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Gamma-ray measurement for classification of fuel debris and waste

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Abstract. The detection of prompt fission gamma-rays and Tl-208 decay gamma-rays, which are unique to nuclear materials, could be applied to sorting nuclear fuel debris and radioactive waste from the Fukushima Daiichi Nuclear Power Plant. To confirm its technical feasibility, particle transport calculations using the Monte Carlo N-Particle code were performed.

1 Introduction

In the decommissioning of the Fukushima Daiichi Nuclear Power Plant, the retrieval and safe containing/storage of fuel debris—comprising the solidified mixture of melted nuclear fuel and other materials generated by the accident—is under consideration. The materials to be extracted from the Primary Containment Vessel (PCV) include not only the fuel debris but also melted structural elements such as gratings and support pillars to which some nuclear fuel is attached. Currently, no technology exists to non-destructively measure and estimate the amount of nuclear fuel material contained within fuel debris and other melted materials. Depending on the accuracy of the estimation, this technology could be used to determine the properties of the retrieved objects and to sort fuel debris from radioactive wastes. Therefore, under the direction of Tousou Mirai Technology Co. Ltd, various Japanese organizations are conducting simulation analyses, tests, and developing estimation algorithms for the active neutron method, passive neutron method, muon scattering method, X-ray CT method, and passive gamma-ray method, evaluating their applicability to fuel debris and other materials. These technologies will ultimately be important for IAEA safeguards. In the case of uranium (U)-containing waste from fuel cycle facilities, used in the IAEA safeguards verification, the U amount may be determined by the detection of gamma-rays at 1001 keV emitted by Pa-234m, a daughter nuclide of U-238 [1]. However, it has been pointed out that such gamma-ray measurement techniques cannot be used for fuel debris that contains many fission products (FPs), which emit various energies of gamma-ray.
In this study, we conduct a feasibility study of the passive gamma-ray method, focusing on the spontaneous prompt fission gamma-rays (PFGs) directly emitted from nuclear materials. PFGs exist in a higher energy region than that of FPs and can avoid the interference by them. Moreover, Tl-208 decay gamma-rays are also taken into consideration. We aim to measure these gamma-rays in simple ways.

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1.1 Prompt fission gamma ray

During the nuclear fission of heavy elements and for a very short time thereafter, various types of radiation are emitted. One of them is prompt gamma radiation. Conventional activation analysis is also well known as prompt gamma-ray analysis (PGA), which analyses elements or isotopes by the specific peak of prompt gamma-rays from (n, γ) reaction, for example, which are emitted within 10^{-14} seconds after the neutron capture. When applied to heavy elements such as U and plutonium (Pu), they also have specific gamma-ray peaks from (n, γ) reactions, which are classified as prompt gamma-rays, however, most of them exist in the high-energy region above 4 MeV and the emission rates are low, which makes measurement difficult. PFGs have higher emission rate and are emitted forming a continuous energy spectrum whose peak is around 300 keV and which continues over 10 MeV. Therefore, we can observe a very clear spectrum only in the energy range of interest where there are no disturbing energy peaks of FPs. One of previous studies by Nauchi measured PFGs in a system of U-Al fuel loaded on a KUCA-C with a HPGe detector [2]. In the case of absence of steel, PFGs with a descending trend toward 6 MeV on the logarithmic axis were clearly observed in the region higher than the 2.2 MeV capture gamma-ray of H-1 and lower than the capture gamma-rays of surrounding structural materials at about 6 MeV.

1.2 Tl-208 decay gamma ray

It was suggested by Shiba et al. that Tl-208 decay gamma ray at 2615 keV is emitted in large numbers from nuclear materials [3]. Figure 1 illustrates how Tl-208, commonly known as natural background, could be present in nuclear materials. The figure is a simplified chart of the nuclides, where arrows for α and β decays are shown as the same length. Since the gamma rays of Ac-228 were not observed in the experiment of [3], which signifies the present Tl-208 is not from Th-232, U-236 or Pu-240. Note that the red lines in the figure indicate that even if the nuclides are present, it is unlikely to be ancestors of Tl-208 due to their long half-lives. The reason presumes Pu-236 existence. The reference also supports this hypothesis [4]. Moreover, in an example of gamma-ray measurement of a dry cask containing spent fuel, Ziock et al. [5] observed the Tl-208 peak at 2615 keV.

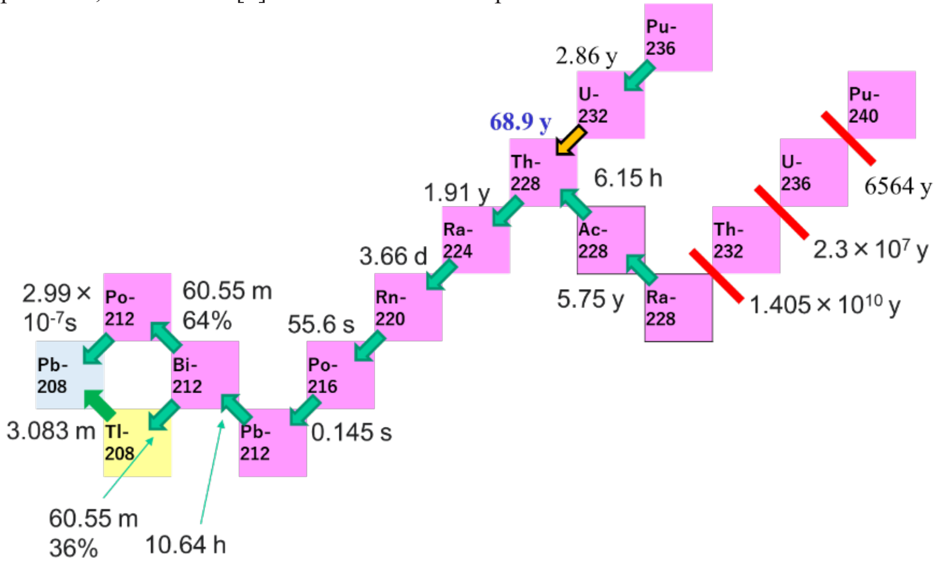

Fig. 1. Simplified chart of the nuclides

2 Simulation

Simulations were performed to measure PFG and Tl-208 decay gamma ray in a system with debris in a storage can. The encapsulation model was referred to the literature [6]. The model consists of a SS storage can of thickness of 5 mm. The debris consists of nuclear material, iron, zircaloy, and oxygen. The debris may be surrounded by water if debris and water are encapsulated together in a can at the time of removal. A lead collimator of 2 cm in length and width and 40 cm in length is cut outside the storage can. A gamma-ray detector was placed outside of the collimator to measure gamma rays; although LYSO was used as the detector in the literature [3], there was a desire to evaluate the peak region of Tl-208 and the PFG region closer together in the high-energy region around 2.5-3 MeV, where statistics do not accumulate easily. In addition, the separation is poor because of the proximity of the peak region of Tl-208 to the captured gamma ray of 2.2 MeV by H-1 that often appears in the said region with LYSO. Based on the above, a HPGe was used as the detector in this study (GC2020-7600-2002C, CANBERRA).

We used the Monte Carlo N-Particle Transport Code, Version 6 (MCNP6) to simulate the detector response of the leakage gamma rays from the can. We used MCPLIB04 as the photon library and el03 as the electron library. F8 pulse height (PH) tally along with the Gaussian energy broadening option in MCNP6 was used to simulate the response of the HPGe detector. In this simulation, the ratio between PFG emission from spontaneous fission by nuclear materials and Tl-208 decay gamma ray emission were set to be about 5600:1.

In order to confirm the geometrical sensitivity of the gamma ray at 2615 keV by the decay of Tl-208, the detectability in different geometries was investigated. The geometries employed are shown in the figures 2-4. Figure 2 shows the case when the debris is dry. The geometries are: extremely centered (1-a), extremely along the can circumference, or cylindrical (1-b), and all equally mixed, or homogeneous (1-c). Figure 3 shows the wet case where debris and water were encapsulated together in a can at the time of removal, and the aforementioned cases (2-a) through (2-c) were investigated in the same manner, respectively. Figure 4 shows the case where nuclear material, structural materials, and water were encapsulated in a checkerboard pattern without homogeneously mixing. Note that the blank area is supposed to be filled with air throut the figure 2-4. All of these patterns correspond to those reported by Nagatani et al [7].

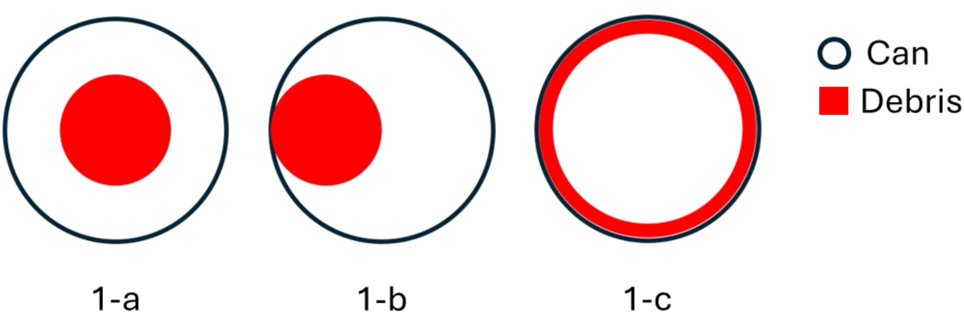

Fig. 2. Geometrical configuration for dry cases

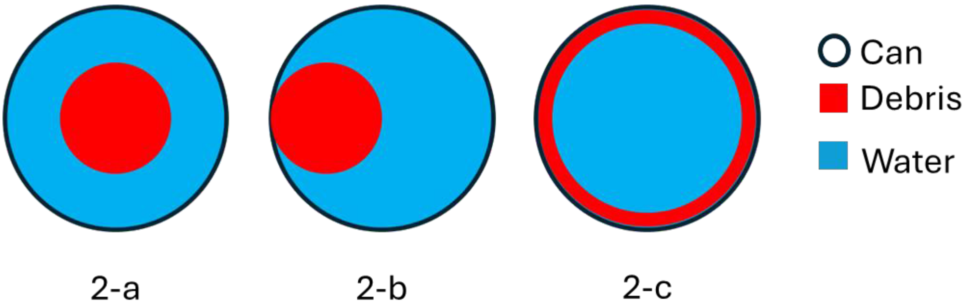

Fig. 3. Geometrical configuration for wet cases

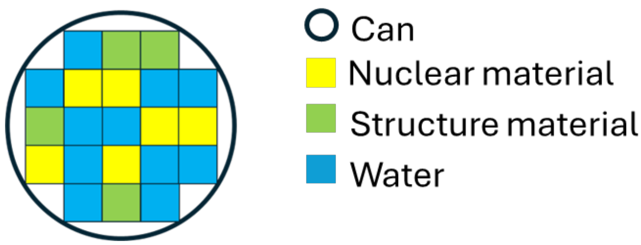

Fig. 4. Geometrical configuration for checkerboard pattern (hereafter referred as 3)

3 Result and discussion

An example gamma-peak result from the case 1-a is shown in figure 5. The Tl-208 peak was clearly observed; the peak was enough sharp that the PFG, which forms the baseline gamma continuous, were found to be low and do not interfere the Tl-208 peak. Note that this is an MCNP simulation result and the unit of pulse height is arbitrary.

The peak net counts of Tl-208 gammay ray at 2615 keV derived by the geometrical sensitivity analysis are shown in Table 1. Like in the figure 5, gamma rays of 2615 keV were clearly observed in all cases, and the difference between the highest and lowest pulse heights was within 300%.

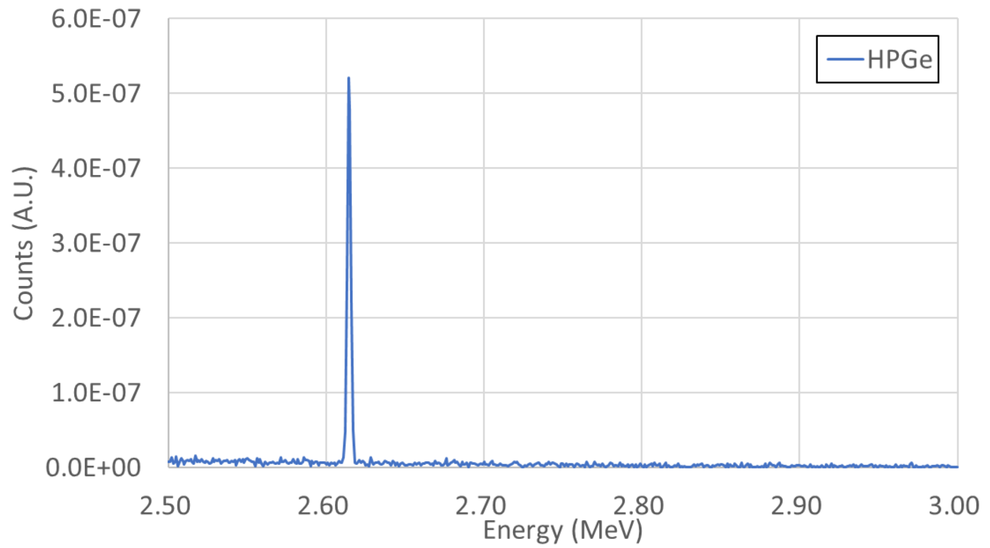

Fig. 5. Detector response by PFG and Tl-208 decay gamma ray

Table 1. Peak net counts comparison

Case	Net counts
1-a	5.13E-06
1-b	6.86E-06
1-c	1.30E-05
2-a	3.50E-06
2-b	4.04E-06
2-c	7.36E-06
3	6.28E-06

4 Conclusion

In this paper, it was shown that the detecting decay gamma ray of Tl-208 and PFG from a storage can could be useful to know nuclear material existence. Simulation of the detection of those gamma rays was performed to examine the feasibility of this approach. In the future, the validity of the simulation will be examined through actual measurement experiments using real spent fuel, and then propose a sorting method for debris and waste using these gamma-ray detection. Ultimately, we will aim to propose this technique for IAEA safeguards verification.

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