Study on High-Temperature Performance of LaBr₃(Ce) Scintillators

Y Hou¹, S Liu¹, H Yuan¹, Q Gu¹, C Zhang¹, Z Fang², M Zhang¹
¹Beijing Glass Research Institute, Beijing, 101111, China
²Beijing Industrial Technology Research Institute, Beijing, 101111, China
Email: liushan@bitri.cn

Abstract. In this paper, the relative light output, energy resolution and count rate of encapsulated LaBr₃(Ce) crystals were tested under the temperature from 25 °C to 175 °C using ¹³⁷Cs γ radioactive source. The variation of high temperature scintillation performance of LaBr₃ and NaI(Tl) crystal packages was measured and analyzed. The results show that the decline of light output of LaBr₃(Ce) scintillation crystal is about 80% of that of NaI(Tl) at 175 °C. The energy resolution of LaBr₃(Ce) at high temperature 175 °C is approximately 8%, which is better than the 9.9% energy resolution of NaI(Tl) scintillation crystals at room temperature 25 °C. The count rate of LaBr₃(Ce) is about 5-6 times more than that of the NaI(Tl) crystal.

1. Introduction
The scintillation properties of most inorganic scintillation crystals are susceptible to temperature effects. When the ambient temperature is significantly higher than room temperature, the dependence of crystal scintillation performance on temperature is particularly prominent. The influence is more significant as the temperature rises, and the temperature effects of different scintillation crystals are completely different. Some industrial application, such as petroleum logging, requires a scintillation crystal with a small temperature effect [1]. Scintillation crystals based on high temperature environment applications mainly include NaI(Tl), CsI(Na), BGO and GSO(Ce), etc.. Among these crystals, NaI(Tl) scintillation crystal is the most commonly used scintillator in high temperature logging due to its high light output and excellent energy resolution in high temperature environments. However, as the detection environment continues to increase the demand for key properties such as scintillation crystal light output, energy resolution, and count rate, new scintillation crystals with superior performance are expected.

Recent years, a series of novel halide scintillation crystals with excellent scintillation properties were developed. All the investigations about these novel halide crystals indicate that they show merits of high light yield and good energy resolution. LaBr₃(Ce) scintillation crystal has a density of 5.0 g/cm³, alight output of ~65000 photons/MeV, a decay time of 20 ns, and a peak emission of 380 nm [2-4]. It has good resolution for both X-ray and γ-ray, and have been gradually applied to high-energy physics, nuclear physics, and nuclear medicine. But LaBr₃(Ce) scintillation crystal is extremely hydroscopic in air environment and must be encapsulated and used like the NaI(Tl) crystal. For the expected substitute material in high temperature application, LaBr₃(Ce) has strong advantages in light energy output, energy resolution, count rate, and decay time compared to NaI(Tl) scintillation crystals.
Thus, investigation on LaBr₃(Ce) scintillation crystal in high temperature environment application require more detailed verification of the experiment.

In this paper, we investigate scintillation performance of LaBr₃(Ce) scintillation crystal in high temperature environment. The traditional NaI(Tl) scintillation crystal was used as a reference to explore LaBr₃(Ce) scintillation crystal as a substitute of NaI(Tl) in the applications of high temperature nuclear detection environment. The energy spectrums of LaBr₃(Ce) and NaI(Tl) scintillation crystals at different temperatures were obtained through the experiments. The variation of relative light output, energy resolution, count rate and deviation of count rate with temperature of LaBr₃(Ce) and NaI(Tl) scintillation crystals are analyzed.

![Figure 1. LaBr₃(Ce) scintillation crystals with high temperature encapsulation.](image)

2. Materials and Method

2.1. Samples

The self-made LaBr₃(Ce) scintillation crystal was measured in the experiment, as shown in Fig.1. The high hydroscopicity and the high temperature environment application require special encapsulation for LaBr₃(Ce) scintillation crystals different from that for room temperature environment application. Titanium alloy and K9 glasses were used as shell and light exit window respectively. A NaI (Tl) scintillation crystal of high temperature encapsulation produced by Saint-Gobain was used as comparative sample. The detailed sizes of all the samples we used in this experiment are listed in Table 1.

| Sample | Crystal     | Size          |
|--------|-------------|---------------|
| LBC-1  | LaBr₃(Ce)   | Φ28 mm × 28mm |
| LBC-2  |             | Φ22mm × 27mm  |
| LBC-3  |             | Φ11mm × 6mm   |
| LBC-4  |             | Φ25mm × 51mm  |

Table 1. The size of all the scintillation crystal samples.
2.2. Measurement device
Figure 2 is the experimental system we set up for the measurement of scintillation performance under high temperature. The photomultiplier used in the experiment is R1288A-07, which Hamamatsu Co. Ltd. developed for high temperature application. Its maximum working temperature was 175 °C. The R1288A-07 is connected with the system through positive high voltage mode. A 100 kΩ resistor and a 4.7 nF high voltage (3 kV) capacitor are used as the load resistance $R_L$ in the external circuit and DC blocking capacitor at the anode output respectively. RC filtering is applied to the high voltage output. The test system uses high voltage power supply Ortec 556 to provide operating voltage for the photomultiplier tube. The photomultiplier output signal is input to the main amplifier Ortec 672 after preamplifier Ortec 113, and the converted data is acquired by multichannel analyzer Ortec 926.

The glass exit window of the crystal sample is coupled with the photomultiplier entrance window by DC200 silicone oil. The coupled detector is placed in a compact dark room which is fixed in electric thermostatic drying oven. $^{137}$Cs radioactive source was placed on the upper end of the crystal. The oven was heated to 175 °C as the heating rate of 2 °C / min from 25 °C and thermostated for 30 minutes at every 25 °C. The working voltage of the photomultiplier tube R1288A-07 is positive high voltage of 1400 V. The input capacitance of preamplifier Ortec 113 is adjusted to 500 pF. The shaped amplifier Ortec 672 is set as a gain of 30 times and a forming time of 0.5 μs. The scintillation performance of the crystal sample at 662 keV γ-ray was mainly studied by the way of perpendicular incidence. The oven heating program is started after the circuit system stabilizes, and the output signal of the detector at every 25 °C is collected.

3. Results and discussion
3.1. Relative light output
The charged particles or gamma rays with energy $E$ are incident on the scintillator to excite $N$ photons with an average energy of $\varepsilon$. The luminous efficiency of the scintillator can be expressed by Eq. (1).

$$\eta = \frac{N \times \varepsilon}{E}$$

(1)
In the actual measurement, the photon number $N$ and the photon average energy $\varepsilon$ are difficult to measure simultaneously. Considering the sensitivity of the measuring instrument and other conditions, the relative light output is usually used to evaluate the luminescent properties of the scintillator [5]. Combined with our high temperature experiments, the definition of relative light output can be briefly described as the ratio of the pulse amplitude of the sample of the scintillator crystal at each measured temperature point to the pulse amplitude at the initial temperature ($25 \, ^\circ\text{C}$). That is to say, we can obtain the change of light output on each measured temperature point by measuring the $\gamma$ ray energy spectrum at 662 keV and calculating the ratio of the all-energy peak position under different measured temperature points to the all-energy peak position the initial temperature.

When the scintillator detector is irradiated by a $^{137}\text{Cs}$ source, the output pulse amplitude spectrum consists of an X-ray peak of 32 keV, a Compton platform, and a gamma-ray peak of 662 keV. The relative light output of the scintillation crystal sample was characterized by the 662 keV gamma ray full energy peak obtained by the detector. Using the Gaussian distribution function defined in the MAESTRO software to fit the region where the 662 keV gamma ray energy spectrum peak is located, the site of the distribution center(channel) is obtained, which is the relative light output value of the crystal sample.

![Figure 3. Pulse amplitude spectrum of LaBr$_3$(Ce) crystal (LBC-3) with temperature.](image)

Figure 3 and Figure 4 show the pulse amplitude and $^{137}$Cs spectrum of sample LBC-3 ($\Phi$22 × 27 mm LaBr$_3$(Ce)) at each temperature point. It can be seen that the light output of the LaBr$_3$(Ce) scintillation crystal gradually decreases as the temperature increases. The temperature effect of the scintillation crystal, the temperature effect of the photomultiplier tube, decrease in the transmittance of the couplant and the reflectance of the reflective material caused by the increase in temperature, all contributes to the reduction of the overall output signal amplitude of the detector.

Figure 5 shows the variation of the full energy peak position (channel) of all the crystal samples with temperature. It can be seen that although the sizes of six LaBr$_3$(Ce) crystal samples are different, the relative light output is substantially close, which indicates that the relative light output changes basically due to the temperature effect of the scintillation crystal. The light output at 25 °C of NaI(Tl) is used as a reference for 100% and the light output results of other temperature are normalized. Thus the relative light output values of all the crystal samples are calculated and comparable. The relative light output of LaBr$_3$(Ce) decreased slightly by 13.95%-25.6% in the temperature range of 25 °C - 100 °C, and decreased sharply by 47.12%-53.97%at the temperature range of 100 °C -175 °C. Correspondingly, the relative light output of NaI(Tl) degraded close to one-third of the initial, and drop more significantly by 66.66% at 175 °C.
3.2. Energy resolution

In the low-energy nuclear physics experiment, the single-energy particle with energy E is incident on the scintillator and is fully absorbed. After the excitation of the scintillator, the amplitude of the output pulse is statistical distribution due to the statistic process of the output signal received by optical transmission and photodetector [5]. $\Delta p$ represents the full width at half maximum (FWHM) of the distribution spectrum, $p$ represent the peak position value of the distribution spectrum, $\sigma$ represents the standard deviation of the Gaussian distribution, the energy resolution of the scintillation detector can be calculated by formula (2) [5-7].

$$\frac{\Delta E}{E} = \frac{\Delta p}{p} = 2.35 \frac{\sigma}{p}$$  \hspace{1cm} (2)

For the energy spectrum distribution of 662keV, the Gaussian function can be directly used to make single-peak fitting, and the standard deviation and distribution center value of the fitted
parameters are obtained. Thus, energy resolution of the measured crystal sample at 662 keV γ-ray can be calculated by formula (2).

From Figure 6, it can be seen that the γ-ray energy resolution of the crystal samples at 662 keV increase with the gradual increase of the ambient temperature. When the temperature in the oven is below 75 °C, the energy resolution of the scintillation crystal sample is relatively less affected by temperature. However, when the temperature rises above 100 °C, the energy resolution of the sample increases linearly, and the energy resolution at 175 °C deteriorates to 1.33 times of the initial time. The average energy resolution of three sizes of LaBr₃(Ce) scintillation crystals in the extreme high temperature environment of 175 °C is 8.38%, 8.15% and 8.27%, which is still better than that (9.9%) of NaI(Tl) scintillation crystal at 25 °C.

![Figure 6. The variation of energy resolution of all the scintillation crystals samples with temperature.](image)

![Figure 7. The variation of count rate of all the scintillation crystals samples with temperature.](image)

### 3.3. Count rate

It is well known that the count rate of scintillation crystals is proportional to the size of the crystal. Figure 7 shows the change in the count rate of each crystal sample in the 0-1.4 MeV spectrum range...
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with temperature. It can be seen from the figure that the average counts of the energy spectrum of Ø28×28 mm and Ø22×27 mm LaBr3(Ce) crystals are 669.61 cps and 506.89 cps, respectively, which is about 6 times and 5 times more than the count rate of Ø25×50 mm NaI (TI). The count rate of Ø11×6 mm LaBr3(Ce) crystals is lower than that of Ø25×50 mm NaI(Tl) mainly due to the large size gap.

The light exit window of encapsulated crystal and the photomultiplier tube housing are made of glasses which contain a certain amount of 40K radioisotope. 40K is unstable and will spontaneously generate 40Ar and 40Ca through β decay and emit γ-ray and β-rays with energy of 1.31 MeV and 1.46 keV, respectively[8]. In order to reasonably compare the 137Cs energy spectrum of each crystal sample, the count rate of the 0–1.4 MeV energy segment in the 137Cs spectrum of the crystal sample was selected for comparison. The count rate of this energy segment at 25 °C is set to 100%, thus the influence of temperature on the count rate can be quantitatively given. The deviation $L_{\text{non}}$ of the count rate at different temperature can be calculated through formula (3).

$$L_{\text{non}} = \frac{N_i - N_0}{N_0} \times 100\%$$  \hspace{1cm} (3)

In the formula, $N_i$ is the count rate measured at each temperature point; $N_0$ is the count rate at the initial temperature of 25 °C.

![Figure 8. Deviation of count rate of all the scintillation crystals with temperature.](image)

The count rate deviation of the crystal sample can be obtained by calculation, as shown in Figure 8. The degree of deviation is within 5%. The count rate of NaI(Tl) at 50 °C and 75 °C has obvious fluctuation, the deviation in the range of 25 °C-100 °C is about 1.41%. As for LaBr3(Ce) crystal samples, The degree of deviation in 25 °C-100 °C is less than 0.2%.

4. Conclusion

In this paper, high temperature performance of LaBr3(Ce) and NaI(Tl) scintillation crystal were compared by a high temperature experiment at 25 °C - 175 °C. The values of relative light output, energy resolution, count rate and its deviation of LaBr3(Ce) (LBC-3 as representative) and NaI(Tl) at different temperature are listed in Table 2.
Table 2. Temperature effect of LaBr₃(Ce) (LBC-3 as representative) and NaI(Tl)

| Temp(°C) | Normalized light output(%) | Energy resolution(%) | Count rate(cps) | Deviation of count rate(%) |
|----------|-----------------------------|----------------------|----------------|----------------------------|
|          | LBC | NI | LBC | NI | LBC | NI | LBC | NI |
| 25       | 145.00 | 100 | 5.92 | 9.90 | 469.38 | 108.01 | 0.00 | 0.00 |
| 50       | 139.70 | 83.36 | 6.14 | 10.30 | 471.36 | 111.45 | -0.17 | 1.38 |
| 75       | 133.26 | 74.99 | 6.14 | 10.00 | 469.43 | 111.81 | -0.27 | 1.43 |
| 100      | 122.85 | 65.64 | 6.40 | 10.50 | 472.00 | 110.33 | -0.52 | -0.10 |
| 125      | 106.41 | 58.58 | 6.81 | 11.10 | 472.39 | 111.69 | -0.91 | -0.51 |
| 150      | 87.59  | 47.69 | 7.60 | 11.50 | 477.86 | 111.65 | -1.84 | -1.37 |
| 175      | 72.19  | 39.39 | 8.29 | 12.40 | 477.17 | 112.95 | -3.45 | -3.63 |

The relative light output of LaBr₃(Ce) at each temperature point is less affected by high temperature, and decline of light output of LaBr₃(Ce) is about 80% compared to that of NaI(Tl) at 175 °C. LaBr₃(Ce) has an energy resolution of about 8% at 175 °C in extreme high temperature environment, and even better than the 9.9% energy resolution of NaI(Tl) scintillation crystal at room temperature of 25 °C. In the high temperature environment, Ø22×27 mm and Ø28×28 mm LaBr₃(Ce) crystals have a count rate of about 5-6 times more than that of the Ø25×50 mm NaI(Tl) crystal. Therefore, high temperature performance of LaBr₃(Ce) crystal is overall superior to that of traditional NaI(Tl) scintillation crystals. That is to say, LaBr₃(Ce) scintillation crystal is a promising substitute material for NaI(Tl) crystal in gasoline logging, space detection and other high temperature nuclear detection environments applications.

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