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ENERGETIC RESOLUTION STUDY ON PURE AND CsBr DOPED CsI UNDER GAMMA EXCITATIONS AND ALPHA PARTICLES

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ABSTRACT

Pure and doped CsI crystals were grown using the Bridgman technique. Bromine was the doping element which was studied in the range of $1.5x10^{-1}$ M to 10^{-2} M. The distribution of the doping element at crystalline volume was determined by neutron activation. Concerning gamma radiation response it was carried out measurements to evaluate the developed scintillators in the energy range of 350 keV to 1330 keV. For alpha particles measurements an ^{241}Am source was used with 5.54 MeV energy. The resolution of 3.7% was obtained for the CsI:Br 10^{-2} M crystal, when excited with alpha particles from an ^{241}Am source. For CsI:Br 10^{-1} M crystal 9.1% resolution was obtainded when excited with gamma radiation from ^{22}Na source, with 1275 keV energy.

Keywords: crystals, energy resolution, CsI, radiation detectors

1. INTRODUCTION

The development of new radiation detectors using scintillation crystals, what permits to increase the speed of response, the accuracy in dose and energy, and at the same time, feasibility to simplify and reduce costs in the production process is always needed.

Inorganic scintillators, are explored in the new fields as positron emission tomography (PET), computed X-ray tomography (CT), spatial physics and astronomy. In the many cases CsI:Tl and NaI:Tl conventional scintillators are used, while barium fluoride (BaF_2) and bismuth germanate (BGO) scintillators are appropriate for PET. Inorganic scintillators are also used in high energy physics, for measurements of gamma energy and electrons/positrons in accelerators. A scintillation detector of large dimensions, named electromagnetic calorimeter of total absorption (EM), carries a lot of crystals, in some cases more than 10^4 crystals and total weight up to 10 ton. The NaI:Tl was the first cintilator material used in calorimeter (EM). Subsequently, CsI:Tl crystals were used and, recently, BGO scintillators have been used. BaF_2 and pure CsI scintillators have been used in radiation detectors of small proportions in HEP and in experimental nuclear physics [1].

There is, hence, constant interest in find new scintillator materials or to improve the characteristics of known scintillators. CsI doped with Tl is one of the most widely used scintillators because of its hight yield (~ 65000 ph/MeV). However, their long decay time (~1300 ns) reduces their application mainly for low count rate measurements [2]. In contrast, undoped CsI has a very shorter decay time (~10 ns), but its scintillation efficiency is also largely reduced (~ 200 ph/MeV) [3]. CsI doped with cesium bromide was proposed as a scintillator that combines advantages of the both above mentioned crystals.

The CsI matrix is relatively low hygroscope, has a high atomic number, facility to handle and a low cost material. The aim of the present study is to grow pure and doped crystals for comparasion. The energy resolution of the undoped CsI crystal was compared with the bromine doped CsI crystals, under gamma excitations and alpha particles.

2. MATERIALS AND METHODS

The pure CsI and those CsBr doped crystals were grown in accordance with the vertical Bridgman techique [4,5].

The subsequent determination of bromine concentration as dopant in the CsI:Br crystals was performed by neutron activation analysis (NAA) [6]. The investigated dopant concentrations were 10^{-2}M, $5x10^{-2}$M, 10^{-1}M e $1,5x10^{-1}$M. This method consists, essentially, of production of artificial radionuclide from steady elements by irradiation under neutron flow and measure of emited gamma radiation, which is characteristic for each element. The sample was irradiated in the IEA-R1 Nuclear Reactor for 8 hours. The applied neutron flow was $10^{12}cm^{-2}s^{-1}$. The sampling for characterization analysis was collected in intermediate sections of crystals (at the same distance of the growth start).

The dopant concentration profile in the CsI:Br crystal (10^{-1} M nominal concentration) was determinated by the same method. The crystal sample was cut in 18 slices, 6mm each, as showed in Fig. 1. One gram of each slice was taken for this analysis.

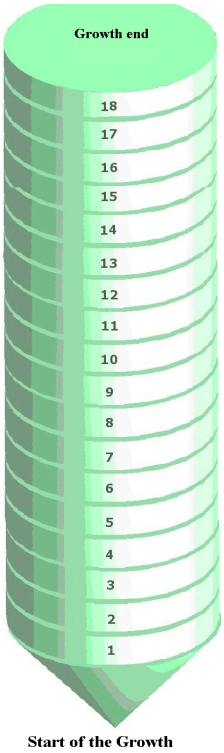

Figure 1. Scheme in court of the CsI:Br crystal (10^{-1} M)

To study the response to gamma radiation and alpha particles, the crystals were machined, polished and coupled directly to the bi-alkali photomultiplier tube (RCA 8575 model, 21-pin). Silicone grease (Dow Corning – viscosity 1.0 McStokes) was the optical interface material. This provides a uniform refraction index at full contact surface between the crystal and the photomultiplier tube. The sides of the crystal which were not in contact with the photosensor were covered with Teflon tape to ensure good reflection of light. The radioactive sources were positioned in the center of the upper face of the crystal. The electronic modules used for processing signals from the photomultiplier tube were as it follows: pre-amplifier (Ortec model 276), amplifier (Ortec model 450), source voltage (Ortec 556), multichannel analyzer (Ortec ADCAM model 918A), Phillips oscilloscope (PM3295A 400MHz) and Pentium III microcomputer.

The energy resolution of the detectors crystal CsI:Br and pure CsI coupled to photomultiplier tube was determined using gamma radiation sources and energies between 355 keV to 1333 keV, and 5.54 MeV for alpha particles source. The operation voltage of photomultiplier was 2700 V for the detection of gamma rays and 2200 V for the detection of alpha particles. The accumulation time during the counting was 600 s. The crystals used in gamma spectroscopy were cut with dimensions of 2 cm in diameter and 2 cm in height and in the spectroscopy for alpha radiation, crystals to 2 cm in diameter and 5 mm thick were used.

3. RESULTS AND DISCUSSION

In this work, the bromine was added to the starting material (salt CsI) with molar fractions of 10^{-2}, $5x10^{-2}$, 10^{-1}, $1.5\ x10^{-1}$. The results of neutron activation analysis represent the concentration of bromine in the crystal CsI: Br and pure CsI grown. They are shown in Table 1.

Table 1. Content of bromine found in an intermediate region of the crystals CsI: Br, determined by neutron activation. Each result is the average of 3 results, with 3 different patterns of Br with the standard deviation.

Sampling of the crystals	Bromine concentration (molar fraction)
CsI:Br 10^{-2} M	$0.75x10^{-2} \pm 0.02x10^{-2}$
CsI:Br $5x10^{-2}$ M	$4.32x10^{-2} \pm 0.12x10^{-2}$
CsI:Br 10^{-1} M	$0.91x10^{-1} \pm 0.02x10^{-1}$
CsI:Br $1{,}5x10^{-1}$ M	$1.41x10^{-1} \pm 0.04x10^{-1}$
pure CsI	ND

ND - not detected under the experimental conditions of analysis.

The obtained results by neutron activation analysis (NAA), Table 1, confirm that the average values found in the grown crystals are consistent with those introduced in the salt CsI, showing the incorporation of Br in the structure of the CsI matrix.

The concentration of dopant in the crystal is a parameter of quality control, important in the production of crystal scintillators, because of the luminescence properties are affected by the concentration of dopant. The bromine concentration results, in eighteen regions of the crystalline block of CsI: Br 10^{-1} M, are shown in Table 2 and represented in Fig. 2.

In the study of distribution of the bromine dopant along the crystalline volume, bromine was added to the starting material (salt CsI) with mole fraction of 10^{-1}. During the crystallization process, that concentration was changed resulting in a concentration gradient The neutron activation analysis results indicated a higher concentration at the top of the crystal, with subsequent decrease in the initial phase of growth. A relative uniformity of Br concentration between slice 14 and slice 3 was found, as shown in Fig. 2 and Table 2, which is the region of the crystalline volume indicated for use as radiation detector. From the slice 14 on, the concentration of bromine increases abruptly. Therefore crystals cutted in this region do not have uniformity of doping. The high concentration of bromine in the top of the crystal is expected, taking into account that the crystal grows from the bottom up and impurities tend to deposit at the end of the growth, due to the phenomenon of segregation.

Table 2. Determination of bromine concentration in 18 slices of 6 mm thick CsI: Br crystal, performed by neutron activation analysis.

Slice of the CsI:Br crystal	Bromine concentration (molar fraction)
1	$0.50x10^{-1} \pm 0.003$
2	$0.52x10^{-1} \pm 0.003$
3	$0.57x10^{-1} \pm 0.004$
4	$0.53x10^{-1} \pm 0.003$
5	$0.60x10^{-1} \pm 0.004$
6	$0.50x10^{-1} \pm 0.003$
7	$0.56x10^{-1} \pm 0.004$
8	$0.61x10^{-1} \pm 0.004$
9	$0.65x10^{-1} \pm 0.004$
10	$0.64x10^{-1} \pm 0.004$
11	$0.66x10^{-1} \pm 0.004$
12	$0.61x10^{-1} \pm 0.004$
13	$0.66x10^{-1} \pm 0.005$
14	$0.81x10^{-1} \pm 0.006$
15	$0.95x10^{-1} \pm 0.007$
16	$0.96x10^{-1} \pm 0.007$
17	$1.01x10^{-1} \pm 0.008$
18	$1.56x10^{-1} \pm 0.012$

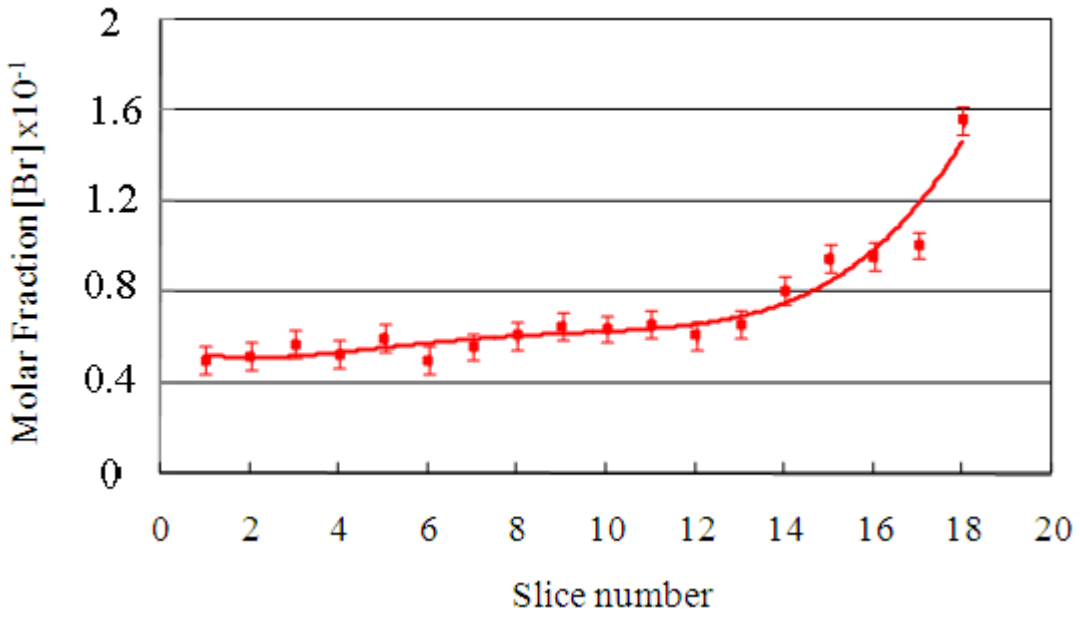

Figure 2. Concentration of bromine according to the height of the CsI: Br crystal.

A homogeneous distribution of dopant in an appreciable region of the crystals is highly desirable in materials for the construction of radiation detectors, since the optimization of the scintillation light emission efficiency is dependent on that homogeneity.

The spectra shown in Fig. 3 illustrate the results of gamma spectroscopy for ^{22}Na radiation, obtained with the crystal CsI: Br 10^{-1} M, and pure CsI crystal, coupled to the photomultiplier tube. The measurements conditions were identical for all crystals, i.e., distance between source and crystal, the counting time, tension in the photomultiplier tube, signal amplification and size of crystals. Resolutions were obtained in 49.1% and 9.1% for energy of 511 keV and 1275 keV, respectively, using crystal CsI: Br 10^{-1}M and resolutions of 51.4% and 8.7%, for energy of 511 keV and 1275 keV, respectively, using pure CsI crystal.

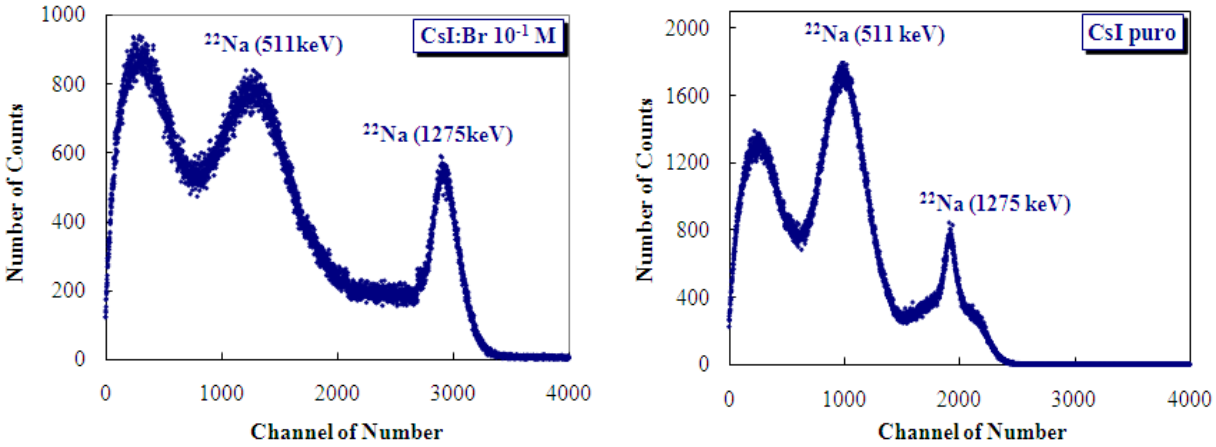

Figure 3. Spectra obtained for ^{22}Na radiation with CsI: Br 10^{-1}M and pure CsI crystals.

The resolutions percentage of the CsI:Br and pure CsI crystals, depending on the energy of gamma radiation in the range of 355 keV to 1333keV, are shown in Fig. 4.

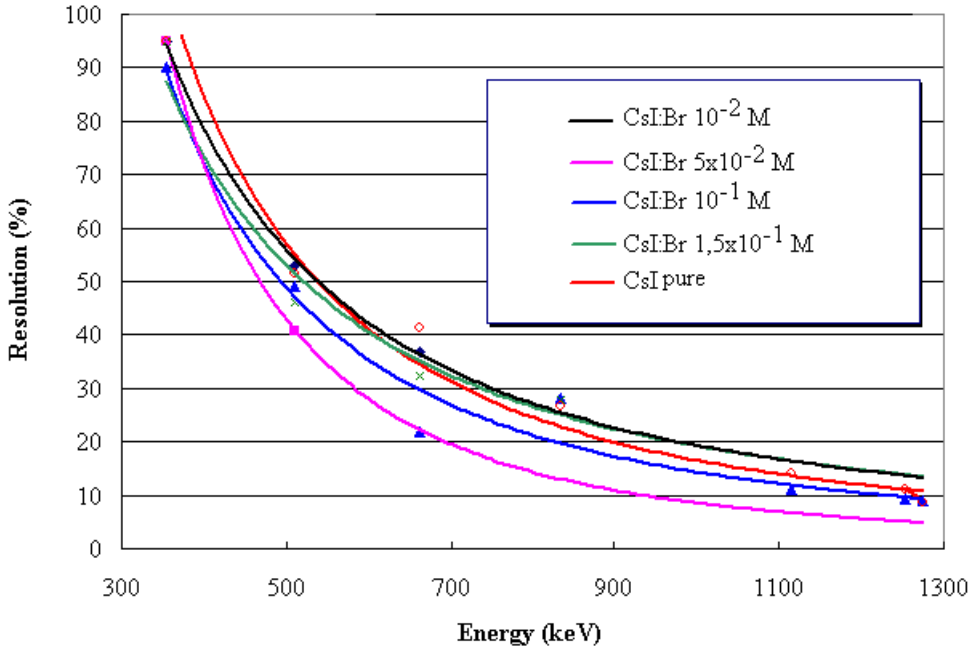

Figure 4. Resolutions percentage of crystals CsI:Br with various concentrations of bromine and pure CsI as a function of gamma radiation energy.

The resolution for all studied energies was able to be determined only for the detector CsI:Br, doped with molar fraction of 10^{-1}. For crystals doped with lower concentrations of Br 10^{-1} it was not possible to determine the resolution for energies higher than 662 keV. In CsI: Br crystals, the luminescence is caused primarily by the presence of Br dopant and its characteristics depend on the nature of electronic transition involved in the emission process. This may explain the differences from obtained results for a resolution of different dopant concentrations. In doped crystals with small amounts of impurities, called extrinsic crystals, although the radiation produces pairs of ions or excitons in the atoms of the host (CsI), they return to ground state, dissipating heat without radioactive emission [7]. Another factor that can be attributed to differences in the results of energy resolution is the intrinsic resolution or quality of the crystal [8]. The intrinsic resolution is influenced by the concentration and uniformity in the distribution of dopant in the crystal, the number of internal reflections of photons in the visible region, generated from the dopant, and conditions of polishing [9]. These factors can interfere with the statistics of light photons detection, emitted inside the crystal, due to the occurrence of the attenuation of produced light.

In Fig.5, the results of alpha spectroscopy for radiation of ^{241}Am (5.54 MeV), are presented. They were obtained with the crystal scintillators CsI: Br and pure CsI.

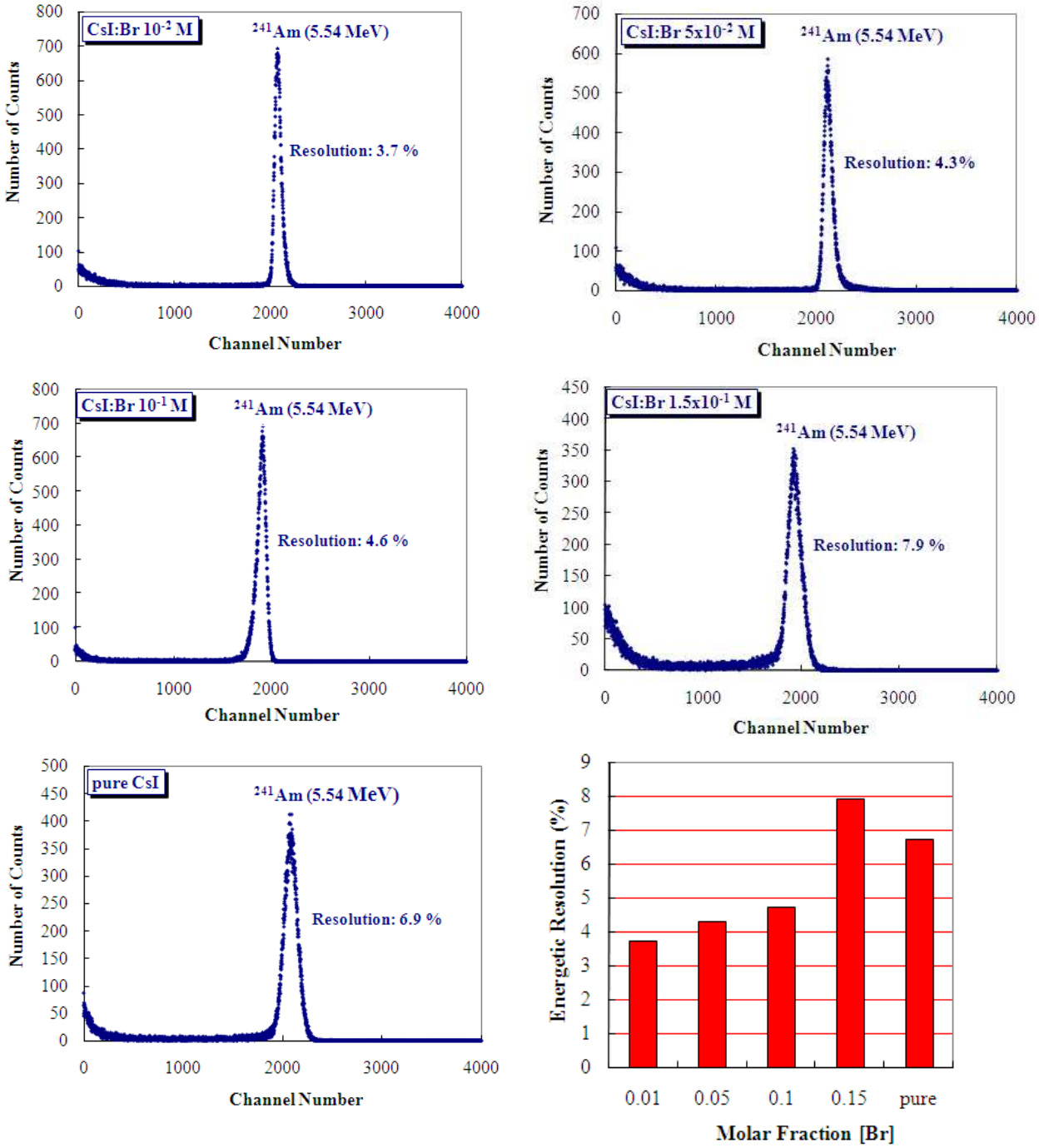

Figure 5. Spectra obtained for the radiation of ^{241}Am with CsI:Br and pure CsI crystals

4. CONCLUSIONS

The analysis of the bromine dopant concentration done by neutron activation showed the stability of bromine incorporated into the CsI structure. Analysis of bromine dopant concentration in eighteen pieces of crystal with 10^{-1} molar fraction showed that 70% of the crystal presented a relatively homogeneous region as concentration of bromine, which is the crystalline volume fraction suitable for use as scintillator.

The CsI:Br crystals showed response to gamma radiation with defined photopeak spectra, and in the studied concentration range the crystal with molar fraction 10^{-1}, presented the best resolution.

The results for the crystal CsI:Br with 10^{-1} molar fraction suggest uniformity of scintillation properties in the crystalline volume, probably due to adequate transparency in the region of luminescence, the emission of the activator and uniform distribution of individual emission centers, responsible by the scintillation in the crystalline volume.

The light production was dependent on the concentration of dopant, when the crystals were excited with gamma radiation. However, regarding to dopant concentration dependence for excitation with alpha particles, no significant differences were observed.

The crystals CsI:Br showed response to radiation of alpha particles demonstrating spectra with defined peaks, and in the concentration range of studied doping, the crystal with molar fraction 10^{-2} exibited the best resolution.

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