

## Study of CsI:CO<sub>3</sub> Crystals for $\alpha$ Detector

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We grew CsI:CO<sub>3</sub> single crystals with 0.1 mol % concentration of CO<sub>3</sub> by using Czochralski method. We studied the feasibility of the CsI:CO<sub>3</sub> crystal as an  $\alpha$ -ray detector which can be used to measure total alpha or beta activity in environmental samples. For the discrimination between  $\alpha$  and  $\beta$  particles, we investigated the fluorescence decay time characteristics of a CsI:CO<sub>3</sub> single crystal. We also investigated the other scintillation properties such as the alpha beta ratio, the pulse height spectrum, and the ability of pulse shape discrimination between the pulse of  $\alpha$  and that of the  $\beta$ . We used 2  $\mu$ m aluminum Mylar foil for the radiation entrance window of a CsI:CO<sub>3</sub> crystal. To estimate the energy loss of  $\alpha$  particle in the aluminum Mylar foil, we simulated by using the GEANT 4 simulation tool kit. We demonstrated the performance of the  $\alpha$  detector with various radiation sources as well as environmental soil sample.

**KEYWORDS:** CsI:CO<sub>3</sub> crystal, alpha detector, scintillation detector, pulse shape discrimination, radon

### I. INTRODUCTION

Scintillation crystals have been being used in the various fields such as high-energy physics, nuclear instrumentation, radiation measurement, medical imaging and etc. For these applications, development of good-performance scintillation crystals are demanded<sup>1</sup>. There have been developed various scintillation crystals for above applications. We grew CsI:CO<sub>3</sub> crystals with different concentrations of CO<sub>3</sub> by using Czochralski method to develop a scintillation crystal to answer the above request<sup>2</sup>. We also characterized scintillation properties of these crystals. Scintillation characteristics such as fluorescence decay time, emission spectrum, energy resolution, linearity of luminescence response to proton beams, pulse height spectrum of various radioactive sources, pulse shape discrimination power between  $\alpha$  and  $\beta$  (or  $\gamma$ ) showed that the CsI:CO<sub>3</sub> single crystals have good scintillation properties<sup>2</sup>.

The  $\alpha$ -ray detection is important because it can be used as an environmental radon (<sup>222</sup>Rn) monitoring equipment. It is also important to detect radon which contaminates environmental air which is one of the potential health hazard radiative materials. Breathing air containing <sup>222</sup>Rn gives health hazard by radiation dose to the lung. The principal radiation dose is due to its decay products such as <sup>218</sup>Po, and <sup>214</sup>Po. Their contribution to the radiation dose to the lung is 2-3 orders of magnitude greater than that of <sup>222</sup>Rn<sup>6</sup>. All of these radioactive isotopes emit  $\alpha$ -ray when they decay. So, we need to monitor the concentration of these radionuclides in air by an  $\alpha$ -ray detection.

Underground spaces are getting more important as a astrophysics experiment requiring low radiation background such as detections of dark matter, neutrino, and etc. The example of underground physical experiments are Korea Invisible Mass Search (KIMS) and Super-Kamiokande in which the

radon contamination is one of the most important background source<sup>4,5</sup>.

In this paper we investigated the feasibility of the CsI:CO<sub>3</sub> crystal as an  $\alpha$ -ray detector which can be used to measure total alpha or beta activity in environmental samples.

### II. GEANT4 Simulation

We grew CsI:CO<sub>3</sub> crystal with CO<sub>3</sub> concentration of 0.1 mol percent by using the Czochralski method. We cut the crystal to make a sample with dimension of 9.5  $\times$  9.5  $\times$  6.2 mm<sup>3</sup>. To use this sample as an  $\alpha$  sensor we wrapped the side surfaces with Teflon tape and the front face was wrapped with 2.0  $\mu$ m Mylar aluminum foil (EJ-590/B10HH ALUMINIZED MYLAR) which was used as an  $\alpha$  entrance window. This aluminum foil was composed of a polyester film of 1.8  $\mu$ m thickness coated on both sides with aluminum metal layers both of 0.10  $\mu$ m thickness<sup>7</sup>. Figure 1 shows the CsI:CO<sub>3</sub> crystal sample with and without wrapping.

We performed computer simulations to estimate the 5.5 MeV alpha energy loss in the Mylar entrance window by using the Geant4 simulation toolkit<sup>8,9</sup>. The results of the simulations are shown in Fig. 2. It was shown that the alpha energy deposited (a) in the 0.2  $\mu$ m thickness of aluminum layers, (b) in the 1.8  $\mu$ m thickness of polyester film, and (c) in the 6.2 mm thickness of CsI:CO<sub>3</sub> crystal. From Fig. 2, we found that 4 % of alpha energy is deposited in the Mylar window and 96 % of alpha energy penetrates the Mylar window for 5.5 MeV alpha particles. It proves that the Mylar aluminium foil work well as an alpha particle window for the alpha-detector. If the alpha energy is higher than 5.5 MeV, the Mylar entrance window works even better, because the portion of the energy deposited in the Mylar window become smaller. From these results, we conclude that the energy loss of alpha particle in the entrance window can be ignored.

### III. Experiments

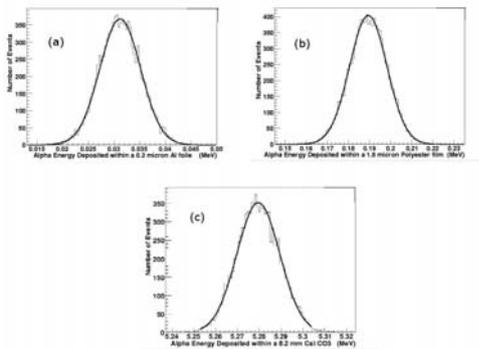
Our experiment was composed of two parts, preliminary part and main part. First, we measured characterization of the

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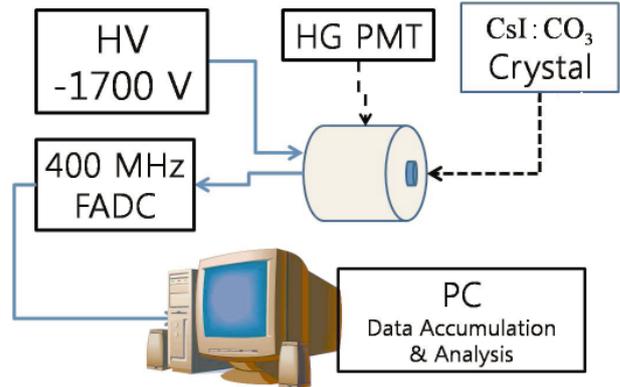
**Fig. 1** The upper figure is a CsI:CO<sub>3</sub> single crystal sample and the lower one is the sample wrapped with Mylar aluminum foil.



**Fig. 2** Alpha energy deposited (a) in the 0.2 μm thickness aluminum layers, (b) in the 1.8 μm thickness polyester film, and (c) in the 6.2 mm thickness CsI:CO<sub>3</sub> crystal.

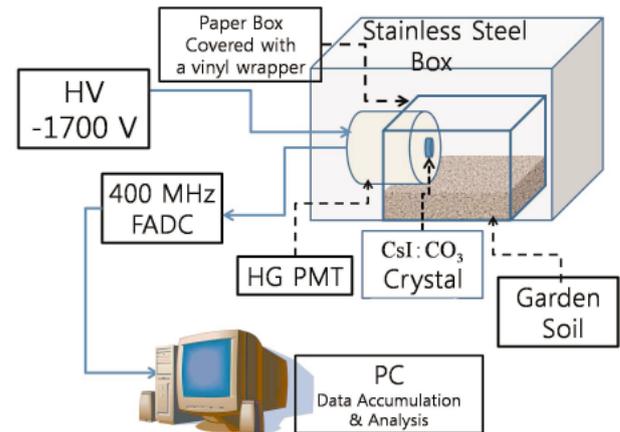
crystal and then studied the alpha-detector part.

We carried these measurements by a pulse height analysis system equipped with a 2-inch high-gain photomultiplier tube (PMT, Photonis XP2260). We made CsI:CO<sub>3</sub> single crystal sample with dimension of 9.5 × 9.5 × 6.2 mm<sup>3</sup>, and wrapped the side surfaces with Teflon tape, the front face with 2.0 μm Mylar aluminum foil as described in section II. One end of the crystal sample was coupled directly with the widow of the PMT by using an optical grease. The output of the PMT was directly feed to the 400-MHz FADC (flash analog-to-digital converter)<sup>10</sup>. The digitized signals were transferred to the personal computer through USB. The 662-keV γ-rays from <sup>137</sup>Cs, the 511-keV γ-rays from <sup>22</sup>Na, and 5.5 MeV α-ray from <sup>241</sup>Am were used for the CsI:CO<sub>3</sub> crystal characterizations. The data were analyzed by using the ROOT package<sup>11</sup>. Single photoelectrons were identified by using a clustering algorithm to reduce noise effect. A schematic is shown in Fig.3.



**Fig. 3** Schematic of the experimental setup for the sample characterization.

To test the alpha-detector performance, we measured environmental α-ray by the pulse height analysis system mentioned above. The alpha-detector and a sample of garden soil were located inside of a stainless airtight container as shown in Fig. 4.



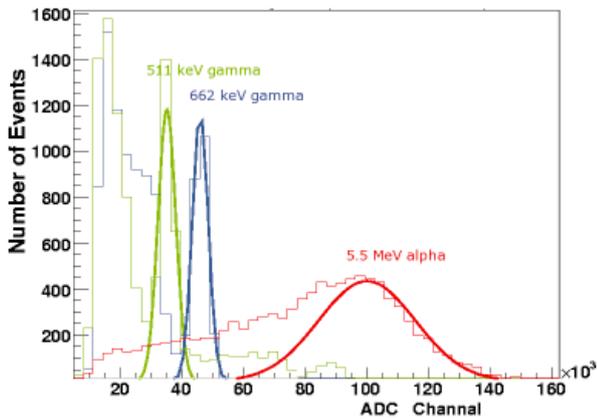
**Fig. 4** Schematic of the experimental setup for for the environmental alpha decay measurement.

#### IV. Results and Discussion

##### 1. Pulse Height Spectrum, Energy Resolution and Alpha Beta Ratio

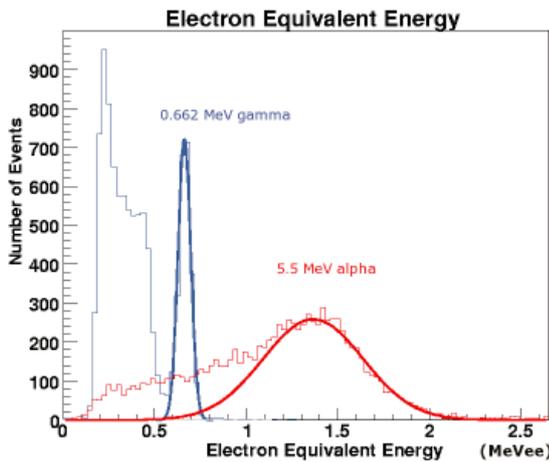
To test the alpha-detector performance, we measured the pulse height spectrum of the CsI:CO<sub>3</sub> crystal irradiated by various radiations, such as 662 KeV γ-ray from <sup>137</sup>Cs, 511-keV γ-rays from <sup>22</sup>Na, and 5.5 MeV α-ray from <sup>241</sup>Am. The results of our measurements are shown in Fig. 5. As shown in Fig. 5, the energy resolution of CsI:CO<sub>3</sub> crystal sample is 12% in FWHM for 662 KeV γ-ray, whereas the α-energy resolution is not so good. Since the CsI:CO<sub>3</sub> crystal is so hygroscopic, its surface can have damaged layer easily. The damaged layer could cause the bad energy resolution for α-ray even if it does not effect to the γ-ray energy resolution. The alpha/beta ratio of the CsI:CO<sub>3</sub> crystal is measured to be 0.25 as shown in Fig. 5, where the alpha is defined as mean

value of analog digital convert channel (ADC channel) number divided the  $\alpha$  particle kinetic energy, and beta is defined as mean value of ADC channel number divided the  $\gamma$  (or  $\beta$ ) particle kinetic energy. The value 0.25 of the alpha /beta ratio means that 75% of the kinetic energy of an  $\alpha$  particle is lost by mechanism due to the thermal quenching in CsI:CO<sub>3</sub> crystal sample<sup>12</sup>). If we take the peak vale of the 662 keV  $\gamma$ -



**Fig. 5** Pulse Height spectrum of 662 KeV  $\gamma$ -ray, the 511-keV  $\gamma$ -rays, and 5.5 MeV  $\alpha$ -ray.

rays spectrum as a calibration energy, we can convert the ADC channel numbers of the spectrums as the electron equivalent energy (Eee). Figure 6 shows the electron equivalent energies of 662 keV  $\gamma$ -ray and 5.5 MeV  $\alpha$ -ray.



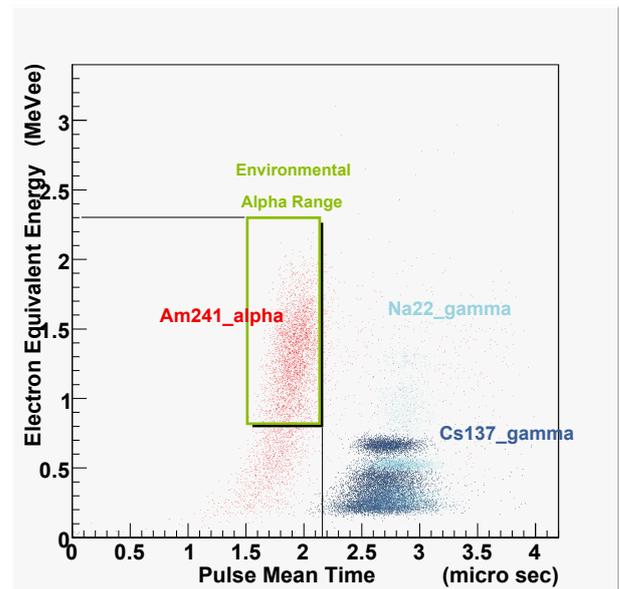
**Fig. 6** Electron equivalent energy spectrum of 662 KeV  $\gamma$ -ray, and 5.5 MeV  $\alpha$ -ray.

We show the two dimensional histogram of pulse mean time vs. electron equivalent energy in Fig. 7 for 662 keV  $\gamma$ -ray and 5.5 MeV  $\alpha$ -ray, respectively. The pulse mean time is the pulse height weighted time average, defined as

$$\langle t \rangle = \frac{\sum t_i \times q_i}{\sum q_i},$$

where  $q_i$  is amplitude of the pulse at the channel time  $t_i$  up to  $4 \mu\text{s}$ <sup>12</sup>). For  $\alpha$  and  $\gamma$  separation, 5.5 MeV  $\alpha$  particles and 662 keV  $\gamma$  radioactive sources were used as shown in Fig.7. We specified the environmental  $\alpha$ -ray range as  $1.5 \mu\text{s} \leq \langle t \rangle \leq 2.15 \mu\text{s}$  in horizontal axis and  $0.8 \text{ MeV} \leq E_{ee} \leq 2.3 \text{ MeV}$  in vertical axis, where  $E_{ee}$  is electron equivalent energy of  $\alpha$ -ray. Our above consideration made us to present the specified two dimensional range of environmental alpha ray as a rectangle which does not overlap the  $\gamma$ -ray band shown as in Fig. 7.

Two clear bands in Fig. 7 enable us to set the energy range of environmental  $\alpha$ -ray. The main environmental  $\alpha$ -ray sources are <sup>222</sup>Rn and its decay products <sup>218</sup>Po, <sup>214</sup>Po. The kinetic energies of  $\alpha$ -rays from these radionuclides are known as 5.591 MeV, 6.115 MeV, and 7.834 MeV, respectively<sup>13</sup>). Using alpha/gamma separation method as shown in Fig. 7, environmental alpha-ray can be measured.



**Fig. 7** Two dimensional histogram of pulse mean time vs. Eee of of 662 KeV  $\gamma$ -ray, and 5.5 MeV  $\alpha$ -ray.

## 2. Environmental $\alpha$ -ray Detection Using the CsI:CO<sub>3</sub> Crystal

To measure the environmental radiations, we put the CsI:CO<sub>3</sub> crystal of 0.1 mol % concentration of CO<sub>3</sub> and sample of garden soil into a stainless airtight box. We accumulated 88,000 events data for 23 hours. The trigger rate was 1.03 Hz. From the measured data we plot the two dimensional histogram of pulse mean time vs. Eee as shown in Fig. 8. To discriminate environmental  $\alpha$  particles from the  $\gamma$  particles, we used box selection criteria specified on the histogram in the previous section. Figure 8 shows two dimensional histogram of pulse-mean-time vs. Eee distribution from an environmental soil sample and a radioactive source of <sup>137</sup>Cs overlapped with the box selection criteria. Clearly, the points in the selection box are separated form the other points out of the box, as shown in Fig. 8. These data points in the box can be interpreted as environmental alpha rays from <sup>222</sup>Rn and its decay

products radionuclides,  $^{218}\text{Po}$ ,  $^{214}\text{Po}$ . Figure 9 shows energy distribution of selected alpha particles after alpha/beta ratio correction. Even though we need to accumulate more sample for conclusive results, events around 8 MeV can be interpreted as 7.834 MeV  $\alpha$  from  $^{214}\text{Po}$  and lower energy bands are alpha particles from  $^{222}\text{Rn}$  and  $^{218}\text{Po}$  decay.

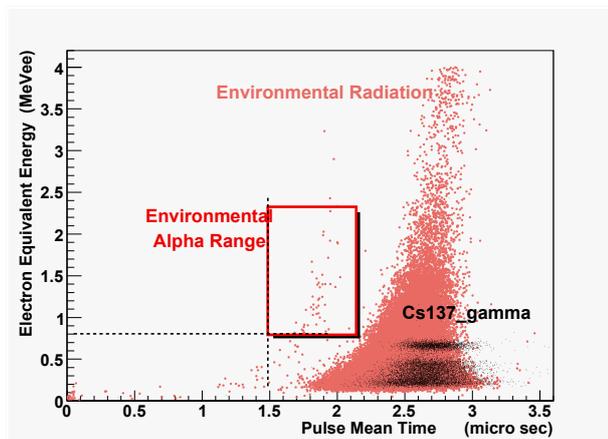


Fig. 8 Two dimensional histogram of pulse mean time vs. Eee distribution from an environmental soil sample and a radioactive source of  $^{137}\text{Cs}$ .

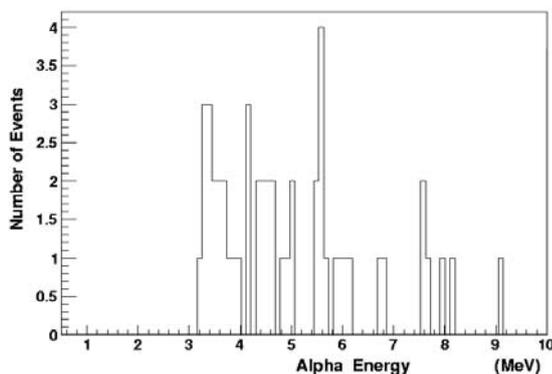


Fig. 9 Energy of  $\alpha$  particles from garden soil.

## V. Conclusion

We grew CsI:CO<sub>3</sub> single crystals with 0.1 mol % concentration of CO<sub>3</sub> by using Czochralski method. We studied the feasibility of the CsI:CO<sub>3</sub> crystal as an  $\alpha$ -ray detector. We measured its scintillation properties such as fluorescence decay time, the energy resolution, and the pulse height spectrum for the various radioactive sources at room temperature. The energy resolution of CsI:CO<sub>3</sub> was measured to be 12 % in FWHM for 662 keV  $\gamma$ -ray radioactive source. We demonstrated clear pulse shape separation between  $\alpha$  and  $\gamma$  ( $\beta$ ) by pulse mean time method.

To study the feasibility of the CsI:CO<sub>3</sub> crystal as an environmental  $\alpha$ -ray detector, we measured environmental radiation using garden soil and showed clear separation between  $\alpha$

particles from  $^{222}\text{Rn}$  and its decay daughters from  $\gamma$  ( $\beta$ )-rays. From our experiment, we demonstrated that the CsI:CO<sub>3</sub> single crystal works as a good environmental alpha ray detector due to its good pulse shape discrimination power. However, this crystal have a disadvantage. It is so hygroscopic that, some one must be careful to protect it from humidity if one tries to a use CsI:CO<sub>3</sub> single crystal as the alpha detector.

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