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INORGANIC SCINTILLATORS OF Li^+ DOPED CsI FOR NEUTRON DETECTION

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ABSTRACT

The scintillation method is still one of the main methods used for the detection of ionizing radiations. The universality of this method is considered to be its major advantage. It may be used for registration of almost all types of radiation in a wide range of energy (1 eV – 10 GeV). In recent years has been an increasing demand for new types of neutron detectors to be used in applications such as spectrometers, photodetectors, materials characterization, and so on. For these applications, inorganic scintillators play an important role in the detection and spectroscopy of neutrons. Scintillation crystals based on cesium iodide (CsI) have relatively low hygroscope, easy handling and low cost, features that favor their use as radiation detectors. In this work, lithium doped CsI crystals were grown using the vertical Bridgman technique. In this technique, the charge is maintained at high temperature for 10 h for the material melting and complete reaction. The temperature gradient 21° C/cm and 1 mm/h descending velocity are chosen as technique parameters. The concentration of the lithium doping element (Li) studied was 10^{-3} M. The optical transmittance measurements were made in the CsI:Li crystal at room temperature, using a spectrophotometer with a wavelength in the range 200 nm to 900 nm. Analyses were carried out to evaluate the scintillators developed, concerning neutron radiation from AmBe source, with the energy range of 1 MeV to 12 MeV. In this paper was investigated, the feasibility of the CsI:Li crystal as a neutron detector to be used for monitoring, due to the fact that in our work environment there are two nuclear research reactors and calibration systems.

1. INTRODUCTION

The radiation detectors are essential in all fields of nuclear energy. Devices that are placed in an environment where there is a radiation field, are able to indicate their presence. Radiation detectors consist of a radiation-sensitive element and a system that makes those effects on a value related to a measured quantity of such radiation. Ionizing radiation covers broad spectrum of energy and different types of interactions with matter. As a result each detector has its field of use defined by the type of radiation, energy range and characteristics of their physical response.

The detection of neutrons is not trivial due to lack of charge of these particles and the peculiarity of their interactions with matter. Neutron sources also generate gamma radiation which can interfere with its measurement. It is necessary that the detector system is able to discriminate these interferences. The main types of neutron detectors are sensitive to: (a) gases, (b) self-powered (self power), (c) scintillators and (d) semiconductors. [1]

Among the types of detectors, scintillators meet the diverse needs in the field of radiation detection.

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 depending on their physicochemical characteristics, namely inorganic scintillators, organic scintillators and gases scintillators. Among the inorganic crystals, the most used as scintillators are made of alkali metals, in particular alkali halide [2,3]. The inorganic crystals have been studied for use as radiation sensors, since the 50s [3]. Since then, various inorganic materials have been studied in various fields of science and engineering. The research of new scintillator materials has become increasingly in recent years, driven by the needs of development of high-energy physics, nuclear tomography and other fields of science and engineering. A better understanding of the various mechanisms scintillation has enabled the use of new materials for detecting various types of radiation. Although many of the foundations of physics, essential to the process of scintillation, have been studied, the need to improve these materials has been important in the research and development of materials scintillators.

Neutron detection with spatial, temporal and energy resolution is important to the improvement of high energy physics, neutron forensic, non-proliferation of special nuclear materials, the search for dark matter, inelastic neutron scattering and it is fundamental to the advancement of nuclear medicine.

In this work, the feasibility of the CsI:Li crystals, as neutron detectors, was investigated. The pure CsI was used for comparison.

2. MATERIALS AND METHODS

Lithium doped CsI crystals were grown using the vertical Bridgman technique. In the process of growing crystals by the vertical Bridgman technique, a number of parameters involved, including the growth velocity, the temperature profile of the furnace, cleaning materials, and the geometric configuration of the material from which the crucible is made are the major. We used a furnace for crystal growth starting materials which have a melting point of below 1000 ° C. The system is open and has been designed to operate with sealed ampoules inside. The starting material used with purity of 99.99%. The concentration of the lithium doping element (Li) was 10^{-3} M

In order to obtain crystals of good quality (thermal and mechanical stability) are needed. Mechanical or thermal fluctuations can produce localized over-colds, leading to irregular growth of the crystal. Mechanical stresses due to thermal expansion and contraction can be reduced by selecting a suitable crucible. Quartz was chosen as the material to be used in the manufacture of crucibles in the process of crystal growth, in this respect the following factors: (a) be thermal and mechanically stable up to at least 100 ° C above the melting point of the material to be crystallized, (b) being chemically inert to the molten material and not influence the properties of the crystal, (c) be resistant to the atmosphere in which is held the crystal growth, (d) be resistant to temperature changes while having low thermal conductivity in high temperature gradients.

The crucibles were subjected to a constriction at its top center, where smaller cylinders were placed quartz diameter slightly smaller than the inner diameter of the crucible, which were retained up to the constriction in order to facilitate the sealing of the same solder oxy acetylene. The other end of the crucibles was molded into a conical shape, so that the initially formed core in this region can serve as a guide for the crystal growth.

The salt of cesium iodide (CsI) and lithium iodide (LiI) were subjected to a dehumidification process under continuous vacuum of 10^{-6} mbar and a temperature of 200° C for 3 h in order to remove residual water, gas, atmospheric and volatile impurities. The dehumidification process involved the following steps: slow heating of the system under continuous vacuum to the temperature of 100° C (temperature range in which no hydrolysis occurs), kept at this temperature for 1 h. From this temperature, still under vacuum, maintained by heating the system with a heating rate of 25° C to 200º C, kept at this temperature for 3 h to remove water absorbed chemically.

Growth of crystals by the Bridgman technique, we used the two-zone oven (hot and cold). The quartz tube containing the dopant salt and CsI:Li was placed in the hot zone of the furnace, and the melt temperature of 650°C. After complete melting of the salt was necessary to stabilize the temperature, and the melt stayed in this condition for 10 hours to ensure the homogenization of the load. Only then started displacement of the crucible, which was made towards the cold zone of the furnace at a speed of 1 mm h^{-1} by means of a DC motor. During this procedure was avoided any vibration or motion of the solid-liquid interface, as this could cause changes in the orientation of the crystal. The temperature was strictly controlled using a microprocessor-based controller.. Finally, after 120 hours, the crucible is busy with the single crystal.

The grown crystals were subjected to heat treatment. In this procedure, vacuum of 10^{-6} mbar and continuous temperature of 350º C, for 24 hours, were employed. The grown crystals were polished with ethylene glycol pA ($C_2H_6O_2$),The polishing of slightly hygroscopic crystals differs from metal polishing or glazing on two aspects: the material is low hardness and can deteriorate in the presence of moisture. The softness of the crystal means that polishing is fast and in a short period of time, a considerable layer of material can be removed. The side surfaces are left not polished to enhance internal reflection.

Transparency is a critical factor for scintillators, since the emitted photon with energy in the visible region, needs to be transported efficiently to the photosensor. The transparency can be evaluated by a direct measurement of light transmission at a wavelength of flicker. Accordingly transmittance tests were performed on samples of CsI: Li crystals with concentrations 10^{-3} M using a UV-visible spectrophotometer (Shimadzu UV-1601 PC). The measurement system is basically a light source, a monochromator and a detector. Between these two elements, the sample is positioned. The magnitude of this study was to measure absorbance (A) which is related to the transmittance (T) through the expression:

$$A = \log(1/T) \quad \Rightarrow \quad T = 10^{(-A)} \tag{1}$$

The spectral coverage was 190 nm to 1100 nm, and the optical path length was 1 cm. The same measurements were performed for the pure CsI crystal for comparison purposes.

In the study of the response to neutron radiation, 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 was not in contact with the photo-sensor, were covered with several layers of Teflon tape to ensure good reflection of light. The electronic modules used for the processing of signals from the photomultiplier tube were: Photomultiplier (RCA 8576), Multichannel analyzer (ADCAM Ortec 918), High voltage (Ortec 556), Pre amplifier (Ortec 276), Amplifier (Ortec 450), Oscilloscope (Phillips PM3295A 400 MHz).The detection efficiency of the scintillator crystal was measured in two different positions: in the first (position I), the AmBe source was positioned at a distance of de 5 cm from the photomultiplier tube. In the second (position II), the AmBe source was positioned at a distance of 5 cm from the photomultiplier tube, using paraffin as the interface.

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. Fluency was 2.6×10^6 neutrons/second. The operating voltage of the photomultiplier tube was 1900 V; the accumulation time in the counting process was 1800 s. The scintillator crystals used were cut with dimensions of 2 cm diameter and 2 cm high.

4.RESULTS AND DICUSSION

In the Fig. 1 show CsI:Li crystals grown by vertical Bridgman technique

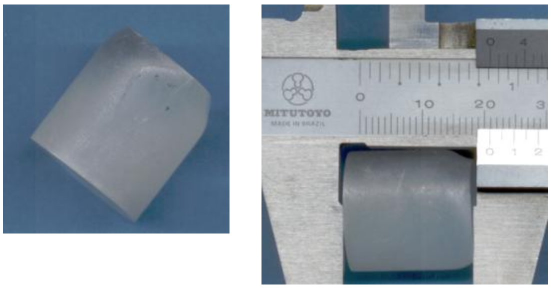

Figure 1: CsI:Li crystals grown by vertical Bridgman technique.

In the Fig.2 show the transmittance curve of the pure CsI and CsI:Li crystals.

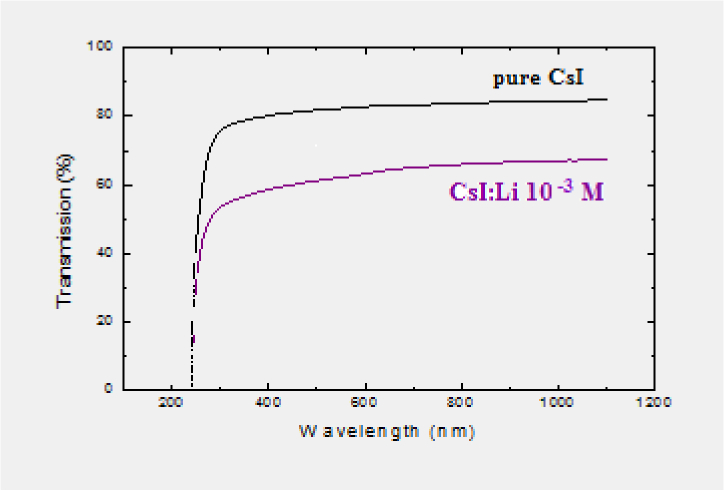

Figure 2: Curves of transmittance as a function of wavelength for the pure CsI crystals and CsI:Li 10^{-3}M with a thickness of 1 cm

To quantify the degree of transparency of crystal scintillators produced, tests were performed optical transmittance. In the samples of the crystals, the transmittance remarkably decreased according to the decrease of the wavelength. The optical transmittance of pure CsI crystal was 85% for a wavelength of 1100 nm, dropping to virtually zero at wavelengths below 240 nm. The CsI: Li 10^{-3} M crystal showed similar behavior pure CsI , approximately 65% for a wavelength of 1100 nm and nearly zero for wavelengths below 240 nm. Higher transmittance was observed for wavelengths above 400 nm, which is the area of sensitivity of the quantum efficiency of the photomultiplier.

Crystals of CsI: Li with nominal concentration of 10^{-3}M when excited by neutron radiation from an AmBe source absorbs neutrons, resulting in ^{3}H and alpha particles; n + ^{6}Li (7,5%) → ^{3}H(2.75 MeV) + α ,as shown in Fig. 3. One major attractive feature of Li is its very low cross section for gamma interactions; however, with a natural abundance of 7.5% for ^{6}Li. [4,5] The neutron line will be in an order of magnitude more intense if ^{6}Li enriched material is used.

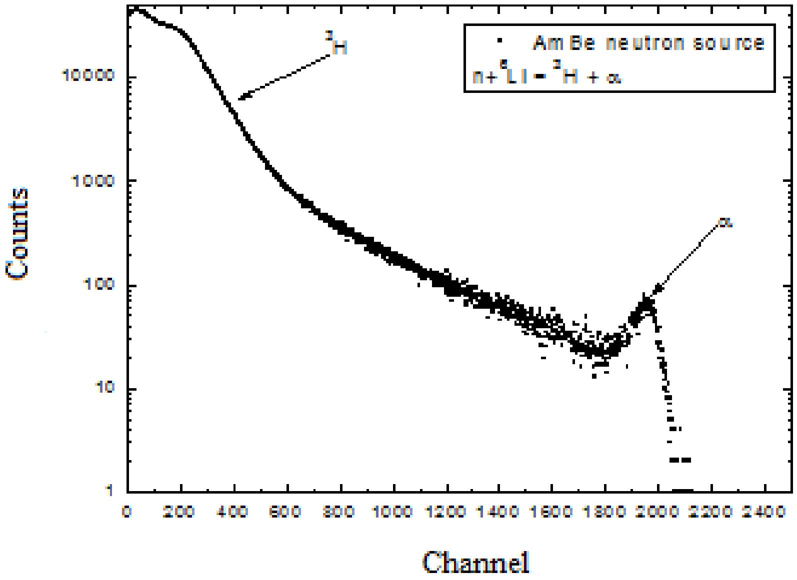

Figure 3: Pulse height spectrum of the CsI:Li inorganic scintillator from the AmBe neutron source.

For neutrons become thermalized is necessary that they undergo collisions when crossing the field, losing part of their energy until they reach thermal range. For this to occur moderators materials are used, which has the function of reducing your velocidade. Materiais hydrogenated, eg paraffin and polyethylene, are rich in hydrogen and are considered good moderators.

The curve shown in Fig. 4 illustrates the results for the neutron radiation from an AmBe source using the CsI:Li scintillator crystal. Paraffin was used for the thermalized of fast neutrons. It can be observed that the CsI: Li crystal shows good discrimination for gamma radiation and neutrons. It may, therefore, be used to detect neutrons in environments with the presence of gamma radiation.[6,7,8]

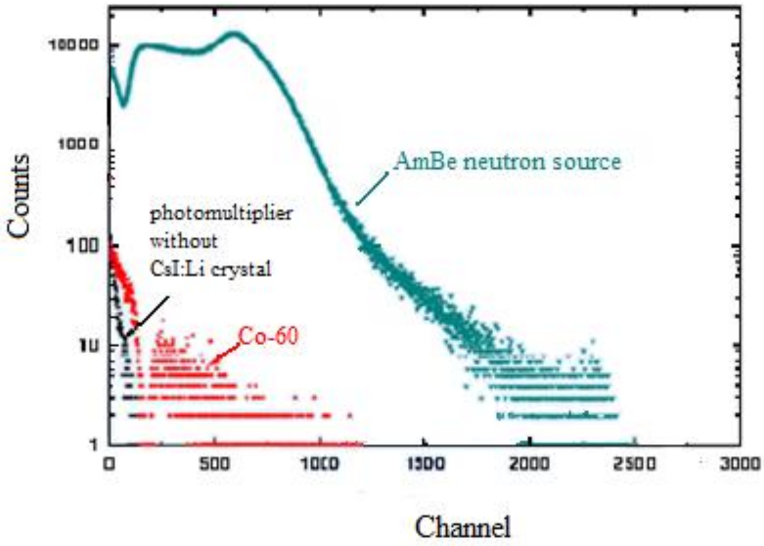

Figure 4: Pulse height spectra of the CsI:Li inorganic scintillator, from radiation of ^{60}Co and AmBe sources.

In Fig. 5, the radiation of the neutron spectrum using CsI:Li and pure CsI crystals, under the excitation of an AmBe neutron source, is shown

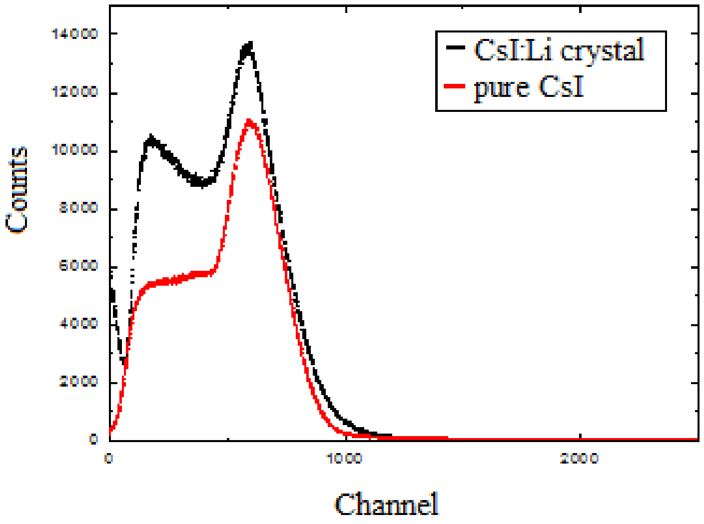

Fig. 5 – Pulse height spectrum of the CsI:Li inorganic scintillator and the pure CsI.

The largest number of counts obtained with the crystal CsI: Li, when excited with radiation from a neutron AmBe source, compared to the number of counts obtained with pure CsI crystal, demonstrates the incorporation of lithium in the crystal structure.

5. CONCLUSIONS

The addition of the Li and Br to the CsI matrix resulted in crystals with promising results, when excited with neutron radiation. The crystals showed sensitive neutron radiation.

The grown crystals showed appropriate optical qualities. The CsI: Li crystal at a concentration of 10^{-3}M exhibited a significant decrease in transmittance compared to the value of pure crystal

Obviously, further work will have to be carried out on these materials, in particular on the concentration of dopant and crystal growth technique parameters.

The crystal doped with lithium (CsI:Li) showed a better efficiency since neutrons have a high cross-section for the reaction (n, α). Even though the crystal is small, the products of this reaction (n + 6Li →3H + α) are detected in this crystalline volume.

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