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Fabrication and characterization of cubic $SrI_2(Eu)$ scintillators for use in array detectors

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ABSTRACT

Strontium iodide ($SrI_2(Eu)$) is a promising spectroscopic detector for use in both nuclear security and medical imaging owing to its excellent energy resolution and low internal background radiation. A cubic form is preferable when coupling with a silicon-based photosensor in order to build an array detector for use in applications such as Compton cameras. Here, cubic $SrI_2(Eu)$ crystals with 10 mm sides were fabricated and evaluated. The cubic $SrI_2(Eu)$ samples coupled to an avalanche photodiode exhibited an energy resolution of approximately 3.6% at 662 keV when using a shaping time of 3 μs. An increase in light output and an improvement of energy resolution were also observed at lower temperatures. The excellent energy resolution of these devices indicates that these crystals are promising potential detectors for use in Compton cameras and other imaging detectors.

1. Introduction and background

Strontium iodide doped with Europium ($SrI_2(Eu)$) is a promising scintillation detector for use in nuclear security and medical imaging applications owing to its high light output (80,000–110,000 photons/MeV), excellent energy resolution (< 4% FWHM), and absence of background radiation [1,2]. Compton cameras are used for gamma ray imaging in contaminated regions such as Fukushima. Achieving better spatial resolution to localize the contamination compared with the existing aerial measurement is required. The good energy resolution and high density (4.6 g/cm^3) of $SrI_2(Eu)$ make it a very attractive alternative to sodium iodide (NaI(Tl)) detectors. In the previous development [3] using 1 cm cube GAGG crystal the energy resolution of approximately 7–8% @ 662 keV limits the final angular resolution. Finer energy resolution is preferable also with the moderate operating count rate around 10 kHz in our practical application although there exists trade-off between sensitivity depending on the geometry and angular resolution. Compared with GAGG or CsI crystals, which are commonly used in Compton imaging, $SrI_2(Eu)$ crystals can achieve finer energy resolution and therefore, better angular resolution [4,5]. Several favorable characteristics of $SrI_2(Eu)$ when combined with a photomultiplier tube (PMT) have previously been reported [6–8], particularly relating to their use in spectroscopic applications. When building an array detector, cubic crystals are preferred, especially when coupling with a silicon-based solid photodetector such as an avalanche photodiode (APD) or a silicon photomultiplier (SiPM). A silicon-based photodetector can be used to ensure a very compact design for a gamma imaging system; however, owing to its hygroscopic properties, such a detector has not yet been fabricated in a cubic form. In this study, we present the fabrication and evaluation of a cubic $SrI_2(Eu)$ detector with 10 mm sides, coupled with an APD, in order to fabricate a novel array detector.

2. Materials and methods

2.1. Crystal preparation

In general, single crystals of $SrI_2(Eu)$ are prepared by the vertical Bridgman method in a quartz crucible. However, $SrI_2(Eu)$ has strong hygroscopic nature, and therefore, we have to remove H_2O from the raw material completely before crystal growth to obtain high quality single crystals. We have applied our original technology named “Liquinert” process for this purpose. The details of this process are mentioned in other studies [8,9]. The Liquinert process is an effective technology for removing H_2O from halide raw materials, and it is suitable to prepare high quality metallic halides scintillation crystals. We prepared several $SrI_2(Eu)$ single crystals with the size of φ15 mm and approximately 80 mm length

Abbreviations: FWHM, Full width at half maximum of a function

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that contain 1.5 mol% of Eu. The samples of cubic shape SrI_2(Eu) were prepared as follows: (A) A 10 mm cubic crystal was cut from aφ15 mm crystal, (B) all surfaces of 10 mm × 10 mm were ground with #600 SiC paper in the glove box with moisture less than 0.1%, (C) the fine powdered SrI_2(Eu) that resulted from grinding was removed by a dried gauze, (D) then, one face of the crystal was polished with #4000 fine MgO powder to obtain a clear surface, (E) the other five faces were wrapped with Teflon tape, (F) the polished surface with a silicone grease was put on the glass window of the aluminum case, (G) the crystal and the aluminum case was closed using epoxy glue after the coverage of the aluminum cap, (H) the preparation procedure was complete when the glue was fixed. Fig. 1 shows a picture of a fabricated SrI_2(Eu) detector encapsulated within an aluminum box.

2.2. Measurements using a APD, PD and PMT

The three samples fabricate in the moisture level of 0.1% were tested in the experiment and compared with the one of 2%. The setup of the experimental equipment used in this study is shown in Fig. 2. An avalanche photodiode (APD S-8664-1010 Hamamatsu Photonics) with an active area of 10 × 10 mm^2 was used for the measurements. S8664-1010 has a high quantum efficiency of up to 80% in the 400–900 nm range, with peak sensitivity at a wavelength of 600 nm [10]. The aluminum-encapsulated crystals were coupled to the APD using OKEN6262 silicone grease and wrapped in white Teflon tape. A charge sensitive preamplifier (CoolFET, Amptek A250CF) and a shaping amplifier (ORTEC 571) were connected to the detector and signals were converted using a digital multichannel analyzer (MCA 8000d, Amptek). The time constant for the shaping amplifier used in the experiments was between 0.5 and 10 μs. The dependence of the energy resolution on temperature was determined in the temperature controller box from − 10 to 30 °C, using steps of 10 °C. A temperature controller (ESPEC SH-242) was used in order to change the temperature from − 10 °C to 30 °C, using 10 °C steps. A bias voltage of approximately 400 V was supplied to the APD, using a high voltage (HV) module power supply (DT5521, CAEN) through the charge amplifier module. In order to characterize the light yield of the device, a 10 × 10 mm^2 photodiode (PD S3590-08) coupled to the crystals using OKEN626 silicone grease was employed. The PD had a sensitivity range from 340 to 1100 nm, with peak sensitivity at a wavelength of 960 nm. The relative light yield was calculated using the ratio P_{Cs}/P_{Am}, where P_{Cs} is the peak channel at 662 keV from the ^{137}Cs source, and P_{Am} is the peak channel of 59.5 keV from the direct excitation of the ^{241}Am source, from the bottom side of the detector.

Prior to the measurements conducted using the APD, the performance of three samples was confirmed using a super bialkali PMT (R6231-100 Hamamatsu Photonics Vop = +992 V), which exhibited a peak sensitivity for a wavelength of 350 nm and a sensitive area of 46 mm^2 for checking the operation. The output signals were processed with a multichannel analyzer (LEMCA1B Leading Edge Algorithm), which corresponds to the shaping time of 8 μs. The measurement was carried out at room temperature (approximately 26–29 °C).

Fig. 1. Sample of cubic SrI_2(Eu) crystal encapsulated in a cubic aluminum box.

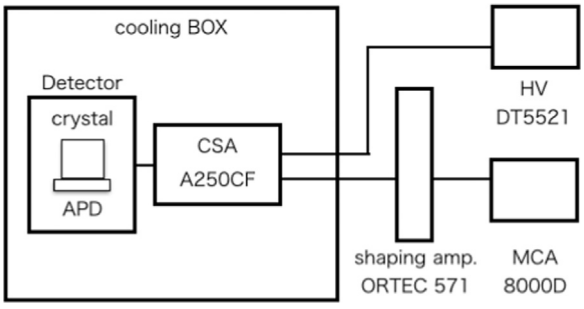

Fig. 2. Experimental setup of SrI_2(Eu) crystal samples coupled to an APD/PD device.

3. Results

3.1. Measurement and energy resolution with PMT

The measured energy resolutions for samples 1–3 prepared with 0.1% moisture condition were 3.49%, 3.48% and 3.44% with a light output of 87.2%, 100% and 97.6%, respectively with PMT. The energy resolution with the crystal prepared with 2% moisture was approximately 11%.

3.2. Measurement and energy spectra with APD

A typical measured energy spectrum obtained for the ^{137}Cs source at a temperature of 0 °C and a shaping time of 3 μs, using the 10 mm SrI_2(Eu) crystal cube coupled to a 10 × 10 mm^2 APD is shown in Fig. 3. The measurements were conducted with the average measured count rate of approximately 45.28 cps. Fig. 4 shows the typical energy spectrum for a ^{22}Na source at a

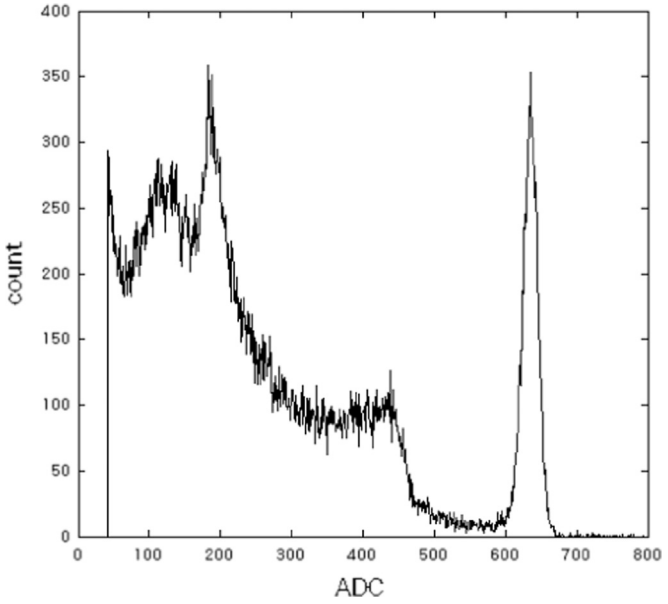

Fig. 3. Energy spectrum of a cubic SrI_2(Eu) detector coupled to an APD (HV = 351 V, 3 μs shaping time, at 0 °C), using a ^{137}Cs source.