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ARTICLE

Application of beta coincidence to nuclide identification of radioactive samples contaminated by the accident at the Fukushima Nuclear Power Plant

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We proposed a new method to determine the activity of each nuclide in a mixture of Cs-134, Cs-137 and Sr/Y-90. The point of this new measurement method is simultaneous counting between beta and gamma ray detection. We assembled a setup that employs plastic and NaI(Tl) scintillation detectors for beta- and gamma-ray detection, respectively. The obtained result shows that discrimination of Cs-134 and Cs-137 was successfully achieved. Radioactivity determined by measurement was in agreement with one prepared as a mixture. The results show that the method can be adapted to measure the radioactivity of samples contaminated by the accident at the Fukushima Daiichi Nuclear Power Plant.

Keywords: beta ray; coincidence; nuclide determination in a mixture of Cs-137 and Cs-134; beta emitters; plastic scintillation detector; NaI(Tl) scintillation detector; Fukushima Daiichi Nuclear Power Plant accident

1. Introduction

At present, a large amount of radioactive materials is known to have been released over a widespread area by the accident at the Fukushima Daiichi Nuclear Power Plant. Cs-134 and Cs-137 were declared as radionuclides of concern based on the results of the actual investigation because other detected nuclides such as radioactive iodine have already decayed [1]. Sr/Y-90 is also concerned.

Measurement by gamma-ray spectrometry was required to protect the general public from non-negligible exposure. A high-purity germanium detector is often adapted for gamma-ray spectrometry because of its preeminent energy resolution. For pure beta radionuclides such as Sr/Y-90, complicated chemical treatment to separate the nuclide creates a bottleneck for sample measurement. These are some of the concerns over current radioactivity measurement.

We propose a new method to measure each radioactivity in a mixture of Cs-134, Cs-137, and Sr/Y-90. The points of this method are (1) to use simultaneous counting between beta and gamma rays for discrimination of Cs-134 from Cs-137, and (2) to apply the $4\pi\beta$–γ coincidence counting method for absolute determination of the beta-ray emission rate. Radioactivity of pure beta emitters such as Sr/Y-90 is deduced by subtract of contribution of Cs-134 and Cs-137, which is deduced from gamma-ray spectrometry, from all activities.

In this paper, we present a prototype experimental setup and its results. The intention is to demonstrate the detection method as a start to development. The setup

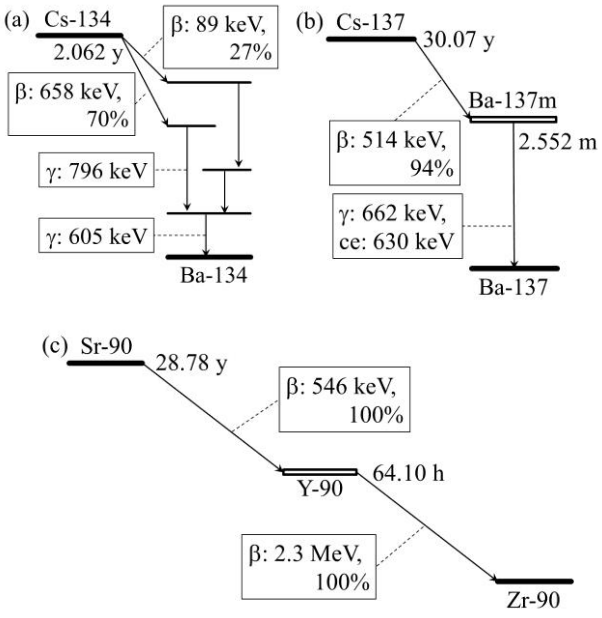

Figure 1. Decay schemes of relevant radionuclides described with types of transition, emission energies and its ratios: (a) Cs-134, (b) Cs-137, (c) Sr/Y-90. Energies of beta rays are expressed in maximum. The given energy of conversion electron (ce) is in average [2].

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was applied to a prepared sample mixed with an aqueous solution of Cs-134, Cs-137, and Sr/Y-90.

2. Experimental setup

Figure 1 shows the principal decay scheme of relevant radionuclides. Sr/Y-90 emits beta-rays without gamma ray emission. Cs-134 emits beta and gamma rays simultaneously. In contrast, Cs-137 emits gamma rays through a meta-stable state of Ba-137 with a relatively long half-life (2.5 min) after beta-ray emission. Discrimination of Cs-134 from Cs-137 is based on detection of the prompt emissions of gamma rays after beta transition in only Cs-134 decay. It means that high efficiency of beta-ray detection is preferable.

Figure 2 shows the layout of the prototype setup. It consisted of a NaI(Tl) scintillation detector for gamma rays and a plastic scintillator for beta rays. The layout of these detectors is suitable for acquiring coincident counting of beta and gamma emissions with a large solid angle. This source preparation provides very high detection efficiency for beta rays because of the low self-absorption and almost 4π solid angle. It also prevents contamination during the removal of the source.

A water sample with a mixture of Cs-134, Cs-137, and Sr/Y-90 (ratio 2:3:5, total of 97.9 Bq equal to 100.5 electron per sec) was dropped on top of the cylindrical thin plate of the plastic scintillator (40 mm diameter × 4 mm in thickness). After being left alone for more than half a day for it to completely dry naturally, the radioactive sample was sandwiched to another plastic scintillator with optical binding grease at the edge of the plastic (**Figure 3**).

The sandwich scintillators were inserted and fixed into an assembly jig made of aluminum that also acted as a shield against outside light. A scintillation light reflector was painted around the scintillator and a light guide. The light guide had a rectangular shape with a hole at the center to set the plastic scintillator. Scintillation light in the plastic was guided into photomultipliers facing each other across the light guide. A coincidence event in both photomultipliers generates a trigger of beta-ray detection with the removal of individual noise in each photomultiplier.

Two cylindrical NaI(Tl) detectors (3 inch diameter × 3 inch in thickness) were placed orthogonally to the plastic scintillator to make the solid angle as large as possible. Lead shields (5 cm thick) were placed on all lateral sides of the NaI(Tl) detectors.

Figure 4 shows schematic drawing of the data acquisition system. An event detected by both beta- and gamma-ray generated a trigger for the data acquisition (DAQ) system. In the measurements conducted in this study, the pulse height of signals of beta and gamma rays through the shaping amplifier were acquired in list mode by the CAMAC DAQ system. The system allowed us to reconstruct the pulse height spectra in coincidence mode in offline analysis.

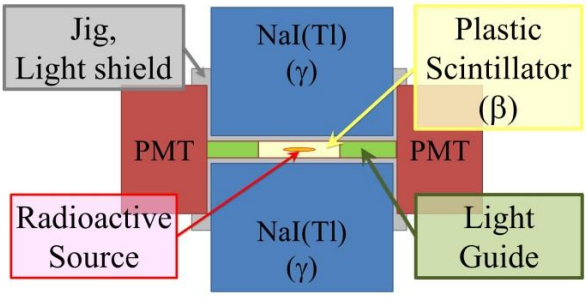

Figure 2. Prototype setup employing a disposal plastic scintillator optically connected to photomultipliers (HAMAMATSU, H3177-50) through a light guide and NaI(Tl) scintillation detectors (BICRON, #3M3/3) surrounding the plastic.

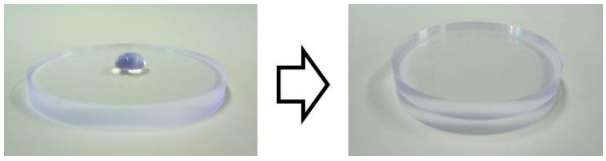

Figure 3. Picture of source preparation on the disposal plastic scintillator (EJ-212). A water sample containing radionuclides (Cs-134, Cs-137, and Sr/Y-90) was dropped, dried, and sandwiched.

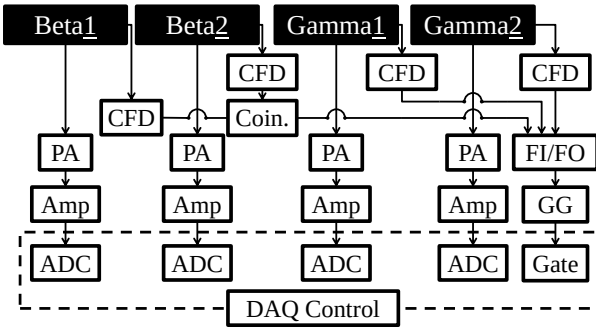

Figure 4. Block diagram of the electronics and the data acquisition system: CFD (constant fraction discriminator, ORTEC 935), PA (pre amplifier, ORTEC 113), Amp (linear shaping amplifier, ORTEC 572A), FI/FO (fan in/fan out, N-TM102), GG (gate & delay generator, KN1500), and ADC (analog to digital converter, C-TM405).

3. Results and discussion

Figure 5 shows the gamma-ray pulse-height spectra obtained using the NaI(Tl) detector. The red line was obtained analytically from raw data (black line) under the condition of γ–β coincidence. The red line clearly shows photo peaks derived from Cs-134. The photoelectron peak of 605 keV was successfully separated from the 662 keV of Cs-137. Furthermore, the sum peak of Cs-134 (1401 keV) was separated from the photo peak of K-40, which is often dominant in natural nuclides.

These results indicate that gamma-ray spectrometry employing a NaI(Tl) detector with beta ray coincidence is useful for measuring the radioactivity of Cs-134 and

Cs-137 individually.

Figure 6 shows beta-ray pulse-height spectra obtained using the plastic scintillation detector. In the beta spectra, the higher pulse height than the conversion electron peak was caused by high-energy beta rays from Y-90 and the lower pulse height overlapped with beta rays from Cs-134, Cs-137, and Sr-90. The photo peak of conversion electrons derived from Cs-137 could be detected in the raw spectrum but was eliminated under the coincident condition.

The beta-ray emission rate of the prepared sample including conversion electrons was deduced by a conventional extrapolation technique with 4πβ–γ counting [3, 4]. Beta-ray emission rates from Cs-134 and Cs-137 were deduced by gamma-ray spectrometry. Sr/Y-90 was deduced from the uplift for the total beta-ray emission rate without the Cs-134 and Cs-137 components. Beta ray emission rate of the sample was determined as 100.2 electrons per sec by the 4πβ–γ coincidence counting method. This value agreed with prepared radioactivity of the sample. This indicates that this measurement methodology is applicable to radioactivity measurement of pure beta emitters such as Sr/Y-90 and even of mixtures of Cs-134 and Cs-137.

In the case of low concentration of Sr/Y-90, determination of beta ray emission rate by 4πβ–γ coincidence counting has relatively large uncertainty, so we conducted extra measurements in which samples separately contained Cs-134, Cs-137, and Sr/Y-90 were prepared. Since their activities when dropped on the plastic were known for each radionuclide, the counting efficiencies were deduced. Samples of Cs-137 and

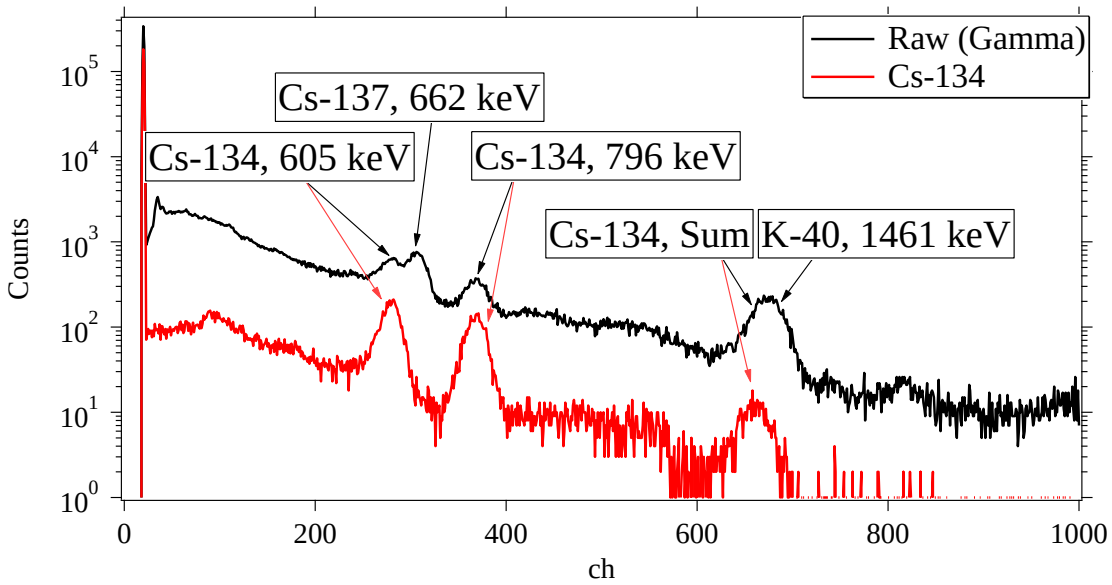

Figure 5. Gamma ray spectra obtained using the NaI(Tl) scintillation detector: (black) raw, (red) derived from Cs-134 under the condition of coincidence with beta rays.

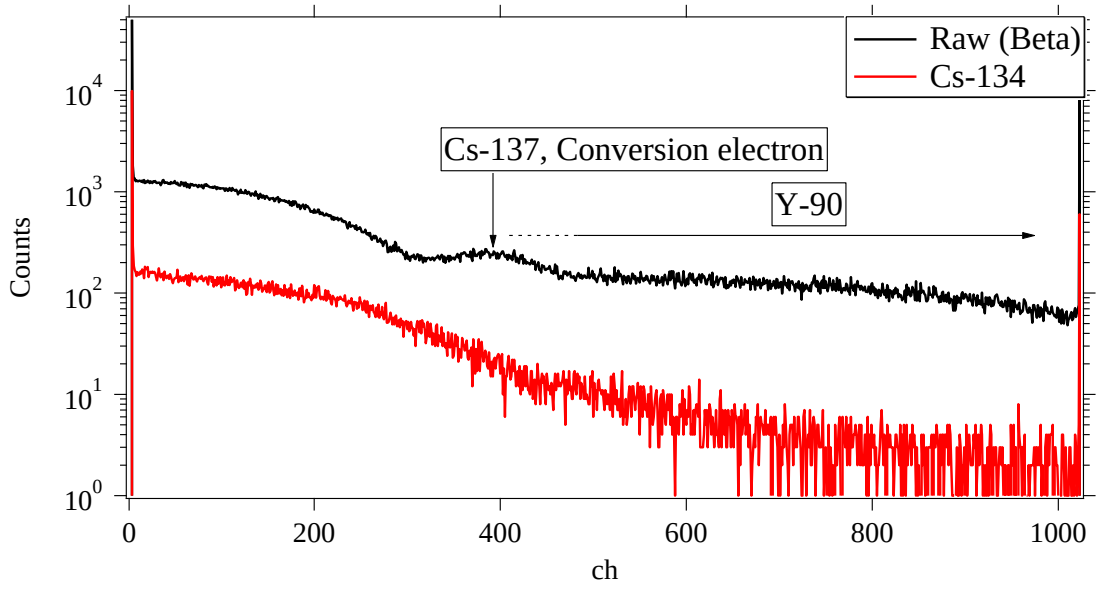

Figure 6. Beta ray spectra obtained using the plastic scintillation detector: (black) raw, (red) derived from Cs-134 under the condition of coincidence with photopeaks of gamma rays.

Sr/Y-90 that emitted high-energy beta rays showed sufficient counting efficiencies even with the present prototype setup. However, the counting efficiency of Cs-134 was slightly low because Cs-134 has a low-energy beta-ray emission branch. Therefore, we found that it was necessary to modify the counting efficiency of low-energy beta rays in the next setup of this measurement method.

4. Conclusion

In this paper, we introduced a new methodology for measuring radioactivity. A prototype setup was assembled to demonstrate the methodology. The obtained results show that application of beta-ray coincidence to gamma-ray spectrometry can successfully discriminate nuclides even in mixtures of several nuclides. Discrimination among Cs-134 and Cs-137 was demonstrated. The present setup employed NaI(Tl) detectors as a gamma-ray detectors. The method can be applied to other types of gamma ray detector, e.g., CsI(Tl).

The beta-ray pulse-height spectrum was also obtained. Even though the scintillation light entered into the photomultiplier at a shallow solid angle, sufficient detection efficiencies were achieved for Cs-137 and Sr/Y-90. Combining beta- and gamma-ray detectors can realize the absolute measurement of beta-ray emission rate by application of $4\pi\beta-\gamma$ counting technique because of the high detection efficiencies for both beta and gamma rays. In next projected setup, it is necessary to further improve the efficiency of beta-ray detection.

The detection method can be applied to determine the radioactivity of pure beta emitters in a mixture of Cs-134, Cs-137, and K-40 without complicated chemical treatment. This is convenient for source preparation and makes implementation more rapid.

We aim to apply beta ray measurement to radioactivity measurement. One potential application is measuring samples contaminated by the Fukushima accident. However, there are some challenges that need to be overcome. We will develop some optimization processes and a toolkit. We expect that the use of this method will then increase.

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