

Identification of RBMK and VVER spent nuclear fuel batches by means of HGRS in course of forensic examination

This paper gives a study of the high resolutions γ ray spectrometry (HRGS) capabilities to define seized spent nuclear fuel (SNF) origin during its forensic examination in the first 24 hours of investigation. Keeping in mind the multicomponent nature of the spent nuclear fuel, during test method development attention was paid on the following set of nuclides: cesium-134, cesium-137, ruthenium-106, cerium-144, antimony-125, europium-154, europium-155, uranium, plutonium, americium and curium.

With this purpose, a detailed gamma-spectrometric study of different VVER and RBMK SNF samples was carried out. Spectral analyses of these samples were made by means of two different instruments: HPGe planar photon detector system GLP-36360/13P4 and coaxial one GEM35P4-76.

One of the greatest problems in SNF spectral analysis by means of HRGS is a continuous background signal in wide area of the spectra which is induced by γ -ray emission of fission products (first of all ^{137}Cs). In the course of this study was found that HPGe planar detector is better for the task on hands then coaxial one. It is due to the difference of their intrinsic efficiency curves. It was found that U or Pu γ -ray emission signal can be reliably detected if ^{137}Cs signal intensity is not more than ~ 600 times greater. This estimation has been made for HRGS spectral data with statistical uncertainty per channel below 3%.

According to calculated relative γ -emission intensities almost all γ -spectral lines of actinides in spectra of normal SNF batches are completely suppressed by background γ -emission of fission products or by spectral interferences. The same situation remains even after long cooling period of 30 years and more. There are only a few exceptions: ^{241}Am , ^{243}Am and ^{243}Cm . Unfortunately, measured (current) activities of ^{241}Am , ^{243}Am and ^{243}Cm are hard to use for SNF identification. In real practice ^{241}Am presence in γ spectrum is good enough only for confirmation of seized sample's nature as SNF. If SNF cooling period is known an up-to-date decay adjusted $^{137}\text{Cs}/^{243}\text{Am}$ activity ratio can be used to evaluate SNF burn-up. A very promising fingerprint signature of very high burn-up SNF batches is a $^{137}\text{Cs}/^{243}\text{Cm}$ activity ratio. The main advantage of this ratio is that it is almost independent towards SNF cooling period. It is because half-life periods of these nuclides are very close to each other and after longest possible SNF cooling period of about 60 years this ratio shifts only for $\sim 5\%$.

Another case is a sample of abnormal SNF batch, which accidentally undergo partial separation of actinides and fission products. Uranium and plutonium nuclide γ -ray signals can be directly detected and measured in γ -spectra of such samples without any sample preparation. Abnormal SNF batches has a great significance as potential objects of forensic examinations, because during INES 6 and INES 7 class incidents many fragments of these batches can become out of regulatory control. In the present study γ -spectra of such abnormal SNF batches was obtained and successfully used to determine their $^{232}\text{U}/^{235}\text{U}$ isotope ratios.

In case of SNF the majority of γ -ray emission comes from γ -emitting fission products that can be also referred as radionuclide markers. These radionuclides can be used itself to determine origin of seized SNF sample. In the present study activities of the following nuclides were measured in SNF samples: ^{125}Sb , ^{134}Cs , ^{137}Cs , ^{154}Eu and ^{155}Eu . Activity ratios of these nuclides can be used as fingerprint signatures. Among all of these ratios the most useful one for SNF batch identification is a $^{137}\text{Cs}/^{134}\text{Cs}$ activity ratio. There is a common practice to measure this ratio for every SNF batch after the end of its irradiation to evaluate fuel burn-up value. Due to this practice, initial value of this ratio is known for every SNF batch. So, this value can be taken from nuclear power plant archival records, up-to-date decay adjusted and used as a perfect fingerprint signature to identify seized SNF sample's origin. If current measured value of seized sample's $^{137}\text{Cs}/^{134}\text{Cs}$ ratio is equal to corresponding up-to-date decay adjusted initial ratio of some SNF batch it can be stated, that this batch is an origin of seized sample. Unfortunately, this method cannot be applied to other activity ratios of mentioned above radionuclides because there is no common practice to measure their initial activity ratios for every SNF batch after the end of its irradiation.

In conclusion of the IAEA project No18989 a test method for RBMK and VVER SNF identification by means of HGRS was developed and proposed. This test method could serve as a guideline for HGRS composition analysis and data interpretation in case of nuclear forensic examination of seized spent nuclear fuel samples.

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Track Classification: MORC: Nuclear forensics