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SCINTILLATION RESPONSE OF CsI:Tl CRYSTAL UNDER NEUTRON, GAMMA, ALPHA PARTICLES AND BETA EXCITATIONS

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ABSTRACT

Among the converters of X and gamma radiation in light photons, known as scintillators, the one which is the most efficient emits photons with a wavelength near 400 nm. Particularly, among them, the cesium iodine doped with thallium (CsI:Tl) crystal is that which matches better between the light emission spectrum (peak at 540 nm) and the quantum sensitivity curve of the photodiodes and CCD (Charge Coupled Device). This explains the renewed interest in using this crystal as scintillator.

Although the CsI:Tl crystal is commercially available, its local development would give the possibility to obtain it in different geometric configurations and coupling. Moreover, there is a special interest in studying new conditions that will alter the properties of this crystal in order to achieve a optimal level of its functional characteristics. Having an efficient national scintillator with low cost is a strategic opportunity to study the response of a detector applied to different types of radiation. The crystal of cesium iodide activated with thallium (CsI:Tl) has a high gamma detection efficiency per unit volume. In this paper, the CsI:Tl crystal, grown by the vertical Bridgman technique in evacuated silica ampoules and with the purpose of use as radiation detectors, is described. To evaluate the scintillator, measures of the thallium distribution in the crystal volume were taken, with overall efficiency score. The scintillator response was studied through gamma radiation from sources of ^{137}Cs, ^{60}Co, ^{22}Na, ^{54}Mn, ^{131}I and ^{99m}Tc; the beta radiation from source of ^{90}Sr/ ^{90}Y, alpha particles from ^{241}Am source and the scintillator response to neutrons from Am/Be source. The energetic resolution for ^{137}Cs gamma rays (662 keV) was 10%. The results showed the validity of using the CsI:Tl crystal developed in our laboratory, in many applications in the area of radiation detectors.

Keywords: scintillator, crystal growth, radiation detector

1. INTRODUCTION

The need to detect radiation after the discovery of X-rays and radioactivity has arisen and it is a challenge that has extended to the XXI century. The detection of radiation and the measure of its properties are required in all facets of nuclear technology, namely, in scientific studies, operation of reactors for power generation, radiation protection, industry and the medical field. The diversity of the physical interaction between radiation and matter is such that it can be said that no detector may be adequate to measure all types of radiation, even the generic application of only one type of radiation: each detector, therefore, has limited applicability to special cases.

The type of detector used depends on several factors; the particles to be observed, the energy of the particle and the environment in which the detector is to be used. So obviously the need to develop processes and tools capable of making apparent the presence of particles and their properties. The development of radiation detectors using scintillation crystals which increase the efficiency, speed of response, accuracy in dose and energy and at the same time allow to simplify and reduce costs in the production process are constant focus of scientific research.

The scintillators are materials capable of producing light when ionizing radiation dissipates its energy in their midst. Due to the existence of different types of scintillators they were classified into three groups according to their physicochemical characteristics, namely: inorganic, organic and gas scintillators. Among the inorganic crystals, such as scintillators most commonly used consist of alkali metals, in particular alkali halide.[1,2,3,4] The inorganic scintillators have been studied for use as radiation sensors from the 50s. [3] Since then, various inorganic materials have been studied in various fields of science and engineering. Currently, among the crystals based on cesium iodide (CsI) crystal doped with thallium is the most common, being one of the more high performance scintillators for detecting gamma rays per unit volume.

A considerable number of papers have been published on the basic physical behavior of scintillation in CsI:Tl. Some work on this theme have been summarised by Kaufman et al. [5], Dietrich and Murray [6], Heath et al. [7] while general features of scintillation processes in inorganic crystals have been described by Birks in 1964 [1]. In recent years, in heavy ion nuclear physics and in high energy astrophysics, attention has been paid to the CsI:Tl scintillator for its pulse shape discrimination [8]. In order to apply CsI:Tl crystals to these fields, it is necessary to investigate the scintillator response through gamma radiation, beta radiation, alpha particles and neutrons.

Thallium doped cesium iodide CsI:Tl, a scintillator crystal, is a high potential detector material, due to its wide applications in photocathode, gamma rays detectors and particle detectors in nuclear experiments. CsI:Tl is considered one of brightest inorganic scintillators whose high wavelength of 540 nm light emission makes it best coupled to red-enhanced photomultiplier tubes (PMT), photodiodes or charge coupled devices (CCDs). It has good scintillation efficiency and is less hygroscopic and less brittle than NaI. Its high light output, good energy resolution and large absorption coefficient to high energy particles make CsI:Tl crystal a popular material for ionization radiation detection. [9]

Although the CsI:Tl crystal is, also, commercially available, its local development is convenient giving the possibility to obtain it in different geometric configurations, coupling it and studying the response of the crystal when excited with different ionization densities. Moreover, there is a special interest in studying new conditions that will alter the properties of this crystal in order to achieve a optimal level of its functional characteristics. The relative simplicity of obtaining, installing and their feasible cost make these crystals attractive for applications in various areas of radiation. The price of raw materials and the method of producing and fabricating the material into the desired size and shape all come into the final cost. These and other factors differ in their relative importance in selecting better materials for a particular application.

The aim of this work was the growth of thallium doped CsI single crystals, in evacuated crucibles by vertical Bridgman technique and scintillation response for a variety of radioactive source including beta, alpha, gamma and neutrons.

2. MATERIALS AND METHODS

Single crystals of cesium iodine doped with thallium (CsI:Tl) with nominal concentration of 10^{-3} M, were grown by two zone vertical Bridgman technique using stoichiometric amounts of thallium iodine (TlI). These compounds were obtained from Metal Gesellschaft K.K., with a purity of 99.99 %. This method is based on the work of Bridgman in 1925 [10,11,12]. This is a technique to grow single crystals of various halides. In this method, temperature gradient move slowly, relative to a crucible until the melt in the crucible solidifies. A crucible containing the material is kept in the upper zone (hot zone) for complete melting; then it is slowly lowered (1 mm/h) to the lower zone (cold zone) through optimized temperature gradient until crystalline solidification completes.

In order to evaluate the homogeneity of the thallium (Tl) concentration in CsI crystals, the analytical technique of flame atomic absorption was used. The crystal was cut into nine slices; each slice with a thickness of 10 mm. The Fig. 1 shows scheme of the CsI:Tl crystal. Fractions (70mg) of the sliced samples were separately placed in beakers and dissolved in HNO_3 solution (2:3). The solutions were heated to 40 °C with stirring, until they were transparent. The solution to the "blank" was prepared with CsI in HNO_3 salt (2:3), at the same concentration of the samples. This solution was, also, used to prepare the standard curve.

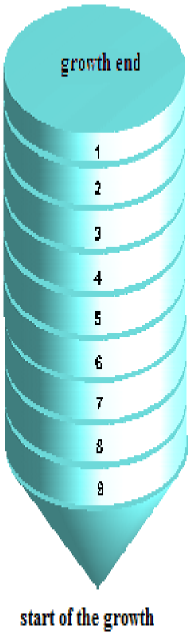

Figure 1. Scheme of the CsI:Tl crystal.

Before starting the measurements of neutrons, beta, gamma and alpha particles with CsI:Tl crystals, the spectrum of the laboratory background radiation was obtained. This measurement was carried out to evaluate the strength and influence of possible natural radioactive sources in the environment measurement.

In the study of the response to neutron radiation, gamma radiation, beta radiation and alpha particles the crystals were directly coupled to the photomultiplier tube (RCA MODEL 8575, 21 pins) using silicone grease (Dow Corning) viscosity of 0.5 McStokes, as optical interface. This ensured uniform refractive index across the contact surface between the crystal and photomultiplier tube. Sides of the crystal, which were not in contact with the photo-sensor, were covered with several layers of polytetrafluoroethylene (PTFE) tape to ensure good reflection of light. The electronic modules used for processing of signals from the photomultiplier tube were conventional Canberra electronic.

Since energy resolution could be determined only by using monochromatic radiations, these characteristics were investigated just for alpha and gamma rays source. Energy resolution of the detector-photosensor system, expressed in percentage, was determined by the ratio between the total width at half maximum (FWHM) and its energy photopeak.

The energy resolution of the CsI:Tl crystal detector system was determined using gamma radiation of the ^{137}Cs (662 keV), ^{60}Co (1173 keV, 1333 keV), ^{22}Na (511 keV, 1275 keV), ^{54}Mn (835 keV), ^{131}I (364 keV) and ^{99m}Tc (140 keV) sources. The operation of the photomultiplier voltage was 1300 V, and the accumulation time in the counting process was 300 s. The scintillator crystal used was cut with dimensions of 20 mm diameter and 20 mm height.

The energy resolution of the CsI:Tl crystal detector using alpha particle of the ^{241}Am (5.486 MeV) source was determined. The operating voltage of the photomultiplier tube was 880 V; the accumulation time in the counting process was 1800 s. The scintillator crystal used with dimensions of 20 mm diameter and 10 mm height.

The scintillator's ability to detect neutrons, was verified with the AmBe source positioned at a distance of de 100 mm from detector, using 70 mm of paraffin as the interface to neutron thermalization. A foil of cadmium (Cd) was placed around the detector and the photomultiplier to avoid the scattered neutrons contribution, as shown in Fig. 2. In response to neutron radiation, an AmBe source with energy range of 1 MeV to 12 MeV was used. The activity of the AmBe source was 1Ci Am. The fluency was 2.6 x 10^6 neutrons/second. The operating voltage of the photomultiplier tube was 800 V; the accumulation time in the counting process was 3600 s. The scintillator crystals used were cut with dimensions of 20 mm diameter and 10 mm height.

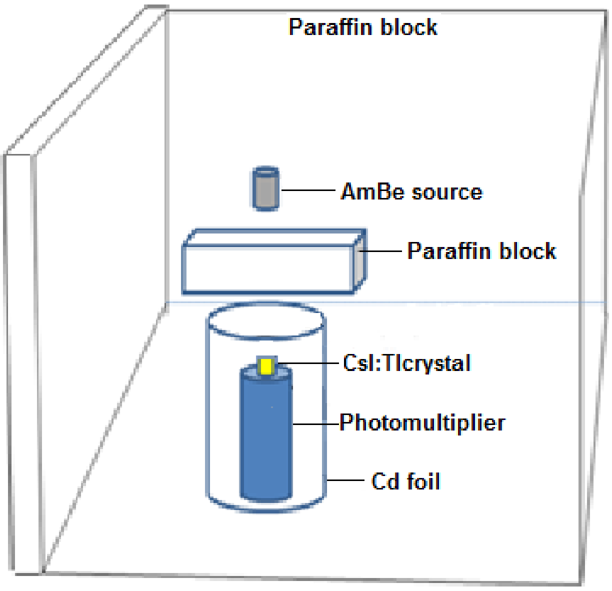

Figure 2: Schematic representation of the array used for measurements.

In response to beta radiation, a ^{90}Sr/ ^{90}Y source was used. The operating voltage of the photomultiplier tube was 880 V; the accumulation time in the counting process was 3600 s. The scintillator crystal used had dimensions of 20 mm diameter and 10 mm height.

3. RESULTS AND DISCUSSION

The results of the thallium concentration in nine regions of the block crystalline CsI:Tl, after the heat treatment are shown in Table 1 and plotted in Fig.3

Table 1: Determination of thallium concentration in slices of 10 mm thick of the CsI:Tl crystal, made by flame atomic absorption.

Slices of the CsI:Tl crystal	thallium concentration (molar fraction)
1	$3.57x10^{-3}$
2	$3.47x10^{-3}$
3	$1.04x10^{-3}$
4	$0.50x10^{-3}$
5	$0.39x10^{-3}$
6	$0.56x10^{-3}$
7	$0.36x10^{-3}$
8	$0.32x10^{-3}$
9	$0.47x10^{-3}$

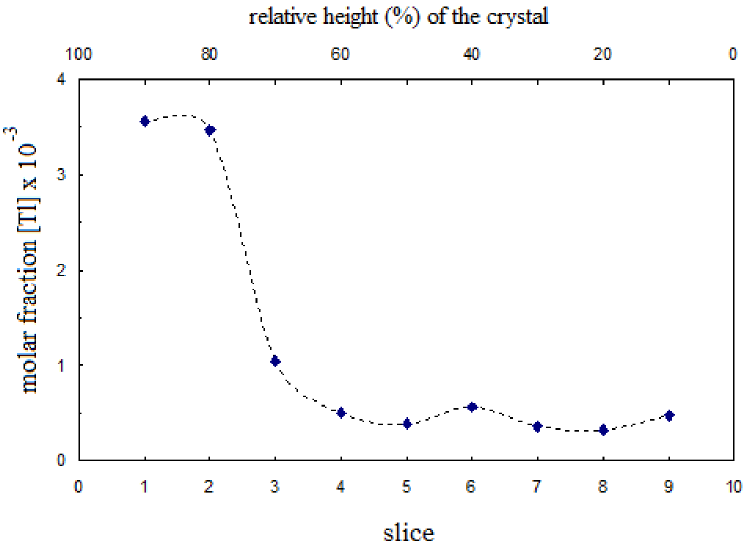

Figure 3: Concentration of thallium according to the height of the CsI:Tl crystal.

A homogeneous distribution of dopants in a significant area of the crystals is highly desirable for materials in the construction of radiation detectors, since the optimization of the scintillation light emission efficiency is dependent on this homogeneity. However the difficulty of obtaining uniformity in the concentration of dopants in the growth direction is due to the fact that, generally, the distribution coefficient is different from the unit, which gives a concentration gradient.

In the study of thallium dopant, distribution along the crystal volume was added to the starting material (CsI salt), with the mole fraction 10^{-3}. During the crystallization process, the concentration is changed, causing a concentration gradient. The results of flame atomic absorption indicate a higher concentration at the top of the crystal, with subsequent decrease in the initial growth phase. A good uniformity of the concentration of Tl from the slice 4 and slice 9 was found, as it can be seen in Fig. 3 and Table 1, which is the region of the crystalline volume indicated for use as radiation detector. From the slice 4, the thallium concentration increases abruptly and, therefore, this region will produce crystals exhibiting homogeneity of the dopant. The high concentration of thallium in the top of the crystal is expected taking into account the fact that the crystal grow upwards and impurities tend to be deposited at the end of the growth process. The control in the distribution of dopants throughout the crystals is related to the segregation phenomenon that occurs during the growth of crystals.

The quality of a converter in scintillation gamma may be assessed by two parameters, the light conversion efficiency of the process and energy resolution. The system ability to discriminate energy detector experiments was conducted using a photomultiplier. In this work, it was assessed the parameter resolution settings evaluated by the ratio of the width at half maximum and their energy photopeak.

The energy spectra shown in Fig. 4 illustrate the results of gamma spectrometry to radiation of ^{22}Na, ^{54}Mn, ^{137}Cs, ^{60}Co, ^{99m}Tc and ^{131}I, obtained from the crystal system of CsI:Tl crystal coupled photomultiplier.

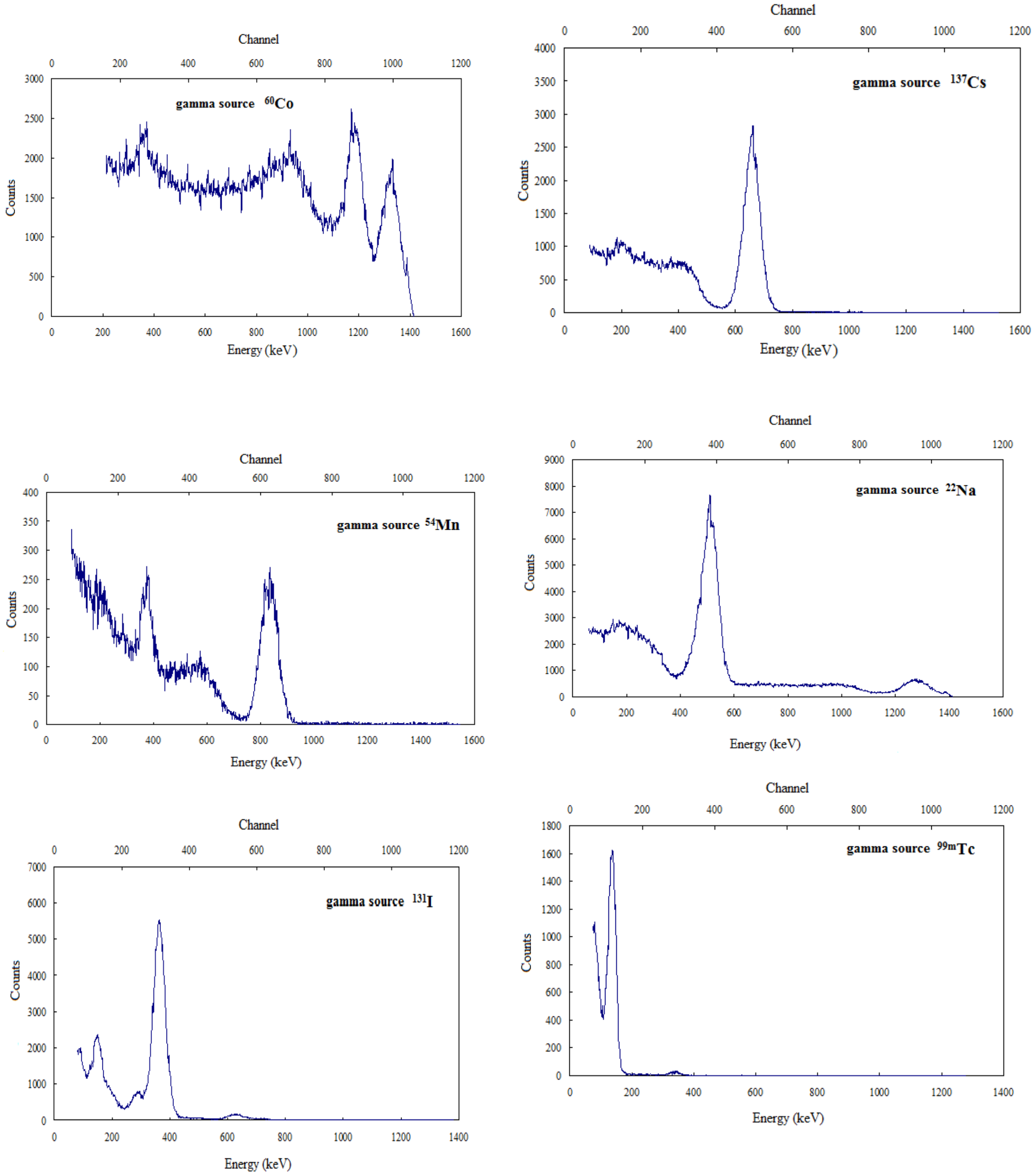

Figure 4: Energy spectra obtained for the radiation from ^{60}Co, ^{137}Cs, ^{54}Mn, ^{22}Na, ^{131}I, ^{99m}Tc with CsI:Tl crystal coupled to a photomultiplier.

The energy resolution percentage of the detector CsI:Tl coupled to the photomultiplier, on the basis of gamma radiation of energy for the six radioactive sources used are shown in Table 2 and plotted in Fig. 5.

Table 2: Percentage energy resolution due to the gamma radiation energy for the crystal CsI (Tl) with dimensions of 20 mm in height and 20 mm in diameter

Radioactive source	Energy level (keV)	Energy Resolution (%) (FWHM)
^{99m}Tc	140	15.5
^{131}I	364	11.9
^{22}Na	511	12.5
^{137}Cs	662	10.0
^{54}Mn	835	8.8
^{60}Co	1173	5.6
^{22}Na	1275	7.9
^{60}Co	1333	5.9

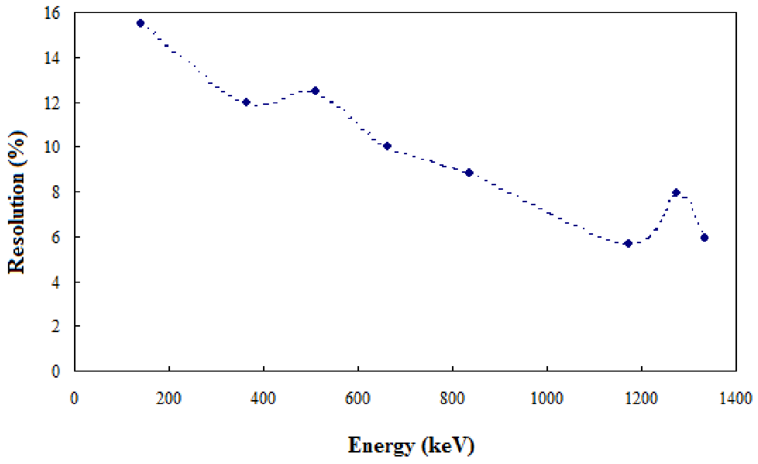

Figure 5: Percentage energy resolution with CsI:Tl crystal coupled to the photomultiplier, according to the energy of gamma radiation.

The CsI:Tl crystal proved to be a good converter of high-energy photons from the gamma rays into light photons able to sensitize the photosensor. To meet this goal the CsI:Tl crystal provides suitable conditions, mainly consisting of chemical elements of high atomic number (Z cesium = 133, Z iodine = 127); thus, favoring the interaction of gamma radiation with multiple electron shell (photoelectric effect and Compton) and, also, having a favorable interaction core structure of the type pair production.[3]

The scintillation response of CsI:Tl crystal to alpha particle with energy of the 5.54 MeV shows resolution of 5.18 %, whose value is in agreement with findings in the literature. [8,13] The Fig. 6 shows alpha spectroscopy results from radiation of ^{241}Am (5.54 MeV) obtained with CsI:Tl crystal.

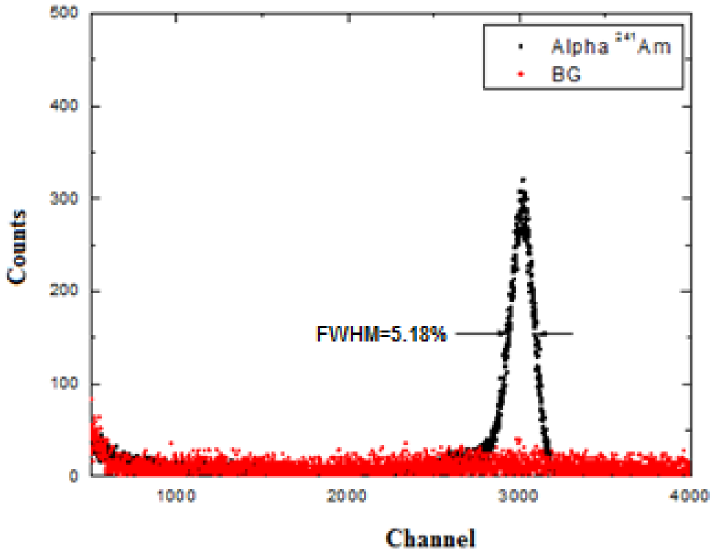

Figure 6: **Energy spectrum of the scintillation light from ^{241}Am (5.54MeV) alpha particles in CsI:Tl crystal coupled to a photomultiplier.**

The Fig.7 show energy spectrum results from radiation of ^{90}Sr/^{90}Y beta radiation obtained with CsI:Tl crystal.

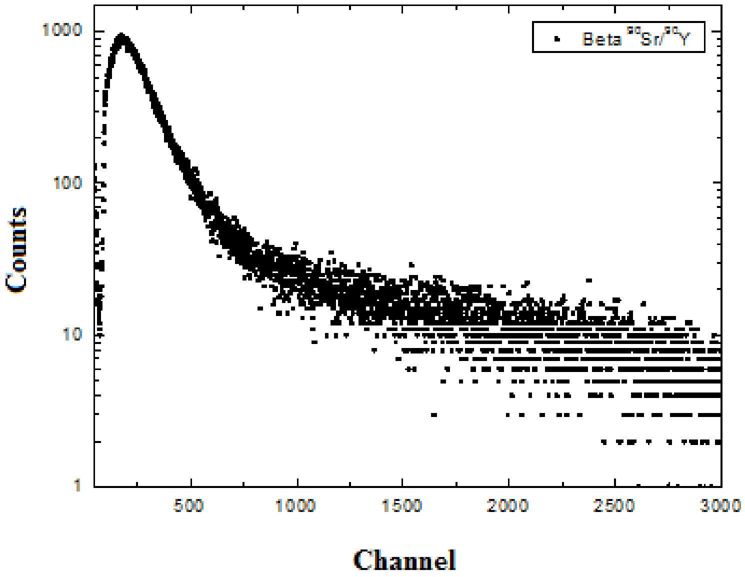

Figure 7: **Energy spectrum of the scintillation light from ^{90}Sr/^{90}Y beta radiation in CsI:Tl crystal.**

The Fig.8 shows the response of the CsI:Tl crystal to the neutrons from a AmBe source.
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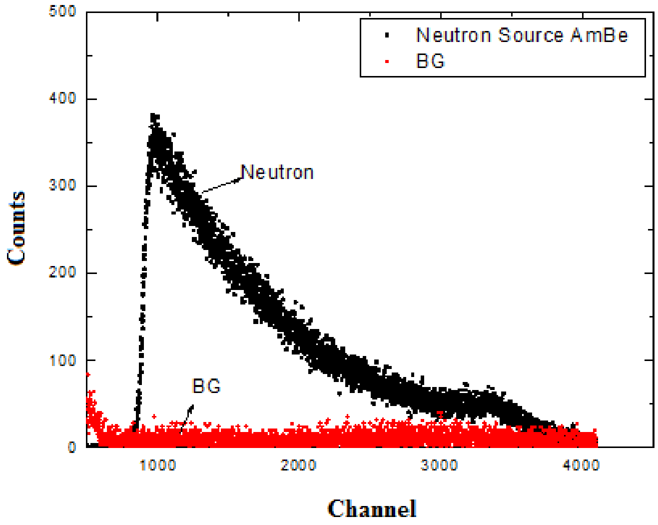

Figure 8: Energy spectrum of the scintillation light from AmBe neutron radiation in CsI:Tl crystal.

This result can be due prompt gamma or beta radiation. The publication of the IAEA: "*Database of prompt gamma rays from slow neutron capture for elemental analysis*", presents in Table 7.2. "Summary of data for radioactive isotopes produced by thermal neutron activation", where the produced mode of ^{128}I can be by β^- or EC and the ^{134}Cs by IT. [14] Table 7.3 "Adopted prompt and decay gamma rays from thermal neutron capture for all elements", show a large range of prompt gamma for the ^{127}I and ^{134}Cs. Evidently, much of the gamma energy cannot be detect due to the thickness of the detector, and the detector does not have resolution to show all gamma energies.

4. CONCLUSIONS

The study showed that the CsI:Tl crystal grown by the Bridgman technique exhibit qualities suitable for use as radiation sensor. Obviously, further work will have to be carried out on these materials, in particular on the concentration of dopants and on the crystal growth technique parameters.

The analysis of the thallium concentration of the dopant in nine slices of crystal, obtained by the Bridgman technique, showed that 60% of the crystal has a homogeneous region in thallium concentration, which is the fraction of the crystal volume more suitable for use as a scintillator.

Crystals of the CsI:Tl with nominal concentrations of 10^{-3} M showed good energy resolution when excited by gamma and alpha particle radiation. The crystals showed, also, sensitivity under neutron and beta excitations.

The results demonstrate the feasibility of using CsI:Tl crystal developed in this work as a radiation detector in nuclear instrumentation area, such as in portable radiation monitors, instrumentation for nuclear medical and equipment for teaching and research.

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