CHARACTERISTICS EVALUATION AND GROWTH OF $BI_4GE_3O_{12}$ SINGLE CRYSTAL BY CZOCHRALSKI METHOD

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The single crystal scintillator of bismuth germinate ($Bi_4Ge_3O_{12}$:BGO) was successfully grown by the conventional Czochraski technique. The characteristics of the grown BGO were evaluated and presented on the excitation, emission responses and energy spectra of the γ-rays from ^{241}Am, ^{133}Ba, ^{57}Co, ^{22}Na, ^{137}Cs and ^{54}Mn radio-isotopes. The energy resolution of grown BGO, $\Delta E/E$, was estimated to be 12.1% at 662 keV of γ-ray for ^{137}Cs nuclide. Compared to the commercial BGO crystal, we confirmed that the grown BGO has a good performance and is comparable to reference one.

Keywords : BGO Single Crystal, Scintillator, Czochralski Method, Energy Resolution

1. INTRODUCTION

The bismuth germinate (BGO) single crystals were first grown by Nitsche in 1965 as a replacement for the bismuth silicon oxide ($Bi_4Si_3O_{12}$:BSO) a rare natural mineral [1]. BGO is a technologically important material and is widely used as a scintillating crystal for the radiation detectors. The advantages of BGO as a scintillator are its high atomic number (short radiation length), non-hygroscopic property and extremely small afterglow. At higher than 2 MeV energy of γ-rays, BGO provides better spectral resolution than NaI(Tl) [1,2]. Because of these factors, it is extensively used for the large electromagnetic calorimeters in high energy physics experiments, a detector of X-rays, gamma rays in the nuclear medicine diagnostic systems, particularly Positron Emission Tomography (PET) and Computed Tomography scanners (CTS) [3]. BGO crystals are generally grown by the Czochralski technique. The Czochralski technique has been applied to the growth of many kinds of single crystals because of its technical superiority for growing large high-quality crystals [4].

We have successfully grown a BGO single crystal of $\Phi 16 \times 33$ mm^3 by the Czochralski method with RF induction heating. In order to evaluate the performance, we have measured the emission, transmission and gamma rays spectra of the grown BGO single crystal and compared properties with those of reference one which is commercially available.

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2. EXPERIMENTAL METHODS

2.1 Crystal Growth

The BGO single crystals were grown by the conventional RF heated Czochralski technique using a self-developed crystal growth system whose rotation rates can be varied from 1 to 50 rpm and pulling rates from 0.225 to 22.5 mm/h. The starting materials used were 99.9995% pure bismuth germanium oxide powders. Pre-sintered BGO powder was melted in an inductively heated platinum crucible, 32 mm in diameter and 32 mm in length, under oxygen atmosphere. This is to prevent decomposition of the melt and corrosion of the crucible [5]. The crucible is surrounded by a RF coil, zirconia and ceramic heat-insulating material, and they were placed in a double-walled water-cooled stainless-steel chamber of 350 mm in diameter and 450 mm in length. The maximum temperature that can be reached up to 1800 ºC with the furnace and the temperature is stabilized within ±0.3 ºC. The temperature of the heating zone was controlled by a user or the temperature controller. The heating elements can be operated at considerably higher temperature than that of furnace. However, the properties of the ceramic refractories, the surrounding thermal shields and the duration of useful life of heater limit the operating temperature. The seed holder assembly was manufactured with molybdenum. The rotation and pulling of the seeds can be controlled by the manual and automatic modes. For crystal growth experiments, the BGO powder was heated in a platinum crucible for 6 hours till the melting point of BGO, 1050 ºC. At melting temperature, it was stabilized for about 30-40 minutes and the BGO seed was dipped into the melt to test the crystallization temperature. The pulling rate of seed and crystal rotation rate were 1.5 mm/h and 25 rpm, respectively. The BGO crystals were grown three times because the impurities and yellow color of BGO crystal were decreased at each time [5,8]. The first and second crystals were grown with platinum crucible of $\Phi 32 \times 32$ mm and the third crystal was grown in the platinum crucible of $\Phi 25 \times 25$ mm. The third crystal of $\Phi 16 \times 21$ mm dimension was grown successfully.

2.2 Scintillation Property Measurements

The crystals for characterizations were cut from as-grown crystals and mechanically polished using sand papers and alumina powder (particle size: 0.05 μm) to mirror finish. In order to evaluate the characteristics of the grown BGO, we compared the properties of the grown BGO and those of the commercial BGO which was bought from AMCRYS and polished at both side on the crystal surfaces. An overview of all the crystals is shown in table 1.

The surfaces of BGO crystal were optically polished to enable transmission measurements. The transmission and absorption spectra were measured using the CARY Eclipse fluorescence and CARY 300 Conc UV-visible spectrophotometer made by VARIAN. The ORTEC model 113 was used for preamplifier and model 572A for pulse shaping amplifier. The pulse shaping time was 1 μsec. The output from the amplifier was directly sent through multi-channel analyzer (MCA) of the ORTEC model 919E for gamma spectroscopy. The BGO crystals were coupled to the photomultiplier (H7415 Hamamatsu), which has a quantum efficiency of about 20% at 480 nm, with the index matching silicon optical grease for the good transmission of scintillation light and the BGO crystals were wrapped with several layers of white Teflon tape as a reflector. The energy resolution of BGO crystals were measured with ^{241}Am, ^{57}Co and ^{133}Ba, ^{137}Cs, ^{22}Na and ^{54}Mn radioisotopes. The radiation sources were located at a 10cm distance from surface of the sample crystal.

3. RESULTS AND DISCUSSION

3.1 Scintillation Property

The transmission and absorption spectra of the grown crystals were shown in Fig. 1. The transmission spectrum ranges of BGO have been generally reported from 60% to 80% transmission rate and its result showed a good agreement with that of BGO report [3,5-7]. The excitation and emission wavelength of BGO crystal were reported about 300 nm and 480 nm and we observed at 302 nm and 480 nm respectively as shown in Fig. 2.

3.2 Energy Resolution

The spectra of gamma rays were measured with ^{241}Am, ^{133}Ba, and ^{57}Co for low energy (<500 keV) and with ^{22}Na,

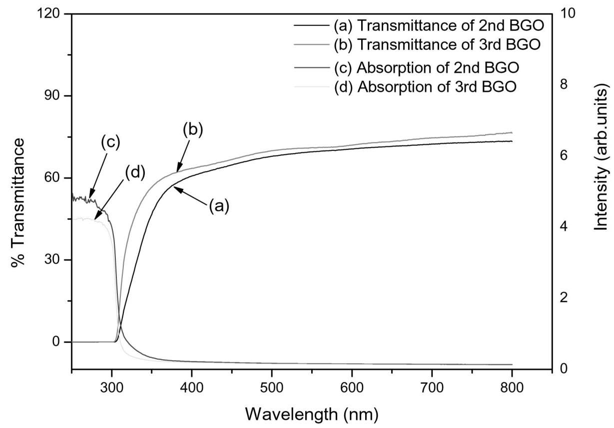

Fig. 1. Transmission and absorption spectra of the second and third grown BGO crystal.

Table 1. The tested BGO crystals.

Crystal	Size [mm^3]	Surface finish	
2nd BGO	$9.5 \times 9.5 \times 7.5$	Both side polished	Grown crystal
3rd BGO	$9.5 \times 9.5 \times 7.5$	Both side polished	Grown crystal
Reference	$10 \times 10 \times 10$	Both side polished	AMCRYS

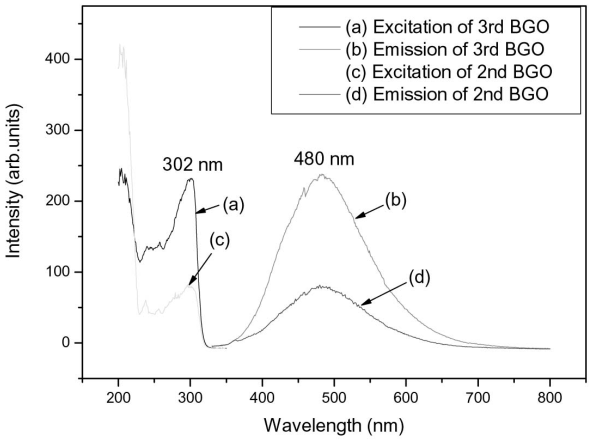

Fig. 2. Excitation and emission spectra of the second and third grown BGO crystal.

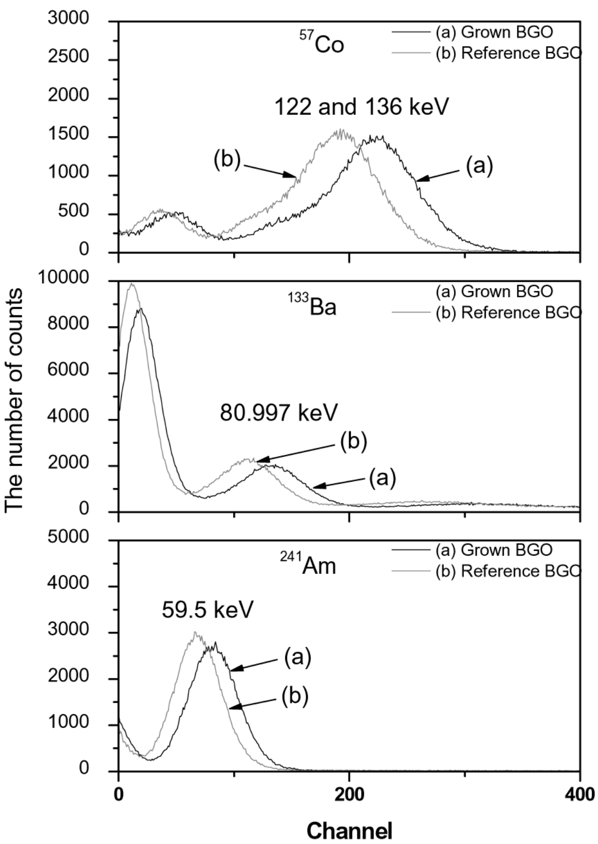

Fig. 3. Gamma ray spectra of ^{241}Am (59.5 keV), ^{133}Ba (81 keV) and ^{57}Co (122 and 136 keV).

^{137}Cs, and ^{54}Mn for high energy (>500 keV). The gamma rays spectra were shown in Fig. 3 - 6. The Fig. 3 shows that the counts and peak position of grown crystal were similar to those of reference crystal under 500 keV. The Figure 4 - 6 show that the comparison of energy spectra over 500 keV and the each pulse height spectra were measured with the

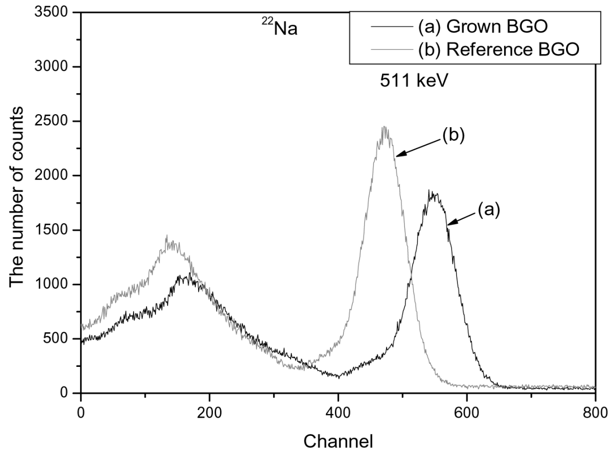

Fig. 4. Gamma ray spectrum of ^{22}Na (511 keV).

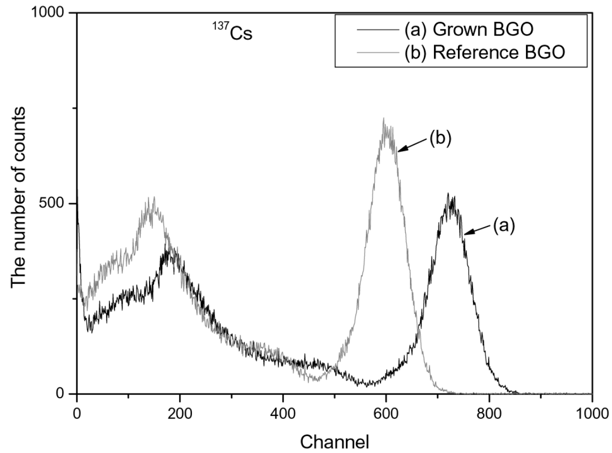

Fig. 5. Gamma ray spectrum of ^{137}Cs (662 keV).

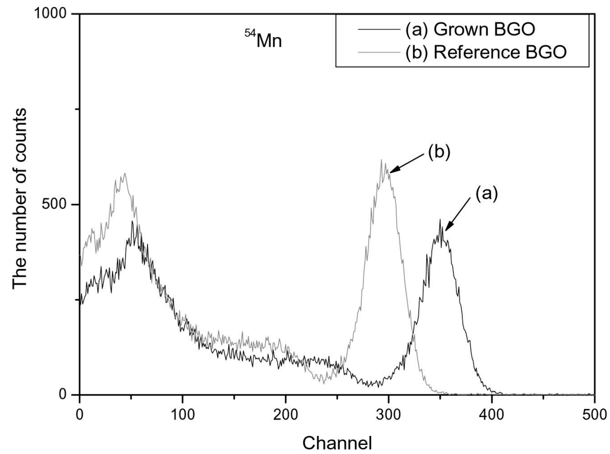

Fig. 6. Gamma ray spectrum of ^{54}Mn (834 keV).

grown BGO and reference crystals under the same gain of spectroscopy amplifier. The measured spectra shows that

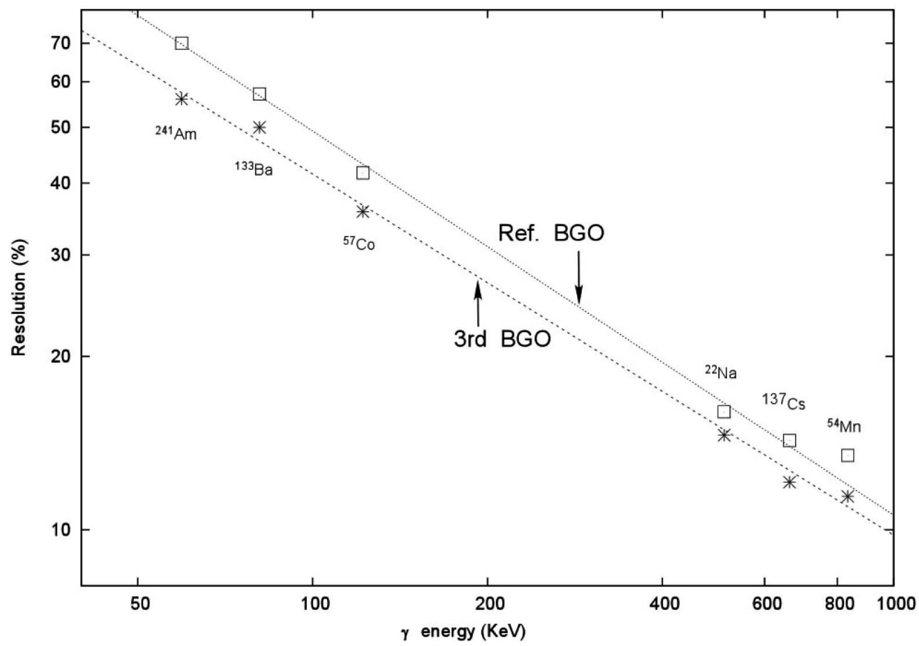

Fig. 7. Energy resolution of our third grown BGO (9.5 × 9.5 × 7.5 mm^3) and reference BGO (10 × 10 × 10 mm^3) as a function of gamma-ray energy.

the peaks from our grown crystal are positioned at the higher channel that from the reference crystal.

The energy resolutions in FWHM (Full Width Half Maximum) of individual BGO crystals are shown in Fig. 7. The energy resolution of the grown third crystal and reference crystal were evaluated 12.1% and 14.3% respectively at 662 keV of ^{137}Cs isotopes. The difference of peak position and energy resolution at same energy is considered to be due to the radiation length effect by the difference crystal size and the quality of the grown and reference BGO.

4. CONCLUSION

The BGO single crystals were grown by the Czochralski technique. For characteristics of the grown crystals, the transmission, absorption, excitation, emission, gamma-ray spectra and energy resolution measurements were carried out. The emission and excitation have a peak at around 480 nm and at 302 nm respectively, and the shape is similar to that of typical BGO. In the gamma ray measurements, six different radioactive sources, ^{241}Am, ^{133}Ba, ^{57}Co, ^{22}Na, ^{137}Cs and ^{54}Mn, were used in order to measure the crystal's radiation responses. The energy resolutions were measured as 12.1% in our BGO crystal and 14.3% in reference crystal at 662 keV γ-ray of ^{137}Cs radiation source. The experimental results indicate that the quality of our BGO single crystal have a good performance, which is comparable to that of reference BGO.

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