Scintillation Properties of CsI:Na, ^{133}Ba Crystal

M. J. Kim, Hong Jooknu Kim, H. Park, K. Tanida, Sunghwan Kim, J. K. Cheon, and K. B. Lee

***Abstract*—In Hyperball-J experiments, the lutetium oxyorthosilicate (Lu_2SiO_5:Ce, LSO) scintillators are constantly used to monitor and calibrate the Ge detectors. The LSO crystal has ^{176}Lu as a natural radioactive source. A new CsI:Na crystal that contains a ^{133}Ba radioactive source was developed in this study. This developed crystal can be used for detector calibration instead of LSO scintillators.**

The crystal was grown using a 2-zone vertical Bridgman method with a pulling speed of 0.5 mm/h. The CsI:Na, ^{133}Ba crystal was cut to ϕ 10 mm x 5 mm. Under X-ray excitation, the emission band of the crystal was observed between 330 nm and 650 nm. The scintillation properties of the CsI:Na, ^{133}Ba crystal such as light yield, energy resolution and decay time, were also measured using a ^{137}Cs γ-ray source.

A High Purity Germanium (HPGe) detector was used to estimate the radioactivity in the crystal. The γ-ray peaks were identified from ^{133}Ba decay in the CsI:Na, ^{133}Ba crystal with a HPGe detector.

***Index Terms*—^{133}Ba crystal, Bridgman method, CsI:Na, decay time, detector calibration, light yield.**

I. Introduction

FOR many years, inorganic scintillators have played an important role in many fields of fundamental research such as high energy physics, nuclear physics, and astrophysics to detect X- and γ-ray radiation. In addition to these basic studies, there have been many applications including medical imaging, industrial systems, and security systems at airports. Most inorganic scintillators have been studied with the aim of a high energy resolution, high light yield, fast response, high density, and high atomic number Z [1]–[3].

The CsI:Na crystal is a good alkali halide crystal. CsI:Na is less hygroscopic with higher stopping power than NaI:Tl [1]–[5]. The scintillation decay curve of a CsI:Na crystal was fitted with a sum of three exponentials and three decay time values were obtained corresponding to the fast ($\sim$ 460 ns), slow ($\sim$1.8 μs), and tail ($\sim$8 μs) in [4]. The peak of the emission spectra of the CsI:Na crystal is 420 nm. This material has a very high light yield ($\sim$ 42,000 photons/MeV) [3].

The X-ray detector system, Hyperball-J, which has been used for hypernuclear γ spectroscopy experiments, consists of approximately thirty Ge detectors, each surrounded by fast lead-tungstate ($PbWO_4$, PWO) counters for background suppression. Ge detectors are monitored and calibrated constantly by a system using lutetium oxyorthosilicate (Lu_2SiO_5:Ce, LSO) scintillators, which include ^{176}Lu as a natural radioactive source. This source emits several γ-rays (mainly 202 keV and 307 keV), and efficient data taking is possible because it is self-triggerable using β-rays. With this feature, calibration and X-ray data can be taken simultaneously from a LSO crystal [6].

In this study, the CsI:Na crystal, which contains a ^{133}Ba radioactive source, was grown using the Bridgman method, and the X-ray emission was measured for this CsI:Na, ^{133}Ba crystal. The pulse height spectra, energy resolution, relative light yield, and fluorescence decay time were also measured using the ^{137}Cs γ-ray source. This newly developed crystal can be used for detector calibration, not only to replace the LSO crystal in the Hyperball-J experiment, but also in other experiments. Compared to LSO crystals, the CsI:Na, ^{133}Ba crystal has a range of γ-rays such as 80 keV, 276 keV, 303 keV, 356 keV, and 384 keV. Moreover, it emits a 30 keV of X-ray, which can be used for calibration too. This type of crystal makes it possible to use a wide range of detector calibration energies. To the best of the authors' knowledge, this is first developed crystal scintillator with radioactive source doping.

II. Experiment

A. CsI:Na, ^{133}Ba Crystal Growing

The CsI:Na single crystal, which contains ^{133}Ba was grown using a two-zone vertical Bridgman method with CsI (Sigma Aldrich 99.99%) and 0.5 mole % of NaI (Sigma Aldrich 99.99%) powders with a liquid ^{133}Ba (99.99%) radioactive source. The mixed CsI:Na powder in sealed quartz tube, which contains radioactive ^{133}Ba in liquid form, was dried at the high temperature in a heating furnace. To prevent moisture and oxygen contamination, the powders were prepared and placed into a quartz ampoule in an argon-purged glove box. The ampoule was cleaned with 10% nitric acid, acetone, ethanol, and de-ionized water, and sealed under a 10^{-6} torr environment. The crystal was grown using a 2-zone vertical Bridgman method with a pulling speed of 0.5 mm/h. The sample of the CsI:Na, ^{133}Ba crystal was grown the cylinder shape, and then cut into three parts with dimensions of ϕ 10 $\times$ 5 mm^3, the top, middle, and bottom of the grown crystal. Each sample included a small amount of the ^{133}Ba radioactive source ($\sim$ 0.2 μCi). The samples were polished with mixed Al_2O_3 powder (grain size of 0.02 μm) in mineral oil with a polishing cloth (Buehler,

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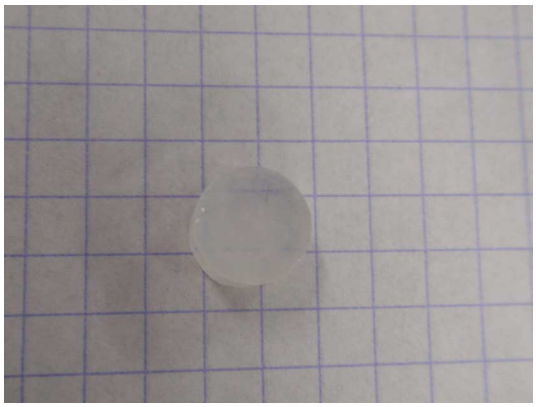


Fig. 1. A photograph of the grown CsI:Na, ^{133}Ba crystal.

No. 40-7218) for optical transmission characterization [7]. Fig. 1 shows the cut and polished CsI:Na, ^{133}Ba crystal sample.

B. Experimental Setup

To measure the X-ray emission spectrum of the scintillation crystal, all faces of the sample crystal were wrapped with Teflon tape except for one side to allow attachment with an optical fiber. To avoid the scintillation leakage, a holder made from Teflon with a hole at the center was used during the attachment of the optical fiber with the surface of the CsI:Na, ^{133}Ba sample crystal. Scintillation light from the crystal due to the X-rays irradiation was transmitted through an optical fiber to the QE65000 spectrometer (Ocean Optics) [8]. The spectrometer was calibrated using calibrated file.

The scintillation properties of the CsI:Na, ^{133}Ba were examined at room temperature. The CsI:Na, ^{133}Ba crystal was optically coupled with a 2-inch bialkali photomultiplier tube (XP2260) using optical grease. A signal from the PMT was fed to a preamplifier and then to a 400 MHz flash analog to digital converter (FADC) board. The FADC board made by NOTICE KOREA [9] was used for signal digitization. The digitized signal was read out using a Linux based computer through USB2 [10]. The data was analyzed using the C++ based data analysis program, ROOT package [11].

III. Scintillation Properties

A. X-Ray Emission Spectrum

The emission spectra for each of the three parts of the CsI:Na, ^{133}Ba scintillation crystal separately measured under X-ray irradiation at room temperature. The emission spectrum of the CsI:Na, ^{133}Ba consisted of broad band from 330 nm to 650 nm. The maximum emission peak wavelength was approximately 420 nm, which matches the results from the bialkali PMT well. Fig. 2 shows the emission spectrum of the CsI:Na, ^{133}Ba crystal. The results from each of the three separate scintillators were the same.

B. Energy Resolution and Relative Light Yield

The energy resolution of the three samples of the top, middle, and bottom parts of the CsI:Na, ^{133}Ba scintillation crystal was

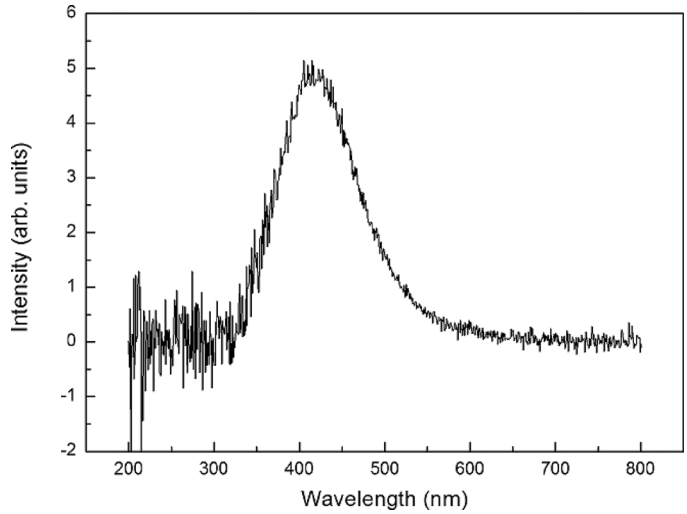


Fig. 2. X-ray induced emission spectrum of the CsI:Na, ^{133}Ba crystal. The peak wavelength of CsI:Na, ^{133}Ba crystal was approximately 420 nm.

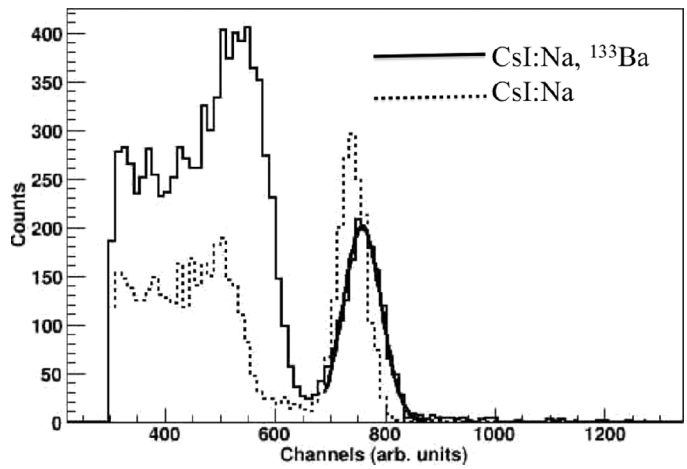


Fig. 3. It shows the energy resolution and relative light yield of the CsI:Na, ^{133}Ba crystal (solid line) for 662 keV from ^{137}Cs radioactive source. Dotted line shows pulse height spectrum of the CsI:Na crystal for 662 keV from ^{137}Cs radioactive source. The energy resolution of CsI:Na, ^{133}Ba crystal was 10% (FWHM).

measured. This measurement was carried out by the optical coupling of scintillation crystals to the PMT. Fig. 3 shows the relative light yield and energy resolution of the CsI:Na, ^{133}Ba and CsI:Na crystal with 662 keV from ^{137}Cs γ-ray source. The light yield of CsI:Na, ^{133}Ba crystal was almost same that of CsI:Na crystal. The energy resolution was 10% (FWHM). In addition, all parts of the results were the same.

C. Fluorescence Decay Time

The pulse shape of PMT output to the 400 MHz FADC was registered directly and the decay time spectrum of the CsI:Na, ^{133}Ba crystal was calculated using the recorded pulse shape information [12], [13] under ^{137}Cs γ-rays excitation, as shown in Fig. 4. The decay time of the CsI:Na, ^{133}Ba crystal consisted of two decay components and the decay curves were fitted with two exponential components. The observed decay time of the short and long decay component was 455 ns (55.6%) and 1853 ns (44.4%) as shown in Fig. 4(a).

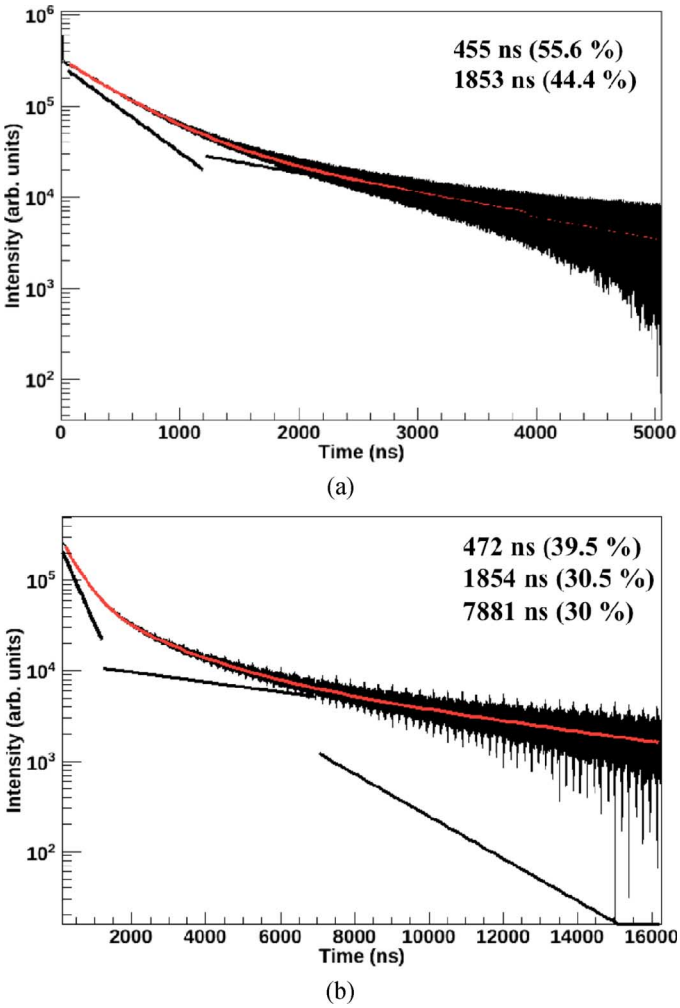


Fig. 4. The decay time of (a) CsI:Na, ^{133}Ba crystal which measured 455 ns (55.6%), and 1853.8 ns (44.4%) (b) CsI:Na, crystal which measured 472 ns (39.5%), 1854 ns (30.5%), and 7881 ns (30%).

The decay time of CsI:Na, ^{133}Ba crystal was compared with that of the CsI:Na crystal with 662 keV photons from the ^{137}Cs radioactive source. The decay time of the CsI:Na crystal consists of three components: 472 ns (39.5%), 1854 ns (30.5%) and 7881 ns (30%) in Fig. 4(b). We compared Fig. 4(a) and (b), the long decay component of CsI:Na was no longer apparent in the CsI:Na, ^{133}Ba crystal. This will be studied further.

IV. Background Identification

A. Gamma-Ray Peak Identificatorm by HPGE Detectorm

The energy peak of the CsI:Na, ^{133}Ba crystal was measured using a High Purity Ge (HPGe) detector (GMX-60220-PLUS-S) which is a N-type coaxial HPGe detector made by Ortec [14], to identify the γ-rays peaks of the ^{133}Ba radioactive source. Most coaxial HPGe detectors have entrance windows 500-μm to 1000-μm thick. The entrance window of this HPGe detector is a 0.3-μm-thick, ion-implanted contact, extending the power range of useful energies to approximately 3 keV. Fig. 5(a) shows the γ-ray transitions from ^{133}Ba to ^{133}Cs. Fig. 5(b) shows, the various γ-rays from the decay of ^{133}Ba to

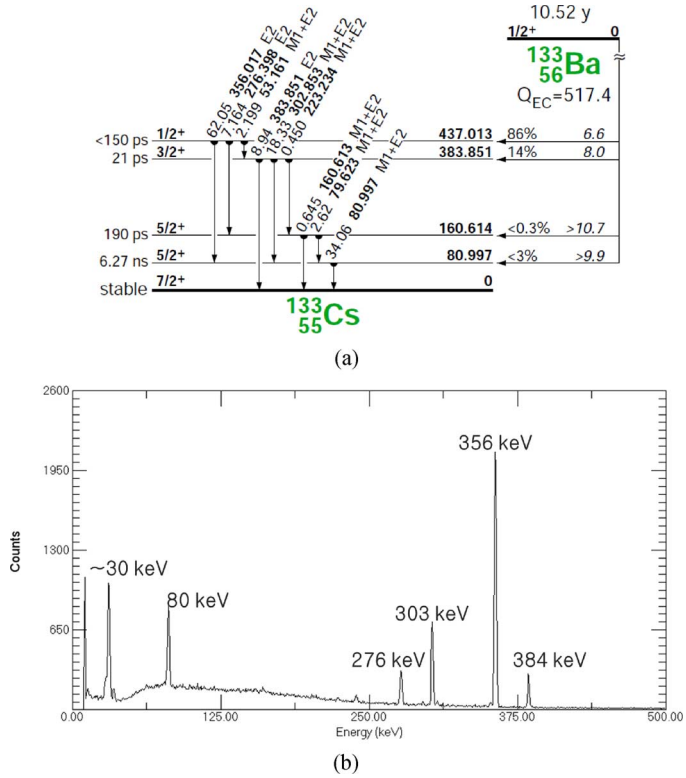


Fig. 5. (a) ^{133}Ba(10.52 y) disintegrates by electron capture to ^{133}Cs via the excited states of 437 keV and of 383 keV [15]. (b) Results of ^{133} Ba radioactive source using HPGe detector.

^{133}Cs. The peaks of the γ-rays in the CsI:Na, ^{133}Ba crystal were identified as approximately 30 keV (X-ray), 80 keV, 276 keV, 303 keV, 356 keV, and 384 keV, respectively [15], [16].

B. Comparison of CsI:Na, ^{133}Ba With CsI:Na Crystal

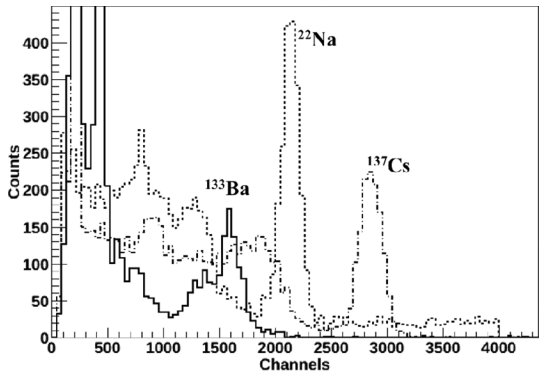
To verify the background of ^{133}Ba radioactive contained CsI:Na scintillation crystal, the background spectrum was compared with the energy spectrum of CsI:Na crystal using an external ^{133}Ba γ-ray source.

First, the pulse height spectrum of CsI:Na crystal was measured using various γ-rays. Photo peaks of 356 keV from ^{133}Ba, 511 keV from ^{22}Na and 662 keV from ^{137} Cs were observed, as shown in Fig. 6(a). Fig. 6(b) shows a comparison plot of the energy spectrum of CsI:Na crystal with a ^{133}Ba (1 μCi) source and the internal background of CsI:Na, ^{133}Ba crystal. Three main peaks were observed in the CsI:Na, ^{133}Ba crystal. The first peak at 30 keV was identified as a K-shell X-ray, the second peak was identified at 80 keV, and the third peak was the sum peak of the first and second from the ^{133}Ba radioactive source.

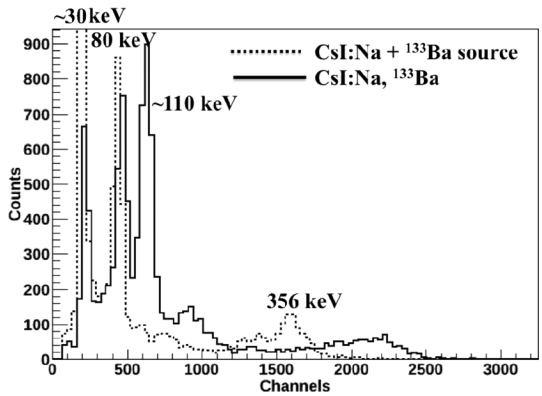
V. Conclusions

A CsI:Na, ^{133}Ba crystal was grown using the Bridgman method and cut it into three parts. The scintillation properties of the CsI:Na, ^{133}Ba crystal were measured at room temperature. The light yield of the CsI:Na scintillation crystal was also compared to identify the background of CsI:Na, ^{133}Ba.

The X-ray induced emission spectrum was measured. The maximum emission peak wavelength was approximately 420 nm, which matches well with the results from the bialkali



(a)



(b)

Fig. 6. (a) Pulse height spectrum of the CsI:Na crystal for various γ-ray source such as 356 keV from ^{133}Ba (solid line), 511 keV from ^{22}Na (dotted line) and 662 keV from ^{137}Cs (dash-dotted line). (b) Comparison of the internal radioactivity decay spectrum of CsI:Na, ^{133}Ba crystal with an energy spectrum of CsI:Na crystal irradiated with ^{133}Ba radioactive source.

PMT. The energy resolution of the CsI:Na, ^{133}Ba crystal measured using a ^{137}Cs γ-ray source was approximately 10%. The decay time of the short decay component was 455 ns (55.6%) and the long decay component was 1853 ns (44.4%).

The energy peaks of the CsI:Na, ^{133}Ba crystal were measured using a HPGe detector to identify the ^{133}Ba peak. The energy peaks of ^{133}Ba were observed at approximately 30 keV (X-ray), 80 keV, 232 keV, 276 keV, 303 keV, 356 keV, and 384 keV, respectively. Various γ-rays can be more accurate detector calibration than LSO crystal. In addition, to confirm the internal background of CsI:Na, ^{133}Ba crystal, the background spectrum of CsI:Na, ^{133}Ba crystal was compared with the energy spectrum of CsI:Na using a ^{133}Ba radioactive source. We found 30 keV (X-ray), 80 keV, and sum peak ($\sim$ 110 keV) from CsI:Na, ^{133}Ba crystal.

From the above results, show that self-triggers can be formed from the CsI:Na, ^{133}Ba scintillator signal with X-rays ($\sim$30 keV) or internal conversion electron due to the cascade transition γ-ray from the electron capture decay of ^{133}Ba to ^{133}Cs. A range of γ-rays can be used for detector calibration with a self-trigger from ^{133}Ba decay. This newly developed CsI:Na, ^{133}Ba crystal makes possible to use instead of LSO crystal in Hyperball-J experiment and another experiments .

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