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Statistical uncertainties of nondestructive assay for spent nuclear fuel by using nuclear resonance fluorescence

Toshiyuki Shizuma [a,*], Takehito Hayakawa [a], Christopher T. Angell [a], Ryoichi Hajima [a], Futoshi Minato [b], Kenya Suyama [b], Michio Seya [c], Micah S. Johnson [d,e], Dennis P. McNabb [d]

[a] Quantum Beam Science Directorate, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
[b] Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
[c] Integrated Support Center for Nuclear Nonproliferation and Nuclear Security, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1198, Japan
[d] Lawrence Livermore National Laboratory, 7000 East Ave. Livermore, CA 94550, USA
[e] Department of Physics and Astronomy, San Jose State University, One Washington Square, San Jose, CA 9519, USA

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ABSTRACT

We estimated statistical uncertainties of a nondestructive assay system using nuclear resonance fluorescence (NRF) for spent nuclear fuel including low-concentrations of actinide nuclei with an intense, mono-energetic photon beam. Background counts from radioactive materials inside the spent fuel were calculated with the ORIGEN2.2-UPJ burn-up computer code. Coherent scattering contribution associated with Rayleigh, nuclear Thomson, and Delbrück scattering was also considered. The energy of the coherent scattering overlaps with that of NRF transitions to the ground state. Here, we propose to measure NRF transitions to the first excited state to avoid the coherent scattering contribution. Assuming that the total NRF cross-sections are in the range of 3–100 eV b at excitation energies of 2.25, 3.5, and 5 MeV, statistical uncertainties of the NRF measurement were estimated. We concluded that it is possible to assay 1% actinide content in the spent fuel with 2.2–3.2% statistical precision during 4000 s measurement time for the total integrated cross-section of 30 eV b at excitation energies of 3.5–5 MeV by using a photon beam with an intensity of 10^6 photons/s/eV. We also examined both the experimental and theoretical NRF cross-sections for actinide nuclei. The calculation based on the quasi-particle random phase approximation suggests the existence of strong magnetic dipole resonances at excitation energies ranging from 2 to 6 MeV with the scattering cross-sections of tens eV b around 5 MeV in ^{238}U.

1. Introduction

Nondestructive assay (NDA) of fissionable nuclei is a key technology in nuclear material management concerning nuclear security and safeguards. A method using nuclear resonance fluorescence (NRF) with an external photon source has been proposed as a promising NDA technique [1] and identified as a potential technology for nuclear safeguards [2–5]. In the NRF method it is possible to assay specific nuclear material by detecting a characteristic NRF signal emitted from the isotope of interest.

The NRF measurement becomes efficient by using the mono-energetic photon beam produced by the laser Compton scattering (LCS) method [6,7]. This has been demonstrated in recent nuclear physics measurements [8,9] and application studies [10–13]. Owing to the recent progress in electron accelerator and high-power laser technologies, enhanced capabilities of the LCS photon beam for flux and brightness are expected. Research projects toward the generation of an intense photon source are being carried out worldwide. An LCS photon source based on the modern technology of an X-band normal conducting linac, MEGa-ray at Lawrence Livermore National Laboratory (LLNL), is under construction for nuclear security applications [14]. An extended MEGa-ray system will be installed at the Extreme Light Infrastructure Nuclear Physics (ELI-NP) facility for photo-nuclear science and its applications [15]. In addition, an LCS photon source based on a superconducting energy-recovery electron linac (ERL) has been proposed [7]. A test facility of the ERL-LCS photon source is being built by a collaborative effort of Japan Atomic Energy Agency (JAEA) and High Energy Accelerator Research Organization (KEK). Such intense LCS photon sources are also useful for hybrid K-edge X-ray fluorescence densitometry (HKED) [16].

One of the most important issues for NDA is to establish accurate technique for assay of ^{239}Pu in spent fuel. In fact, in the next generation safeguards initiative of United States, Department of Energy, the NDA of Pu is the first priority in technological development. Hayakawa et al. [17] have proposed an NRF-based

* Corresponding author.
E-mail address: shizuma.toshiyuki@jaea.go.jp (T. Shizuma).

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NDA system utilizing an ERL, and have estimated the measurement accuracy of Pu by using the Monte Carlo simulation code GEANT4 extended so as to handle the NRF processes [10]. It has been concluded that 1% fraction of ^{239}Pu in a fuel assembly can be detected with a statistical error lower than 2% within a measurement time of 4000 s using a high flux LCS photon beam based on the ERL.

The performance of the NRF-based NDA system, such as statistical uncertainties, is affected by radiation background from spent fuel as well as background from coherent scattering such as Rayleigh, nuclear Thomson, and Delbrück scattering. In this paper, we present results of analysis of the NRF intensity and the backgrounds from both the spent fuel and the coherent scattering. We also discuss the NRF strength based on previous experimental data as well as theoretical predictions based on the quasi-particle random phase approximation (QRPA).

2. Detection system

We have previously proposed an NDA system with an intense LCS photon source based on the ERL accelerator technology [17]. In the ERL system, electrons are accelerated by the time-varying radio-frequency (RF) field stored in a superconducting linac, and are subsequently transported to a recirculation loop. The electron beam retains low emittance, which is important for the generation of an intense LCS photon beam with a small energy width. Such a photon beam can be generated by combining the ERL and a high power mode-locked laser [7].

The heat loaded from the radioactive decay of fission products requires cooling the spent fuel in a water pool. While neutrons and low-energy X rays are scattered and absorbed by the cooling water, high energy photons can penetrate it and reach the spent fuel. Scattered γ-rays from actinide nuclei in the spent fuel are measured with a multi-detector array system consisting of several high-purity germanium (HPGe) detectors placed outside the water pool.

Fig. 1 shows a schematic view of the detection system consisting of 24 HPGe detectors. Six sets of four HPGe detectors are placed at $\pm 120^\circ$, $\pm 130^\circ$, and $\pm 140^\circ$ with respect to the incident LCS photon beam. These detector angles are different from those used in our previous simulation study [17]. The backward angles are chosen to reduce coherent scattering contributions (see Section 3.3). The energy resolution of HPGe detectors is typically better than 0.2% (full width at half maximum (FWHM)). The distance between the detector and the fuel assemble is about 50 cm. The solid angle is defined to be $\approx 1.7 \times 10^{-2}$ sr by a lead collimator placed in front of each HPGe detector. In addition, lead γ-ray filters with a thickness of about 5 cm are placed between the detector and the fuel assembly. These filters optimize counting rates of the detectors by effectively absorbing photons with energies lower than 1 MeV. More details of the detection system are presented in Ref. [17].

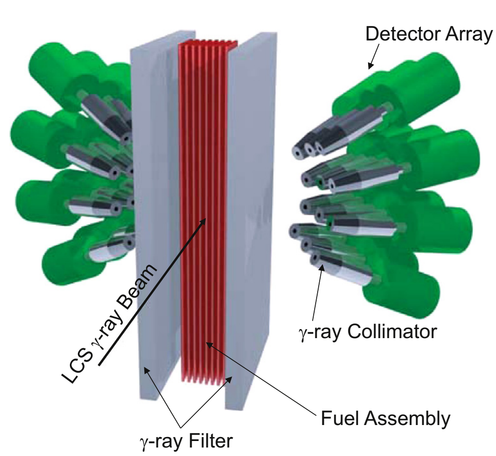

Fig. 1. Schematic view of the detection system comprising of 24 HPGe detectors. Two sets of 12 HPGe detectors are placed at both sides of the fuel assembly.

3. Estimation of counting rates and precision

3.1. NRF counting rates

NRF is a process of resonant excitation of nuclear levels by absorption of photons and subsequent de-excitation to lower-lying levels by γ-ray emission. It occurs if the energy of the incident photon is identical to the resonance energy of the nucleus of interest. The energy and angle integrated NRF cross-section is given [18] as

$$I_s = 2g\left(\frac{\pi\hbar c}{E_\gamma}\right)^2 \frac{\Gamma_0 \Gamma_i}{\Gamma} \tag{1}$$

where E_γ is the resonance energy, and Γ, Γ_0, Γ_i are the total decay width, the ground-state decay width, and the partial decay width to the final state, respectively. The spin factor g is defined as $(2J_1+1)/2(2J_0+1)$ with J_0 and J_1 being spins of the ground state and the excited state, respectively. By measuring the energies of the NRF γ-ray peaks, nuclear species can be identified. In addition, the isotopic concentration can be determined by measuring the intensity of the NRF peak in the γ-ray spectrum. In the following discussion, we restrict ourselves to consider dipole excitation in even-even nuclei for simplicity.

The most probable states observed in actinide nuclei are those with $J^\pi = 1^\pm$ [19]. It follows that de-excitations to either the $J^\pi = 0^+$ ground state or the $J^\pi = 2^+$ first excited state are primarily observed. Assuming pure dipole nature for both transitions to the first excited and ground states, the ratio R of the reduced transition probabilities B is given by

$$R = \frac{B(1^\pi \to 2^+)}{B(1^\pi \to 0^+)} = \frac{\Gamma_1}{\Gamma_0}\left(\frac{E_0}{E_1}\right)^3 \tag{2}$$

where Γ_0 and Γ_1 represent the decay width to the ground and excited states, and E_0 and E_1 are the corresponding transition energies. Since the first excited states are observed at excitation energies between 10 and 50 keV in actinide nuclei (*e.g.*, 45 keV for ^{238}U, 44 keV for ^{238}Pu, and 8 keV for ^{239}Pu [20]), *i.e.*, $E_0 \approx E_1$ MeV, Eq. (2) can be reduced to

$$R \approx \frac{\Gamma_1}{\Gamma_0}. \tag{3}$$

On the other hand, according to the Alaga rules [21], R equals to 1/2 for $K=1$ or 2 for $K=0$ where K is the projection of the total angular momentum vector of the resonant state on the nuclear symmetry axis. Therefore, Γ_0/Γ and Γ_1/Γ values in Eq. (1) become 2/3 and 1/3 for $K=1$ or 1/3 and 2/3 for $K=0$, respectively.

In practice, the NRF yield is obtained from the following equation:

$$Y_{\rm NRF} = \phi \times I_s \times N_t \tag{4}$$

where ϕ is the intensity of the incident LCS photon beam, and N_t is the number of the target nucleus. In the present estimation, we assume that the diameter of the fuel rod is 10 mm and the density of the spent fuel is 10.96 g/cm^3. The spent fuel includes 1% plutonium of which the isotopic ratios are 52% for ^{239}Pu and 48% for ^{240}Pu. Assuming $\phi = 10^6$ photons/s/eV, the NRF yield is calculated using Eq. (4).