Assessment of the Nuclide Inventory for Uranium-Contaminated HEPA Filter Wastes

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1. Introduction

Around 3,000 units of spent HEPA filter waste at KAERI have been dismantled into a filter frame and filter media part, where most radioisotopes are captured, for efficient management. The filter media generated from the same facility were then compacted into a 200-L drum, after taking the representative samples for the assessment of the nuclide inventory of them according to the sampling procedure [1, 2].

In general, uranium existing in the waste could be analyzed using radiochemical methods such as alpha spectrometry, a liquid scintillation counting (LSC) system, and mass spectrometry. Although these methods can be processed with low detection limits by using the chemical pre-treatment of samples and the specific detector, time and cost constrains compared with their accuracy dictates the use of a simpler method of application. High resolution gamma spectrometry can simply and easily provide a reasonable estimate of the uranium activity as a less time and cost consuming technique [3].

In this study, a high purity germanium (HPGe) detector was employed to determine the activity of uranium-contaminated HEPA filter waste. The activity of U-235 and U-238 was identified by the analyses of a gamma ray peak of 143.76 keV belong to U-235 and of the daughters of U-238 such as Th-234 and Pa-234m with a secular equilibrium. To confirm their feasibility using gamma ray spectroscopy, the analysis results were compared with the gross alpha activity in the same sample.

Methods and Results

Representative samples of about 2 cm x 3 cm taken from the HEPA filter media were collected into a 1-L Marinelli beaker. In the beaker, these samples had an apparent density of about 0.15 g/cm^3.

2.1 Identification of U-235 and U-238

The U-235 activity could be directly calculated from the detected gamma ray peaks. Several gamma ray energies and intensities of naturally occurring uranium and actinium series are shown in Table 1. Since 185.715 keV of U-235 which had the highest intensity, interferes with 186.211 keV of Ra-226, which is a daughter of U-238, the gamma ray peak of U-235 was selected as 143.76 keV, in which the interference peak does not exist around that energy range. The U-238 activity cannot be directly determined as it has only a weak intensity of about 0.064 % at 49.55 keV. However, due to the secular equilibrium between U-238 and daughter nuclides such as Th-234 and Pa-234m, it is possible to take daughter nuclide activity as the U-238 activity.

Table 1. Several gamma ray energies and intensities of naturally occurring uranium and actinium series

Nuclide	Energy (keV)	Intensity (%)	Nuclide	Energy (keV)	Intensity (%)
U-235	143.76	10.96	U-238	49.55	0.064
	163.33	5.08	Th-234	63.29	4.5
	185.715	57.2		92.38	2.81
	202.110	1.08		92.80	2.77
	205.311	5.01	Pa-234m	1001	0.838

As shown in Table 1, for analyzing a gamma ray of 63.29 keV in Th-234, there are some drawbacks in terms of the interference with 63.9 keV of Th-232 and the bad resolution of a p-type HPGe detector. On the other hand, the analysis of Pa-234m has an advantage of non-interference around 1001 keV in the gamma ray spectrum, however, this method has a high minimum detectable accuracy (MDA) because of its week gamma ray intensity. Therefore, gamma ray peaks of 92.38 and 92.80 keV were used to calculate the activity of Th-234. Because it is impossible to separate these two peaks in the gamma ray spectroscopy, these two peaks were assumed as one peak with an energy of 92.59 and intensity of 5.58 %.

2.2 Density correction of the representative samples

Calibration in gamma ray spectroscopy is generally conducted using a reference standard source with a density of 1 g/cm^3. However, since the densities of the samples in the 1-L Marinelli beaker were shown to be around 0.15 g/cm^3, the density correction factor [4], which means the ratio of the detected count rate at the sample density to the reference density of 1 g/cm^3, was calculated using the MCNP code. In the simulation process, the geometry of an HPGe detector was designed according to the detector specifications, as shown in figure 1. The results of several gamma ray energies are shown in figure 2.

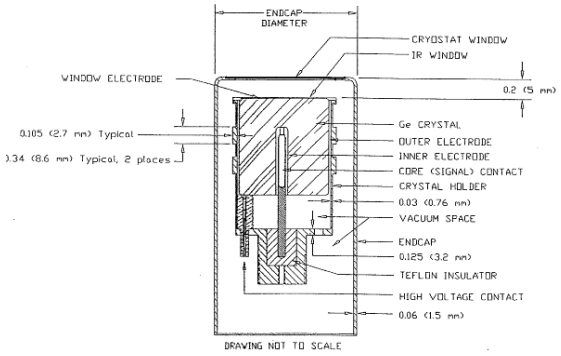

Figure 1. Cross-sectional view of the HPGe germanium detector

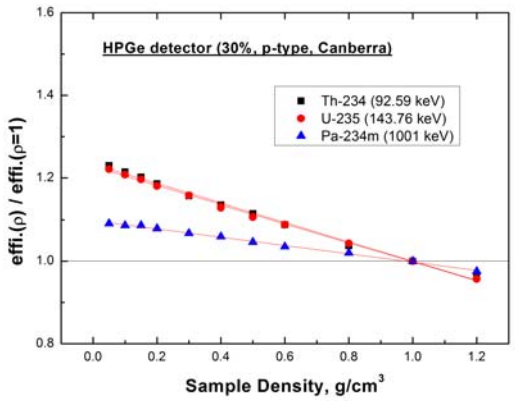

Figure 2. Density correction factor of the HPGe detector according to the sample density

2.3 Verification of uranium activity

Based on the density correction, the activities of Th-234 and Pa-234m were made to have good agreement, particularly above several Becquerel per gram in the Th-234 activity. This means it is possible to use the Th-234 activity of 92.59 keV with an intensity of 5.58 % as the activity for U-238.

To check other possible alpha nuclides in the representative sample, analyses of the gross alpha and gross beta were conducted by taking samples of the 12 planchets about 1 % of the original sample in a 1-L Marinelli beaker, as shown in figure 3.

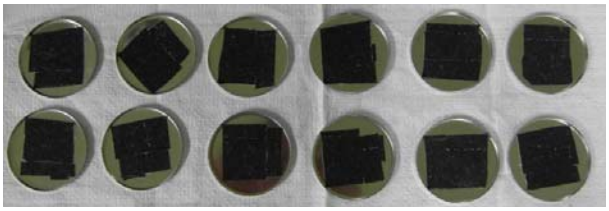

Figure 3. Samples for gross alpha and gross beta analysis that were taken from a 1-L Marinelli beaker

The results on the gross alpha and gross beta are shown in figure 4, which is illustrated in log-log scale. A straight line in the graph indicates the uranium activity including U-235 and U-238. The broken line, which indicates the gross beta activity, is expected to originate from beta-gamma nuclides in the uranium decay chain, such as Th-234 and Pa-234m. Since the slopes of the three lines really have the same values, it could be estimated the other alpha and beta nuclides do not exist in the representative samples.

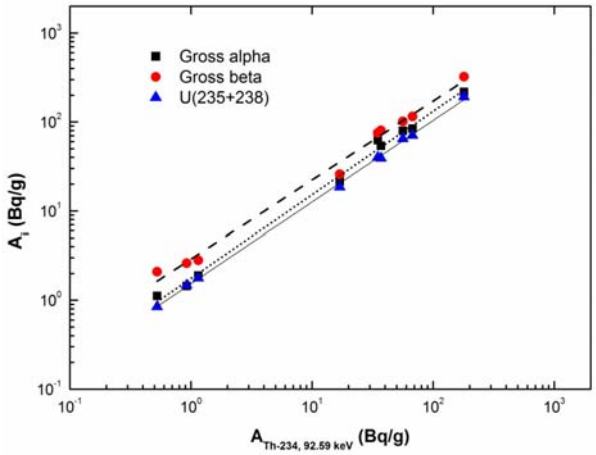

Figure 4. Gross alpha, gross beta, and uranium activities according to Th-234 activity at 92.59 keV

3. Conclusion

The activity of uranium-contaminated HEPA filter waste was determined using an HPGe detector. The activity of U-235 was directly analyzed from a gamma ray peak of 143.76 keV, while that of U-238 was identified using an analysis of its daughters such as Th-234 and Pa-234m with a secular equilibrium. To determine its feasibility using a gamma ray spectroscopy, the analysis results are compared with the gross alpha and gross beta activities of the sample. As a result, the slopes for the gross alpha, gross beta, and uranium activities based on an HPGe detector and depending on the activity of Th-234 at 92.59 keV had almost the same value. This means it is possible to use the activity of Th-234 with 92.59 keV energy and an intensity of 5.58 % as the activity for U-238.

REFERENCES

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