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# DEPENDENCE OF SCINTILLATION CHARACTERISTICS IN THE CsI:Br CRYSTALS ON $Br^-$ CONCENTRATIONS UNDER GAMMA EXCITATIONS

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## ABSTRACT

In recent years, a great interest in inorganic scintillators with fast luminescence decay time (about ten ns), high density and high light output has emerged, for application in nuclear physics, high energy physics, nuclear tomography and other fields of science and engineering. In this work, the growth of pure CsI crystals and CsI:Br using the Bridgman technique, is described. The concentration of the bromine doping element (Br) was studied in the range of $1.5x10^{-1}$ M to $10^{-2}$ M. To evaluate the scintillators developed, systematic measurements were carried out for luminescence emission and luminescence decay time for gamma radiation, optical transmittance assays, Vickers microhardness assays and analysis of crystals response to the gamma radiation in the energy range of 350 keV to 1330 keV. It was obtained 16 ns to 19 ns for luminescence decay time for CsI:Br crystals. These results were very promissing. The results obtained for microhardness showed a significant increase in function of the doping elements concentration, when compared to the pure CsI crystal, increasing consequently the mechanical resistance of the grown crystals. The validity of using these crystals as radiation sensors may be seen from the results of their response to gamma.

## 1. INTRODUCTION

For 50 years, scintillators have played a major role as nuclear radiation detectors. They are widely used in the experimental nuclear physics, high energy physics, nuclear medicine, nuclear tomography, environmental studies and many other fields of use. This is because of their high detection efficiency for nuclear radiation, capability to measure energy spectra and the possibility to work with a very high counting rate up to $10^7$ $s^{-1}$ counts. Capability to detect a wide assortment of radiations, including gamma and X-rays, charged particles and neutrons, as well as the great variety in size and constitution, make the scintillators the best choice for different applications.

In recent years, the progress in high energy physics and nuclear physics has caused the necessity to develop electromagnetic calorimeters capable of working under high counting rates, requiring the use of multiple detectors with high density [1] and short decay time (about ten ns), thus stimulating the scintillation materials science.These detectors are usually built

with thousands of scintillator crystals and in some experiments the total volume of the detector can reach more than $1m^3$ [2]. 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 facility to handle.

The CsI crystals, commercially available are doped with 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 ~ 10ns. However, their light yield is only of 0.06-0.02 of NaI:Tl crystals [3]. Recently, promising results have been found for the crystal of CsI doped with bromine (Br) for its use as radiation detectors. However, few studies have been found in the literature on the development and scintillation characteristics of the CsI:Br crystals. In this work, Cesium Iodide (CsI) doped with Cesium Bromide (CsBr) were grown by Bridgman technique, varying the concentrations of the Br from $5x10^{-4}$ to $1.5x10^{-1}$ mole. The mechanical and scintillation characteristics of the developed CsI:Br crystals were studied.

## 2. MATERIALS AND METHODS

CsI crystals with [Br] of $1.0x10^{-2}$, $5.0x10^{-2}$, $1.0x10^{-1}$, $1.5x10^{-1}$ mole used in this work were grown in accordance with the vertical Brigdman technique, using a quartz crucible in vacuum atmosphere. The starting material used with a purity of 99.99% was obtained from Metal Gesellschalt K.K., Germany. Before starting the crystal growth, the powder was purified by evacuating and heating in the quartz crucible at about 200°C, for 3 h, to remove mainly residual water. After the purification process, the quartz crucible was filled with argon gas and sealed. Crystals around 110 mm long were obtained with a growth rate of 1 mm/h. Pure CsI crystal was also grown for comparison.

The emission spectra of these crystals were measured with a monochromator (JASCL FP55A) for 511keV 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 662 keV gamma rays from a $^{137}Cs$ source by oscilloscope [4]. 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).

The optical transmission measurements were made in the CsI:Br crystals. This measurement was performed at room temperature, using a Shimadzu spectrophotometer with a wavelength varying in the range between 200 to 800nm.

The mechanical properties of the crystals were evaluated by microhardness measurements. Microhardness was measured by a version of the Vickers method, using a Micromet 2100 electronic micro durometer (Buehler Lake Bluff).

## 3. RESULTS AND DISCUSSION

Transparent and uniform CsI:Br crystals, 25 mm in diameter and 110 mm high, were reproducibly grown at a concentration of (Br) in the range of $1.5x10^{-1}$ M to $10^{-2}$ M. Above these concentrations, the crystals were non-uniform in composition, and presented large opaque regions. Zaslavsky et al., growing CsI(Tl) crystal with high [Br] concentration, have found the same results, which they attributed to a possible decomposition of the solid solution and non-uniformity of the composition [5].

A predominant luminescence band near 450 nm and a single broad band around 320 nm were found with the addition of the Br, as showed in Fig. 1. The maximum emission luminescence wavelength around 450 nm presents a good match with the quantum efficiency spectra of the photomultiplier, making the CsI:Br suitable to be used as radiation detectors.

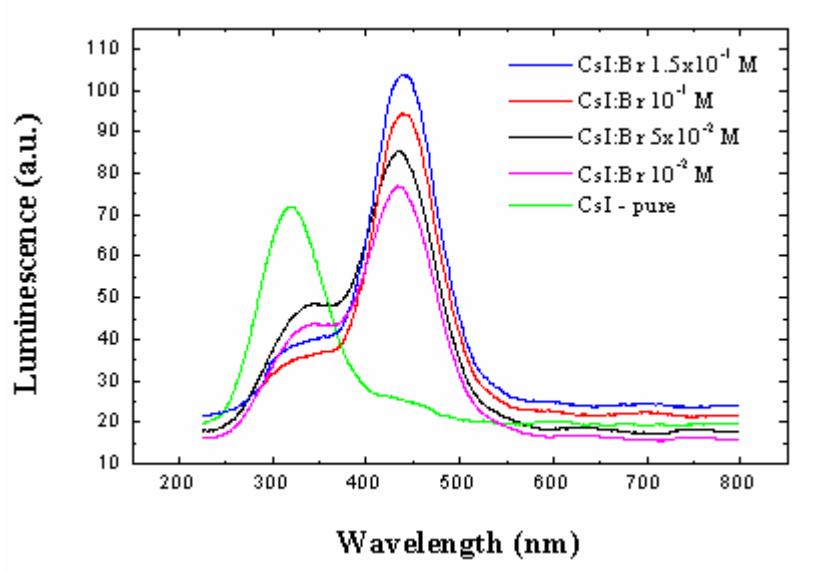


**Figure 1. Emission spectra of the CsI:Br at different concentrations and pure CsI crystals.**

Table 1 summarizes the luminescence decay time results of the CsI:Br crystals at different concentrations. No significant difference was observed in the decay time values in the range from $1.0x10^{-2}$ to $1.5.0x10^{-1}$ mole for CsI:Br. Moreover, the addition of the dopant Br did not change significantly the decay time value in the CsI crystal [6].

**Table 1. Decay time values of the pure CsI and CsI:Br. Experimental uncertainties are 10%.**

| Crystal | Molar Fraction | Luminescence Decay time (ns) |
|---|---|---|
| pure CsI | | 12 |
| CsI:Br | $10^{-2}$ | 16 |
| | $5x10^{-2}$ | 17 |
| | $10^{-1}$ | 19 |
| | $1.5x10^{-1}$ | 19 |

Fig. 2 presents the optical transmission measurement for CsI:Br crystals. The transmittance decreases as the dopants concentrations increase for crystals.

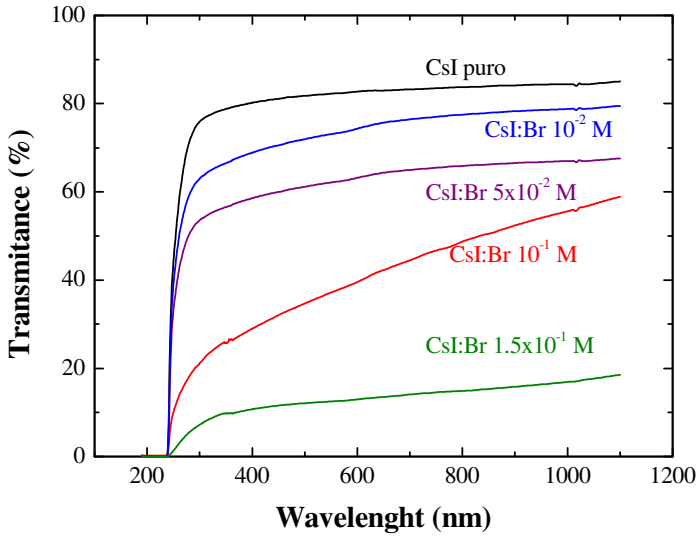


**Figure 2. Transmittance curves of the CsI:Br at different concentrations.**

The microhardness results for CsI:Br crystals at different dopant concentrations are showed in Fig.3. It is clearly demonstrated that alloying CsI crystal with Br increases its microhardness as a function of the dopant concentration increase in the studied range. Alloying CsI with Br concentrations above $5x10^{-2}$ mole results in a further two times increase of its microhardness. Zaslavsky et al found similar results alloying CsI(Tl) with Br [5].

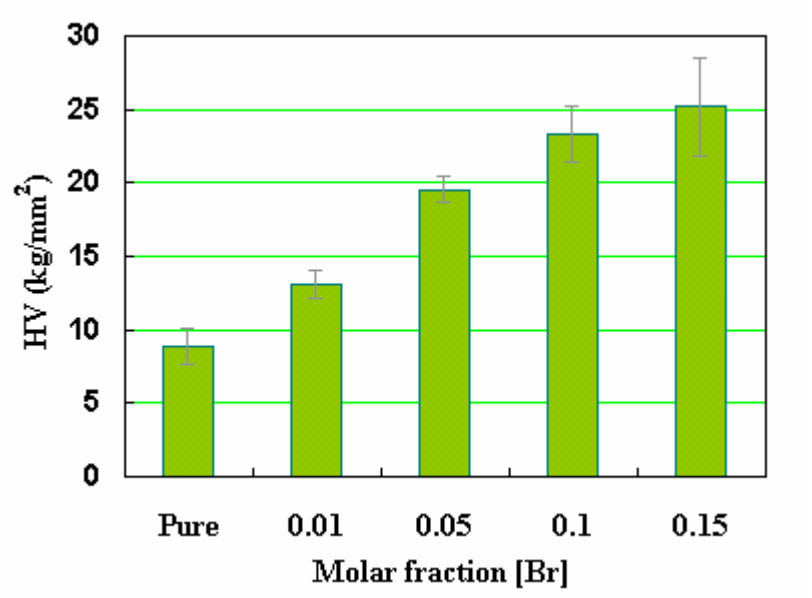


**Figure 3. Vicker microhardness of the pure CsI crystal and for CsI:Br crystals at different dopant concentrations.**

The energy spectra and the energy resolution for CsI:Br, with molar fraction $10^{-1}$ when excited with $^{137}Cs$, $^{54}Mn$, $^{22}Na$ and $^{65}Zn$ are showed in Fig. 4. Nevertheless, 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. Table 2 summarises the energy resolution results for pure CsI and CsI:Br crystals at different concentrations of dopants using different gamma radiation energies. The best energy resolution was found for CsI:Br doped with $10^{-1}$ M of Br, compared to the pure CsI and crystals with other concentrations of Br.

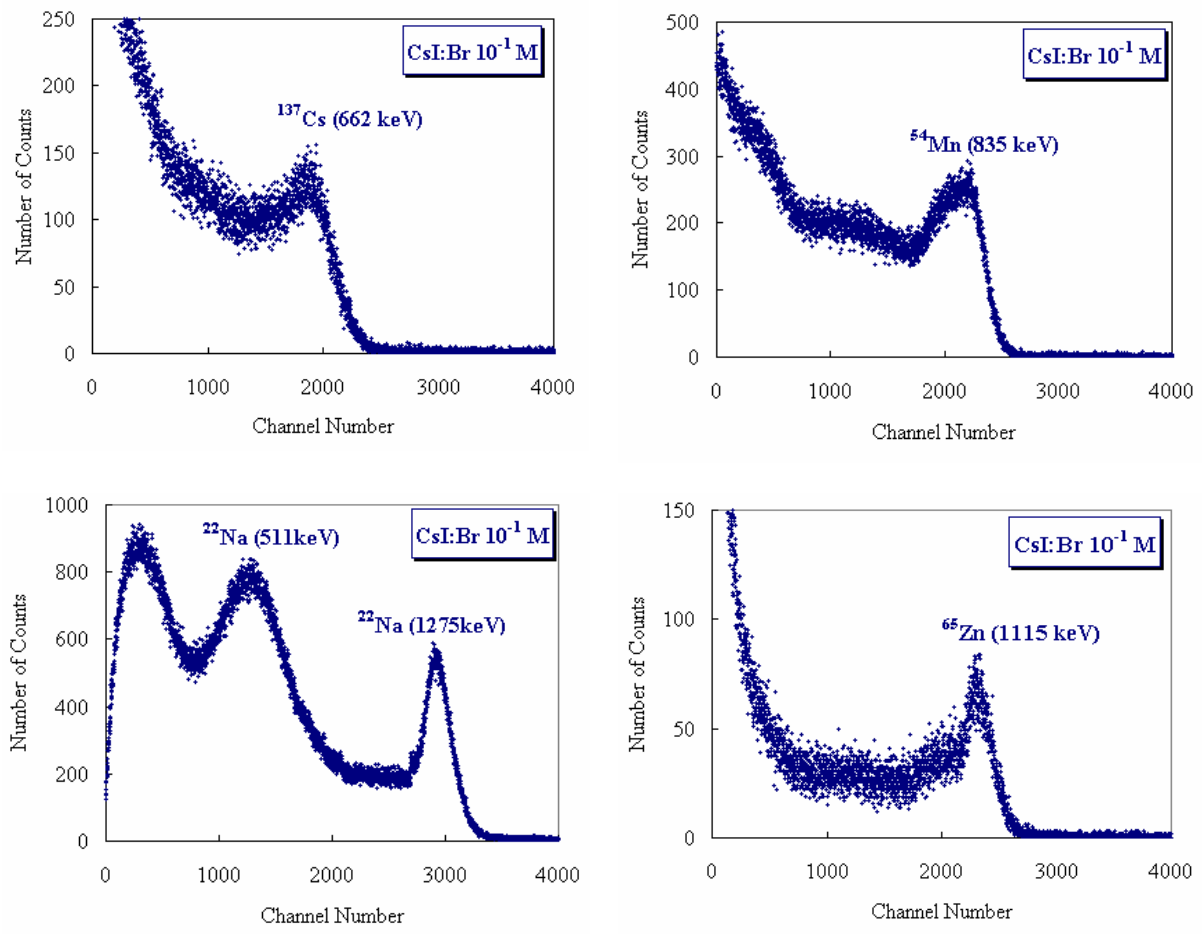


**Figure 4. Energy spectra of the CsI:Br $10^{-1}$ M.**

As it can be observed in Table 2, the resolution for all energies studied may be determined for the CsI detector doped with Br with a molar fraction $10^{-1}$. For the crystals doped with concentrations smaller than $10^{-1}$ M of Br, it was not possible to determine the resolution for energies higher than 662 keV. In the CsI:Br crystals, the luminescence observed at 450 nm is originated predominantly by the (Br) dopant presence and the luminescence spectrum plus its characteristics depend on the nature of the electronic transition involved in the emission process. This may explain the differences obtained in the resolution results, for different dopant concentrations. Another factor likely to be attributed to the differences observed in the energetic resolution results is the intrinsic resolution or the quality of each crystal.

**Table 2. Energy resolution of CsI:Br and pure CsI crystals for different concentrations of dopants. (NI: photopeak not identified)**

| Radioactive source | Energy (keV) | CsI:Br crystal $10^{-2}$ M Resolution (%) | CsI:Br crystal $5x10^{-2}$ M Resolution (%) | CsI:Br crystal $10^{-1}$ M Resolution (%) | CsI:Br crystal $1.5x10^{-1}$ M Resolution (%) | pure CsI crystal Resolution (%) |
|---|---|---|---|---|---|---|
| $^{133}$Ba | 355 | >90 | >90 | >90 | >90 | >90 |
| $^{22}$Na | 511 | 53,1 | 40,7 | 49.1 | 46 | 51.4 |
| $^{137}$Cs | 662 | 36,7 | NI | 22 | 32,4 | 41.2 |
| $^{54}$Mn | 835 | NI | NI | 28.2 | 27,8 | 26.5 |
| $^{65}$Zn | 1115 | NI | NI | 11.2 | NI | 14.1 |
| $^{60}$Co | 1173 | NI | NI | 9.3 | NI | 11.1 |
| | 1333 | | | | | |
| $^{22}$Na | 1275 | NI | NI | 9.1 | NI | 8.7 |

## 4. CONCLUSIONS

Concluding, the addition of the Br to CsI crystal resulted in a crystal with fast decay time, close to the pure CsI, what is a suitable characteristic to be used for detection in high counting rate detectors, in high energy physics applications. The crystal CsI:Br, doped with $10^{-1}$ M of Br, presented the best detection efficiency of the radiation gamma and energy resolution in all studied energy ranges.

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