

Investigation of radioisotope source masking effects on uranium gamma-ray spectra in determining of ^{235}U abundance in uranium samples

Abstract

In nuclear technology, one key parameter is the uranium enrichment of the uranium material for the characterization. For quick and accurate determination of uranium enrichment at border check-points to prevent illegal trafficking and contraband of special nuclear materials, such as uranium or plutonium, gamma-ray spectrometry is a fast and cheaper one than other analytical tools due to its ease of use, portability, non-destructive nature for determination of the isotopic uranium abundance. From point of nuclear material security in the nuclear security activities, it is well known that gamma-ray spectrometry a versatile technique and it allows analysts or FLOs(front of lines) or first responders on-site to measure and identify the radiation sources and materials. However, the uranium enrichment necessitates the use of specifically designed instrumentation and methods. In this study, for the correct identification of depleted uranium(DU), natural uranium(NU) and low enriched uranium(LEU) samples, the masking effects of the medically/industrially used radioisotopes have been investigated on uranium gamma-ray spectra, which were taken by LaBr₃:Ce, CdZnTe and HPGe detectors, respectively. In practice, especially in transportation of radioactive materials or radioisotope sources, the nuclear materials can be masked by other radioisotopes such as ^{241}Am , ^{57}Co , ^{133}Ba , ^{152}Eu , $^{99\text{m}}\text{Tc}$ and other medically used radioisotopes. In such circumstances, the presently available algorithms used for the de-convolution of the peaks in the acquired gamma-ray spectra have some deficiencies to de-convolute the close lying peaks and thus the calculated peak areas for the analytically used peaks can result in erroneous results in the uranium enrichment determination.

In this study, two intermediate energy resolution detectors that are a 38.1mmx38.1mm LaBr₃:Ce scintillator with a resolution of FWHM=2.7%@662 keV(^{137}Cs) and a 15mm x 15mm x 7.5mm CdZnTe semiconductor with a resolution of FWHM=2.5%@662 keV(^{137}Cs) and a high resolution(FWHM=0.575keV@122 keV(^{57}Co)) portable, LN₂ cooled and HPGe (its crystal has 37.7mm in diameter and 16.4 mm in thickness) were used in the experiments. As a uranium material, depleted, natural and low enriched uranium(U₃O₈) samples having the enrichment of about 0.32 to 4.51% atom ^{235}U certified reference materials (EC-NRM171 sealed cans) obtained from EU-JRC IRRM were used. From the acquired uranium spectra, the enrichment values of the samples were determined two different analytical approaches: one is uranium enrichment meter principle(EMP) that uses 185.7 keV peak of ^{235}U and the other is multi-group gamma ray analysis (MGA) that uses very closely appeared x-ray and gamma-rays, which are lying in 80-130 keV low energy region of uranium spectrum. In the experimental setup parameters, each detectors was installed in a radiometric bench in which the uranium source sample and the detector was aligned with a laser light and measured the source-to-detector distances. In each detector setup, firstly, lead collimators and interleaved absorbers(aluminium and iron) between sample and detector were placed to observe the effects on the accuracy ^{235}U isotopic abundance in uranium samples. Then, at a given source-to-detector geometry, the different radioisotope sources (such as ^{241}Am , ^{57}Co , ^{133}Ba , ^{152}Eu , $^{99\text{m}}\text{Tc}$, ^{232}Th) are, in turn, counted together with uranium sample in a sealed aluminium can to simulate the masking conditions, where the chosen radioisotope source was, respectively, placed at the lateral sides, back side and front side of it in order to observe the scattering effect of uranium in the Al-can. The spectra analyses of the uranium materials masked by other radioisotope sources were made COLEGRAM and with a newly developed spectrum de-convolution algorithm based on the modified Figure-of Merit (m-FOM) under MATLAB platform. This algorithm can be easily applicable to EMP. In MGA analysis, U-235View(Ortec) and Genie MGAU (Canberra) commercial softwares were used. These programs are applicable to the uranium gamma-ray spectra taken by a high resolution HPGe detector. The differences between the calculated enrichment values and the reference values of ^{235}U at different worst measuring conditions were generally found to be less than 15%. The differences become much lower in favorable conditions such as long counting time, ideal detector-sample distance. However, in case of the uranium spectra masked by other radioisotope source or materials, the interpretation of discrepancies in the ^{235}U enrichment results will be discussed in detail in the presentation.

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