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Feasibility study of ^{235}U and ^{239}Pu characterization in radioactive waste drums using neutron-induced fission delayed gamma rays

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ABSTRACT

This paper reports a feasibility study of ^{235}U and ^{239}Pu characterization in 225 L bituminized waste drums or 200 L concrete waste drums, by detecting delayed fission gamma rays between the pulses of a deuterium-tritium neutron generator. The delayed gamma yields were first measured with bare samples of ^{235}U and ^{239}Pu in REGAIN, a facility dedicated to the assay of 118 L waste drums by Prompt Gamma Neutron Activation Analysis (PGNAA) at CEA Cadarache, France. Detectability in the waste drums is then assessed using the MCNPX model of MEDINA (Multi Element Detection based on Instrumental Neutron Activation), another PGNAA cell dedicated to 200 L drums at FZJ, Germany. For the bituminized waste drum, performances are severely hampered by the high gamma background due to ^{137}Cs, which requires the use of collimator and shield to avoid electronics saturation, these elements being very penalizing for the detection of the weak delayed gamma signal. However, for lower activity concrete drums, detection limits range from 10 to 290 g of ^{235}U or ^{239}Pu, depending on the delayed gamma rays of interest. These detection limits have been determined by using MCNPX to calculate the delayed gamma useful signal, and by measuring the experimental gamma background in MEDINA with a 200 L concrete drum mock-up. The performances could be significantly improved by using a higher interrogating neutron emission and an optimized experimental setup, which would allow characterizing nuclear materials in a wide range of low and medium activity waste packages.

1. Introduction

Radioactive wastes can be sorted depending on their activity and the time scale needed for their radio-toxicity to reach an acceptable level. This paper deals with the characterization of 225 L bituminized waste drums produced in France and 200 L concrete waste drums produced in Germany, in the frame of collaboration between the Nuclear Measurement Laboratory of CEA Cadarache and the Institute of Energy and Climate Research – Nuclear Waste Management and Reactor Safety of FZJ. The latter holds a PGNAA facility called MEDINA (Multi Element Detection based on Instrumental Neutron Activation) able to accommodate these drums, and for which performances have already been validated on reference matrices [1].

The bituminized waste is made of radioactive sludge, originating from spent fuel reprocessing operations, homogeneously incorporated in bitumen. It contains plutonium, americium, uranium, curium and various beta emitters, among which ^{137}Cs is responsible for a very high gamma radiation level. CEA has already studied a nondestructive measurement system to characterize these drums [2], including gamma radiography, gamma spectroscopy, passive neutron coincidence counting, and active neutron interrogation with the differential die-away technique (DDT). The estimation of the alpha activity was an important objective, the main contributions in these waste being due to ^{241}Am (measured by gamma spectroscopy), ^{239}Pu (measured by DDT, see hereafter), and ^{240}Pu (measured by passive neutron coincidence counting). The amount of fissile material can be estimated using the DDT and, by measuring prompt and delayed fission neutrons, it is also possible to separate the ^{239}Pu and ^{235}U contributions.

Another approach presented in this paper could be to detect fission delayed gamma rays emitted by fission products beta decays. This approach provides isotopic information on fissile

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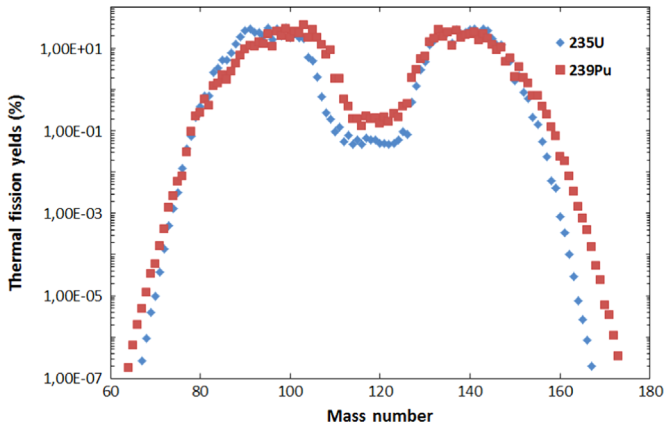

Fig. 1. Distribution of the cumulative thermal fission yields (fraction of fission fragment produced directly and via decay of precursors par 100 fissions induced by thermal neutrons) as a function of mass number, from JEFF3.1 database.

elements thanks to the differences between actinide fission yields illustrated in Fig. 1 for ^{235}U and ^{239}Pu. A significant difference can be observed in the 80–130 mass number region of the fission products. The possibility to quantify ^{235}U and ^{239}Pu using these differences was exposed through several studies [3,4], and especially in the case of radioactive waste packages [5]. In this work, measurements were first performed at the Nuclear Measurement Laboratory (CEA Cadarache) on metallic U and Pu samples to identify delayed gamma rays of interest for ^{235}U and ^{239}Pu differentiation. The corresponding delayed gamma yields have been estimated and compared to tabulated data. These yields are then used to calculate with MCNP the expected delayed gamma signal in case of 225 L drum filled with bituminized, or with a concrete matrix, using the MCNPX model of MEDINA [6]. Finally, the neutron activation gamma background has been measured in MEDINA with a concrete waste drum mock-up, in view to calculate ^{235}U and ^{239}Pu detection limits.

2. Delayed gamma ray measurements in REGAIN

The present study is based on the experimental results obtained in REGAIN graphite cell (Fig. 2) equipped with a 14 MeV D-T neutron generator (GENIE16 from SODERN) with a nominal emission of 10^8 n/s, a 30% relative efficiency n-type coaxial high-purity germanium (HPGe) detector with a transistor reset

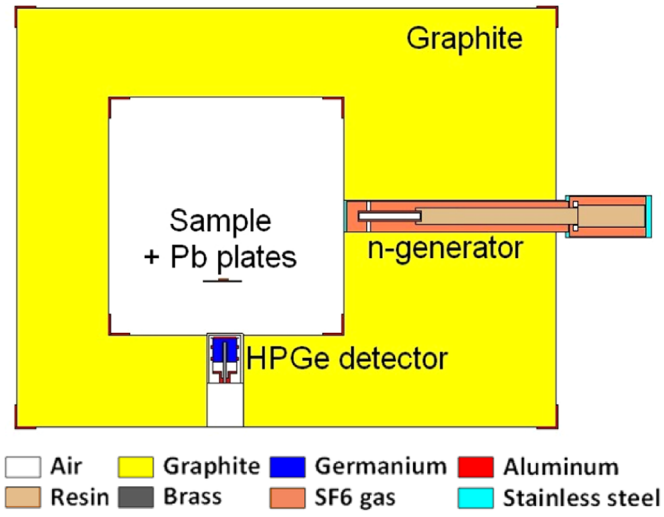

Fig. 2. MCNP model of the U and Pu samples measurements in REGAIN.

preamplifier (GR3018). A gamma spectroscopy analog electronics including a CANBERRA 2024 amplifier and 8715 Analog-to-Digital Converter (ADC) is used. The measurements were performed using two thin samples of metallic U and Pu enclosed in 55 mm × 20 mm × 1.15 mm zirconium alloy plates, placed at a distance of 12.5 cm in front of the detector (Fig. 2), and containing respectively ≈ 8.9 g of ^{235}U and ≈ 7.6 g of ^{239}Pu. A 4 mm thick lead plate was placed between the samples and the detector to lower the background associated to ^{241}Am gamma rays and to Pu and Am X-rays. The delayed gamma rays emitted by fission fragments with half-lives up to about 10 min are preferentially measured using a cyclic irradiation as shown in Fig. 3. The average neutron emission was 8×10^7 n/s during the 7200 s total cyclic irradiation time. Gamma rays are detected between two neutron pulses after a preset delay of 10 ms following the pulse, to avoid the detection of capture gamma rays. The average dead times (in fact count rate and dead time are not constant between the pulses) were determined using a ^{60}Co point source placed near the ^{235}U and ^{239}Pu samples, and were respectively about 4.4% and 14%, leading to an effective measurement time of 4130 s for the ^{235}U sample, and 3715 s for the ^{239}Pu. The delayed gamma rays following the radioactive decay of fission fragments having a half-life larger than 15 min are measured after irradiation, in a lead-shielded low background gamma spectrometer, after a one minute delay for sample transfer. In the shelter, the distances between the ^{235}U/^{239}Pu samples, the lead plate and the HPGe detector are the same as in REGAIN, thus guarantying the same geometrical efficiency than for the measurement in REGAIN. The detector used in this low-background spectrometer is a p-type 25% relative efficiency coaxial HPGe (EGPC25). The 46800 s post-irradiation measurement was segmented into 900 s cycles to check the half-lives of the identified gamma-ray emitters. Gamma-ray spectra are reported in Figs. 4 and 5. In order to identify possible interferences, the active background of the REGAIN cell was acquired for inter-pulses measurements. In addition, the passive backgrounds have been also recorded with the samples in REGAIN (Fig. 4) and in the low background spectrometer (Fig. 5). To optimize the signal-to-noise ratio in the post-irradiation measurements, each delayed gamma-ray net area is determined from the spectrum recorded with an acquisition time as close as possible to 6 times the radioactive period of the corresponding precursor. In case of isotopes with half-life larger than 7800 s, the longest 46800 s acquisition spectrum is used. Count rates are summarized in Table 1. For post-irradiation measurements, the dead time monitored by the electronics was lower than 1%. In Table 1, the relevance of the detected gamma-rays for ^{239}Pu/^{235}U discrimination is also reported, based on the ratio between the ^{239}Pu and ^{235}U emission rates, taking into account that the fission rates in these samples are similar (within 10–15% as estimated by experiment or by simulation, as explained further).

The relative uncertainties (1σ) mentioned in Table 1, given by Visugamma spectrum analysis software [7], are quite large for some peaks due to low statistics, poor signal-to-noise ratio (e.g. 830 keV, 952 keV, 973 keV gamma rays in Fig. 6, and 1032 keV Pu and 1614 keV gamma rays in Fig. 7), and interfering peaks (e.g. 952 keV and 1427 keV gamma rays in Fig. 6). One can note that the ^{235}U/^{239}Pu ratios are consistent with the ones measured in a similar experiment [8], e.g. 0.415 (±17%) for ^{89}Rb vs. 0.342 in [8], 0.462 (±4%) for ^{92}Sr vs. 0.486 in [8], 0.568 (±14%) for ^{94}Sr vs. 0.610 in [8].

3. Estimation of fission delayed gamma yields from REGAIN measurements

The count rate S^{REGAIN} (in s^{-1}) of a fission delayed gamma-ray