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Radiation Damage of Cerium Doped Lanthanum Bromide Crystal

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In this paper, the radiation damage of the $LaBr_3$: Ce crystals with different cerium doped concentration are investigated. One is the $LaBr_3$: 3 % Ce crystal. The sizes of this crystal reach to 11 mm × 11 mm × 13 mm. One is the $LaBr_3$: 5 % Ce crystal. This crystal is a cylindrical crystal with sizes Φ 25 × 15 mm^3. The optical and scintillation properties of the $LaBr_3$: Ce crystal, such as light output and energy resolution, were measured before and after γ-ray irradiations at different doses. The crystals both display lower photoelectron output and poorer energy resolution after irradiation

Keywords: $LaBr_3$ crystal, Cerium concentration, Energy resolution, Light output, Radiation damage.

INTRODUCTION

Cerium activated lanthanum bromide, $LaBr_3$: Ce, discovered in 2001 by Delft University of Technology[1], has already been established as one of the most promising candidates because of its high light output (61,000 ph/Mev), superb energy resolution (2.8 %) and fast decay time (19 ns)[2]. Those properties are superior to NaI: Tl crystal which is one of the most commonly used scintillators.

The radiation hardness is also very important for the scintillator. So far, there are few articles reported radiation damage of the $LaBr_3$: Ce crystals. The $LaBr_3$: Ce crystals have excellent resistance to proton irradiation damage[3]. When exposed to simulated solar proton flare spectra with fluencies up to 1012 protons cm^{-2}, integrated above a 60 MeV threshold, the crystals are radiation tolerant showing no measurable degradation effects. When the $LaBr_3$: Ce crystal is irradiated by γ-ray, it also shows good radiation hardness. In case of 3.4 kGy, $LaBr_3$: Ce crystals have no permanent modification of energy resolution and color change. After 72 h, the properties of the crystal will return to the previous[4,5]. In the packed crystal, it also reduces the light output by almost one quarter and increases the energy resolution by 1.6 %.

Many factors will affect the radiation damage. One of the important factors is the concentration of the doping ions. In the $LaBr_3$:Ce crystal, the concentration of the cerium should not be very low because the scintillation emission comes from the cerium. But the concentration of the cerium should not be very high because of the concentration annealing. In this paper, the radiation damage of $LaBr_3$: 3 % Ce (sample 1) and $LaBr_3$: 5 % Ce (sample 2) crystal were investigated. The light output and energy resolution before and after irradiation were showed. The crystals both display obviously lower photoelectron output and poorer energy resolution after irradiation.

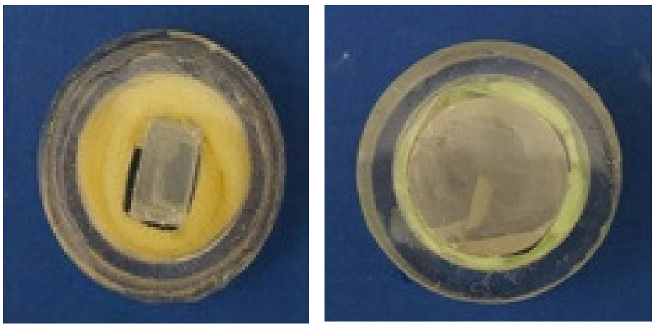
EXPERIMENTAL

The crystals were grown by the vertical Bridgman method in a resistance-heated furnace[6]. The sealed quartz ampoule was put in the furnace which was composed of three temperature zones, including high temperature zone, temperature gradient zone and low temperature zone. The high temperature zone was used to melt the raw materials. When the temperature field reached to a steady state, the quartz ampoule started to descend slowly. The crystal was grown in the temperature gradient zone at a predetermined rate of 8 mm/day. After completing growth, the furnace was cooled down to room temperature at a speed lower than 30 K/h. The as-grown crystal is shown in Fig. 1.

The Ce^{3+} molar ratio in the sample 1 is 3 %. The size of the sample 1 is 11 mm × 11 mm × 13 mm. The Ce^{3+} molar ratio in the sample 2 is 5 %. The sample 2 is a cylinder crystal. The diameter is 25 mm. The length is 15 mm. The sample 2 cracked during the machining process. The picture of samples 1 and 2 are shown in Fig. 2.

In order to collect the light effectively and prevent deliquescence, four sides of the crystal were covered by the high reflective films firstly. Then it is wrapped with sponges. Then the packed crystal was sealed in the glass tube with transparent quartz slice at both sides.

Fig. 1. Sample 1 (left) and Sample 2 (right) before processing



(a) (b)

Fig. 2. Sample 1 (a) and Sample 2 (b)

Scintillation light output was measured before and after irradiation by using a Hamamatsu R1306 PMT with a bi-alkali photo-cathode and a borosilicate glass window for samples 1 and 2. A collimated ^{137}Cs source was used to excite the crystal. The energy resolution was obtained by the Gaussian fit. The measurement results were calibrated by the Saint-Gobain crystal. Some main scintillation properties of the sample 1 and the sample 2 before irradiation are presented in Table-1.

The γ-ray irradiations were carried out respectively under a 50 curie ^{60}Co source for irradiations at 30 rad/h and a 7000 curie ^{137}Cs source for irradiations at 7,000 rad/h. The dose rates were calibrated. Before the irradiation, the crystals were placed in a dark environment for cooling down. After each round of the irradiation, the crystals were also placed in a dark environment for cooling down. The cooling time is different according to the different dose.

RESULTS AND DISCUSSION

Light output: Fig. 3 shows the light output of sample 1 and sample 2 irradiated with γ-ray source at different doses. The light output was normalized. In Table-2, it can be observed that the light output of sample 1 decreases gradually with the increase of irradiation dose. It reflected the changes of the light output clearly. When the dose is 10^2 rad, the light

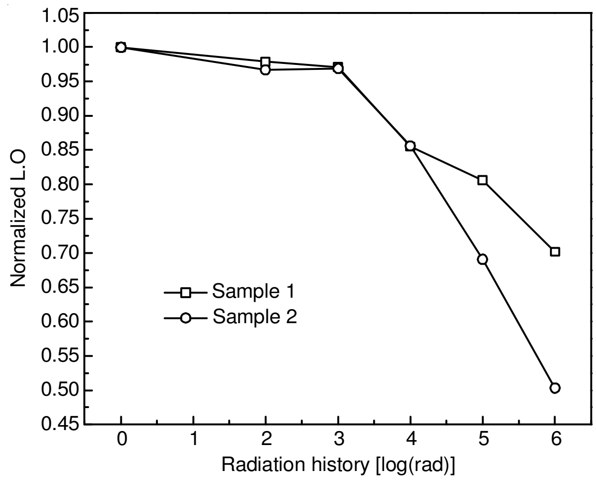


Fig. 3. Normalized light output of sample 1 and sample 2 after different radiation

TABLE-1
SOME INITIAL PROPERTIES OF THE SAMPLE 1 AND SAMPLE 2

$LaBr_3$/Properties	Size (mm)	Ce^{3+} concentration	L.O (Pe/Mev)	Decay time (ns)	Energy resolution @662 keV (%)
Sample 1	11 × 11 × 13	3%	5051	19.6	5.5
Sample 2	Φ25 × 15	5%	4866	19.7	7.3
Saint-Gobain.	Φ 25 × 25	5%	6290	19.0	3.7

TABLE-2
LIGHT OUTPUT AND ENERGY RESOLUTION OF SAMPLE 1 AND SAMPLE 2

Dose(rad)	Sample 1		Sample 2	
	L.O (p.e./Mev)	Resolution@ 662 keV	L.O (p.e./Mev)	Resolution@ 662 keV
0	5,136	5.5	4,765	7.3
10^2	5,028	11.7	4,609	7.7
10^3	4,985	11.8	4,620	9.4
10^4	4,392	14.8	4,091	10.1
10^5	4,142	19.4	3,292	11.1
10^6	3,609	21.4	2,397	17.1

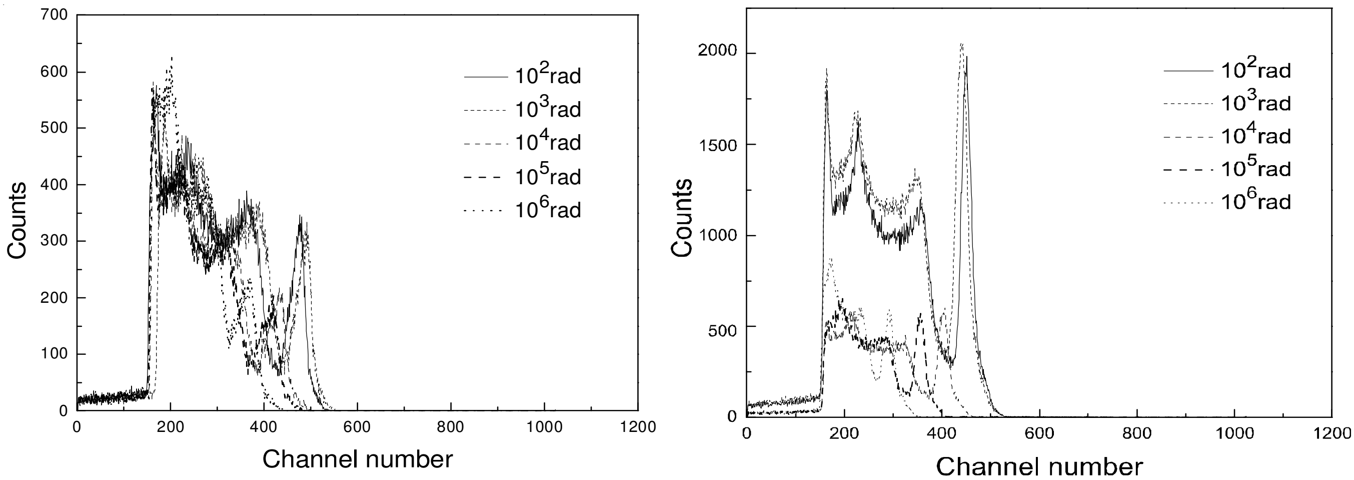


Fig. 4. Energy resolution of sample 1 (left) and sample 2 (right)

output of sample 1 only decreases by 3 %. The second exposure, delivering a next 10^3 rad dose, produces relatively more additional damage. Also the further irradiations, terminating at a cumulative dose of 10^6 rad, as much as 29.8 % of the output is lost. In the end, the light output has decreased to 70.2 % of the initial.

The light output of the sample 2 after different radiation was also showed in the Table-2. In the same way, the light output of sample 2 gradually decreases with the increase of the irradiation dose. When the dose is under 10^2 rad, the light output only drops 3.3 % and is similar to sample 1. However, the total loss of its output after 10^6 rad gamma irradiation is close to 50 %, which is indeed a huge value compared to 29.8 % measured in the sample 1.

The light output of sample 1 and sample 2 show a noticeable damage under 10^6 rad dose. Comparing sample 1 and sample 2, the sample 1 has better radiation hardness in the light output.

Energy resolution: In the whole process of irradiation, no change of the colour of the crystal, of the interface or the reflector was observed under the irradiation.

In Fig. 4, we have plotted the energy resolution of sample 1 at 662 keV for an increasing integrated dose. The experimental data are also shown in the Table-2. The most significant change of energy resolution takes place after the first irradiation, *i.e.*, the 10^2 rad dose. The resolution increases by 6.2 % percentage points (pp) for sample 1. The next irradiation, supplying a ten times higher dose of 10^3 rad, produces less additional damage, particularly the resolution goes up by only 0.1 %. The last dose of 10^6 rad seems to be more harmful to both materials, increasing resolution to 21.4 % for sample 1. Seemingly after a kind of a "damage shock" following the first irradiation, the damage increases linearly with the delivered dose.

In Fig. 4 and Table-2, they also show the energy resolution of sample 2 at 662 keV under the different doses. The energy resolution increases by 9.8 % from 7.3 to 17.1 %. The result that sample 2 exposed to γ-ray source differs from sample 1. Under the last dose, crystal also has heavy damage. The energy resolution increases by 6 %. The other doses all increase by 1-2 %.

Both crystals show obvious radiation damage after the γ-ray irradiation. And the sample 2 has larger loss in the light output.

Several factors affect the measurement results of the scintillation properties, such as the way of the crystal package, the relative position of the crystal to the PMT, the sizes of the crystal, *etc*. The quality of the crystal is the dominating factor among all the factors. More crystal defects will capture more electrons and holes that are inevitably induced into the crystal lattice by the irradiations. The cerium concentration in the $LaBr_3$:Ce crystal inevitably affect the crystal quality. But too low cerium concentration means the low light output of the $LaBr_3$:Ce crystal. And too high concentration lead to the self-absorption of the cerium.

Conclusion

The γ-ray induced radiation damage in $LaBr_3$: Ce crystals are evaluated. After receiving a dose of at least 10^2 rad from γ-ray, both the $LaBr_3$:3 % Ce crystal and $LaBr_3$:5 % Ce crystal show the deterioration of light output and energy resolution. Under the irradiation dose of 10^6 rad, the light output reduced by 29.8 and 49.7 %, respectively. Meanwhile, the energy resolution of each crystal increases by 15.9 and 9.8 %, respectively.

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