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Development of crystals based on cesium iodide for measurements of gamma radiation and alpha particles*

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Abstract. During the past 50 years, scintillators have been among the most important nuclear radiation detectors. Inorganic scintillators are widely used in experimental nuclear physics, high energy physics, nuclear medicine, nuclear tomography, environmental studies and many other fields of use. Scintillation crystals based on cesium iodide (CsI) are matters of relatively low hygroscopy, high atomic number, easy handling and low cost, characteristics that favor their use as radiation detectors. In this work, the growth of pure CsI crystals, CsI:Br and CsI:Pb, using the Bridgman technique, is described. Ions of divalent lead (Pb^{2+}) doped in the crystal structures are efficient emission centers and their application as scintillators is still the reason for intensive studies. Recently, promising results have been found for crystals of CsI doped by bromine (Br) for their use as radiation detectors. The concentration of the bromine doping element (Br) was studied in the range from 1.5×10^{-1} M to 10^{-2} M and the lead (Pb) in the range from 10^{-2} M to 5×10^{-4} M. To evaluate the scintillators developed, systematic measurements were carried out for luminescence emission and luminescence decay time for gamma radiation, Vickers microhardness assays, and analysis of crystals response to the gamma radiation, in the energy range from 350 keV to 1330 keV, and alpha particles from a ^{241}Am source, with energy of 5.54 MeV. The obtained values for luminescence decay time for CsI:Br and CsI:Pb crystals, were from 13 to 19 ns.

Key words: scintillator • crystals • gamma radiation • alpha particles • luminescence

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Introduction

In recent years, the search for new scintillators has renewed, increasing the needs for the development of high-energy physics, nuclear tomography and other fields of science and engineering. For high energy particle accelerators, it is common the use of multiple detectors capable of working under high counting rates, requiring detectors with short decay time (about 10 ns) and high density [3, 8]. These detectors are usually built with thousands of scintillator crystals and, in some experiments, the total volume of the detector can reach more than 1 m^3. Therefore, in the crystal choice it should be considered the simplicity to obtain assembly and feasible cost. These requirements make CsI based scintillation crystals promising materials for this application because they exhibit a relatively low hygroscopy, high atomic number, a low cost and easy handling [4, 5]. The CsI crystals commercially available are doped by Tl or Na, although they present restriction in the measurement of the high counting rates due to their slow luminescence decay time. The pure CsI crystals have a fast decay time of ~10 ns. However, their light yield of the two components is 0.06 and 0.02 of NaI:Tl

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crystals [5]. Ions of divalent lead (Pb^{2+}) doped in the crystal structure are an efficient emission center and their application as scintillators is still the reason for intensive studies doping Pb ion in different compounds. Recently, promising results have been found for crystals of CsI doped by bromine (Br) for their use as radiation detectors [2]. However, few studies have been found in the literature on the development and scintillation characteristics of CsI:Br and CsI:Pb. In this work, cesium iodide (CsI) doped by cesium bromide (CsBr) and lead iodide (PbI) were grown by the Bridgman technique, varying the concentrations of the Br and Pb from 5×10^{-4} to 1.5×10^{-1} M. The mechanical and scintillation characteristics of the developed CsI:Br and CsI:Pb crystals were studied.

Materials and methods

CsI crystals with Br of 1.0×10^{-2} M, 5.0×10^{-2} M, 1.0×10^{-1} M, 1.5×10^{-1} M and CsI crystals with Pb of 1.0×10^{-2} M, 1.5×10^{-3} M, 1.0×10^{-3} M, 5.0×10^{-4} M used in this work were grown in accordance with the vertical Brigdman technique, using a quartz crucible in vacuum atmosphere. The starting procedure used with the purity of 99.99% was obtained from Metal Gesellschalt K.K., Germany. Crystals around 120 mm long were obtained with a growth rate of 1.2 mm/h. Pure CsI crystal was also grown for comparison.

The mechanical properties of the crystals were evaluated by microhardness measurements using a version of the Vickers method, with a Micrometer 2100 electronic microdurometer (Buehler Lake Bluff).

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 luminescence decay time curves were carried out under excitation of 662 keV gamma rays from a ^{137}Cs source by an oscilloscope. The scintillation pulse decay was measured. The decay data were decomposed and analysed with the least squares fitting technique.

The scintillation light output was measured at room temperature using an RCA 8575 photomultiplier (PMT).

Results and discussion

Transparent and uniform CsI:Br and CsI:Pb crystals, 25 mm in diameter and 120 mm in height, were reproducibly grown at a concentration of Br in the range of 1.5×10^{-1} M to 10^{-2} M, while the concentration of the Pb used was of 10^{-2} M to 5×10^{-4} M. Above these concentrations, the crystals were non-uniform in composition and presented large opaque regions. Zaslavsky *et al*., while growing a CsI(Tl) crystal with high Br concentration, have found the same results, which they have attributed to a possible decomposition of the solid solution and non-uniformity of the composition [10].

No significant change was found in the concentration of Br and Pb in crystals one year after their growth, showing the stability of the Br, incorporated in the CsI structure.

In the crystal growth, small concentrations of the dopant distribute in the crystalline matrix, substituting the lattice constituent or occupying no defined position in its structure. As the ionic radius of Br^- (1.96 Å) is close to that of I^- (2.20 Å) and both ions have the same co-ordination number (IV) and approximate electronegativity values, it is expected that Br^- ion substitutes the position of I^- ion in the crystalline lattice. On the other hand, the Pb^{2+} ions have different electronegativity from that of competitive Cs^+ ions in the matrix, (Pb^{2+} = 1.8 eV and Cs^+ = 0.7 eV) and the ionic radius between them differ by about 25%, (Pb^{2+} = 1.29 Å and Cs^+ = 1.74 Å) [7], making the incorporation substitution limited. This explains the difficult incorporation of Pb in crystal growth for Pb concentrations above 10^{-2} M, which showed a Pb composition not constant and a large opaque section. On the other hand, for the Br element it is possible to incorporate up to 1.5×10^{-1} M concentration, to obtain uniform and transparent crystals.

The microhardness results for CsI:Br and CsI:Pb crystals, at different dopant concentrations, were increased as a function of the dopant concentration, in the studied range. Alloying CsI with Br concentrations above 5×10^{-2} M, results in a further two times increase of its microhardness. Zaslavsky *et al*. found similar results alloying CsI(Tl) with Br [10]. In the present study, the effect of addition of Pb^{2+} ions in the CsI matrix resulted in the hardness improvement of the grown crystals. An increase in hardness for the CsI:Pb crystals was found for Pb 10^{-2} M of the order of 1.5 times, in comparison with the pure CsI crystal. In some studies on the hardness of alkali halide crystals doped by divalent ions, an increase in hardness was found [9]. However, there is no reported study of the effect of doping on the hardness of the CsI:Pb crystal. Figure 1 shows the microhardness results for CsI:Br and CsI:Pb crystals.

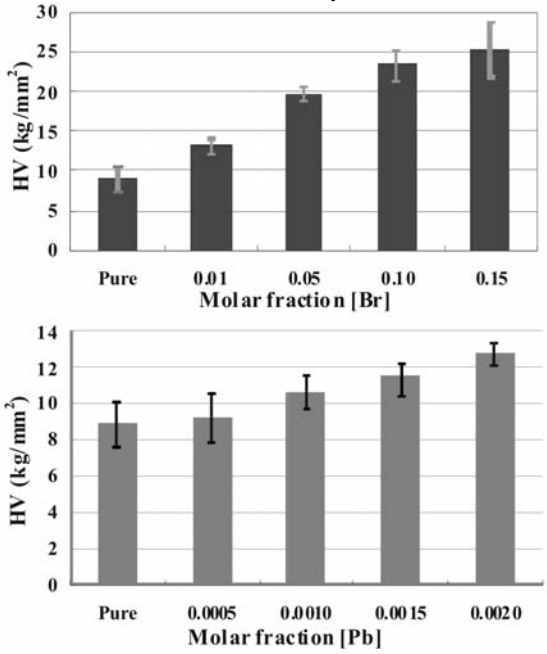

Fig. 1. The microhardness results for CsI:Br and CsI:Pb crystals at different dopant concentrations.

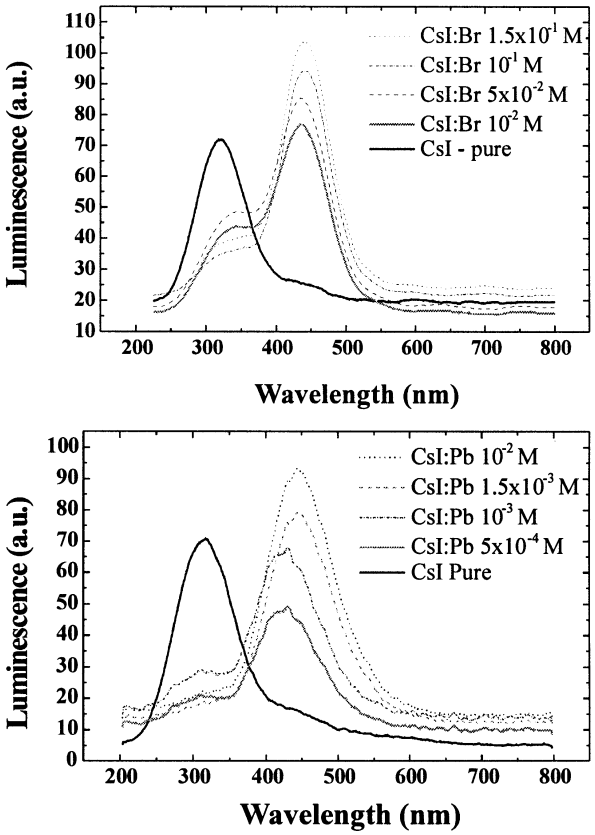

Fig. 2. Luminescence emission spectra of the CsI:Br, CsI:Pb at different concentrations and pure CsI crystals.

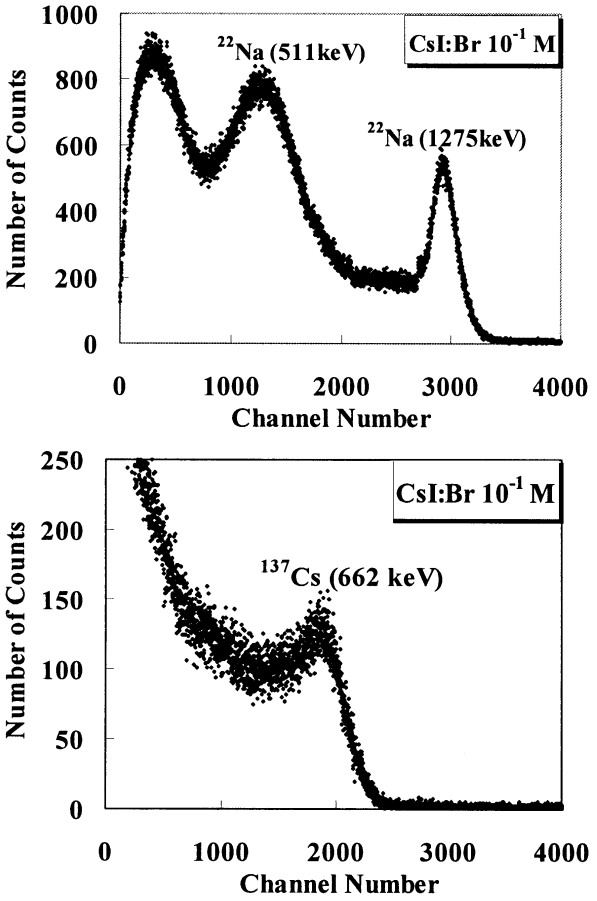

Fig. 3. Energy spectra of CsI:Br 10^{-1} M.

A predominant luminescence band near 450 nm and a single broad band around 320 nm were found with the addition of Br or Pb. The maximum emission luminescence wavelength around 450 nm presents a good match with the spectral sensitivity of photomultipliers, making both CsI:Br and CsI:Pb crystals suitable to be used as radiation detectors. Figure 2 shows the luminescence emission spectra of the CsI:Br, CsI:Pb at different concentrations and pure CsI crystals.

The energy spectra and the energy resolution for CsI:Br, doped with 10^{-1} M of Br when excited with ^{22}Na and ^{137}Cs, are shown in Fig. 3. Although the energy resolution values are poorer compared to those of CsI:Tl, the CsI:Br crystal is more suitable for applications that require fast decay time detectors. On the other hand, CsI:Pb presents a good radiation response, although the photopeak was not defined in the spectra. The best energy resolution was found for CsI:Br doped by 10^{-1} M of Br, compared to the pure CsI and crystals with other concentrations of Br. This crystal showed resolution of 22% when excited with gamma radiation of ^{137}Cs (662 keV) and resolution of 49.1% and 9.1% when excited with gamma radiation of ^{22}Na (511 keV, 1275 keV), respectively.

Figure 4 shows the pulse height spectra of the CsI:Br crystals at different Br concentrations. As it can be observed, CsI:Br at 10^{-1} M presented the best result of the pulse height for gamma radiation when compared to pure CsI crystal and CsI:Br at other concentrations in the studied energy range.

Table 1 summarizes the luminescence decay time results of the pure CsI crystal and CsI:Br, CsI:Pb crystals at different concentrations. The pure CsI crystal has been grown in the present work for comparison between luminescence decay time performances, once this crystal was taken as a reference material. The result evidenced 12 ns luminescence decay time for the pure CsI crystal. This figure is in accordance with the literature data [1]. No significant difference was observed in the luminescence decay time values, in the range from 1.0×10^{-2} M to 1.5×10^{-1} M for CsI:Br, and from 5.0×10^{-4} M to 1.0×10^{-2} M for CsI:Pb. When dopant elements are introduced during crystal development, they may imply very different material behavior. For example, taking thalium as dopant for CsI matrix it resulted in 1300 ns luminescence decay time for CsI:Tl crystal [5]. Thus, the addition of bromide and lead as

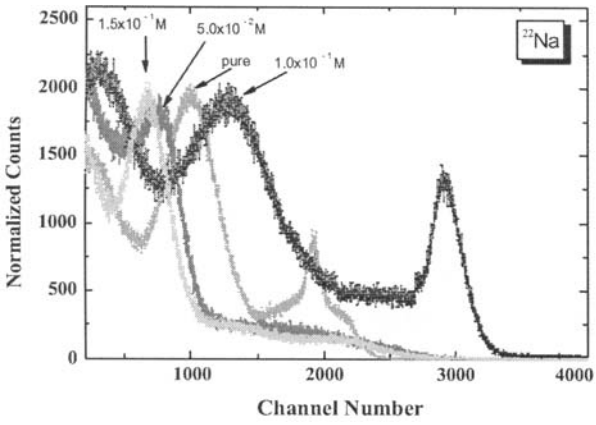

Fig. 4. Pulse height spectra of CsI:Br and pure CsI crystals.

Table 1. Luminescence decay time values of the pure CsI, CsI:Br and CsI:Pb crystals. Experimental uncertainties are 10%

Crystal	Molar fraction	Luminescence decay time (ns)
pure CsI		12
CsI:Br	10^{-2}	16
	5×10^{-2}	17
	10^{-1}	19
	1.5×10^{-1}	19
CsI:Pb	5×10^{-4}	13
	10^{-3}	14
	1.5×10^{-3}	14
	10^{-2}	17

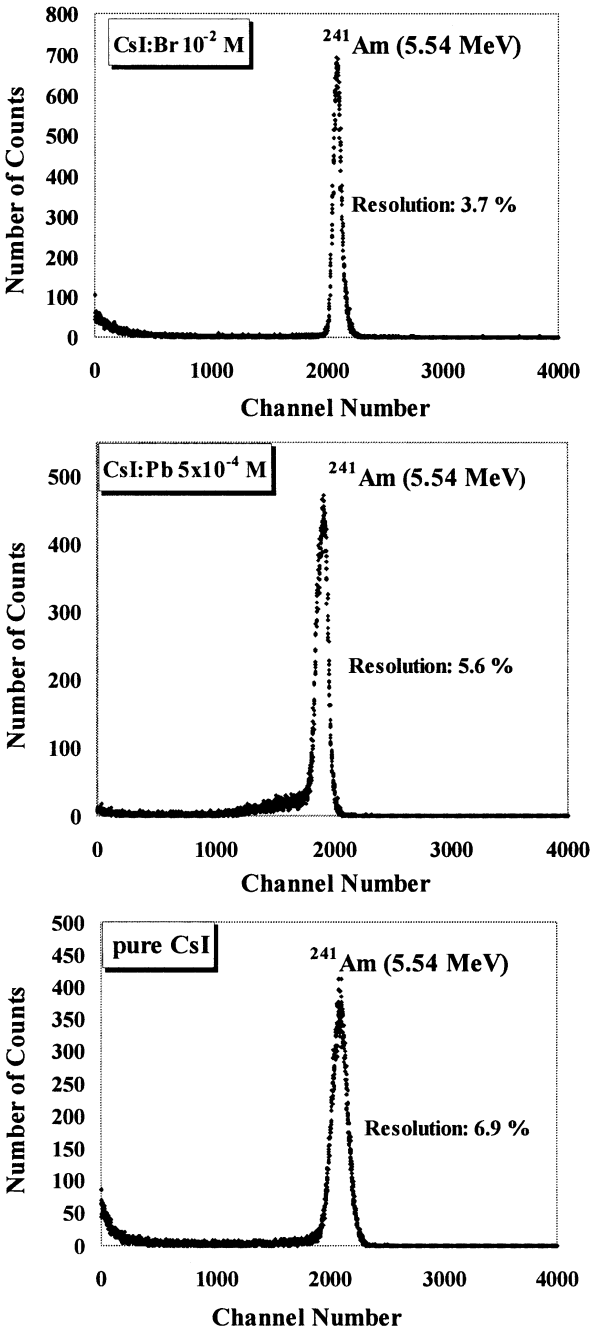

Fig. 5. Energy spectra of the scintillation light from ^{241}Am 5.5 MeV alpha particles in CsI:Br, CsI:Pb and pure CsI crystals.

dopants did not introduce significant changes in the pure CsI crystal luminescence decay time, if compared to thalium dopant.

The spectra of the scintillation from ^{241}Am 5.5 MeV alpha particles, obtained from CsI:Br, CsI:Pb and pure CsI crystals are shown in Fig. 5. CsI:Br and CsI:Pb crystals showed better alpha radiation resolution, when compared with the pure CsI crystal.

The main processes, in which alpha particles lose energy, are excitation and ionization. But, if the alpha particle loses energy in inelastic collisions, before those processes, the resolution is reduced. The addition of the dopants in the crystalline matrix tries to optimize the main processes and reduces loss energy by inelastic scattering; therefore, it results in a better resolution.

Better results of the resolution were found for the CsI:Br and CsI:Pb crystals when excited with alpha particles, in comparison with the results obtained with gamma radiation. It has been suggested that the difference between the response for gamma radiation and alpha particles is associated with the difference in the transport mechanism of electrons and holes towards the luminescence centers [6].

Conclusions

The addition of Br and Pb to the CsI crystal resulted in a crystal with fast decay time, close to the pure CsI, which is a suitable characteristic to be used for detection in high rate detectors, in high energy physics applications.

The CsI:Br crystal doped by 10^{-1} M of Br presented a better gamma radiation detection efficiency and energy resolution, in all the studied energy ranges.

The CsI:Br and CsI:Pb crystals presented a good detection of alpha radiation in all the studied dopants concentration range. These crystal performance evidenced a better resolution, when compared to pure CsI crystal.

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