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Luminescence and scintillation characteristics in the CsI:Br crystal on Br concentrations

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Abstract

Cesium Iodide (CsI) doped with Cesium Bromide (CsBr) was grown, using the Bridgman technique, varying the concentration of the Br from 10^{-2} to 2×10^{-1} mol. The Br dopant composition was analyzed by chemical and optical methods. A good uniformity of the Br dopant distribution over the height was observed inside each ingot. To evaluate the crystal obtained [CsI(Br)], systematic measurements of optical transmittance, emission spectra, pulse heights and luminescence decay curves were carried out.

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Keywords: Scintillator; Bromide; Luminescence; Cesium iodide detector

1. Introduction

In recent years, much interest in inorganic scintillators having a short decay time (about 10 ns), high density and high light yield have emerged, for application in nuclear physics, high-energy physics, nuclear tomography and other fields of science and engineering. Among the inorganic scintillators, the pure CsI and BaF_2 crystals have a fast decay time of ~10 and 1 ns, respectively [1,2]. However, their light yield amounts only to 0.05–0.08 of NaI(Tl).

On the other hand, appreciable attention has also been given to develop cheap heterogenous EM- calorimeters, which are capable of working under high counting rates (up to $10^7\,s^{-1}$) and high secondary particle multiplicity conditions, displaying high levels of radiation resistance and energy resolution [3]. Heavy scintillators have been proposed for the improvement of the detection efficiency of soft γ quanta in sampling EM-calorimeters and, consequently, for the improvement of their energy resolution [3].

CsI-based scintillation crystals are considered promising materials for these applications, because

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they exhibit a high plasticity, relatively low hygroscopicity and low cost [1–3]. These characteristics make them suitable to be used as detectors of ionizing radiation under hard operation conditions, such as heavy mechanical and thermal loading. The use of the CsI crystals doped with Tl or Na, proposed as a scintillation detector has been mentioned in the literature since the 1950s [2]. Although they have a high light yield compared to pure CsI, their slow decay times of 1000 μs have limited their use for high counting rates. It is known that alloying with bromide (Br) increases the microhardness [4] and the yield limit [3] of CsI crystal. However, few studies have been found in the literature on the development and scintillation characteristics of the CsI(Br). In this work, Cesium Iodide (CsI) doped with Cesium Bromide (CsBr) was grown by Bridgman technique, varying the concentration of the Br from 10^{-2} to 1 mol. The scintillation characteristics of the developed CsI(Br) crystal were studied.

2. Experimental methods

CsI crystals with [Br^-] of 1×10^{-2}, 1.5×10^{-1}, 2.0×10^{-1} mol used in this work were grown in accordance with the vertical Brigdman method using a quartz crucible in vacuum atmosphere. The starting procedure used had a purity of 99.99% and was obtained from Metal Gesellschalt K.K., Germany. Crystals around 100 mm long were obtained with a growth rate of 1.2 mm/h. Pure CsI crystal was also grown for comparison.

The grown crystals were used for measurements of the scintillation emission, efficiency and decay curves. As an auxiliary experiment, the variation of the [Br^-] along these crystals was measured. The [Br^-] was determined using the neutron activation analysis technique.

The stability of the Br incorporation in the CsI crystal structure was evaluated, measuring the [Br] concentration by neutron activation analysis technique after 6 months.

The emission spectra of these crystals were measured with a monochromotor (JASCL FP55A) by 511 keV annihilation gamma rays from a ^{22}Na source excitation. The signal from the monochromator was detected with a UV-sensitive quartz photomultiplier (Hamamatsu Photonics R 1668). The spectral response of the detection system was not corrected.

Measurements of the decay curves were carried out under excitation of 551 keV annihilation gamma rays from a ^{22}Na source by a single photons counting [1]. The scintillation pulse decay was measured. The decay data were decomposed and analyzed with the least-squares fitting technique.

3. Results and discussion

Transparent and uniform over the volume CsI(Br) crystals 25 mm in diameter and 100 mm in height were reproducibly grown at a concentration of CsBr not exceeding 2.0×10^{-1} mol. The results of the Br concentration analysis over the crystal height by neutron activation analysis technique showed that the non-uniformity of the Br distribution over the height did not exceed 10% inside each ingot. The CsI(Br) ingots with a Cesium Bromide concentration above 2.0×10^{-1} mol were non-uniform in composition, and presented large opaque regions. Zaslavsky et al. [5], growing CsI(Tl) crystal with high [Br] concentration, have found the same results, that they attribute to a possible decomposition of the solid solution and non-uniformity of the composition. No significant change was found in the concentration of Br in the crystal 6 months after crystal growth, showing the stability of the Br incorporated in the CsI structure, which was determined by neutron activation analysis technique.

The results of the CsI(Br) transmittance measurement are presented in Fig. 1. For CsI crystal with 10^{-2} mol, the transmittance in a broad range of the transmittance has shown that in the case of a careful treatment of the surface, optical losses in the ultraviolet range are practically the same as those in the visible range, where the transmittance of pure CsI and conventional CsI(Tl) and CsI(Na) is observed [1]. However, for 1.5×10^{-1} and 2.0×10^{-1} mol, a strong decrease in the transmittance was observed in the luminescence emission region of the CsI(Br), as shown in Fig. 1.

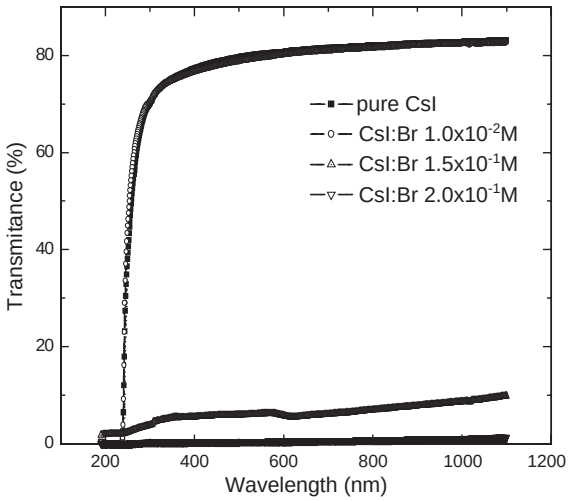

Fig. 1. Transmittance curves of CsI pure and doped with 10^{-2}, 1.5×10^{-1} and 2.0×10^{-1} mol.

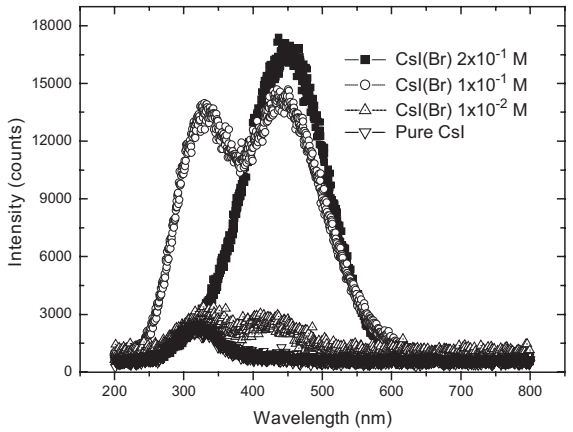

Fig. 2. Luminescence spectra (b) of CsI pure and doped with 10^{-2}, 10^{-1} and 1 mol.

Fig. 2 shows the emission spectra of the CsI crystals with [Br^-] from 10^{-2}, 1.5×10^{-1}, 2.0×10^{-1} mol and pure CsI measured under the excitation of 667 keV gamma rays from a ^{173}Cs source. It is clealy seen from Fig. 2 that the luminescence bands are observed near 310 nm and near 450 nm for 10^{-2}, 1.5×10^{-1} mol. Similar emission spectra for the [Br^-] are reported in Ref. [3]. However, for CsI(Br) 2.0×10^{-1} mol only the band near 450 nm was observed. For pure CsI, the luminescence band was found at 310 nm, which is the luminescence expected for pure CsI crystal with perfect structure. An increase in the intensity of luminescence at 310 nm was observed for CsI doped with 10^{-2}, 1.5×10^{-1} mol compared to the pure CsI. The luminescence intensity of CsI (Br) 1.5×10^{-1} mol was around 6 times higher than that of pure CsI, whereas the increase observed in the intensity of CsI(Br) 10^{-2} mol was 1.5. It can be seen from Fig. 2 that besides the intensity around 310 nm band, there is a luminescence band around 450 nm, with almost the same intensity as for the 310 nm band. In this work, pure CsI was found only for the peak at 310 nm.

A fast decay time around 15 ns was found for 10^{-2}, 1.5×10^{-1} mol. Moreover, it should be noted that the behavior of the curve testifies the presence of a slow component. Hamada et al. [6] described a similar behavior for pure CsI crystal and the study performed to suppress its slow component by heating. This luminescence was attributed to the emission from excitons annihilating at the site of such crystal defects like I-vacancy or F-center. Further experimental studies to suppress the slow component and verify the luminescence origin of the CsI(Br) crystal are recommended. However, it should be emphasized that the addition of the Br in CsI crystal resulted in an increase in the light output of the crystal, with fast decay times closed to the pure CsI, which are the characteristics suitable to be used in detection for high-rate detectors in high-energy physics, nuclear physics and high-resolution medical applications, as well as in high resolution electromagnetic calorimetry and for positron tomography [1,3]. The conventional CsI doped with Tl or Na presented higher output. However, their use is limited for these applications due to the increase in their decay time to 1000 ns. To CsI(Br) 2×10^{-2} mol a similar result i.e. decay time was found.

References

[1] A.V. Gektin, A.I. Gorelov, V.I. Rykalin, V.I. Selivanov, N.V. Iran, V.G. Vasilchenko, Nucl. Instr. and Meth. A 294 (1990) 591Sh.

[2] S. Kubota, S. Sakuragi, S. Hashimoto, J. Ruan, Nucl. Instr. and Meth. A 268 (1988) 275.
[3] G.I. Brivitch, I.G. Brivitch, V.G. Vasilchenko, V. Lishin, V.F. Obraztsov, V. Polyako, S. Solovjev, V.D. Ryzhikov, Nucl. Instr. and Meth. A 469 (2001) 77.
[4] L.P. Antoniv, I.V. Garapyn, N. Tsal, Izv. Acad. Sci. USSR, Inorg. Mater. 24 (10) (1988) 1749.
[5] B.G. Zaslavsky, S.I. Vasetsky, A.M. Kudin, V.Yu. Gres', L.N. Shpilinskaya, T.A. Charkina, L.V. Kovaleva, A.I. Mitichkin, A.N. Boyarintsev, S.Yu. Sumarokov, J. Crystal Growth 222 (2001) 751.
[6] M.M. Hamada, F.E. Costa, M.C.C. Pereira, S. Kubota, IEEE Trans. Nucl. Sci. 48 (1) (2001).