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#7-63 Determination of uranium and plutonium isotopic composition with medium resolution gamma-ray detector

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Determination of uranium and plutonium isotopic composition is a key challenge in several application fields of the nuclear industry: MOX fuel fabrication, safeguards, dismantling and decommissioning (D&D) applications, reprocessing plants. Isotopic composition measurements are generally carried out using a High Purity Germanium (HPGe) detector, which requires cooling, imposing constraints in terms of cost, deployment, and maintenance. The measurements are then analyzed using a dedicated software such as the IGA, MGA, or FRAM industrial codes.

The goal of this study is to determine the isotopic composition of plutonium and uranium by analyzing medium resolution spectra obtained using CZT and LaBr3 detectors. The measurements were conducted using a Polaris gamma camera from H3D (H420 model) and two LaBr3 detectors of different sizes. At 120 keV, there is a factor of four between the CZT camera resolution (2.3 keV) and the HPGe resolution (0.5 keV) and a factor thirteen between the LaBr3 resolution (6.8 keV) and the HPGe resolution.

MAGIX-UPu, a code developed by CEA LIST, is based on the automatic analysis of X and gamma rays emitted by the different radionuclides contained in the material to be analyzed, especially the isotopes of plutonium and uranium. The energies of the peaks in the spectrum indicate which isotopes are present, and the number of counts in the peaks provides information on the respective quantity of each isotope, allowing the isotopic composition to be calculated.

The main challenge arises from the complexity of the uranium and plutonium spectra to be analyzed, due to the medium resolution of the CZT and LaBr3 detectors. Despite these difficulties, the objective of this study is to determine whether it is possible to determine the isotopic composition from medium resolution gamma spectra.

The analysis of a spectrum with MAGIX-UPu consists of several stages: the fine energy and resolution calibration, the region fitting to extract peak areas and the simultaneous estimation of relative efficiency and mass fractions of the isotopes.

The study of uranium spectra was conducted on several enrichment levels of 235U, between 0.3% to 9.6% and the study of plutonium spectra on several samples, with 239Pu fractions contained between 69% to 94%.

An isotopic composition was determined from the uranium spectra provided by the two LaBr3 detectors, but a specific analysis had to be developed based on the size of the detectors. A notable impact of counting statistics related to the distance between the source and the detector was observed. For the plutonium samples, isotopic composition results were also obtained (Deviations between 239Pu mass fractions obtained by the software and the reference are less than 20%), which is a significant achievement considering the degraded resolution of this family of detectors compared to an HPGe semiconductor. It should also be noted that, the analysis starting at 129 keV, several plutonium isotopes can only be characterized by analyzing a single peak, which poses a significant constraint in data processing. As with the germanium detector, analysis performance is also affected by the fractions of 239Pu content of the sample (the lower the fraction, the more the performance of the code is degraded).

The results obtained from the analysis of spectra provided by the Polaris H420 gamma camera from H3D are much better, mainly due to the less degraded energy resolution of the CZT detector compared to that of a LaBr3. The mass fractions of 239Pu can be obtained with a deviation of less than 1% compared to the reference value. The results for uranium spectra are also convincing, although the tested enrichment range does

not exceed 9.6%.
It is important to note that the industrial perspectives for this new code are good, as the use of medium resolution detectors reduces the constraints associated to the deployment of an HPGe detector.

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