2 cell, as reported in Table I. The measured decrease of the monitor signal is 0.519 ± 0.001 [1σ], while the ratio obtained by numerical simulation is 0.635 ± 0.020 [1σ], i.e. a relative difference of 18.3% ± 2.5% [1σ]. Concerning the direct comparison of experimental and calculated absolute monitor signals, which shows a 5% overestimation by calculation for the empty cell and about 20% with the drum, one can first note that the main experimental uncertainty comes from the precision on the absolute neutron emission, which is announced by the manufacturer with an approximate 15% uncertainty. It is important to note that all measurements are normalised using a dedicated ^{3}He monitor located outside the measurement cell, and therefore this uncertainty only applies to the absolute signals, but not to the ratio of the signals recorded with and without the drum. Another important uncertainty is the exact quantity of water inside the drum (hydraulic damper water and water box in the bottom, see section IV), which cannot be ascertained with a precision better than a few litres. A complementary MCNP sensitivity study (not reported here) showed that the effect may exceed 10% on the drum monitor signal (as well as on the prompt neutron ^{235}U signal, and a bit less on the delayed neutron signal). On the other hand, the statistical relative uncertainty on all MCNP calculations is better than 3%, but the model of the measurement cell, though established from as-built drawings, may still suffer from small uncertainties like details on the neutron generator design or boron traces in graphite.

TABLE I. EXPERIMENTAL AND CALCULATED DRUM MONITOR SIGNALS IN ACTIVE MEASUREMENT WITHOUT ^{235}U (PROMT NEUTRON TIME WINDOW)

Drum monitor signal	**Results in active mode without ^{235}U**		
	EXP	***MCNP***	***Δ(%)***
S_{MI} with drum (a)	2201.2 ± 2.4 c/s	2818.0 ± 83.1 c/s	-21.9 ± 2.3%
S_{MI} without drum (b)	4240.9 ± 3.4 c/s	4436.2 ± 40.4 c/s	-4.4 ± 0.9%
Ratio (a/b)	0.519 ± 0.001	0.635 ± 0.020	-18.3 ± 2.5%

Moreover, the net prompt and delayed neutron signals, $(S_p)_{net}$ and $(S_r)_{net}$ respectively, were assessed with the ^{235}U samples in the transport basket, see results in Table II. As for the monitor signal recorded during the prompt neutron time window, simulation overestimates the prompt neutron useful signal of ^{235}U. On the contrary, it slightly underestimates the delayed neutron ^{235}U signal, which probably evidences an underestimation of thermal neutron absorption, to which the delayed neutron signal is less sensitive because of the contribution of fast neutron fissions. This underestimation is probably due to the precision on the water quantity in the drum, as mentioned above.

TABLE II. EXPERIMENTAL AND CALCULATED USEFUL SIGNALS OF ^{235}U IN THE DRUM IN ACTIVE MODE WITHIN PROMPT AND DELAYED NEUTRON TIME WINDOWS

Detection blocks signal	Results in active mode with drum and ^{235}U		
	EXP	***MCNP***	***Δ(%)***
$(S_p)_{net}$	2522.4 ± 9.1 c/s	3059.6 ± 38.7 c/s	-17.6 ± 1.1%
$(S_r)_{net}$	63.1 ± 0.9 c/s	59.3 ± 0.2 c/s	6.3 ± 1.5%

Considering the complexity of the experimental set-up and of the numerical model, relative EXP-CALC differences are satisfactory enough for a first validation of the performances previously assessed by simulation for the implementation of the matrix correction method on the industrial P0-2 station [7]. Further experiments are planned in the future with different test matrices to fully calibrate the method and the simulation model.

VII. CONCLUSION

The matrix effect correction method based on drum flux monitors has been proposed to reduce the uncertainty on the fissile mass measured in the P0-2 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 R&D measurement cell has shown that experiment and MCNPX calculations was in good agreement for a large set of test matrices, allowing to consider that numerical simulation could be used to predict the performance of the method for the industrial station. Then, the assessment of the performances of the method on the industrial station was performed by modelling matrices with MCNPX to establish the correlation between the prompt calibration coefficient of the ^{239}Pu signal and the drum monitor signal.

First industrial experiments on P0-2 were reported in this paper to validate the numerical simulation model. Dedicated calibration measurements have been performed with a representative industrial drum filled with ^{235}U fissile samples. Two major quantities were assessed in active mode: (1) the signal of the drum monitor in the prompt neutron time window, with and without the empty drum, and (2) the prompt and delayed neutron signals of ^{235}U within the drum. For both quantities, the agreement between simulation and experiment is satisfactory, considering the complexity of the experimental set-up and modelling uncertainties, which confirms previous numerical performance assessment of the matrix correction method for the industrial P0-2 station. It also show that the numerical model can reliably be used to support next calibration steps of the matrix correction method, in view of the future recovery of historic radioactive waste.

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