Recent R&D on Oxide Scintillation Crystals for Radiation Detectors

M. Ishii[1)], M. Kobayashi[2)], K.Hara[3)], M. Tanaka[3)], I. Yamaga[4)], K. Miwa[4)],
H. Ishibashi[5)], Y. Usuki[6)], Y. Hirose[1)] and K. Harada[1)]
1) SIT, Shonan Institute of Technology, Fujisawa 251, Japan
2) KEK, National Laboratory for High Energy Physics, Tsukuba 305, Japan
3) Institute of Physics, University of Tsukuba, Tsukuba 305, Japan
4) Futeck Furnace Inc., Kanazawa, Yokohama 236, Japan
5) Hitachi Chemical Co., Ltd. Tsukuba Research Laboratory, Tsukuba 305, Japan
6) Furukawa Co., Ltd. Yoshima, Iwaki 970-11, Japan

1. Introduction

Scintillation crystals such as NaI:Tl and $CdWO_4$ are adapted for medical imaging apparatuses of XCT, PET and others to which X-ray have been applied X-ray diffraction apparatus for materials. They demand the development of a further up-to-dates scintillators in the nuclear and high energy physics experiments.

The scintillation crystals has a role of transmitting fluorescence to a photon-electron transducer in addition to the function of luminescence upon absorbing radiation energy. For this reason, the scintillator must be made of such transparent and optically high quality material free form absorption and scattering against fluorescence and having a full length to stop radiation to be entered. Such a scintillator can be realized by single crystal manufactured under optimum growth condition.

Table 1 shows the characteristics for scintillation crystals to be used for making studies on nuclear and high energy physics experiments. NaI:Tl was utilized for crystal balls of SPEAR of SLAC and BGO ($Bi_4Ge_3O_{12}$), for L3 of CERN. At present, CsI:Tl is being utilized for the manufacture of detector of SLAC(BaBar) and KEK(BELLE). Furthermore, they are investigating the possibility of adopting high quality PWO($PbWO_4$) of 13 m^3 for CMS(Compact Muon Solenoid) in LHC of CERN, recently.

Table 1 Characteristics of typical scintillators for nuclear and high energy physics applications.

Quantity	NaI:Tl	CsI:Tl	$Bi_4Ge_3O_{12}$ (BGO)	$Bi_4Si_3O_{12}$ (BSO)	Gd_2SiO_5:Ce (GSO:Ce)	$PbWO_4$ (PWO)
Density(g/cm^3)	3.67	4.53	7.13	6.8	6.71	8.2
Radiation length X0 (cm)	2.59	1.86	1.12	1.15	1.38	0.92
Decay constant (ns)	230	1050	300	100	30-60	<10/<40
Peak emission (nm)	415	550	480	480	440	430
Light yield (relative)	100	85	7-10	2	20	0.26/0.04
Peak excitation (nm)	290	300	280	285	310,350	325
Melting point(°C)	651	621	1050	1030	1900	930
Radiation hardness (rad)	10^3	10^3	10^{4-5}	10^5	$>10^8$	$>10^8$
Cleavage	(100)	none	none	none	(100)	(101)

Making a research collaboration, we have proceeded with R&D of oxide scintillation crystals. We are making studies on BSO($Bi_4Si_3O_{12}$), GSO(Gd_2SiO_5:Ce) and PWO recently and proceeding with our development work of such a large crystal in possible to use practically.

2. BSO ($Bi_4Si_3O_{12}$)

The representative material in practical use of oxide scintillator is BGO. BGO scintillation crystal is large absorption coefficient for radiation and in light output of fluorescence. For this reason, BGO was applied to XCT at an early stage and is now used for industrial sensing apparatus and nuclear and high energy physics experiments ones besides PET.

Disadvantage of BGO is the slow decay of fluorescence such as 300ns. We has been studied on the crystal growth by Bridgman method for BSO crystals which is fast decay and so small as approx. 1/3, as a similar other characteristics of BGO.

Bi_2O_3 and SiO_2 are used as raw materials for crystal growth. After mixing and sintering them, we produced $Bi_4Si_3O_{12}$ of poly crystal and then, fed it in a Pt crucible and grew crystal. As for crystal growth, it descends at 0.5 mm/h in the crucible with a temperature gradient of approx. 10℃/cm.

A segregation phases of $Bi_{12}SiO_{20}$ was yield on the surface of grown crystals and a SiO_2 phase, yielded at the last stage of crystallization. There is no segregation phase inside ingot and transparent crystal was observed. Fig.1 is a photograph of a crystal from which a scintillation module to be used in high energy physics experiments has been cut and polished from an ingot. Uniformity of light output in emission by γ -ray was a problem in the performance of scintillator. The value was in the ±1.5% which is a satisfactory value for high energy physics experiments.

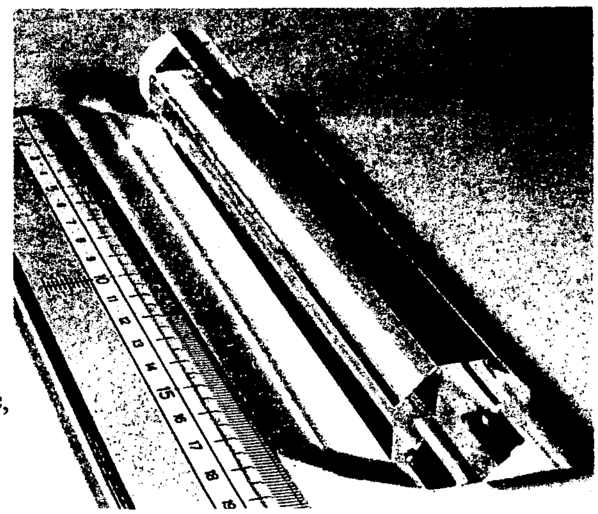

Fig. 1 Photograph of a large BSO crystal.

3. GSO (Gd_2SiO_5:Ce)

Ce doped GSO was a scintillation crystal found by Takagi and Fukazawa of Hitachi and developed by Hitachi Chemical. GSO is relatively larger absorption coefficient as compared with NaI:Tl and higher light output and faster decay as compared with BGO. Over ally, scintillation characteristics are very superior to others. GSO has been put into practical use for γ - ray detector for oil logging sensors. We are making studies on the crystal growth for the purpose of application of GSO to nuclear and high energy physics experiments and further, in the field of medical imaging apparatus.

The melting point of GSO is 1950 ℃ and a Ir crucible is used for crystal growth. Crystal is grown by CZ method. A long scintillation crystals 2x2x38 cm^3 is required for GSO to be used in high energy physics experiments. As for the studies on the crystal growth as a scintillator, investigation was made at first on the Ce concentration for optimization of an light output by luminescence. The largest problem in crystal growth is that the anisotropy of thermal expansion coefficients is large because the crystal structure of GSO is monoclinic, resulting in having a cleavage fracture and therefore, cracks tend be caused in the ingot under growth and cooling of crystal. The cracks of an ingot are a further difficult problems in the production of longer and larger scintillation crystals. In order to prevent cracks from being generated in an ingot, we made calculation on thermal stress generated in an ingot from the temperature distribution during growth of crystal and adopted the optimum growth station for the purpose of prevention of cracks from being caused.

Consequently, we could grow such a large crystal as free from cracks, having 8cm in diameter and 28cm in length, as shown in Fig. 2. Collecting four modules of GSO for high energy experiments of 2x2x20 cm^3 from a grown crystal. In addition to above, we made studies on the radiation damage by high energy particles and γ -ray with 10^8 rad from ^{60}Co and confirmed that GSO has a high resistance to radiation. In view of the results of this studies, GSO was found possible to be applied to the γ -ray detector in nuclear and high energy physics experiments field.

Fig. 2 Photograph of a large GSO:Ce crystal.

4. PWO ($PbWO_4$)

PWO is under study recently as a scintillation crystal for detector of high energy particles in LHC of CERN. PWO is also larger density as 8.2 g/cm^3 and so faster as 15 ns in decay of luminescence. However, the demerits of this materials were small light output and weakness in radiation damage. At present, we are making studies on the improvement of scintillation characteristics of this PWO.

In this studies on improvement on characteristics of scintillation crystal, we developed a technology of high purity not including impurities at first. Consequently, we confirmed that there was emission wavelength (510 nm) due to impurities of Mo. in grown crystal. There is an absorption in 430 nm close to 440 nm in the emission wavelength, proving that the deterioration due to radiation damage. Thinking that the cause of this absorption from Pb vacancies, we tried to improve optical absorption in the vicinity of emission wave length by addition of La^{3+}. PWO:La improves markedly the transmittance associated with scintillation characteristics as shown in Fig.3.

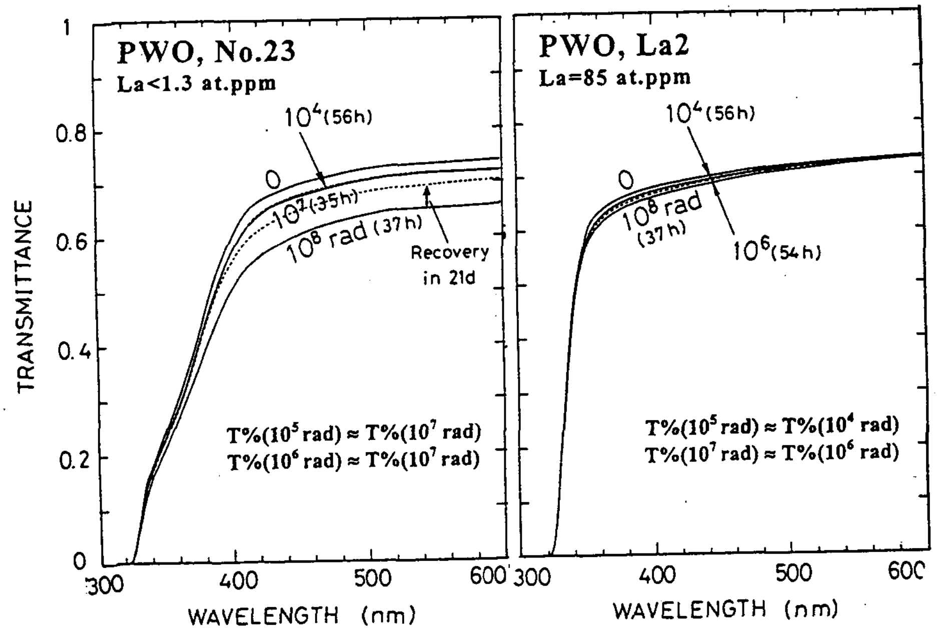

Fig. 3 Improvement in transmittance and radiation damage of La doped $PbWO_4$ crystals.

PWO:La has improves the radiation damage too. Fig.3 shows the transmission spectra after irradiation of ^{60}Co γ -ray. For a scintillator for high energy particles detectors, a large crystal of 2x2x20 cm^3 is required for PWO crystal. We making studies on the process technology of lattice defect and high purity as mentioned here above and development of growth technology of ϕ 80x280mmL large crystals by CZ method at present. As for the studies on the crystal growth of PWO:La, we are making studies on also Bridgman method besides the above.

5. Summary

Scintillation crystals for industrial field are used in fundamental physics i.e. nuclear and high energy physics experiments besides the medical imaging, process control and gauging, container inspection, mineral process etc. For the reason of limited marketability, there are not so many studies with emphasis placed on the crystal growth . The scintillation crystal is an important theme in the studies in the fundamental physics and researchers for crystal growth are expected participate it.

The present work is partly supported by a Grant-in-Aid from the Japanese Ministry of Education, Science and Culture.

References

1) M. Ishii and M. Kobayashi, Prog. Crystal Growth and Charact. 23(1991) 245.
2) M. Kobayashi et al., Nucl. Instr. Meth. 206(1983)417 and ibid. A372(1996)45.
3) M. Ishii et al. SPIE Proc. Inter. Soc. Optical Eng., 2305(1994) 68.
4) K. Kurashige, Y. Kurata, H. Ishibashi and K. Susa, Jpn. J. Appl. Phys. 36(1997)2242.
5) M. Kobayashi et al. Nucl. Instr. Meth. A 333(1993)429 and A373(1996)333.