

Low-temperature CdZnTe Detector for Quantification of Ni-59 in Radioactive Waste

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1. Introduction

According to the Notice No. 2024-13 of the Nuclear Safety and Security Commission on the Acceptance Criteria for Low- and Intermediate-Level Radioactive Waste, waste generators are required to determine the radioactivity concentrations of 14 radionuclides [1]. To this end, radioactive waste samples are chemically separated through various pretreatment procedures depending on the target nuclides, and subsequently measured using a liquid scintillation counter (LSC), gas proportional counter, or high-purity germanium (HPGe) detector according to the type and energy of the emitted radiation.

Among these radionuclides, Ni-59 emits low-energy electrons and X-rays with limited penetration power, making LSC suitable for its measurement. However, Ni-59 cannot be chemically separated from Ni-63, a low-energy beta emitter, and the concentration of Ni-63 in radioactive waste is typically approximately 100 times higher than that of Ni-59. Therefore, quantification of Ni-59 using LSC is extremely challenging. Consequently, low-energy high-purity germanium (LeGe) detectors are generally employed. However, these detectors are very expensive and require cryogenic cooling systems due to significant thermal noise at room temperature [2].

CdZnTe is a semiconductor detector that exhibits satisfactory performance at room temperature and has been widely utilized in various fields, including nuclear power plant operation and decommissioning, medical applications, nondestructive testing, astronomy, and environmental monitoring [3,4]. This semiconductor exhibits significantly reduced noise even at temperatures around -30 °C, enabling system implementation with a compact cooling device compared to conventional LeGe detectors. In this study, a detection system based on a small Peltier module was developed, and its feasibility was verified by measuring various gamma- and X-ray-emitting radionuclides as well as Ni-59 samples.

2. Methods and Results

Figure 1 presents a schematic diagram of the overall system. The CdZnTe detector was manufactured by eV Products, with dimensions of 10 × 10 × 5 mm³. The sample holder was fabricated in a rectangular

parallelepiped shape to ensure effective contact with the Peltier cooler, and the entrance window was composed of 250- μ m-thick Be to minimize radiation attenuation. A CP-040HT cooler manufactured by TE Technology was employed, and thermal grease was applied at the interface between the holder and the cooler to maximize the cooling efficiency. The detection assembly was placed inside a low-temperature chamber to eliminate the influence of ambient temperature variations. As a result, the surface temperature of the holder was measured to be approximately -25°C.

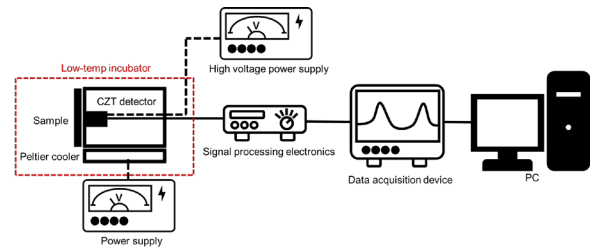


Fig. 1. Schematic diagram of the detection system.

2.1 Detector Optimization

The signal processing circuit consisted of a low-noise preamplifier, shaping amplifier, high-voltage supply, and multichannel analyzer, and optimization of the applied voltage and shaping time was performed in advance. Figure 2 shows the pulse-height spectra of Am-241 and Ba-133 as a function of applied voltage at a shaping time of 1 μ s. At 600 V bias, low-energy tailing was observed, or the peaks were not properly formed, which is attributed to the position dependence of the signal. In CdZnTe detectors with a planar electrode configuration, the signal amplitude is proportional to the sum of the drift distances of electrons and holes. While electrons are fully collected regardless of the interaction position, holes exhibit significant position dependence due to their very low mobility. Moreover, as the temperature decreases, the mobility–lifetime product of holes further decreases [5], which can exacerbate the position dependence. In addition, the current detector structure does not ensure uniform cooling across the entire CdZnTe crystal, further contributing to position-dependent effects. Although these issues can be mitigated by increasing the applied voltage, higher bias results in increased shot noise due to

leakage current. Therefore, 700 V was selected as the optimal condition, at which peak shape was improved without a noticeable increase in electrical noise.

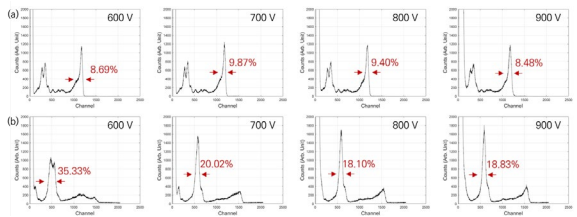


Fig. 2. Pulse-height spectra of (a) Am-241 and (b) Ba-133 as a function of applied voltage.

The energy resolution depends on the shaping time due to the trade-off between serial and parallel noise contributions. Figure 3 shows the pulse-height spectra of Am-241 as a function of shaping time at 700 V bias. The 59.5 keV peak was clearly observed at all shaping times except 0.5 μ s; however, peak broadening became significant at shaping times of 2 μ s or longer. Accordingly, a shaping time of 1 μ s was selected.

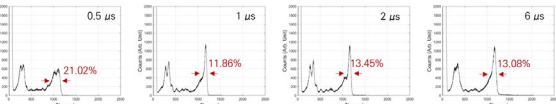


Fig. 3. Pulse-height spectra of Am-241 as a function of shaping time.

2.1 Measurement of Ni-59 sample

The liquid Ni-59 source was precipitated and subsequently deposited onto a glass fiber filter to prepare the measurement sample. The precipitate was distributed over the filter with a diameter of 16 mm, and the activity of the prepared sample was 199.64 Bq. The filter was positioned in front of the entrance window, and the distance between the detector and the filter surface was approximately 4 mm. The entrance window, with a diameter of 15 mm, is slightly smaller than the precipitate distribution, resulting in a loss of detection efficiency. Figure 4 (a) and (b) shows the results of six repeated measurements of background radiation and the Ni-59 sample, each for 10 min. The system showed stable performance over one hour, and the photopeak of Ni-59 was distinguishable from the background region. Figure 4 (c) and (d) presents the summed spectra of the background and Ni-59 measurements, as well as the difference between the two spectra. A distinct photopeak was observed at approximately 7 keV.

The detection efficiency was calculated as the ratio of the number of counts to the total number of emitted radiations. The evaluated efficiency was $3.65 \pm 0.27\%$, and the uncertainty of the efficiency could be influenced by uncertainties in the activity, half-life, emission probability, counting statistics, and other factors. It is expected that the contribution of counting statistics was dominant due to the short measurement time and low activity.

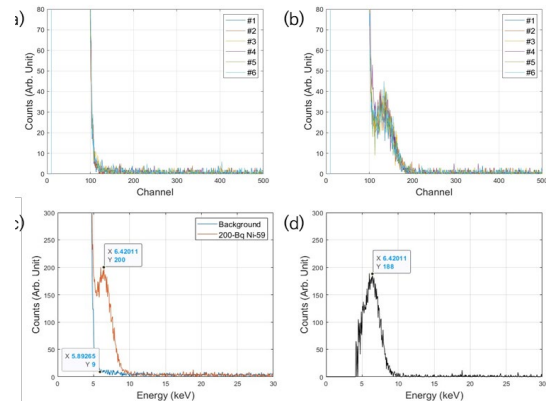


Fig. 4. (a) and (b) Results of six repeated 10-min measurements of background radiation and Ni-59, respectively; (c) summed spectrum over 1 h; and (d) difference between the two spectra.

3. Conclusions

In this study, a low-energy spectroscopic system based on a cooled CdZnTe detector was established. The proposed system was systematically optimized under various applied voltages and shaping times, demonstrating stable operation and the capability to resolve the low-energy photopeak of Ni-59 from background radiation. These results indicate that the developed system has strong potential as a cost-effective and compact alternative for Ni-59 quantification in low- and intermediate-level radioactive waste.

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