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Properties and Application Studies of $LaBr_3$(Ce) Scintillation Detector

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Abstract The $LaBr_3$(Ce) detector is a new type of scintillator detector which offers better energy resolution and higher efficiency for high-energy gamma ray in comparison with traditional detectors，for instance，NaI (Tl) detector. The LaBr3(Ce) detector has merits of a high scintillation light output with a fast decay time，a high temporal and spatial resolution，good temperature characteristics and good radiation resistance. So it was researched and applied widely since it was commercially available years ago. This paper reports not only its above-mentioned properties but also its typical application studies. In the nuclear resonance fluorescence，the detector benefitted the nondestructive assay (NDA) method by shortening the detecting time while the energy resolution was still excellent. In the prompt gamma neutron activation analysis，the advantages of it were shown by comparing with the BGO detector. In the nuclear medicine imaging，the detection with the $LaBr_3$ (Ce) detector in the myocardial perfusion in mice with sufficiently precision. It had a better ability to distinguish between tumors and normal tissues than NaI(Tl) detector. In the space radiation detection，the detector was used to detect the rays with high energy，and it shows excellent radiation resistance. In neutron detection，it has competitive properties in low neutron energy measurement. After investigating the applications of the detector，we confirm the excellence of this detector and affirm it has a good prospect in the further.

Keywords The $LaBr_3$(Ce) detectors；Energy resolution；Detection efficiency；Prompt gamma neutron activation analysis；Nuclear resonance fluorescence analysis

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Introduction

The $LaBr_3$(Ce) detector is a new type of scintillation detector with high energy resolution (about 2.8% at 662 keV)，high light output(~60 000 phe · MeV^{-1}) with a fast decay time (~16 ns)，and high energy efficiency[1]. Table 1 shows the properties of the $LaBr_3$(Ce) detector(BrilLanCe™ 380[2]) and the NaI(Tl) detector[3]. Both of them were produced by the Saint-Gobain. This detector was researched and developed by E. V. D van Loef and others from Delft and Bern Universities. In-depth researches were carried out and a large number of applications had been done since it was reported in 2001.

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This article reviewed the properties of the detector and illustrated its typical applications in nuclear resonance fluorescence detection, prompt neutron activation analysis, nuclear medical imaging and space radiation measurement. Its excellent performances make it possible to replace traditional detectors like NaI(Tl) detector, HPGe detector in some cases. We believe that the detector will play an important role in ray-measuring in the future.

Table 1　Properties of the $LaBr_3$(Ce) detector (BrilLanCe™ 380) and the NaI(Tl) detector[2-3]

Detector	Density /(g · cm^{-3})	Melting point /K	Primary decay time/ns	Light yield /(photons · keVg^{-1})	Wavelength of emission max/nm
$LaBr_3$(Ce)	5.08	1 116	16	63	380
NaI(Tl)	3.67	924	250	38	415

1　Characteristics of $LaBr_3$(Ce) detector

1.1　Energy resolution

$LaBr_3$(Ce) detector has excellent energy resolution, which can be as high as 2.8% at 662 keV better than the energy resolution of NaI(Tl) detector (6%～7%)[1,4]. In some previous studies, researchers tested the energy resolutions of a $LaBr_3$(Ce) detector and a NaI(Tl) detector in the same condition with some point sources. The results are shown in Table 2. It shows that the energy resolution of $LaBr_3$(Ce) detector is much better than that of NaI(Tl) detector. Besides, we noticed that the energy resolutions of the two detectors have scarcely any difference between each other below 100 keV when we measured them with ^{137}Cs. In other words, the energy resolution of $LaBr_3$(Ce) detector is superior in high energy detection compared with the NaI(Tl) detector.

Table 2　Comparison of energy resolutions of $LaBr_3$(Ce) detector and NaI(Tl) detector[1,4]

Point source	Eenergy /keV	$LaBr_3$(Ce) /%	NaI(Tl) /%
^{137}Cs	662	2.8	7.0
^{57}Co	122	6.6	8.9
^{60}Co	1 332	2.1	5.4
^{133}Ba	356	3.8	9.1
^{208}Tl	2 620	1.6	4.5

Table 3　Energy resolutions of $LaBr_3$(Ce) crystals in different size[5]

size/cm length×wide×high or diameter×high	Volume /cm^3	Volume increase factor	Energy resolution $\Delta E/E$/% at 662 keV
0.3×0.3×1.0	0.9	1.0	3.2±0.2
1.3×1.3	1.73	1.9	3.4±0.2
1.9×1.9	5.39	6.0	3.4±0.2
2.5×2.5	12.27	14	2.8±0.2
3.8×3.8	43.10	48	2.8±0.2
5.1×5.1	103.0	144	3.0±0.2

At the same time, the $LaBr_3$(Ce) detector has a better linearity than NaI(Tl) detector from low energy region to high energy region. The energy resolution of $LaBr_3$(Ce) detector changes from about 3% to 6% when the energy of incident photon decreased from 700keV to 100keV. As a contrast, the energy resolution of NaI(Tl) detector increased from 6% to 12% under the same condition[4]. In addition, the size of the $LaBr_3$(Ce) crystal has little effect on the energy resolution[5]. Table 3 shows the energy resolutions of $LaBr_3$(Ce) crystals in different sizes at 662 keV.

1.2　Detection efficiency and light yield of $LaBr_3$(Ce) detector

The detection efficiency of a detector is the ratio of the amount of radiation recorded by the detector to the particle number of radiation emitted by the radiation source[6]. That is to say the higher the number of radiation recorded by the detector, the better the detection efficiency is. Because the $LaBr_3$(Ce) crystal has the characteristics of fast decay time and high light output, $LaBr_3$(Ce) detectors have a superior absorbance of the incident photon[6]. So the $LaBr_3$(Ce) detector can record more radiation, and have better detection efficiency.

A. Favalli from Institute for the Protection and Security of the Citizen of European Commission Joint Research Centre, used point sources(^{22}Na, ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu and ^{228Th}) to calibrate the $LaBr_3$(Ce) detector below 3 MeV, and used ^{66}Ga($T_{1/2}$=9.5 h) to provide γ ray with energy above 3MeV. The results of these calibration showed that the detection efficiency of the $LaBr_3$(Ce) detector better than a NaI(Tl) detector. For example, the intrinsic efficiency of the $LaBr_3$(Ce) detector is 60% higher than it of the NaI(Tl) detector for the 1 332 keV gamma line of ^{60}Co[7]. The efficiency of $LaBr_3$(Ce) detector would bring down from 10^{-3} to 10^{-4}, when energy of incident photon increases from 10^2 to 10^4 keV.

The light output of the $LaBr_3$(Ce) detector is not only higher, but also more stable than NaI(TL) detector. Theoretically, the light yield would not change along with the change of incident photon[8], but the nonlinear response of the scintillation detectors is always existed. Researches show that the change of light yield of $LaBr_3$(Ce) detector is within 3% while it of the NaI(Tl) detector is more than 20% between the energy of 30 keV～1 MeV[9]. The nonlinear response of the $LaBr_3$(Ce) detector is also better than it of the NaI(Tl) detector when the energy varies from 60 to 1 000 keV[2].

1.3 Temperature characteristics of $LaBr_3$(Ce) detector

Researchers in Saint-Gobain measured the light yield response function curves of the $LaBr_3$(Ce) crystal with ^{137}Cs excitation at amplifier, when the nuclear pulse forming time was 1 μs and the temperature varied from -65 to 175 ℃. Fig. 1 shows the effect of temperature on the light yield the $LaBr_3$(Ce) detector.

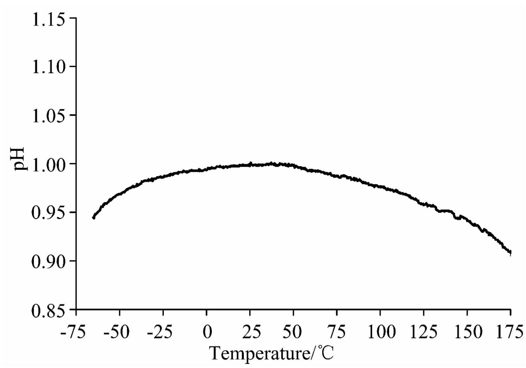

Fig. 1 The light yield response of the $LaBr_3$(Ce) detector with temperature[2]

Fig. 1 shows that the change of the $LaBr_3$(Ce) crystal light output rate is less than 1% when the temperature changes from 0 to 55 ℃. And the change of it is less than 5% when the temperature changes from -65 to 140 ℃[2]. Besides, the work temperature can also affect the energy resolution of the $LaBr_3$(Ce) detector. However the effect of temperature on the energy resolution of the $LaBr_3$(Ce) detector is smaller than it of the NaI(Tl) detector. It means that the $LaBr_3$(Ce) crystal has a better thermal stability in comparison with the NaI(Tl) detector over a wide temperature range. The energy resolution of $LaBr_3$(Ce) detector at 662keV went up from 3.3% to 3.5%, while the temperature went up from -30 to 60 ℃. At the same time, the temperature curves of energy resolution of NaI(Tl) detector were distorted in the same condition[10]. So the $LaBr_3$(Ce) detector will be the better choice when we need to work in extreme weather or in other detection with the temperature changing.

1.4 Radiation damage effect on $LaBr_3$(Ce) detector

S. Normand evaluated the effect of the radiation dose on the energy resolution of $LaBr_3$(Ce) detector and NaI(Tl) detector. The research showed that the energy resolution of $LaBr_3$(Ce) detector at 662 keV fluctuated between 2.14% and 5.83%, while the fluctuation of NaI(Tl) detector was 4.46% to 12.17%, when the expose dose went up from 0.01 to 3 000 Gy. And the energy resolution of $LaBr_3$(Ce) detector can reverse from radiation in a few hours[11]. The spectra after increasing radiation doses are shown in Fig. 2.

According to Fig. 2, we can see that gamma radiation leads to the change of the peak position and the decrease of the light yield in both $LaBr_3$(Ce) detector and NaI(Tl) detector[12-14]. And we can also see that $LaBr_3$(Ce) detector performs better than NaI(Tl) detector. Thus, the $LaBr_3$(Ce) detector is more suitable to use in high radiation environments such as the space γ detection.

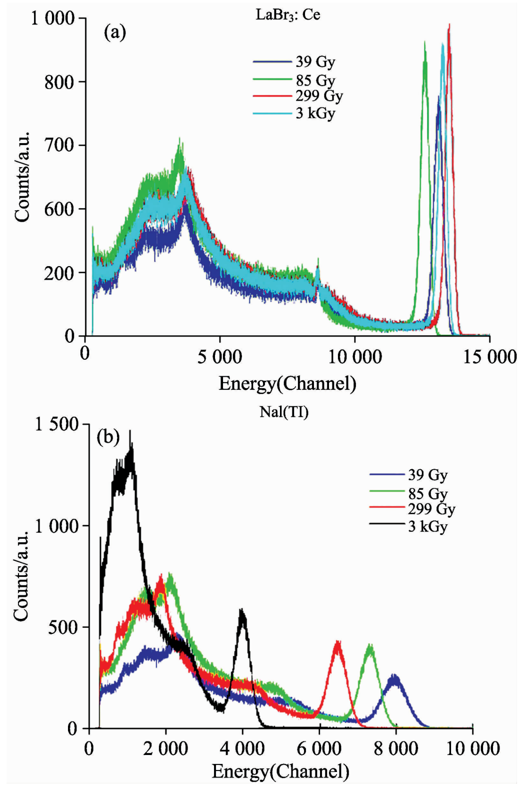

Fig. 2 (a) ^{137}Cs γ spectra observed for several high doses obtained by $LaBr_3$(Ce) crystals[11]; (b) ^{137}Cs γ spectra observed for several high doses obtained by a NaI(Tl) single crystal[11]

1.5 Intrinsic radiation of the $LaBr_3$(Ce) detector

The intrinsic radiation of the $LaBr_3$(Ce) detector is mainly caused by the radionuclides in the $LaBr_3$(Ce) crystal including ^{138}La and ^{227}Ac. The ^{138}La is a kind of radionuclide with the activity of 1.45 counts $\cdot$ (s $\cdot$ cm^3)$^{-1}$ which approaches to its natural abundance[15]. Its effect to the spectrum in 0~1.5 MeV principally comes from beta decay and EC coincidence effect. Furthermore the beta decay is the mainspring of radiation background in the low energy range. In addition, the ^{138}La emits γ rays with the energy of 789 and 1 436 keV whose characteristic peaks overlaps the one of ^{40}K in 1 460 keV[16]. The ^{227}Ac mainly contributes the intrinsic radiation in high energy range by alpha decay. Its decay products contain ^{227}Th, ^{223}Ra, ^{219}Rn, ^{215}Po and ^{211}Bi. The energy of the alpha particles from these alpha decays fluctuated between

4 860～8 000 keV[17].

Its intrinsic radioactivity may limit its application in low-level radioactivity measurement. Especially, if the characteristic rays emitted by nuclides locate in low energy region, or near 789 and 1 400 keV, the detector should be used with caution.

2 Application studies of $LaBr_3$(Ce) detector

2.1 Application study in the nuclear resonance fluorescence

As a kind of nondestructive assay (NDA) method, the nuclear resonance fluorescence (NRF) has been used to investigate the nuclear structure. The nuclear resonance fluorescence refers to the resonant excitation of an excited state by absorption of electromagnetic radiation and the subsequent decay of this level by emission of radiation[18]. Since each nucleus has characteristic resonant energies, the detection of resonant gamma rays provides a unique fingerprint for identification[19]. Therefore, NRF provides a method to NDA of materials.

Because of the narrow band-width of the NRF level (on the order of eV or less), only a very small fraction of the incident gamma ray contributes to the NRF reaction[20]. NRF measurement takes long time to collect sufficient incident gamma rays to reduce uncertainty of results. At the same time, High energy resolution of the detection system is also important in NRF measurement. So high purity germanium (HPGe) detectors and $LaBr_3$(Ce) detectors are the most essential detector for NRF measurements.

In the experiments of a joint project between the Triangle Universities Nuclear Laboratory (TUNL) and Duke University Free Electron Laser Laboratory (DFELL), Mohamed Omer had applied $LaBr_3$(Ce) detector in NRF measurement for ^{235}U. In the NRF measurement, quasi-mono-energetic photon beams with energy of 1 730 keV was use to provide excitation sources and the scattered photons at 90°, with respect to the incident beam, were detected by $LaBr_3$(Ce) detector. Broad and clearly visible NRF peaks were obtained from the de-excitations of ^{235}U nuclei at 1 733 within 77 minutes of beam time even under the high background due to the self-activity of $LaBr_3$(Ce)[19]. On the other hand, the NRF peak at 1 733 keV detected by the HPGe detector with a collection time of 530 minutes is hard to be seen.

The intrinsic radioactivity of $LaBr_3$(Ce) detector is a significant drawback in low count-rate measurement. Thus, in the NRF measurement with $LaBr_3$(Ce) detector, we should pay more attention to the effect of the intrinsic radioactivity of this detector. Mohamed Omer used the detectors to measure the NRF excitation at 2.12 MeV scattered from ^{11}B in B_4C and got clear and statistically satisfying NRF peaks. In order to improve the limit of detection, the statistics-sensitive nonlinear peak-clipping (SNIP) algorithm has been used. And the minimum detectable mass of ^{11}B is on the order of milligrams.

2.2 Application study in prompt gamma-ray neutron activation analysis

The $LaBr_3$(Ce) detector also was used in prompt gamma-ray neutron activation analysis (PGNAA) technique. The PGNAA is also a nondestructive analysis method. It's widely used in determining the existence and amount of the isotopes in the samples since 1970s[21]. Because its detection sensitivity mainly depends upon energy resolution and detection efficiency of detection system, a detector with excellent properties can get accurate result. The $LaBr_3$(Ce) detector with high energy resolution and efficiency has the potential to be applied in PGNNA.

A. A. Naqvi from King Fahd University of Petroleum and Minerals used a 76 mm×76 mm $LaBr_3$(Ce) detector and 102 mm×102 mm(height×diameter) BGO detector to measure prompt gamma rays, with a maximum energy of 6.13 MeV, produced through thermal neutron capture and inelastic scattering of 14 MeV neutrons from propanol, methanol, water, benzene and ethanol bulk samples.

The results are shown in Fig. 3.

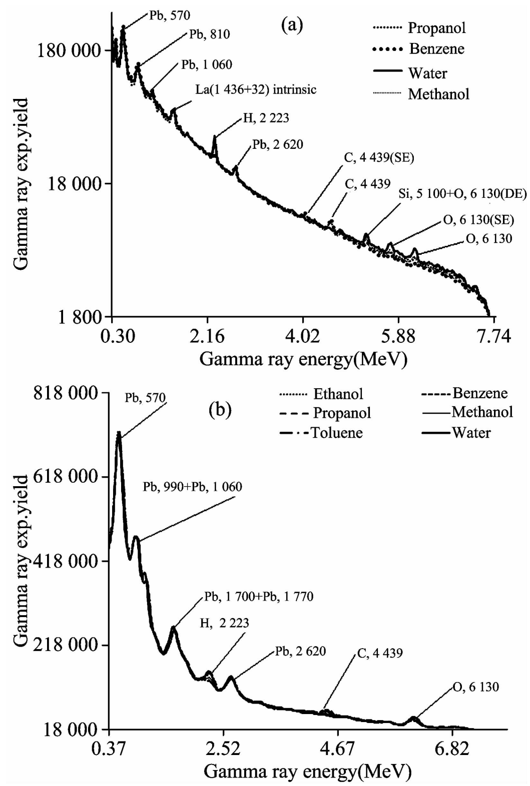

Fig. 3 (a) Full prompt γ ray spectra of $LaBr_3$(Ce) detector from activated benzene, propanol, water, methanol and lead shielding; (b) full prompt gamma ray spectra of 102 mm×102 mm (diameter×height) BGO gamma ray detector for benzene, ethanol, toluene, water and

methanol samples[22]

Because the $LaBr_3(Ce)$ detector has a faster light decay time, a better energy resolution and a higher capacity of counting rate, the $LaBr_3(Ce)$ detector can measure more peaks and these peaks are clearer than the BGO detector. Because the $LaBr_3(Ce)$ detector does not contain oxygen element, so it has better performance than the BGO to detect oxygen peak. Due to the high density, resolution and detection efficiency of the $LaBr_3(Ce)$ detector, it can be widely applied in the PGNAA.

2.3 Application study in the nuclear imaging

Molecular imaging with radionuclides is a suitable tool to study human diseases using animal models, due to its intrinsic high sensitivity. Two representative methods in this field are single photon emission computed tomography (SPECT) and photon emission tomography (PET). They both image the γ-rays emitted from the patients or else. So the detector impacts the performance of the system crucially. Of course, the images are crucial to the diagnostic or research. Therefore, a good γ-rays detector is essential. The $LaBr_3(Ce)$ detector with an excellent energy resolution, high light yield and good time resolution can be a good choice to obtain high quality γ images. F. Cusannoet al from Superiore di Sanita in Rome-built a prototype with tungsten pinhole and $LaBr_3(Ce)$ detector coupled to Hamamatsu Flat Panel PMTs. They used it for measuring the images from the mice. The results of their experiments showed that the $LaBr_3(Ce)$ detector could determine the myocardial perfusion in mice sufficiently precision. Application of $LaBr_3(Ce)$ scintillator coupled to photosensor with high photon detection efficiency and excellent energy resolution will allow dual-label imaging to monitor simultaneously the cardiac perfusion and the molecular targets under investigation during the heart therapy[23].

R. Paniet al from Department of Experimental Medicine "La Sapienza" University made the $LaBr_3(Ce)$ detector connected with a Hamamatsu H8500 Flat Panel PMT to get a γ-camera. With this camera they get a scanning image contour more clear than the γ-camera made up by the NaI(Tl) detector. In their experiment, the imaging sensitivity of $LaBr_3(Ce)$ detectors is higher than 400 cpm/uCi while the imaging sensitivity of NaI (Tl) detector is only slightly higher than 200 cpm/uCi[24]. And the better image signal-to-noise ratio and spatial resolution make the $LaBr_3(Ce)$ detector able to detect tumors as small as 1 cm[25]. Hence, it is more suitable to distinguish between tumors and normal tissues.

2.4 Application study in space radiation detection

The advances in the crystal growth technology makes the $LaBr_3(Ce)$ detector popular in many areas involving detections. As mentioned, there's no measurable radiation damage effect on the $LaBr_3(Ce)$ detector after exposed to proton fluences up to 10^{12} protons cm^{-2}[14]. Because of the good performance, especially the radio resistance, the $LaBr_3(Ce)$ crystal is suitable for space based γ-ray spectroscopy.

To assess the suitability of $LaBr_3(Ce)$ detector for space radiation detections, E. J. Buis et al. investigated the proton induced activation of the $LaBr_3(Ce)$ detector. They measured the activation both internally and externally by measuring the count rate in the scintillator itself and with a germanium detector[26]. Firstly, $LaBr_3(Ce)$ crystal was irradiated by a proton beam. After a few days, measurements showed that bromide is the source for the highest activation; proton induced activation of ^{79}Br and ^{81}Br results in the production of ^{76}Br, ^{77}Br and ^{79}Kr isotopes with lifetimes of 16, 57 and 35 h, respectively[26]. And the decay of these isotopes may interfere with the observables.

F. G. A. Quarati et al. investigated the potential of the $LaBr_3(Ce)$ detector to be applied in some space missions, such as BepiColombo mission to Mercury , and the Jovian system proposed by the ESA/NASA Laplace mission[27]. The $LaBr_3(Ce)$ crystal was coupled with two Photons XP5300B PMTs. And the PMTs were equipped with borosilicate windows. The energy range of incident gamma rays is from 150 keV to 15 MeV. They operated their 3 in. ×3 in. and 2 in. × 2 in. spectrometers with energy resolution of at least 3.4% at 662 keV and 1.3% at 8 997 MeV. This surpasses BepiColombo requirements (4.5% at 662 keV) [27]. Researches confirm $LaBr_3(Ce)$ detectors are suitable for high energy gamma-ray detection. Due to its radiation tolerance, the $LaBr_3(Ce)$ detector is an ideal choice for space application, especially for which with a high need for radiation tolerance.

2.5 Application study in neutron detection

The $LaBr_3(Ce)$ detector has been used for neutron detection in recent years. A. Oberstedt et al. measured the γ-rays from the spontaneous fission of ^{252}Cf with a 2 in. ×2 in. $LaBr_3(Ce)$[28]. The $LaBr_3(Ce)$ detector was reliable in distinguishing the γ-rays from different reactions by detecting their characteristic time-of-flight. Because the time-of-flight distribution was associated with the fission neutron spectrum from $^{252}Cf(sf)$, it was converted to an energy neutron spectrum. Thus, the researchers measured the neutron spectroscopy with the $LaBr_3(Ce)$ detector successfully. The result showed this detector can be used for neutron detection by detecting the time-of-flight of γ-rays.

The neutron detection efficiency also was measured by some other researchers such as A. Ebran and O. Roig. In their experiments they calculate the neutron detection efficiency of a 2″×2″ $LaBr_3(Ce)$ detector with mono-energetic neutrons in the energy range 0.1～2 MeV[29]. With its 5% neutron detection efficiency for the 261～275 keV γ-rays doublet at 700 keV neutron, researches showed that $LaBr_3(Ce)$ detector can compete with other neutron detectors for low neutron energy measurement.

Other studies investigated the sensitivity of $LaBr_3$(Ce) detector to low energy neutrons and came to the conclusion that it was qualified in the neutron detection in the energy range from 40 keV to 2.5 MeV [30]. From all the above, the $LaBr_3$(Ce) detector has great potential in the neutron detection.

3 Conclusion and outlook

In conclusion, the excellent properties of $LaBr_3$(Ce) detector such as the high light yield, high detection efficiency, high time resolution, high energy resolution make it promising in many aspects including the nuclear resonance fluorescence detection, the prompt gamma neutron activation analysis, nuclear imaging, space radiation detection, and neutron measurements. Good spatial resolution and high energy resolution of the $LaBr_3$(Ce) detector gives it great potential in medical imaging, especially in tumor imaging. The excellent temperature characteristic and radiation tolerance performance o f the detector make it applicable in unstable temperature conditions and space detections. Because of its neutron sensitive performance, it has been used neutron detections. So we can infer that the $LaBr_3$(Ce) detector will be widely used in industry, homeland security, environmental protection, medical, space missions, and other fields.

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溴化镧闪烁体探测器性能及应用研究

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摘　要　$LaBr_3$(Ce)探测器是一种新型闪烁体探测器，具有高光产额，高探测效率，高时间和空间分辨率，高能量分辨率，温度特性良好，抗辐射性能良好，操作简便等优点。从2001年以来，该探测器得到了迅速的研究和应用。$LaBr_3$(Ce)探测器在核共振荧光检测、瞬发γ中子活化分析、爆炸物检测、核医学成像、环境辐射监测、空间辐射探测等方面的应用研究中取得了非常良好的效果。该探测器表现出优于以往用于这些领域的探测器的性能(例如NaI(Tl)探测器、BGO探测器、HPGe探测器等)。介绍了$LaBr_3$(Ce)探测器的性能及其应用研究进展，对代表性文献进行了简析和综述，阐明了其良好的应用前景。

关键词　$LaBr_3$(Ce)探测器；能量分辨率；探测效率；瞬发γ中子活化分析；核共振荧光分析

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