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STUDY OF PURE AND Pb^{2+} IONS DOPED CsI CRYSTALS UNDER ALPHA PARTICLES EXCITATIONS

Maria da Conceição Costa Pereira, Tufic Madi Filho and Margarida Mizue Hamada

Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
macoper@ipen.br

ABSTRACT

Scintillation crystals have been used in various fields, such as high energy physics, nuclear instrumentation, radiation measurements, medical imaging, nuclear tomography, astrophysics and other fields of science and engineering. For these applications, the development of good performance scintillation crystals is required. Scintillation crystals based on cesium iodide (CsI) matrix are matters with relatively low hygroscope, easy handling and low cost, characteristics that favor their use as radiation detectors. In this work, pure CsI crystal and lead doped CsI crystals were grown using the Bridgman vertical technique. The concentration of the lead doping element (Pb) was studied in the range of 10^{-2} M to $5x10^{-4}$ M. The distribution of the doping element in the crystalline volume was determined by flame atomic absorption. The CsI:Pb crystal with nominal concentration of 10^{-3} M was cut into 14 slices of 6 mm. The results show a higher concentration at the top of the crystal with a decrease in the initial phase of growth. The dopant concentration of Pb showed good uniformity from the slice 2 to the slice 12: the region is, therefore, suitable for use as radiation detector. The luminescence emission of these crystals were measured. A predominant luminescence band near 450 nm and a single broad band around 320 nm were found with the addition of the Pb^{2+} ions. Analyses were carried out to evaluate the developed scintillators, concerning alpha particles. The resolution of 5.6% was obtained for the CsI:Pb $5x10^{-4}$ M crystal, when excited with alpha particles from a ^{241}Am source, with energy of 5.54 MeV.

Keywords: radiation detectors, alpha particles, crystals growth, Bridgman technique, CsI:Pb

1. INTRODUCTION

Scintillators based on cesium iodide are the leading ones now, among the materials available for solid-state detectors. The progress in high energy physics and nuclear physics has stimulated the science of scintillation materials in two directions. First, there is the search and development of new perspective materials for scintillators. Second, there is a further improvement of the traditional scintillators, including alkali halides. The scintillators created on basis of alkali iodides, such as CsI, find wide applications in the areas of high energy physics, nuclear physics, medicine, industry, security and environmental control devices, geology, astrophysics and other fields of science and engineering. The crystals based on alkali halides are often preferred to other materials due to their low price and the support of developed technologies and equipment for the growth of large crystals with good quality [1,2].

Ions of divalent lead Pb^{2+} built in some crystal structures are efficient emission centers and their application in scintillators is still the goal of an intensive study of emission properties in different compounds containing these ions [3,4].

The development of new radiation detectors using scintillation crystals is important, since it will permit to increase the speed of response, the accuracy in dose and in the energy determination: at the same time, the feasibility to simplify and reduce costs in the production process is another benefit.

There is, hence, constant interest in finding new scintillating materials or in improving the characteristics of known scintillators. The CsI matrix has relatively low hygroscope and a high atomic number; it, also, presents facility to handle and low cost [5].

The aim of the present study is to grow pure and doped crystals for comparison. The energy resolution of the undoped CsI crystal was compared with the lead doped CsI crystals, under alpha particles excitations.

II. MATERIALS AND METHODS

CsI crystals with [Pb]/[CsI] mole fraction $5x10^{-4}$, 10^{-3}, $1.5x10^{-3}$, 10^{-2} used in this work were grown in accordance with the vertical Brigdman technique [6], using a quartz crucible in vacuum atmosphere. The starting material (CsI) 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 sealed. Crystals around 100 mm long were obtained with a growth rate of 1.5 mm/h. Pure CsI crystal was also grown for comparison.

The lead dopant concentration profile in the CsI:Pb crystal with 10^{-3} M nominal concentration was determined by flame atomic absorption. Samples of the crystal were cut in 14 slices of 6 mm each. From each slice, 1 g sample was obtained for analysis.

Samples of CsI:Pb crystals, in various concentrations of the dopant element and pure CsI, were prepared in 1 mm thick slices, polished and properly subjected to the exposure of an incident beam of X-rays from a molybdenum tube (20 mA, 40 kV), at a distance of 50 mm. The exposure time was 30 minutes, thus obtaining the spectrum of Laue.

The luminescence emission spectra of these crystals were measured with a spectrofluorometer (JASCO FP 55A) for 511 keV annihilation gamma rays, from a ^{22}Na source excitation. The signal from the spectrofluorometer was detected with a UV sensitive quartz photomultiplier (Hamamatsu Photonics R1668).

The energy resolution of the CsI:Pb crystal and pure CsI coupled to a photomultiplier tube, was determined using a Am-241 source of alpha particles with energies of 5.54 MeV. The operation voltage of the photomultiplier was 2200 V for the detection of alpha particles and the time of accumulation in the counting process was 600 s. The crystals used in the spectroscopy for alpha radiation were cut with dimensions of 2 cm in diameter and 5mm thick.

III. RESULTS AND DISCUSSION

CsI:Pb crystals, with 25 mm in diameter and 110 mm high were reproducibly grown by the Bridgman technique, at a concentration of (Pb) in the range of $5x10^{-4}$ M to 10^{-2} M. The crystals are shown in Fig. 1.

Figure 1. Pure and Pb^{2+} ions doped CsI crystals.

The results of the analysis by flame atomic absorption, to determine the concentration of lead in the pure CsI and CsI:Pb crystals, are show in Table 1.

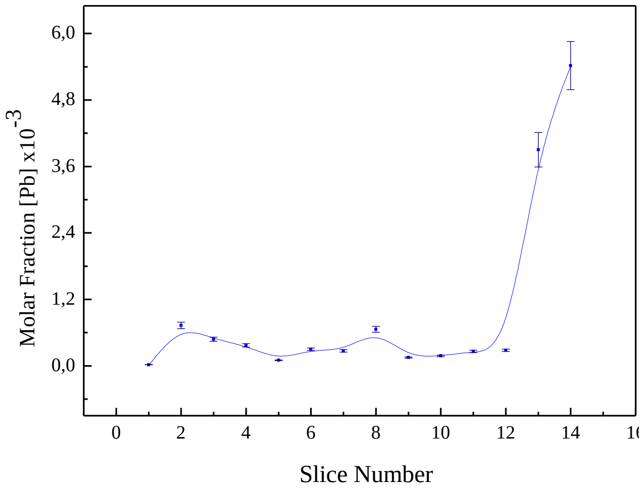
Table 1. Content of lead found in an intermediate region of the CsI:Pb crystals, determined by flame atomic absorption.

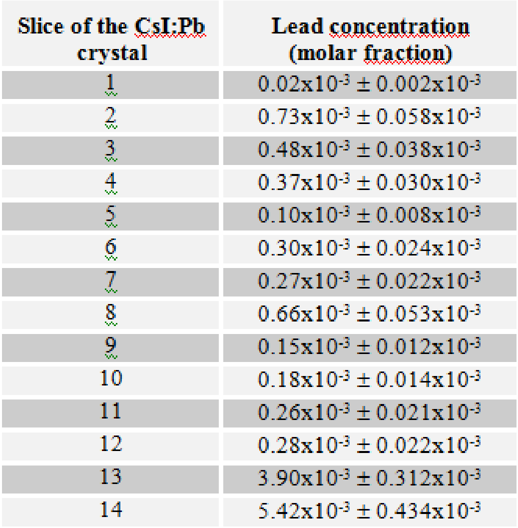
Crystals Samples	Lead concentration (mole fraction)
CsI:Pb $5x10^{-4}$ M	$0.35x10^{-4} \pm 0.028x10^{-4}$
CsI:Pb 10^{-3} M	$0.20x10^{-3} \pm 0.016x10^{-3}$
CsI:Pb $1.5x10^{-3}$ M	$0.68x10^{-3} \pm 0.05x10^{-3}$
CsI:Pb 10^{-2} M	$0.18x10^{-2} \pm 0.01x10^{-2}$
pure CsI	ND

ND: not detected under the experimental conditions of analysis.

The doping element lead was added to the starting material (salt CsI) with a mole fraction of $5x10^{-4}$, 10^{-3}, 1.5 $x10^{-3}$ and 10^{-2}. For result of analysis by flame atomic absorption showed that the incorporation of lead occurred in the matrix of CsI (Table 1). Crystals with Pb concentrations above 10^{-2} M exhibited an uneven composition of Pb and a large opaque area. Zaslavsky and et al. [7] have found similar results, increasing CsI (Tl) with a high concentration of dopant: they attributed the fact to a possible decomposition of the solid solution and lack of uniformity in the composition.

The results of lead concentration, in fourteen regions of the crystalline block of CsI:Pb 10^{-3} M, after thermal treatment are represented in Fig. 2.





Slice of the CsI:Pb crystal	Lead concentration (molar fraction)
1	$0.02x10^{-3} \pm 0.002x10^{-3}$
2	$0.73x10^{-3} \pm 0.058x10^{-3}$
3	$0.48x10^{-3} \pm 0.038x10^{-3}$
4	$0.37x10^{-3} \pm 0.030x10^{-3}$
5	$0.10x10^{-3} \pm 0.008x10^{-3}$
6	$0.30x10^{-3} \pm 0.024x10^{-3}$
7	$0.27x10^{-3} \pm 0.022x10^{-3}$
8	$0.66x10^{-3} \pm 0.053x10^{-3}$
9	$0.15x10^{-3} \pm 0.012x10^{-3}$
10	$0.18x10^{-3} \pm 0.014x10^{-3}$
11	$0.26x10^{-3} \pm 0.021x10^{-3}$
12	$0.28x10^{-3} \pm 0.022x10^{-3}$
13	$3.90x10^{-3} \pm 0.312x10^{-3}$
14	$5.42x10^{-3} \pm 0.434x10^{-3}$

Figure 2. Concentration of lead according to the height of the CsI: Pb crystal.

In the study of the Pb dopant element distribution, with a mole fraction of 10^{-3} in the CsI matrix, the results show a higher concentration at the top of the crystal, with subsequent decrease in the initial phase of growth. A relative uniformity of Pb concentration between slice 2 and slice 12 was found, as shown in Fig. 2: this is the region with the crystalline volume indicated for use as radiation detector.

A homogeneous distribution of the dopant in an appreciable region of crystals is highly desirable in materials for the construction of radiation detectors, since the optimization of the scintillation light emission efficiency is dependent on this homogeneity. But the difficulty of obtaining uniformity in the doping concentration in the growth direction is due to the fact that the distribution coefficient is, generally, different from the unit, what creates a concentration gradient.

In the process of crystal growth, when a small amount of impurities is added to the system (dopant), they are distributed in the crystalline matrix by replacing the constituents of the lattice, taking the name of substitutional impurities; or they occupy positions which are not defined in the structure, and then they known as intersticial impurities. When the contestants in a matrix of ions have different electro-negativities (Pb^{2+} = 1.8 eV) and (Cs^{+} = 0.7 eV) with ionic radii which differ by approximately 25% (Pb^{2+} = 1.29 Å) and (Cs^{+} = 1.74 Å), both with coordination number VIII, [8] this occurrence is limited to the substitutional incorporation. This explains the difficulty in growing crystals with Pb concentrations above 10^{-2} M, showing an uneven composition of Pb and a large opaque area.

The intensity of light produced and the wavelength of maximum luminous efficiency are two important properties of scintillators. The light emerging from the crystal affects the number of photoelectrons generated at the entrance of the photomultiplier tube, what, in turn, affects the pulse height produced at the output of the detector system (crystal and photo-sensor). Therefore, knowing the wavelength of maximum luminescence of the crystal is extremely important when selecting the most suitable photo-sensor assembly of radiation detectors, that is, the photo-sensor should have a maximum sensitivity region close to the center of the crystal band issuance.

The luminescence spectra, as a function of the wavelength for pure CsI crystal and CsI:Pb 10^{-2} M, 1.5 x10^{-3} M, 10^{-3} M and 5x10^{-4} M, excited by gamma radiation from a ^{60}Co source, are shown in Fig. 3.

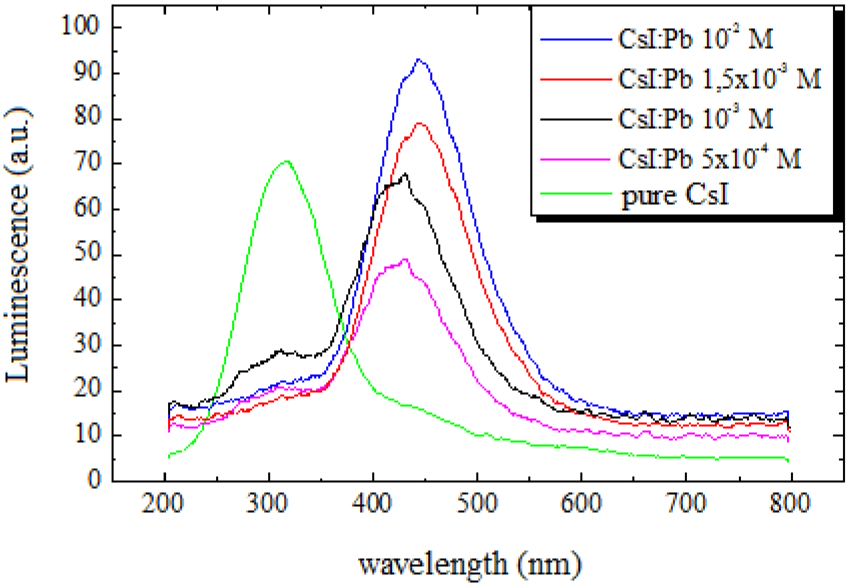


Figure 3. Luminescence spectra for the crystals of pure CsI and CsI:Pb 10^{-2} M, 1.5 x10^{-3} M, 10^{-3} M and 5x10^{-4} M, using gamma radiation from Co^{60}.

Fig. 3 shows the luminescence spectra of the CsI:Pb crystals at various concentrations, with a peak of maximum light emission at a wavelength around 450 nm. However, a decrease in the luminescence intensity of the CsI:Pb $5x10^{-4}$ M crystal was observed when compared to the pure crystal, while, in the case of the CsI:Pb 10^{-2} M crystal, the intensity of the emission peak around 450 nm was significantly higher than that of pure CsI crystal.

Each material has its own scintillation characteristics of emitted light photons and on the other hand, each photo-sensor is more efficient for certain range of wavelength of the incident photon. Therefore, it is convenient to use an appropriate mix between the characteristics of the scintillator material and its photo-sensor quantum efficiency. Babin and et al. [9] mention the interest in crystals CsI:Pb as possible materials for scintillators, since their luminescence is practically not studied. Some data reported by different authors are contradictory. This indicates that a complex system and a strong dependence of the optical characteristics of crystals may be related to the method used for the growth. According to Landor, Keszthely et al. [10], the characteristics of scintillation crystals are also influenced by the growth method used.

As the structure of the lattice is influenced no only by the conditions of the crystal growth and subsequent thermal treatment but also by the presence of impurities, some emission bands are more intense or better separated just in the doped crystals.

In Fig. 4 and 5, Laue spectra achieved for pure CsI and CsI:Pb crystals are shown, indicating that the materials obtained correspond to single structures.

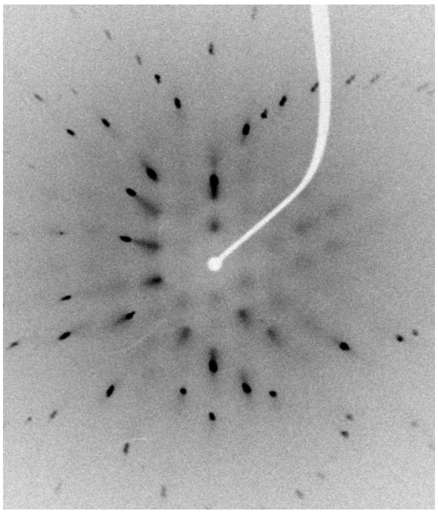
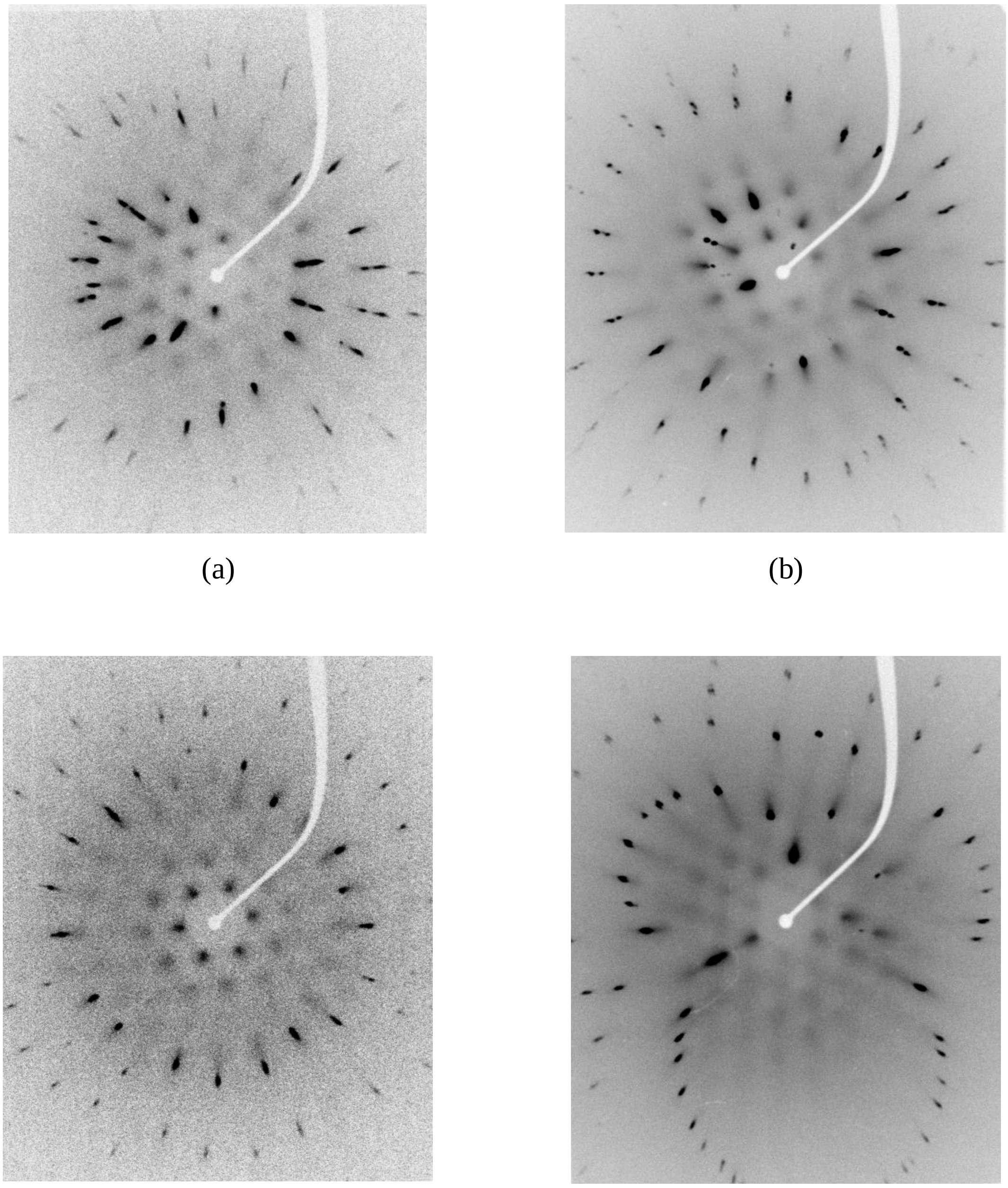


Figure 4. Lauegrama obtained for the pure CsI crystal.



(a) (b)

(c) (d)

Figure 5. Lauegramas obtained for the CsI:Pb crystals. (a) $5\text{x}10^{-4}$ M, (b) 10^{-3} M, (c) $1.5\text{x}10^{-3}$ M, (d) 10^{-2} M.

The X-ray difraction is a useful tool in the study of materials. In this paper we used the Laue technique. Through this technique it is possible to make a qualitative analysis of the degree of crystallinity of the crystal obtained by observing the diffraction spots (presence or absence of deformation) in the Laue figure of the crystal photographed.[11]

As for the system ability to discriminate energy, detector experiments were conducted using alpha radiation with energy of 5.54 MeV to evaluate the parameter resolution, defined as the ratio between the total width at half maximum (FWHM) and the location of the photopeak..

In Fig. 6, the results of alpha spectroscopy for radiation of ^{241}Am (5.54 MeV), obtained with the CsI:Pb scintillator and pure CsI crystals, are present.

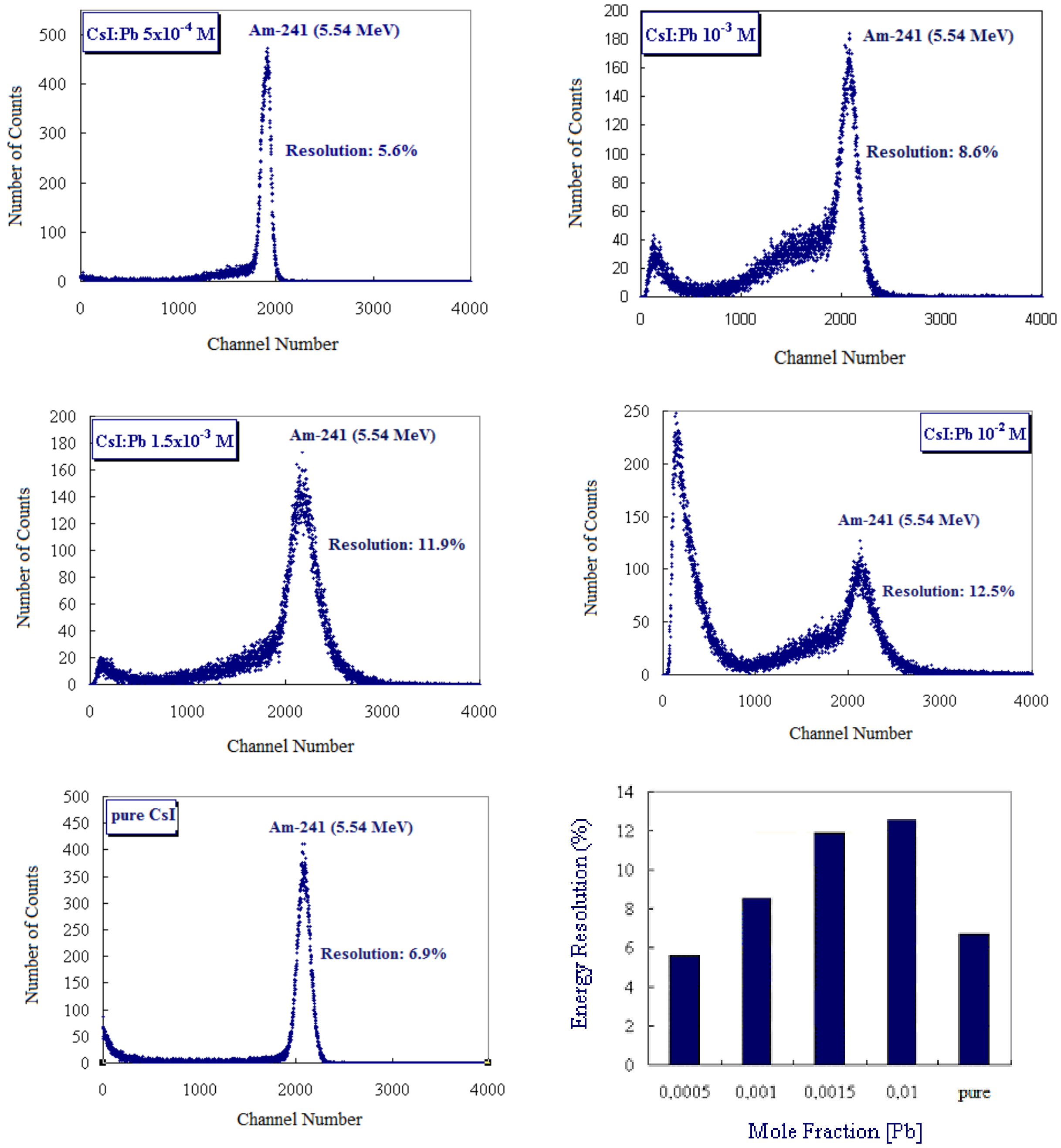


Figure 6. Energy spectra of the scintillation light from ^{241}Am 5.54 MeV alpha particles in CsI:Pb and pure CsI crystals.

The sensitivity of a detection system depends on the efficiency with which light generated in the scintillator is transmitted by internal reflection to the photo-sensor. In this sense, the optical coupling between the crystal and the photo-sensor was made with silicone grease 0.5 McStockes, matching the indices of the crystal refraction used and the surface of the window photo-sensor. The photons of light that reach the crystal interact with the photo-sensor: in this study, a photomultiplier tube was used.

IV. CONCLUSIONS

Undoped and Pb^{2+} doped CsI crystals were grown by the Bridgman technique. The lead element dopant was suitable to be incorporated in the CsI matrix.

The result of the analysis by flame atomic absorption in crystals of CsI:Pb, at various concentrations, showed the incorporation of the dopant element lead in the CsI matrix. The analysis of the lead dopant concentration in fourteen slices of crystal, with 10^{-3} mole fraction, revealed that 70% of the crystal had a relatively homogenous concentration of lead, which is a crystalline volume fraction suitable for use as scintillator.

The luminescence spectra, with peaks of maximum emission around 450 nm show good overlapping with the spectral quantum efficiency of bialkali photomultipliers, demonstrating the feasibility of using crystals of CsI:Pb as radiation detectors.

Laue figures showed characteristic points, indicating that the materials obtained, CsI:Pb and pure CsI, correspond to single crystal structures of good quality.

The CsI:Pb crystals presented response to the radiation of alpha particles, showing spectra with defined peaks: in the concentration range of doping studied, the crystal with mole fraction $5x10^{-4}$, had the best resolution.

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