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Reduction of the uncertainty due to fissile clusters in radioactive waste characterization with the Differential Die-away Technique

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ABSTRACT

AREVA NC is preparing to process, characterize and compact old used fuel metallic waste stored at La Hague reprocessing plant in view of their future storage ("*Haute Activité Oxyde*" HAO project). For a large part of these historical wastes, the packaging is planned in CSD-C canisters ("*Colis Standard de Déchets Compacté s*") in the ACC hulls and nozzles compaction facility ("*Atelier de Compactage des Coques et embouts*"). . This paper presents a new method to take into account the possible presence of fissile material clusters, which may have a significant impact in the active neutron interrogation (Differential Die-away Technique) measurement of the CSD-C canisters, in the industrial neutron measurement station "P2-2". A matrix effect correction has already been investigated to predict the prompt fission neutron calibration coefficient (which provides the fissile mass) from an internal "drum flux monitor" signal provided during the active measurement by a boron-coated proportional counter located in the measurement cavity, and from a "drum transmission signal" recorded in passive mode by the detection blocks, in presence of an AmBe point source in the measurement cell. Up to now, the relationship between the calibration coefficient and these signals was obtained from a factorial design that did not consider the potential for occurrence of fissile material clusters. The interrogative neutron self-shielding in these clusters was treated separately and resulted in a penalty coefficient larger than 20% to prevent an underestimation of the fissile mass within the drum. In this work, we have shown that the incorporation of a new parameter in the factorial design, representing the fissile mass fraction in these clusters, provides an alternative to the penalty coefficient. This new approach finally does not degrade the uncertainty of the original prediction, which was calculated without taking into consideration the possible presence of clusters. Consequently, the accuracy of the fissile mass assessment is improved by this new method, and this last should be extended to similar DDT measurement stations of larger drums, also using an internal monitor for matrix effect correction.

1. Introduction

AREVA NC is preparing to process, characterize and compact old used fuel metallic waste stored at La Hague reprocessing plant in view of their future storage ("Haute Activité Oxyde" or HAO facility project). The packaging intended for a large part of these historical wastes must be done in CSD-C canisters "Colis Standard de Déchets Compactés" on the ACC hulls and nozzles compaction facility [1]. The measurement of the residual fissile materials must take into account differences in the waste matrix between historical and currently processed metallic waste. All these components are mainly made of stainless steel, nickel-rich steel and zirconium. However, the presence of Ion Exchanging Resins (IER), though in minor proportions, gives to the historical waste matrix a moderating capacity that is not encountered in currently processed waste. In this context, the Nuclear Measurement Laboratory (LMN) of CEA Cadarache is studying a matrix effect correction for the industrial neutron measurement stations "P0-2" and "P2-2", which are based on the Differential Die-away Technique (DDT) [2,3]. Among different methods [4–6], This correction is based on the use of an Internal flux Monitor (IM, namely a ^{3}He proportional counter or boron-coated chambers) which is sensitive to matrix materials [7]. Previous studies [8] have shown the feasibility of the method and the ability of MCNP simulations [9] to reproduce experiments and to estimate the performances of the proposed correction [10].

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This paper presents the assessment of final performances of the matrix effect correction estimated with MCNP calculations for a fractional factorial design composed of 36 matrix cases representative of the variety of historical waste, on the measurement station P2-2 at the output of the facility. In addition, the above-mentioned approach based on the internal monitor to predict the useful signal (^{239}Pu$_{eff}$ prompt neutron calibration coefficient, i.e. count rate per gram of ^{239}Pu$_{eff}$) could valuably be completed by a "Drum Transmission" (DT) monitor, which consists in the passive signal of the whole ^{3}He detectors when an AmBe point source is introduced in the measurement cell. On the other hand, the potential presence of fissile material clusters in partially dissolved hulls induces a potential self-shielding effect, which is currently taken into account by a penalty coefficient larger than 20% in order to prevent an underestimation of the fissile mass. So far, this penalty has been treated outside the fractional factorial design, by taking into account a worst case scenario. A major difference in the newly proposed approach lies in the direct consideration of the fissile material heterogeneity in the fractional factorial design, through the use of a specific variable factor, namely the "fissile mass fraction in clusters" representing the fraction of fissile mass distributed between clusters and uniformly contaminated hulls and nozzles.

2. Differential die-away technique

Two Deuterium Tritium pulsed generators, positioned at different heights, are used to emit 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. Thermalized neutrons induce fissions on isotopes like ^{235}U, ^{239}Pu and ^{241}Pu, also referred to as "fissile materials" in this paper. The fast prompt neutrons emitted during fissions are then measured by fast neutron detection blocks made of ^{3}He counters, first surrounded by polyethylene to slow down fission neutrons and increase the probability of the (n, p) detection reaction in ^{3}He, and secondly by cadmium to discriminate fast fission neutrons from thermal interrogating neutrons.

This measurement is performed over a specific time window (fission prompt neutron area) starting a few hundred microseconds after the end of the pulse and lasting a few milliseconds. The net prompt fission neutron signal in this area is obtained by subtracting the background noise, which includes ^{244}Cm and ^{240}Pu spontaneous fission neutrons, photo-neutrons produced by photonuclear reactions in lead and, for a slight part, fission delayed neutrons. For a given matrix, this net prompt signal is proportionally linked to the fissile material mass, when uniformly distributed in the drum, by a "prompt calibration coefficient" (namely "CP9" in the case of ^{239}Pu isotope, in counts per second and per gram of ^{239}Pu).

3. Industrial measurement cell

The industrial neutron measurement cell, called "P2-2" station (the "P2-1" station being an array of gamma spectroscopy germanium detectors), is located in the output of the ACC compaction facility, see Fig. 1. It is mainly used to determine the fissile mass in the final drums filled with compacted discs of metallic waste, in order to ensure sub-criticality in their storage facility. The P2-2 neutron measurement cell includes two high flux "GENIE 36" neutron generators manufactured by SODERN, with a total nominal emission of about 4.10^{9} ns^{-1} [4π], 140 kV high voltage, 100 Hz pulse frequency and 200 μs pulses. They are parallel with the canister axis (Fig. 2) and shifted in height to homogenize vertically the interrogating flux. Three fast neutron detection blocks are covering three walls of the cell, each containing eighty three ^{3}He proportional counters manufactured by Mirion-Canberra, "150NH100" type with 1 m length, 2.5 cm diameter, and 4 bars filling pressure. The neutron detection efficiency measured with a ^{252}Cf source in the centre of the empty cell (without waste drum) is about 22%. The signal of the ^{3}He detectors is processed by current amplifiers to limit pulse length, and consequently pile-up effects and sensitivity to gamma rays [11]. The fission prompt neutron acquisition window extends from 880 to 2427 μs after the start of the neutron generator pulse [12]. Measurement time (clock time) is typically 15 min. The drum is uniformly rotated during acquisition at a speed of about 3 rotations per min. In addition, an AmBe source of about 4.10^{5} ns^{-1} is used to check the proper functioning of detection blocks. This source, as discussed above, allows an additional measurement called "drum transmission (DT)" to improve the matrix effect assessment.

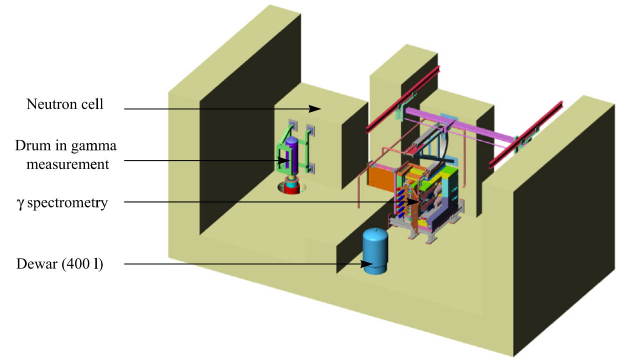

Fig. 1. General layout of the P2 measurement station in ACC facility.

The internal drum monitor IM is composed of two CPNB45 boron-coated chambers manufactured by Photonis (nominal diameter: 2.54 cm, sensitive length: 56.2 cm), containing a boron deposit of 0.2 mg cm^{-2}, with a 92% ^{10}B enrichment. They are located vertically behind a thick lead shielding present in the measurement cavity (see further Fig. 2) to protect detectors from the high gamma flux emitted by the waste.

The CSD-C drum is filled with typically height compacted discs of metallic wastes. The drum is made of stainless steel and the discs are a mixture of stainless steel and zirconium, with a total mass of about 850 kg.

4. Numerical simulation

The layout of the measurement cell and drum modelled in MCNP6.1 [9] is shown Fig. 2. The inner matrix is subdivided into eight discs of compacted hulls and nozzles (hulls and nozzles are mixed in a single material). Each pellet is subdivided into 40 cells with 8 angular sectors, and 5 radii. Note that the two extreme discs (bottom and top) have been partitioned into two sub-discs so as to model a thin pallet on the top and on the bottom, which contains a different material (stainless steel of the compacted drum envelops). Fissile clusters are modelled as an inner cylinder in each cell, sized according to the fissile mass in one cluster. In the extreme sub-discs, however, each cluster is partitioned in the two sub-discs, see Fig. 2. The rest of the cell is filled with hulls and nozzles, and with a homogeneously distributed fissile material representing the usual contamination of the waste. This numerical approach results in 400 cells with a uniform distribution of fissile material in hulls and nozzles, and 320 cells for the fissile clusters.

The IM signal (S_{IM}) measured on the prompt neutron area is obtained with MCNP in a single calculation step described in reference [13]. On the other hand, the DT signal (S_{DT}) measured in passive mode is calculated with an AmBe point-source located in a flexible cable facing the drum (see Fig. 2) at mid-height of the drum.

The prompt calibration coefficient is carried out in two MCNP calculation steps, which is necessary to correctly model the fission rate (τ_f) and detection efficiency ε in a heterogeneous waste, and permits a large gain in computing time compared to a one-step calculation. The mesh grid defined above is used for calculations related to hull and nozzle cells (matrix). The same two-step calculation is performed

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