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How to Measure Fissile in Spent Fuel

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1. Introduction

Since the 2000s, the generation, management, and disposal of spent nuclear fuel have emerged as social problems, and with the international development of sustainable fourth-generation nuclear reactors, interest and research on the recycling of spent nuclear fuel have been actively conducted. In particular, Korea Atomic Energy Research Institute (KAERI) developed DUPIC (Direct Use of PWR Spent Fuel In CANDU) technology as part of the development of nuclear fuel cycle, followed by an international joint study with the United States on the development of pyro-linked SFR (Sodium Fast Reactor) technology. The measurement of nuclear material in the development of nuclear fuel cycle and management of spent nuclear fuel plays a very important role in enhancing the safety and transparency of nuclear material. In addition, international safeguards must be satisfied through nuclear material measurement, and it has become a very important factor in detecting the transition and diversion of nuclear material.

Therefore, various methods for measuring nuclear material have been studied internationally. Recently, technical verification of methods such as Tripple bubbler, Voltametry, Actinide sensor, LIBS (Laser Indirect Break-down System), Mass Spectrometer (IDMS), and Chemical Analysis is being performed for the measurement of nuclear material in the pyro process using spent nuclear fuel. In addition, Los Alamos National Lab (LANL) published a report on the characteristics of various technologies for nuclear material measurement through radiation detection[1]. Recently, KAERI also performed technical analysis and simulation on various nuclear material measurement methodologies for the application on the current pyro process and monitoring of nuclear material, and technical possibility for the application. In addition, recently, for nuclear material safeguards, efforts have been made to derive continuous and reliable information on nuclear material through the development of an integrated nuclear monitoring system[2,3].

2. Measurement Technologies

In order to analyze the content of nuclear material in spent fuel assembly and bulk type in fuel cycle process, simulations on various non-destructive radiation measurement technologies were performed and evaluated. The passive method of measuring gamma rays and neutrons naturally generated from spent nuclear fuel was mainly used for nuclear measurement. In general, measurement of spontaneous fission neutron by Cm244 is internationally well known method to derive the content of plutonium in TRU and spent fuel. However, the direct assay of isotopic plutonium is limited and plutonium assay is indirectly obtained from the help of burnup code and chemical analysis. Major passive measurements include PNAR (passive neutrino reactivity), SINRD (self-interrogation neutron resistance density), Cs137, anti-coincidence, and X-ray fluorescence[1].

Active measurements using external sources include prompt neutron measurement, neutron resonance transmission analysis (NRTA), lead slow down spectrometer (LSDS), differential die-away (DDA), delayed neutron (DN), and delayed gamma (DG) technologies[1]. Some of them have dominant feature to assay isotopic fissile material, however, others have limitation in obtaining distinguished isotopic signal. These methodologies also show difficulties in analyzing the content of each isotope in spent fuel assembly or large scale bulk type fuel, such as U235 and Pu239, which are the main targets for analysis. However, chemical analysis shows high accuracy in the isotopic content analysis, but it requires an analysis procedure, needs a large amount of samples for different location, and there are limitations in real-time analysis. However, in some measurement technologies, assay of isotopic fissile content was obtained from several fuel rods.

3. Radiation Measurements

For Cm coincidence measurement, PNAR using albedo theory, and CIPN(Cf252 interrogation with prompt neutron) using californium neutron source, which are mainly used for neutron measurement, effective plutonium content is obtained from further analysis[1]. When uranium and plutonium isotopes exist in the sample, effective plutonium is expressed like below.

$$^{239}\mathrm{Pu}_{\mathrm{eff}} = C_1 M_{\mathrm{U235}}, + M_{\mathrm{Pu239}} + C_2 M_{\mathrm{Pu241}} \qquad (1)$$

where C_1, and C_2 represent relative contribution of U235 and Pu241 to Pu239 fission. In DDA, which measures induced prompt neutron in fissile fission, plutonium fission has a feature having 1.3 times greater than that of uranium in thermal energy. Therefore, $\mathrm{Pu}_{\mathrm{effective}}$ in thermal energy region is expressed like

$$^{239}\mathrm{Pu}_{\mathrm{eff}} = C_1 M_{\mathrm{U235}}, + M_{\mathrm{Pu239}} + C_2 M_{\mathrm{Pu241}}. \qquad (2)$$

C_1 represents delayed neutron of U235 per unit mass/delayed neutron of Pu239 per unit mass and C_2 represents delayed neutron of Pu241 per unit mass/delayed neutron of Pu239 per unit mass.

NRTA is using resonance energy of nuclear material. Resonance is an intrinsic feature of isotopic fissile material[4]. Fissile material has different resonance characteristics. The transmitted radiation is expressed like below

$$I = I_o(E)\exp[-\Sigma\ (N\sigma(E))] \qquad (3)$$

where N is nuclear content, σ(E) is reaction probability with neutron. NRTA can assay isotopic nuclear material, because each fissile material has its own resonance. It has advantage on fuel rod base sample. However, intense neutron source is required for large nuclear sample, ~E12n's/sec. In addition, NRTA technique has very limited application in water. For spent fuel case, cooled down fuel is more adoptable. Fig. 1 represents characteristics of fissile material, U235, Pu239, and Pu241 based on rod. However, for spent fuel assembly assay, self-shielding and scattering effect must be considered.

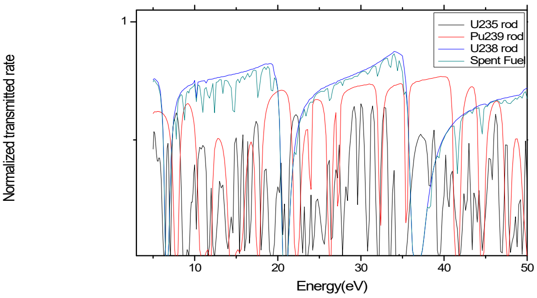


Fig. 1. Neutron transmission on nuclear material.

LSDS technology is also using neutron resonances of fissile material and has advantage to distinguish fission signal from isotopic fissile[5]. Pu239, Pu241 and U235 have different neutron reaction in resonance region. Fig. 2 represents the total neutron cross section for U235, Pu239 and Pu41 in epi-thermal and fast resonance energy region. The fissile fission occurs by interrogation neutron.

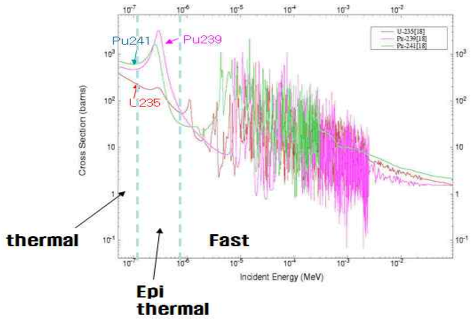


Fig. 2 Total neutron cross section.

The detected signal of induced fission neutron is expressed as

$$D = (N\boldsymbol{\sigma_f}(E)\nu(E)\phi(E))_{235} + (N\boldsymbol{\sigma_f}(E)\nu(E)\phi(E))_{239} + (N\boldsymbol{\sigma_f}(E)\nu(E)\phi(E))_{241}\,. \qquad (4)$$

N is the content of U235, Pu239, Pu241, and $\boldsymbol{\sigma_f(E)}$ is fission cross section, and $\nu(\boldsymbol{E})$ is fission neutron and ϕ(E) is source neutron. LSDS has prominent feature to analyze isotopic fissile material, specially for U235, Pu239, Pu241. However, for analysis of fuel assembly type, intense neutron source is also required. Fig. 3 shows the resolution of neutron in LSDS system.

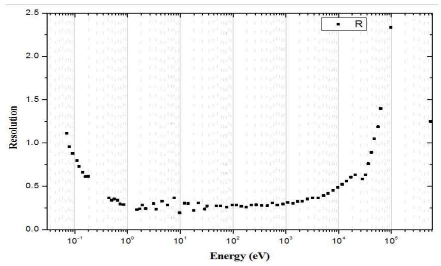


Fig. 3. Energy resolution in LSDS system.

In the gamma ray measurement, anti-coincident technology has advantage to detect hidden peaks in the Compton scattering region[6]. However, spent fuel emits very intense gamma rays below cesium peaks, there is limitation in plutonium measurement, even though Compton area is degraded by using anti-coincidence technique. As anti-coincidence technique is applied, normal peaks are also tend to be reduced with Compton scattering reduction. Therefore, if several fission products, emitting intense gamma rays, are removed, anti-coincidence will be operated. The simulation on the detection signal was performed using the MCNP code[7]. The detected signal is simply expressed as

$$\varepsilon \int_t \int_E h(E)\ \varphi\ (r,E,t)\ dEdt, \qquad (5)$$

where h(E) is energy deposition at the detector, φ is the photon arriving at the detector and ε is detector efficiency. Fig. 4 represents the detector configuration for Compton suppression.

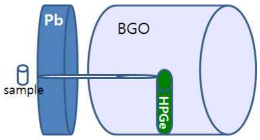


Fig. 4. Detector configuration for Compton suppression.

DDA and DN can be applied together in the fissile assay. As shown in Fig. 5, detected signal is classified by prompt and delayed neutron, as time elapse. DDA and DN are utilized for the fissile assay in the process waste and salt nuclear waste.

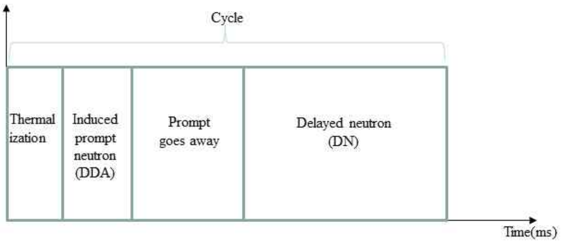
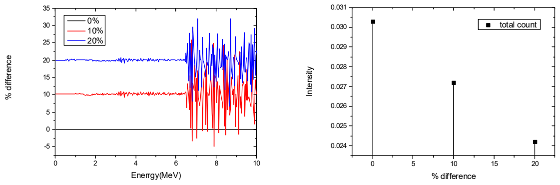


Fig. 5. Characteristics of DDA and DA.

In addition, neutron measurement was examined for applicability of monitoring fissile material in the spent fuel process. In the pyro-process, volatile and semi-volatile fission products are extracted and it is disposed as a waste. Co and Zr in structure and cladding material, which emit intense gamma rays, are also removed. TRU, rare-earth(RE) and some noble metal nuclei are theoretically existing in salt. The simulation was done for the existing materials in electro-reduction process by measuring neutron. The content of nuclear material was changed up to 20%. Fig. 6 represents detected neutron signal. The detected signal was normalized to reference. The content change was clearly appeared and the monitoring of process operation could be possible.



a) neutron energy b) total neutron

Fig. 6. Neutron signal difference.

4. Conclusion

Many technologies have been performed and evaluated for nuclear material management of spent fuel. Radiation measurement is well known and effective way to obtain nuclear material information. In special, NRTA and LSDS has advantage on isotopic fissile assay. However, for assembly type assay, very intense neutron source is required. If several intense gamma emitters are removed before the main fuel cycle process, it is advantage on the application of gamma ray measurement for plutonium analysis. In addition, the neutron measurement could monitor the fissile and the movement of fissile material could be clearly detected. However, the uranium detection was very limited in the spent fuel.

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