Study on Scintillation Properties of Several Non-fluoride Halides under Low Energy $^{241}$Am radiation

S Liu$^1$, Y Hou$^1$, H Yuan$^1$, Q Gui$^1$, C Zhang$^1$, Z Fang$^2$, M Zhang$^1$

$^1$Beijing Glass Research Institute, Beijing, 101111, China

$^2$Beijing Industrial Technology Research Institute, Beijing, 101111, China

Email: liushan@bitri.cn

Abstract. A few of halide inorganic scintillation crystals have been applied as radiation detection materials and are playing important roles in nuclear medicine, high energy physics, security inspection, petroleum logging and so on, but most of them are on the way of research and development. In this paper, a series of novel halide scintillation crystals, LaBr$_3$(Ce), LaCl$_3$(Ce), SrI$_2$(Eu), Cs$_2$LiYCl$_6$(Ce) and NaI(Tl) were studied using low energy $^{241}$Am radioactive source. The effect of reflective film thickness on low-energy ray transmittance and light output of encapsulated scintillators was investigated, Teflon thickness of 0.48 mm as reflective film is best choice to obtain higher light collection and better radiation transmittance. Among all these crystals, LaBr$_3$(Ce) crystal has excellent energy resolution above 25 keV gamma ray radiation, 9.3% at 26.34 keV, 8.4% at 59.54 keV and 2.6% at 661.66 keV. Only NaI(Tl) crystal works fine below 25 keV X-rays radiation, with energy resolution of 17.68% at 13.9 keV, 14.16% at 17.8 keV and 13.42% at 20.8 keV.

1. Introduction
Several hundreds of scintillation crystals have been discovered since the first synthetic scintillation crystal was recognized seventy years ago, and about half of them are halide crystals. A few of halide inorganic scintillation crystals have been applied as radiation detection materials in nuclear medicine, high energy physics, security inspection, petroleum logging and soon [1]. The most commonly used scintillator is NaI(Tl), which has a peak emission of 415 nm, a light output of 38,000 photons/MeV, a density of 3.7 g/cc, and a decay time of 230 ns.

Recent years, a series of novel halide scintillation crystals with excellent scintillation properties were developed, which made halides scintillators the new research hotspot in this field. Typical types of novel halide scintillation crystals include Ce-type rare earth halide crystals (represented by LaBr$_3$(Ce)), Eu-type alkaline earth halide crystals (represented by SrI$_2$(Eu)), and Ce$^{3+}$ activated rare earth complex halide crystals (represented by Cs$_2$LiYCl$_6$(Ce)). Most of the investigations are focused on scintillation properties under high energy radiation, and the results reveal that all these halide crystals showed merits of high light yield and good energy resolution. But there’s very little research on scintillation properties of these novel halide crystals under low energy radiation which is mostly used in nuclear medical imaging. Radioactive $^{241}$Am source emits high intensity of low energy $\gamma$-rays (59.54 keV and 26.35 keV) during the $\alpha$-decay and low energy X-rays (11.9 keV, 13.9 keV, 17.8 keV and 20.8 keV) by the L shell [2-3], so that it is very suitable for the measurement of scintillation properties at 0~100 keV spectrum.
In this paper, $^{241}$Am source is used to investigate the energy spectra of halide crystals LaBr$_3$(Ce), LaCl$_3$(Ce), SrI$_2$(Eu), Cs$_2$LiYCl$_6$(Ce). Energy resolution of these four crystals at low energy of $^{241}$Am source was measured with that of the most widely used NaI(Tl) crystal.

2. Materials and Method

2.1. Samples

Five high-performance scintillation crystal samples were selected for low-energy experiments, four crystals grown by our research group including LaBr$_3$(Ce), LaCl$_3$(Ce), SrI$_2$(Eu), and CLYC(Ce), and one crystal NaI(Tl) purchased from Saint-Gobain, USA.

Scintillation crystals are usually coated with reflective materials on the incident end and the cylinder surface to reduce the light loss of the crystal and improve the light collection efficiency [4], as shown in Figure 1. Teflon film is a commonly used reflective material in scintillation crystal detection and encapsulation. The reflectivity of 0.15 mm-0.5 mm thickness of Teflon can reach to 98%. Considering the limited penetration ability of low-energy rays, in order to balance the accuracy of low-energy segment energy spectrum measurement and the controllability of crystal light loss, it is necessary to select the thickness of the reflective material suitable for low-energy ray measurement.

![Figure 1. the encapsulation structure of scintillation crystal.](image)

The encapsulation of scintillation crystal must be carried out in extremely low oxygen-content and extremely low water-content glove box. The Teflon reflective film with thickness of 0.125/0.25/0.375/0.5/0.625mm was coated on the cylinder surface and the radiation incident end. The effect of thickness of Teflon film on crystal properties was studied. In order to evaluate the optimal low-energy scintillation performance of all the samples we choose, the incident end and the cylinder surface of the scintillation crystal sample will be coated with Teflon reflective film of a specified thickness according to the optimum thickness given by Teflon thickness study.

2.2. Measurement device

The standard energy spectrum measurement system consists of high voltage power supply (Ortec 556, USA), preamplifier (Ortec 113, USA), main amplifier (Ortec 672, USA) and multichannel analyzer (Ortec 926, USA). All scintillation crystal samples were combined into a detector with the same super-alkali photomultiplier tube (R6231-100, Hamamatsu, Japan) to measure the low energy ray spectrum of the $^{241}$Am source. Detectors were placed on the base holder of photomultiplier tube in the glove box, and protected from light by an aluminum alloy dark room. Photomultiplier tube holder and external test equipment were connected with high voltage cable(R G59) and signal cable (RG58). The measurement system is shown as Figure 2.
3. Results and discussion

3.1. Thickness of reflective materials
The effect of the thickness of the Teflon film on the crystal properties is shown in Figure 3. The count rate reflects the transmission under different coating thicknesses. The number of peaks shows the light output performance. From the results, it can be seen that the coated reflective material has a opposite effect on the transmittance and light output, and is mutually constrained. Taking into account, 0.48 mm was chosen as the thickness of the reflective material suitable for low energy ray measurement.

3.2. $^{137}$Cs spectrum of halide crystals
All types of crystals we plan to evaluate their low-energy scintillation performance in this paper show high light yield and good energy resolution under high energy radiation [5-9]. Thus, we can choose high quality crystals through checking their high energy performance under $^{137}$Cs radiation source. The normalized spectrum of scintillation crystal samples are shown in Figure 4, and the crystal size and the energy resolution corresponding to 661.66 keV gamma ray are given in Table 1. The shaping time

![Image of circuit diagram](Figure 2. Circuit diagram of measurement system.)

![Image of response curve](Figure 3. Response curve of transmittance and light output with Teflon film thickness for LaBr$_3$ (Ce) under $^{241}$Am.)
constants of Ce-type rare earth halide crystal LaBr$_3$ (Ce), LaCl$_3$ (Ce), Eu-type alkaline earth halide crystal SrI$_2$ (Eu), and NaI (Tl) were 0.5 μs, and that of Ce$^{3+}$ activated rare earth complex halide crystal Cs$_2$LiYCl$_6$(Ce) crystals was 3 μs. Five scintillation crystals, LaBr$_3$(Ce), LaCl$_3$(Ce), SrI$_2$(Eu), Cs$_2$LiYCl$_6$(Ce) and NaI(Tl), all show excellent energy resolution as 2.6%, 3.3%, 3.5%, 4.7% and 6.4%. These results prove that the crystals we choose are high quality for the corresponding materials.

![Figure 4. The normalized $^{137}$Cs energy spectrum of scintillation crystals.](image)

Table 1. The size and energy resolution of scintillation crystals

| Sample          | Size       | Energy resolution |
|-----------------|------------|-------------------|
| LaBr$_3$(Ce)    | Φ38mm×38mm | 2.6%              |
| LaCl$_3$(Ce)    | Φ25mm×25mm | 3.3%              |
| SrI$_2$(Eu)     | Φ15mm×10mm | 3.5%              |
| Cs$_2$LiYCl$_6$(Ce) | Φ25mm×25mm | 4.7%              |
| NaI(Tl)         | Φ25mm×50mm | 6.4%              |

3.3. $^{241}$Am spectrum of halide crystals

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 photo detector [10]. $\Delta$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 (1) [10-12].

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

For the energy spectrum distribution of 59.54 keV, 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 59.54 keV $\gamma$-ray can be calculated by formula (1). But in the energy spectrum of $^{241}$Am, there are multiple low-energy ray overlapping peaks with similar energies at 0-40 keV. For the analysis of overlapping peaks, a
Gaussian multi-peak fitting method is needed to perform Gaussian fitting on multiple overlapping peaks [12]. The Gaussian multimodal fitting function is expressed as formula (2).

\[
y_c = y_0 + \sum_{n=1}^{\infty} \frac{A_n}{w_n \sqrt{\pi/2}} \exp\left(-2\frac{(x-x_{cn})^2}{w_n^2}\right)
\]

In the formula (2), \(y_0\) is the baseline, \(A_n\) is the area of the peak, that is, the integrated intensity, \(w_n\) is the full width at half maximum of the peak, and \(x_{cn}\) is the peak position. In this experiment, the multi-peak fitting function of Origin software is used to perform Gaussian fitting on the superimposed peak distribution, and the standard deviation and peak center value of each peak of 0-40 keV are obtained. The fitting parameters of the crystal samples corresponding to each peak has been given in Table 2.

**Table 2.** Multi-Gaussian fitting results of spectral distribution at 0-40 keV low energy rays.

| Energy Segment/keV | LaBr₃(Ce) μ | LaCl₃(Ce) μ | SrI₂(Eu) μ | Cs₂LiYCl₆(Ce) μ | NaI(Tl) μ |
|-------------------|-------------|-------------|------------|-----------------|-----------|
|                   | σ           | σ           | σ          | σ               | σ         |
| 0-12              | 5.17 ±0.06  | 4.25 ±0.02  | 14.49 ±0.01| 15.03 ±0.16     | 2.54 ±0.05|
| 12-16             | 13.64 ±0.04 | 13.77 ±0.03 | 18.26 ±0.01| 18.01 ±0.05     | 1.32 ±0.08|
| 16-20             | 17.06 ±0.02 | 17.22 ±0.03 | 18.26 ±0.01| 18.01 ±0.05     | 1.32 ±0.08|
| 20-25             | 21.11 ±0.04 | 21.14 ±0.02 | 22.24 ±0.02| 21.01 ±0.02     | 3.20 ±0.36|
| 25-30             | 25.96 ±0.04 | 26.20 ±0.06 | 26.74 ±0.04| 28.00 ±0.14     | 2.71 ±0.14|
| 30-40             | 30.85 ±0.12 | 31.13 ±0.06 | 31.47 ±0.09 | 31.35 ±0.05     | 1.47 ±0.03|

**Figure 5.** The ²⁴¹Am energy spectrum of Φ38mm×38mm LaBr₃(Ce) scintillation crystals.
Figure 6. The $^{241}$Am energy spectrum of $\Phi25\text{mm}\times25\text{mm} \text{LaCl}_3(\text{Ce})$ scintillation crystals.

Figure 7. The $^{241}$Am energy spectrum of $\Phi15\text{mm}\times10\text{mm} \text{SrI}_2(\text{Eu})$ scintillation crystals.

Figure 8. The $^{241}$Am energy spectrum of $\Phi25\text{mm}\times25\text{mm} \text{Cs}_2\text{LiYCl}_6(\text{Ce})$ scintillation crystals.
The $^{241}$Am energy spectrum of $\Phi 25\text{mm}\times 50\text{mm}$ NaI(Tl) scintillation crystals.

The $^{241}$Am spectrum composed of 59.54 keV and 26.34 keV $\gamma$-ray peaks by experiment and by Gaussian fitting is given in Figures 5-9. It can be seen that three scintillation crystals of LaBr$_3$(Ce), SrI$_2$(Ce) and NaI(Tl) have $\gamma$-ray energy resolution of less than 9% for 59.54 keV, which is better than other scintillation crystals. The energy resolutions measured by LaBr$_3$(Ce), SrI$_2$(Eu) and NaI(Tl) were 8.41%, 8.53% and 8.81%.

By fitting the overlapping peaks, it can be seen that SrI$_2$(Eu) and NaI(Tl) crystals have obvious energy spectrum distribution in the energy range of 30-40 keV, and the energy resolution is 11.65% and 11.10%, respectively. The other three crystals, LaBr$_3$(Ce), LaCl$_3$(Ce), Cs$_2$LiYCl$_6$(Ce), have no spectral distribution in this energy range. For $\gamma$-ray of 26.34 keV, the energy resolution of LaBr$_3$(Ce), NaI(Tl) and SrI$_2$(Eu) scintillation crystals is 9.32%, 12.04% and 14.26%, while that of LaCl$_3$(Ce) and Cs$_2$LiYCl$_6$(Ce) is quite poor, 14.55% and 22.74%, respectively. Four kinds of low-energy X-rays emitted by the $^{241}$Am L shell were measured in this experiment. Only LaBr$_3$(Ce) and SrI$_2$(Eu) crystals could measure the X-ray spectrum of 11.9 keV and their energy resolution is 46.36% and 104.51%, respectively. NaI(Tl) scintillation crystal has an energy resolution of 17.68%, 14.16% and 13.42% corresponding to the X-rays of 13.9 keV, 17.8 keV and 20.8 keV, which is better than the other four crystals. The results show that LaBr$_3$(Ce) scintillation crystal has the best energy resolution of $\gamma$-rays with energy of 26.34 keV and 59.54 keV, and NaI(Tl) scintillation crystal is more suitable for detection of X-rays below 25 keV. The energy resolution values corresponding to energy peaks of $^{241}$Am are listed in Table 3.

Table 3. The energy resolution corresponding to energy peaks of $^{241}$Am.

| Sample          | 59.54keV | 26.34keV | 20.8keV | 17.8keV | 13.9keV | 11.9keV |
|-----------------|----------|----------|---------|---------|---------|---------|
| LaBr$_3$(Ce)    | 8.41%    | 9.32%    | /       | /       | /       | 46.36%  |
| LaCl$_3$(Ce)    | 10.98%   | 14.55%   | /       | /       | /       | /       |
| SrI$_2$(Eu)     | 8.53%    | 14.26%   | /       | /       | /       | 104.51% |
| Cs$_2$LiYCl$_6$(Ce) | 13.63%  | 22.74%   | /       | /       | /       | /       |
| NaI(Tl)         | 8.81%    | 12.04%   | 13.42%  | 14.16%  | 17.68%  | /       |

4. Conclusion

In this paper, $^{241}$Am spectrum of five non-fluoride halide scintillation crystals is studied. It is known that LaBr$_3$(Ce) scintillation crystal has 2.6% energy resolution for $^{137}$Cs at high-energy $\gamma$-ray 661.66keV $\gamma$-ray. In this paper, it is verified that LaBr$_3$(Ce) scintillation crystal still have the best energy
resolution of 8.41% and 9.32% at low-energy $\gamma$-rays 26.34 keV and 59.54 keV respectively, and this makes LaBr$_3$(Ce) scintillation crystal a potential candidate for positron emission tomography (PET), environmental detection, nuclear physics experiments and instruments with high performance requirements on energy resolution, light output and count rate. Compared with other four scintillation crystals, NaI(Tl) scintillation crystal has the best resolution for X-rays below 25 keV, and the energy resolutions for 13.9 keV, 17.8 keV and 20.8 keV are 17.68%, 14.16%, and 13.42%, which make NaI(Tl) be the preferred scintillation crystal for low-energy X-ray detection, applied in X-ray imaging, port security and space X-ray detection.

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