Simulation of an Interrogation Method for Radioactive Waste by Using Nuclear Resonance Fluorescence

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Prior to disposal of the radioactive waste, it is necessary to evaluate the radioactivity of radionuclides. The purpose of our work is to design and evaluate a non-destructive detection system by using nuclear fluorescence resonance (NRF) reactions. In this work, some simulations are performed with a Monte Carlo simulation code. The simulation result showed that a peak of the scattered photons is satisfactorily large. It is concluded from the result that the NRF method can be applied to the radioactive waste management. This paper describes an energy-recovery linac (ERL) based gamma-ray source and the simulation results.

I. INTRODUCTION

Recently, interrogation systems that exploit nuclear resonance fluorescence (NRF) to detect specific isotopes were proposed.[1, 2] Since the NRF by photons depends on excitation levels of a radio isotope, we can identify a radio isotope by detecting scattered photons which have a discrete energy distribution unique to the isotope. The interrogating photons, which range from 1 to 3 MeV, are the most penetrating probes. We propose that the detection method be used for management of radioactive wastes.[3] The non-destructive interrogation method by using the NRF make it possible to measure the discrimination of clearance levels of a concrete solidification radioactive waste.

Prior to disposal of the radioactive waste, it is necessary to evaluate the radioactivity of radionuclides, which contain transuranic or fissile radionuclides. In this process an indirect method such as the scaling factor method (SF method) is used for evaluating 'difficult-to-measure' nuclides. However, the correlation is an unknown factor between 'easy-to-measure nuclides' that are readily measured nondestructively and 'difficult-to-measure' nuclides such as the pure β radionuclides. As other direct method, fission reactions by neutrons or photons are used for evaluating the amount of uranium and plutonium. However, we can not identify nuclear species by these assay techniques.[4]

The purpose of our work is to design and evaluate of the NRF-based detection system for the radioactive waste. For this evaluation, we are developing a Monte Carlo simulation code by using the GEANT4 code.[5] Some simulations have been performed with simple models of a concrete solidification radioactive waste drum and detectors. The simulation results showed that a peak of the scattered photons is satisfactorily large. As a result, it is found that the NRF method can be applied to the radioactive waste management. In this paper, we present a recent result of the simulation.

II. HIGH-FLUX GAMMA-RAY SOURCE

Performance of the NRF-based detection systems depends sensitively on the properties of the beam of interrogating photons. The high-flux and monoenegetic gamma-ray source that we consider here is Compton backscattered gamma-rays. In the gamma-ray sources high intensity laser light collides head on with a relativistic electron beam. This electron beam is produced by an energy-recovery linac (ERL). The ERL is a promising device for next-generation X-ray light sources, because ERL is able to generate an electron beam of high-average current with ultimately small emittance.

Table I shows the system parameters of the laser and electron beam.[6] A laser supercavity increases the laser power to 3000 times and makes a small laser beam size at the collision point with the electron beam. Total flux of the laser Compton gamma-ray is obtained by

$$F = \frac{f N_e N_L \sigma_C}{A} \qquad (1)$$

where f is the collision frequency, N_e is the number of electrons in an electron bunch, N_L is the number of photons in a laser pulse, σ_C is the cross section of Compton scattering, A is effective sectional area of beams at the collision point and given by $A = \pi w^2/2$ for a Gaussian bean. In the case of gamma-ray production by laser photons of 1064 nm wavelength and electron energy of 350 MeV, the maximum energy of gamma-ray is found to be 2.2 MeV, and the scattering cross section can be approximated by Thomson scattering cross section: $\sigma_C = (8\pi/3)r_e^2$, where r_e is the classical electron radius.

With the above assumption, the gamma-ray flux is estimated at 2.7×10^{10} ph/sec,[6, 7] which is enhanced by 8-9 orders of magnitude from the existing facilities. This significant enhancement of the gamma-ray flux is due to the higher density of electrons and photons at the collision point, which is accomplished by modern accelerator and laser technologies.

TABLE I. Parameters of the ERL gamma-ray source

Electron beam	
Repetition	130 MHz
Energy	350 MeV
Bunch charge	100 pC
Normalized RMS emittance (x/y)	2.5 / 1.0 mm-mrad
RMS beam size at the collision (x/y)	37 / 24 μm
Pulse length (RMS)	3 ps
Energy spread (RMS)	0.03 %
Laser	
Repetition	130 MHz
Wave length	1064
pulse energy	1.8 μJ
RMS size at the collision	30 μm
pulse length (RMS)	2 ps
Enhancement of supercavity	3000

We have also made a numerical estimation of gamma-ray flux by using a simulation code CAIN,[8] which is a FORTRAN Monte Carlo code for the interaction involving high energy electron, positron, and photons. In the numerical simulation, we investigated the effect of oblique collision and inhomogeneous distribution of laser photons and electrons, which are neglected in the analytical formula.

Figure 1 shows a gamma-ray spectrum obtained by

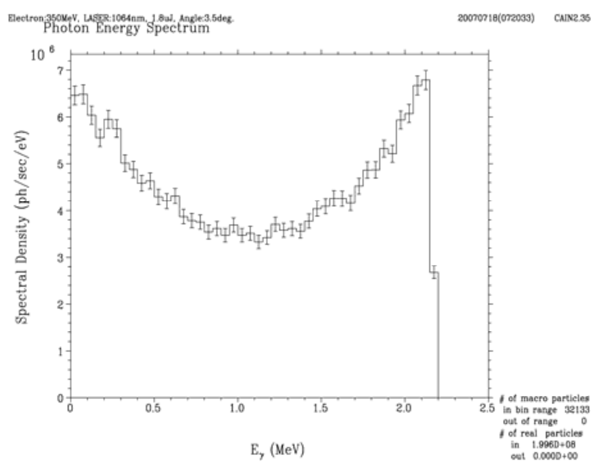

Fig. 1. Numerical simulation result by the CAIN.

CAIN with parameters listed in Table I. The gamma-ray flux near the peak energy E_γ ~ 2 MeV is F = 0.7×10^{10} /sec/keV for a crossing angle of 3.5 degree.

To realize the gamma-ray source based on the ERL, exhaustive R&D on its key technologies is required. These R&D include developing an electron gun that can produce a high average beams of 100 mA with extremely low emittance, a high-power drive laser for the electron gun, and superconducting cavities that can accelerate high-current beams under CW operation.[9, 10, 11]

III. SIMULATIONS

III.A. Simulation model

In this study, some simulations were performed with a GEANT4-based Monte Carlo simulation code. GEANT4 is widely used in applications of high energy, nuclear and accelerator physics, as well as studies in medical and space science. However, nuclear resonance fluorescence is not supported in the original package of GEANT4. We have, therefore, modified GEANT4 to calculate nuclear resonance fluorescence in cascade interactions of photons and particles triggered by incident gamma-rays.

The NRF resonances have peak cross sections given by $2\pi G(\lambda/2\pi)^2$. Here, λ is the photon wavelength and G is the statistical factor $(2J_1+1)/(2J_0+1)$ for a transition from the ground state J_0 to the excited state J_1.[2, 12] The peak cross sections are very large and in the range of 1500 barns for 2.176 MeV photons in a J_0 = 0 to J_1 = 1 transition. Another important factor is the radiative width, Γ, of these excited states to the ground state. For NRF states of interest, Γ is 0.058 eV. The cross section observed in an experiment is broaden by thermal motion of the nucleus, which is Doppler shift. For a typical case of radioactive nuclide, the Doppler width becomes ~0.5 eV.[3] This energy broadening is still small enough in comparison with energy resolution of experimental apparatus.

III.B. Detectors layout of the detection system

We assumed that the radio active waste drum is a concrete solidification uniformly and a bulk density of 2 g/cm^3. The value of the specific radioactivity of ^{238}U is 1000 Bq/g. The specific flux of the gamma-rays which do not scatter in the drum of density ρ at depth x, $F(x)$, can be calculated using the equation:

$$F(x) = F_0 \exp(-\mu_m \rho x) \qquad (2)$$

where F_0 is the incident flux of the gamma-rays and the μ_m is the gamma mass attenuation coefficient. For 2 MeV photons, the flux at the center of the drum is attenuated to

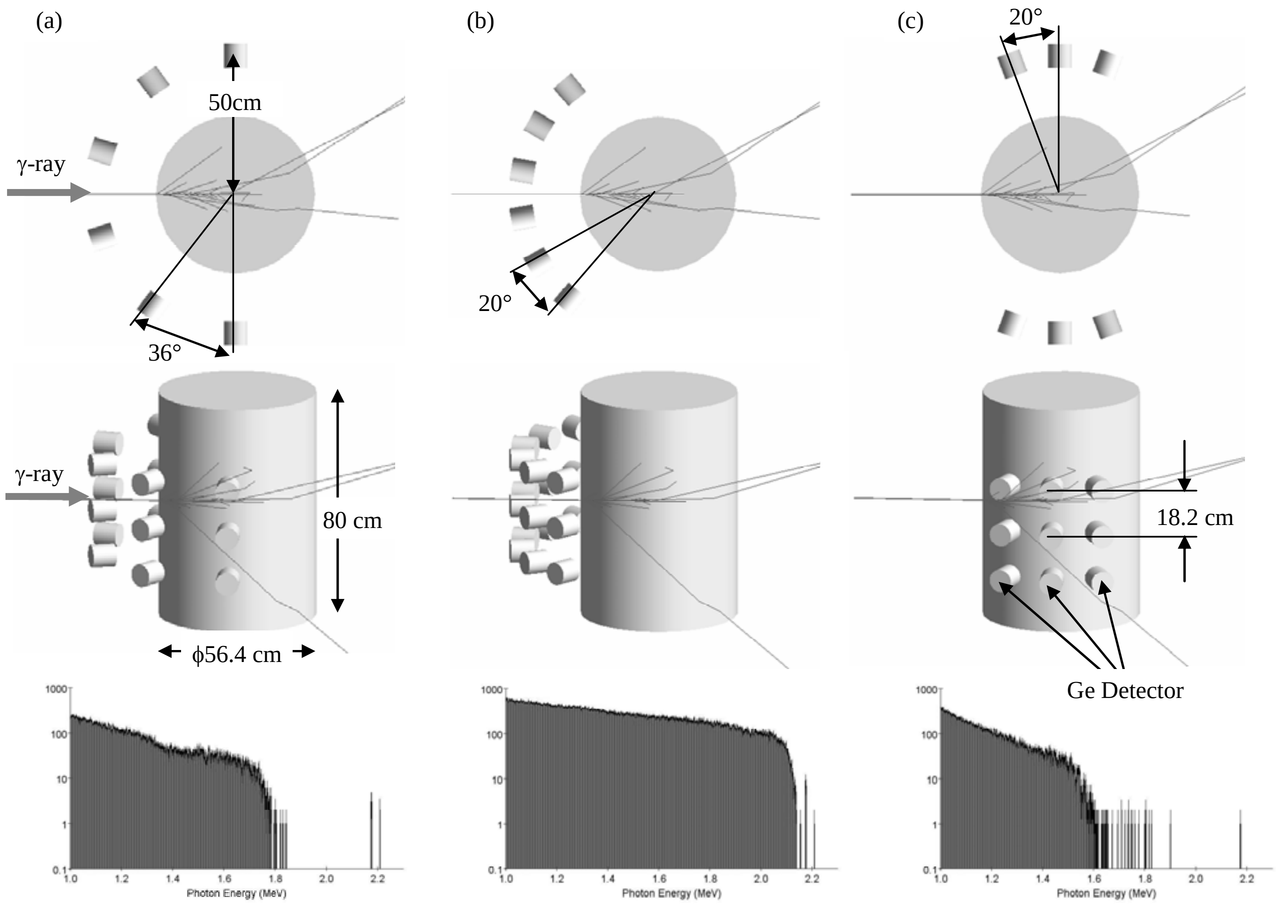

Fig. 2. Three cases of the geometry of the 200-liter drum and the detection system. Incident and scattered gamma-rays are indicated. The gamma-ray energy spectrums are shown for each case.

about 10 %. Since the majority of NRF reactions take place between surface and the center of the object, the detection sensitivity improves by locating the detectors backward.

Three cases of the geometry of schematic outline of the detection system for the simulation are shown in Fig. 2. In the simulation, 18 Germanium detectors are located 50 cm away from the drum center. Assuming the detector size of ϕ84mm×85mm, we can estimate the geometrical efficiency about 3 %. The energy and energy spread of the irradiation gamma-rays are 2.176 MeV and about 1.5% (FWHM), respectively. We assume ^{238}U at a concentration level of 1000 Bq/g is contained in a 200-liter drum and the NRF cross section is 28 mbarn-keV. In this case, the number of gamma-rays is 1×10^9 which is equivalent to irradiation time of about 3 msec.

A large part of background noise seen in the detector signal is due to Compton scattering from nuclei inside the drum, and these scattered gamma-rays have energy lower than the incident gamma-ray.

In the simulation, we can not see the 2.176 MeV line from ^{238}U. Because the number of irradiation gamma-rays is not able to be increased because of the computation time, it is assumed that the value of NRF cross section is enlarged to 100 mbarn-keV in the simulation after here. We select case (a) about the location of the detectors because of influence of inelastic scattered gamma-rays.

III.C. ^{238}U detection by NRF method

We consider detection of ^{238}U by the NRF method. We assume ^{238}U at a concentration level of 1 Bq/g is contained in a 200-liter drum, and the drum is irradiated by gamma-ray at a flux of F = 10^{10} /sec/keV. Energy-resolved gamma-ray detectors, which have a geometrical efficiency of ε = 3%, are prepared for the gamma-ray detection, and the energy of the gamma-ray is tuned at a NRF resonance of ^{238}U, 2.176 MeV. Using these assumptions, we can estimate the number of NRF gamma-rays detected by the detectors for ^{238}U of 1 Bq/g:

$$N_\gamma = \varepsilon \sigma L \frac{\rho}{A_s} \frac{N_A}{238} FT \qquad (3)$$

where σ is a NRF cross section, L a interaction length, ρ a density of material, A_s specific activity, N_A Avogadro constant, T a irradiation time. Putting numbers in to the equation, $\sigma = 100$ mbarn-keV, $L = 0.56$ m. $\rho = 2$ g/cm^3, $A_s = 1.2\times10^4$ Bq/g, $F = 1\times10^{10}$, $T = 10$ sec, we have $N_\gamma = 7\times10^3$.

In the Monte Carlo simulation, we assume ^{238}U at a concentration level of 1000 Bq/g is contained and cross section is 100 mbarn-keV. In this case, the number of gamma-rays is 2×10^9 which is equivalent to irradiation time of about 6 msec and the energy spread of the gamma-rays is about 1.5% FWHM. Figure 4 shows the energy spectrum of the scattered gamma-rays into the Ge detector regions. The spectrum clearly shows the 2.176 MeV line from the ^{238}U with a very low background in spite of the large volume of the drum. The number of NRF gamma-rays is found to be $N_\gamma = 230$ for two bins in total. The number of NRF photons measured by the detectors in the simulation is about 6 % of the analytical estimation by Eq.(3). This difference can be explained by a fact that the analytical estimation ignores inelastic scattering of gamma-rays inside the drum, which decreases gamma-ray population within the resonance width of the NRF.

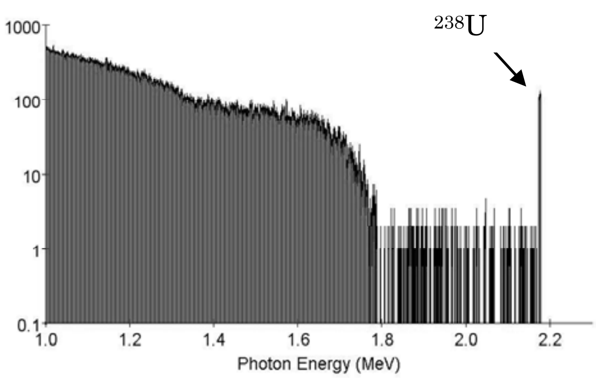

Fig. 3. A result of GEANT4 simulation. The sharp peak corresponds to the NRF signal from ^{238}U.

III.D. Identification of radioisotopes

We considered identification of two or more radioisotopes by the NRF method. It is assumed that the ^{238}U and ^{232}Th at each concentration level of 1000 Bq/g are uniformly contained in the drum. In the simulation, irradiation photon energy is uniform random numbers between 2.0 to 2.2 MeV and the number of gamma-rays is 2×10^9 which is equivalent to irradiation time of about 1 msec. NRF cross section of ^{238}U and ^{232}Th is 100 mbarn-keV respectively. Figure 4 shows the energy spectrum of the NRF signals by ^{238}U and ^{232}Th. The spectrum clearly shows the 2.176 MeV line from the ^{238}U and 2.043 MeV line from ^{232}Th.

The number of NRF gamma-rays is found to be 26 counts of ^{238}U and 123 counts of ^{232}Th in total. The ratio of the number of the NRF signals is larger than the existence ratio (8 wt% of ^{238}U and 25 wt% of ^{232}Th). This difference can be explained by a fact that the gamma-ray population increase within the lower resonance level by inelastic scattering of higher energy photons.

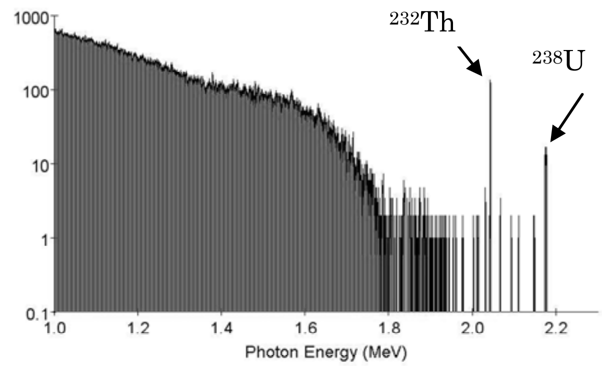

Fig. 4. A result of GEANT4 simulation. We can see the peaks corresponding to the NRF signal from ^{232}Th and ^{238}U.

IV. DISCUSSIONS

The simulation result suggests that detection of ^{238}U by NRF method is possible in principle. A large part of background noise seen in the detector signal is due to Compton scattering from nuclei inside the object to measure, and these scattered gamma-rays have energy lower than the incident gamma-ray. Thus, the signal-to-noise ratio in the NRF-method can be significantly improved by choosing the incident gamma-ray energy equal to the NRF energy of target nuclides. Laser Compton scattering is the most suitable to provide quasi-monochromatic energy-tunable gamma-rays for this purpose.

If the difference of resonance energy for two nuclides is larger than the resolution of experimental apparatus, the simulation result suggests that we can clearly distinguish two nuclides by using gamma-rays of appropriate energy spread. Adjustment of the energy spread of the gamma-rays is possible by putting a collimator at downstream of the collision point. In our gamma-ray source, the energy spread of the gamma-rays obtained by a collimator of 1 mm in diameter at 10 m from the collision point is found to be $\Delta E/E = 10$ % (full width).[6]

The yield of NRF signal is proportional to the number of incident gamma-rays, the number of nucleus in the irradiation region, and the NRF cross section. The NRF cross section has the same order of magnitude for all radionuclides. The detection limit, thus, becomes lower for a long-lived nuclide at small concentration level.

Separation of background noise and NRF signals may be improved using non-isotropic nature of gamma-ray emissions by irradiation of polarized gamma-rays. We have started intensive numerical studies for the polarized gamma-rays.

Although it is necessary to increase the number of incident particles to calculate by a more detailed model, it is impossible because of the computation time. To improve the calculation speed, we have started to develop a parallelized simulation code for the optimization of the NRF gamma-ray assay system.

V. CONCLUSIONS

A nondestructive assay system for radioactive nuclides has been proposed. Some simulations have been performed with simple models of a concrete solidification radioactive waste drum and detectors by Monte Carlo simulations. The simulation result showed that a peak of the NRF signals is satisfactorily large. As a result, it is found that the NRF method can be applied to the radioactive waste management.

Our results indicate that a nondestructive gamma spectrometric method can replace the more time-consuming radiochemical technical traditionally used in measuring “difficult-to-measure” radionuclides.

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