ENS PhD Award – Extended Summary of the PhD thesis

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Title of the PhD thesis: Development of a characterization method for concrete radioactive waste packages using photofission

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The nondestructive characterization of large concrete radioactive waste packages is a major challenge for nuclear safety and long-term management purposes. On the one hand, the fissile mass has to be estimated to retrieve, transport and store legacy wastes in the safest way. On the other hand, a precise knowledge of their alpha activity enables sending them towards the appropriate final repository, which implies significant cost savings. The 870 liters concrete drums are legacy wastes produced by CEA and stored in Cadarache Center, France. They cannot be characterized using passive nuclear measurements, nor by Active Neutron Interrogation. Therefore, we study Active Photon Interrogation to localize and estimate the fissile mass in these large concrete waste drums. Photofission of actinides can be induced with photons having an energy higher than 6 MeV. Since all actinides have similar photofission cross sections, the method cannot directly estimate the fissile mass (e.g. ^{235}U, ^{239}Pu) or the alpha activity (mainly Pu isotopes), and a discrimination method is needed to assess the proportions of the nuclides of interest. The aim of the PhD thesis is to contribute to the development of a method using the photofission technique in order to localize, differentiate and quantify actinides inside large concrete radioactive waste drums. The next sections will detail the developments conducted during the PhD.

In the first place, an extensive study of nondestructive nuclear measurement techniques was conducted in order to highlight the framework within which they can be used to characterize radioactive wastes. Passive methods relying on the analysis of spontaneously emitted radiations (like gamma spectroscopy or neutron counting) become insufficient for low-energy or low-intensity radiations, dense matrices or when the signal is masked by that of other emitters. In these cases, active methods requiring an external radiation source to induce fissions inside the radioactive waste drums are implemented. The aim is then to detect the particles emitted during fission reactions or by fission products to obtain information on the nature and amount of actinides inside the drum. These fissions can be induced via neutron irradiation (Active Neutron Interrogation) or high-energy photon irradiation (Active Photon Interrogation) usually with a *Bremsstrahlung* beam. In the case of 870 liters concrete waste drums, the use of Active Neutron Interrogation is not suitable given the high hydrogenous content of the package leading to penalizing uncertainties in the fissile mass estimation. Therefore, Active Photon Interrogation coupled to the detection of delayed gamma rays emitted by fission products has been studied during this PhD in order to characterize the actinide content inside these large concrete packages.

All actinides can undergo photofission when irradiated with photons of energy greater than 6 MeV and the reaction threshold is similar for all isotopes of uranium and plutonium. In contrast to active neutron interrogation with thermal neutrons, the photofission technique does not allow to directly estimate the mass of fissile material (e.g. ^{235}U, ^{239}Pu…) since all actinides have similar photofission cross sections. Accordingly, a differentiation method was developed during this PhD in order to estimate the proportion of fissile and fertile nuclei. To that extent, the differences in photofission products yields

according to the actinide were exploited via the measurement of photofission products delayed gamma-ray ratios with a high-resolution germanium detector. To that degree, photofission yields of the actinides of interest must be known precisely. Even though nuclear data related to photofission yields of ^{238}U exist, they sometimes present significant discrepancies, even in recent studies. Photofission yield data are even scarcer for fissile isotopes such as ^{235}U and ^{239}Pu, hence the need to perform new measurements during this PhD.

Two experimental campaigns were conducted by using a *Bremsstrahlung* photon beam produced by a Saturne LINAC located in the CINPHONIE facility at CEA Cadarache. The LINAC could accelerate electrons up to 21 MeV. A lead collimator was used to focus the photon beam on the actinide samples, surrounded by a shield made of polyethylene to limit the photoneutron flux (neutrons produced by photonuclear reactions in the materials) at the samples, and thus to minimize neutron fissions. The photon flux at the output of the collimator aperture being not known precisely, nor the endpoint-energy of the *Bremsstrahlung* spectrum expected around 16 MeV, a characterization of the photon beam produced by the LINAC has been performed using photon activation of reference materials and complementary simulation work with the MCNP 6.1 Monte-Carlo code. The photon fluxes were estimated as well as the photon beam end-point energy, being respectively 15.8 MeV [1] and 17.5 MeV [2] for the first and second experimental campaigns. The characterization of the photon beam enabled to perform an evaluation of the photoneutron flux by means of MCNP simulations and neutron activation analysis of irradiated indium pellets. All these characteristics of the photon and neutron beams cross validated via experiments and simulations were used for the measurement of photofission products cumulative yields.

During the first experimental campaign that occurred in 2019, depleted and highly enriched uranium samples were irradiated to measure photofission products yields of ^{238}U and ^{235}U, respectively. The second campaign took place in 2021 and a more massive highly enriched uranium sample as well as a plutonium sample were used besides the depleted uranium sample used in 2019. As a result, this second campaign enabled to measure photofission products yields of ^{235}U, ^{239}Pu and ^{238}U. The method used in both campaigns is similar. The samples were irradiated with the *Bremsstrahlung* beam during two hours and moved in front of a high-purity germanium detector afterwards to perform high-resolution spectroscopy on delayed gamma rays emitted by fission products. The MCNP code was used to estimate the photofission and neutron fission rates in the samples. The MAGIX software developed by CEA List was employed to analyze the spectra and estimate gamma-ray peak areas [3] which were used to estimate the cumulative yields of various photofission products. In the framework of the first campaign, the cumulative yields of 49 and 26 photofission products were measured, respectively for ^{238}U and ^{235}U. Among these, 8 yields for ^{238}U and 3 yields for ^{235}U were unpublished so far. The second experimental campaign enabled to measure 28 photofission product yields for ^{238}U and 26 for both ^{235}U and ^{239}Pu, of which 4 had no record in the literature for ^{239}Pu.

The photofission products yields measured during the second experimental campaign were used to develop a differentiation method to discriminate uranium and plutonium isotopes undergoing photofission reactions. A differentiation information can be obtained between uranium or plutonium isotopes thanks to their different photofission product yields, through the measurement of the relative intensities of specific delayed gamma lines emitted by photofission products having different yields for the different actinides. The proportion of the actinide(s) of interest in the mixture, can thus be computed from the ratio of two net peak areas and from the ratios of the photofission yields of their two emitting photofission products. The objective is to select specific photofission products pairs showing the largest differences, according to the actinide, in their gamma ratios. The most efficient photofission product couples to consider were identified for different actinide mixtures and the experimental demonstration was performed on bare actinide samples.

In the case of actinides inside a matrix, as it is the case for radioactive waste packages, one must consider matrix effects resulting in the attenuation of radiations. To address this issue, the localization

actinides is necessary in order to perform the differentiation of actinides and later their quantification, as well as to estimate the associated uncertainties. Experiments with a concrete matrix of 1 meter diameter and uranium samples were conducted during this PhD to develop and test a localization method [4]. The technique presented in this work uses the ratio of two gamma rays emitted by the same photofission product (^{138}Cs) to exploit the differential attenuation of its gamma rays, when crossing different thicknesses of concrete, that is to say in two different measurement positions. The localization combines experimental data with MCNP simulations performed with the MCNP code. As a result, a theoretical distortion map of the gamma-ray ratio is obtained via simulation and compared to the experimental ratio distortion in both positions of the matrix to retrieve the location of the actinides. The technique showed encouraging results on uranium samples and opened prospects to use gamma rays that are not necessarily close in energy for actinide identification, contrary to the methods previously developed. The localization technique was also used in combination with the differentiation method mentioned before. The latter showed interesting results in the case of a massive uranium sample inside the matrix since its enrichment could be estimated. In the case of a low mass of uranium located in the matrix, only a qualitative enrichment range could be evaluated. For future work, the selection of photofission couples efficient for differentiation of actinides should be re-evaluated by including the energy and intensity of their gamma rays as a selection criterion when projecting forward to the analysis of actinides inside a dense matrix.

The final part of this PhD consisted in studying the detection performances and limitations associated with the Active Photon Interrogation characterization method. First, the detection limits of uranium in the concrete matrix used for the experiments were evaluated. Then, in order to offer a more realistic insight, the detection limits were estimated by simulating an 870 liters waste drum model combined with the analysis of an experimental active background spectrum related to the concrete matrix. At best, detection limits of 144 g of ^{238}U and 45 g of ^{235}U at the center of the 870 liters waste package (i.e. at 50 cm depth) have been evaluated for a high-energy gamma ray emitted by the ^{84}Br photofission product. Previous work conducted in the same laboratory concluded that the Active Neutron Interrogation method could not provide any viable signal arising from actinides when located in the center of an 870 liters waste drum. Therefore, the work conducted during this PhD highlights the importance of the development of Active Photon Interrogation for such radioactive packages. At the end, perspectives were drawn to improve detection limits in the future. Notably, efforts could be directed to lessen the active background by limiting the activation of the detector components and shielding. Another possibility to explore seems to be the detection of gamma rays of energy greater than 3.4 MeV which are only emitted by fission products. To that extent, fast and efficient scintillation detectors could be used between LINAC pulses to measure high-energy gamma rays emitted by short-lived photofission products. This could be used to perform a package scanning and provide localization information on potential hot spots inside the package. Further developments using multiple germanium detectors and a Bayesian framework and a combination of various imaging techniques could also be of particular interest for characterizing 870 liters radioactive waste packages.

References

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