Inorganic Scintillation Crystals for Neutron Detection

Maria da Conceição Costa Pereira, Tufic Madi Filho and
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Abstract: Inorganic scintillators play an important role in the detection and spectroscopy of gamma and X-rays, as well as in neutrons and charged particles. For a variety of applications, new inorganic scintillation materials are being studied. New scintillation detector applications arise continuously and, consequently, the interest in the introduction of new fast scintillators becomes relevant. 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 and bromine 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. After growth is finished, the furnace is cooled at a rate of 20° C/h to room temperature. The concentration of the lithium doping element (Li) studied was 10^{-3} M and the concentration of the bromine was 10^{-2} M. Analyses were carried out to evaluate the scintillators developed concerning the neutron from the AmBe source, with energy range of 1MeV to 12 MeV. Lithium can capture neutrons without gamma-ray emission, thus, reducing the back-ground. The neutron detection reaction is $^{6}Li(n,\alpha)^{3}H$ with a thermal neutron cross section of 940 barns. In this paper, it was investigated the feasibility of the CsI:Li and CsI:Br crystals as neutron detectors for monitoring, due to the fact that in our work environment there are two nuclear research reactors and calibration systems.

Key Words: scintillators, radiation detectors, neutron detection, crystals

I. INTRODUCTION

The need to detect radiation appeared soon after the discovery of X-rays and radioactivity and it is a challenge that has extended to the twenty-first century. The radiation detection and measurement of properties are required in all facets of nuclear technology, i.e., the scientific studies in the operation of reactors for energy production, radiation protection, in industry and in the medical field. The diversity of physical interactions between radiation and matter is such that it can be assumed that no detector may be applied to the measurement of all types of radiation, even the generic application of only one type of radiation, so that each detector has limited applicability to particular cases.

The type of detector used depends on several factors; particles to be observed, the energy of the particle and the environment in which the detector is used. So it is clear the need to develop processes and tools to make perceptible the presence of particles and their properties.

Neutron 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 its presence. Neutron detectors are constituted of a radiation-sensitive element and a system that transform these radioactive effects into a value related to a measured quantity of this radiation. Ionizing radiation covers a broad spectrum of energy and different types of interactions with matter. Consequently, each detector has its field of use defined by the type of radiation, energy range and characteristics of the physical response. 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.[1]

The scintillation method is still one of the mostly used for the detection of ionizing radiation. The universality of this method is considered to be its main advantage. It can be used for registration of almost all types of radiation in a wide range of energy (varying from several eV to tens GeV).

Neutron detection with spatial, temporal and energy resolutions is important to the improvement of high energy physics, neutron forensics, non-proliferation of special nuclear materials, nuclear energy, oil-well logging, the search for dark matter, inelastic neutron scattering, astrophysics, structural biology and it is fundamental to the advancement of nuclear medicine, nuclear chemistry and magnetism.

In this work, the feasibility of the CsI:Li and CsI:Br crystals, as neutron detectors to be used for monitoring in nuclear research reactors and in calibration systems, was investigated. The pure CsI was used for comparison.

II. MATERIALS AND METHODS

Lithium and bromine doped CsI crystals were grown using the vertical Bridgman technique in the Instituto de Pesquisas Energéticas e Nucleares IPEN/CNEN-SP. In this technique, the charge is maintained at high temperature for 10 h for the material melting and complete reaction, using a quartz crucible in vacuum atmosphere. The temperature gradient

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21° C/cm and 1 mm/h descending velocity were chosen as technique parameters. After the growth had finished, the furnace was cooled at a rate of 20° C/h to room temperature. The starting material used with a purity of 99.99% was obtained from Metal Gesellschalt K.K. The concentration of the lithium doping element (Li) studied was 10^{-3} M and the concentration of the bromine (Br) was 10^{-1} M.

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.

In the study of the response to neutron radiation, the crystals were polished with ethylene glycol and 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 are shown in Fig. 1. 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 x 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.

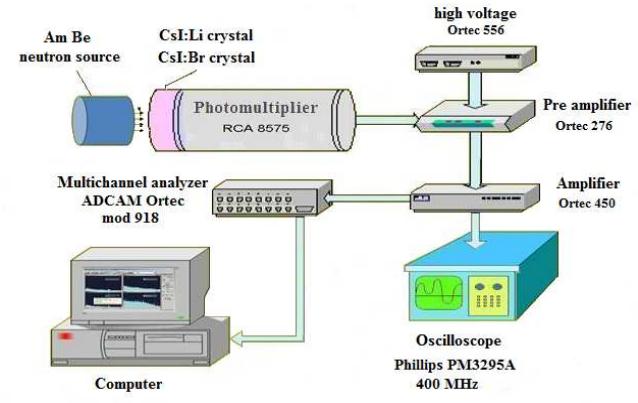

Fig. 1 – Schematic representation of the electronics associated with CsI: Li and CsI:Br scintillator crystals.

III. RESULTS AND DISCUSSIONS

Before starting the measurements of neutrons with CsI: Li, CsI: Br and pure CsI 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. The measurement conditions were the same for all crystals, namely, the distance between the source crystal and the counting time, the photomultiplier tube voltage, the signal amplification and the volume of crystals. In Fig. 2, the result of the laboratory background radiation spectrum is shown.

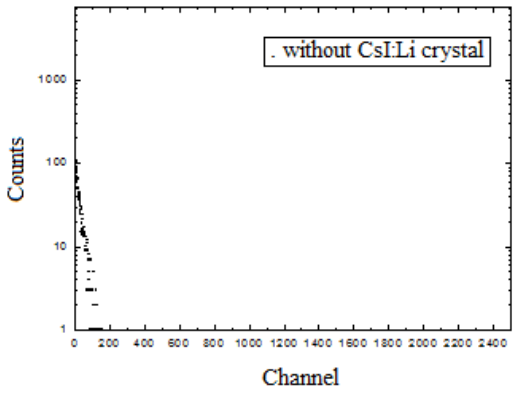

Fig. 2 – Spectrum of the background radiation laboratory

Crystals of CsI: Li with nominal concentration of 10^{-3}M when excited by neutron radiation from an AmBe, ^{6}Li 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. [2] The neutron line will be in an order of magnitude more intense if ^{6}Li enriched material is used.

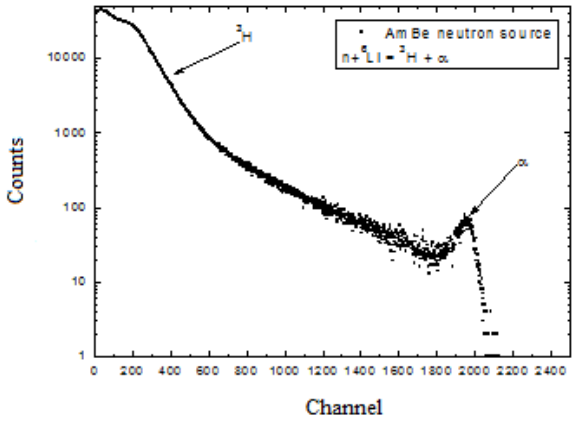

Fig. 3 – Pulse height spectrum of the CsI:Li inorganic scintillator from the AmBe neutron source.

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.[3,4,5]

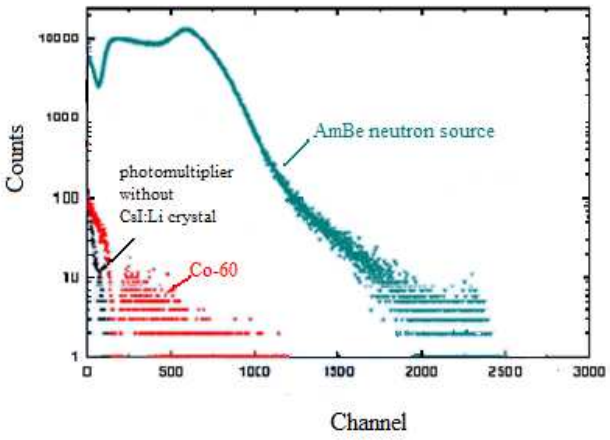

Fig. 4 – Pulse height spectra of the CsI:Li inorganic scintillator, from radiation of ^{60}Co and AmBe sources.

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.

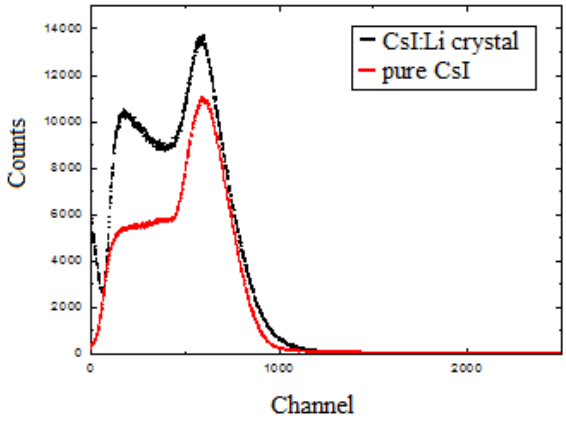

Fig. 5 – Pulse height spectrum of the CsI:Li inorganic scintillator and the pure CsI.

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.

In Fig. 6 and 7, the neutron radiation spectra using CsI:Br crystals, with and without paraffin, are shown.

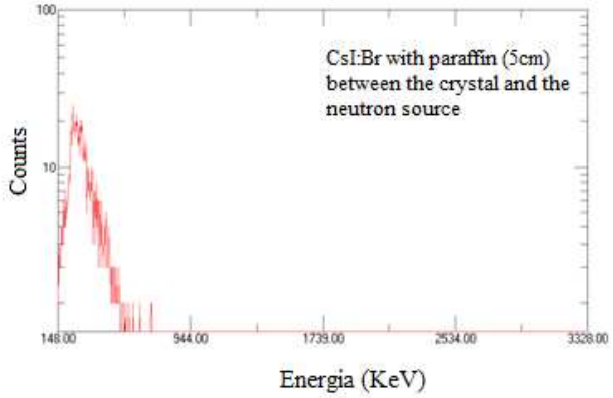

Fig. 6 – Pulse height spectrum of the CsI:Br inorganic scintillator, with paraffin.

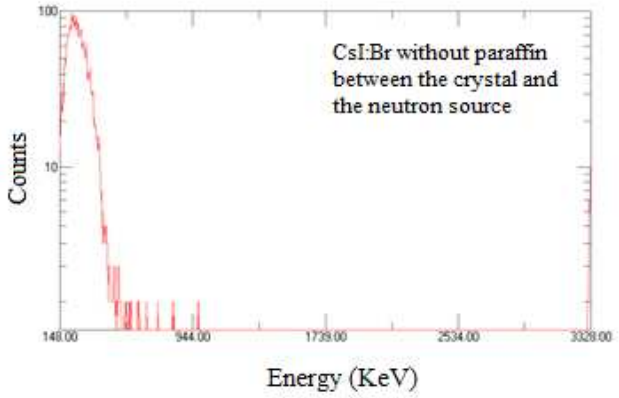

Fig. 7 – Pulse height spectrum of the CsI:Br inorganic scintillator, without paraffin.

It should be noted that Br containing scintillator materials become radioactive under neutron irradiation due to the neutron capture in ^{79}Br, resulting in ^{80m}Br, which decays to ^{80}Br with half-life of 17.6 min. The main decay product is a beta continuum of 2 MeV endpoint. [6,7,8]

In Fig. 8 and 9, the neutron radiation spectra using pure CsI crystals, with and without paraffin, are shown.

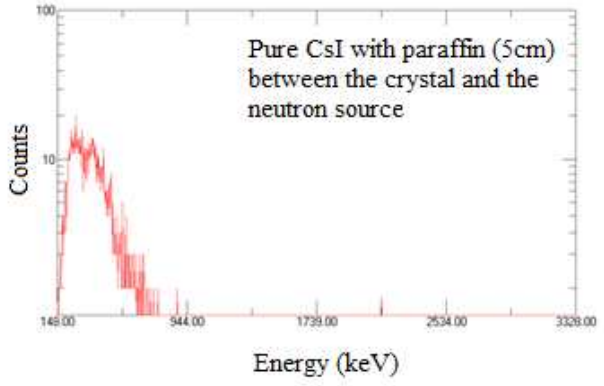

Fig 8 – Spectrum of the pure CsI crystal from radiation of an AmBe source, with paraffin.

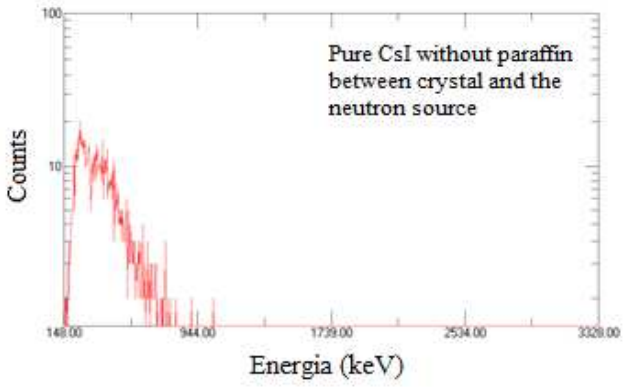

Fig. 9 – The spectrum of the pure CsI crystal from radiation of an AmBe source, without paraffin, can be observed.

As it can be seen in these curves, there are not significant differences. The use of paraffin did not modify the shape of the spectrum obtained for pure CsI crystal, when excited with radiation from the neutron AmBe source.

IV. CONCLUSION

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. Obviously, further work will have to be carried out on these materials, in particular on the concentration of dopants 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 ($n + {}^{6}Li \rightarrow {}^{3}H + \alpha$) are detected in this crystalline volume.

The crystal doped with Br (CsI:Br) showed neutrons with lower efficiency due to the reaction with thermal neutrons be (n, γ) and the gamma generated not be fully absorbed in the crystalline volume.

V. REFERENCES

[1] KNOLL, G. F. Radiation Detection and Measurement. 3rd ed. Hoboken, NJ. John Wiley & Sons, 2000.

[2] Tufic Madi Filho et all, "Experimental and Monte Carlo Evaluation of the Neutron Flux of an Assembly with two AmBe Sources," *Radiation Protection Dosimetry*, vol. 115, no. 1-4, pp. 412–414, 2005, doi:10.1093/rpd/nci150.

[3] LIMA, R. B. *Avaliação experimental do fluxo de nêutrons de um irradiador com fontes de AmBe e sua possibilidade de uso em análise de materiais*. Dissertação de mestrado. IPEN – São Paulo, 2003.

[4] Zamboni, C. B; Filho, T. M.; *Fundamentos da Física de Nêutrons*, cap. 2, ed. Livraria da Física, São Paulo, 2007.

[5] Eijk. C. W. E. ; Inorganic-scintillator development. *Nucler Instruments and Methods in Physics Research*, vol. 460A, pp.1-14, 2001.

[6] Eijk, C. W. E. ; Inorgancic scintillators for thermal neutron detection. *IEEE Transactions on Nuclear Science*, vol. 59, no.5, pp. 2242-2247, 2012, doi: 10.1109/TNS.2012.2186154.

[7] Bartle, C. M. ; Haight, R. C.; Small inorganic scintillator as neutron detectors. *Nuclear Instruments and Methods in Physics Research*, vol.422 A, pp. 54-58, 1999.

[8] Eijk, C. W. E. ; Neutron detection and neutron dosimetry. *Radiation Protection Dosimetry*. Vol. 110, nos 1-4, pp. 5-13, 2004, dói: 10.1093/rpd/nch155.

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