Inorganic Scintillation Crystals for Neutron Detection

Maria da Conceição Costa Pereira, Tufic Madi Filho, and José Patrício Náhuel Cárdenas

***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, are of 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 1 MeV to 12 MeV [1]. Lithium can capture neutrons without gamma-ray emission, thus, reducing the back-ground. The neutron detection reaction is $^{6}\mathrm{Li}(n,\alpha)^{3}\mathrm{H}$ with a thermal neutron cross section of 940 barns. In this paper, the feasibility of the CsI:Li and CsI:Br crystals as neutron detectors for monitoring was investigated, due to the fact that in our work environment there are two nuclear research reactors and calibration systems.**

***Index Terms*— Crystals, neutron detection, radiation detectors, scintillators.**

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 21st 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 there is not detector which may be applied to the measurement of all types of radiation, not even to 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. Therefore, the need to develop processes and tools to make the presence of particles and their properties perceptible is very clear.

The radiation monitoring system is an important requirement in the premises of a nuclear reactor. A variety of types of radiation (neutrons, gamma, beta and fission products) existent in a reactor is associated to the broad energy spectrum of these radiations, implying the need to use a range of detectors to run in the reactor system and security and in the radiological monitoring. As the neutron sources are associated to gamma radiation, it is necessary that the neutron detecting system may be capable to discriminate the gamma interference. Due to the absence of charge in the neutron, some material converter that generates radiations capable to produce signals in the detector should be used. Materials with high cross section, as lithium, are used for this purpose.

The detection of neutrons is not trivial due to lack of load of these particles and the peculiarity of their interaction with matter. The neutron sources, also, generate gamma radiation, which may interfere with their measurement. It is necessary that the detector system may be able to discriminate these interferences.

The main types of neutron detectors are sensitive to: a) gases, b) self-powered, c) scintillators, and d) semiconductors [2].

Some scintillators are manufactured with neutron active material added to achieve enhanced neutron detection capability. The purpose is to achieve more located and more rapid detection of neutrons than it would be possible with gas counters. Gadolinium, ^{10}B and ^{6}Li are typical materials loaded into the scintillator. One major attractive feature of Li is its very low cross section for gamma interactions. The neutron active material initiates the light production by releasing energetic charged particles or gamma rays, when neutron is captured. After the initial interaction with the neutron occurs, the detection process is the same as if the light had been produced by gamma ray. Because the scintillator is, also a gamma ray detector, its gamma ray sensitivity is generally very high. There are, however, several possible configurations with good neutron detection efficiency and low gamma ray sensitivity.

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

---

Manuscript received July 18, 2013; revised May 23, 2014; accepted March 29, 2016. Date of current version June 21, 2016. (*Corresponding author: M. d. C. C. Pereira.*)

All the authors are with the Nuclear and Energy Research Institute, IPEN–CNEN/SP, Instituto de Pesquisas Energéticas e Nucleares–IPEN-CNEN/SP, Av. Prof. Lineu Prestes 2242 Cidade Universitária CEP: 05508-000–São Paulo SP, Brazil (e-mail: macoper@ipen.br; tmfilho@ipen.br; ahiru@ipen.br.

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TNS.2016.2551379

materials capable of producing light when ionizing radiation dissipates its energy in their midst [2].

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 of GeV) [3].

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 [4].

The paper presents the development of a small-sized thermal neutron detector based on cesium iodide scintillator doped with natural lithium and bromine. The ^{6}Li-scintillator can distinguish neutrons and gamma rays by the difference in pulse height. It also has large detection efficiency for thermal neutrons and can detect fast neutrons (thermal cross section = 941 barns and resonance cross section = 423 barns) [5].

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 to reach material melting and complete reaction, using a quartz crucible in vacuum atmosphere. The temperature gradient 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 350 °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. The faces of the crystal that were not in contact with the photo-sensor were covered with several layers of politetraflouroethylene (PTFE or commercially known as Teflon TM) 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.

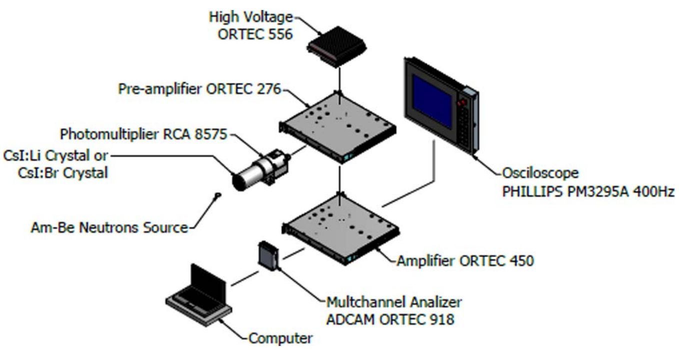

Fig. 1. Schematic representation of the electronics associated with CsI:Li and CsI:Br scintillator crystals.

In response to neutron radiation an AmBe source with energy range of 1 MeV to 12 MeV [1] was used. The activity of the AmBe source was 1 Ci Am and 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 CsI:Li, CsIBr, and pure CsI scintillator crystals used were cut with dimensions of 2 cm diameter and 2 cm high.

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.

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; $n +^{6}\mathrm{Li}(7,5\%) \rightarrow^{3}\mathrm{H}(2.75\ \mathrm{MeV}) + \alpha$, as shown in Fig. 2. 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 [6]. The neutron line will be in an order of magnitude more intense, should ^{6}Li enriched material be used.

The curve shown in Fig. 3 illustrates the results for the neutron radiation from an AmBe source using the CsI:Li scintillator crystal. Paraffin was used for the thermalized the 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 [7]–[9].

In Fig. 4, the radiation of the neutron spectrum using CsI:Li and pure CsI crystals, under the excitation of an AmBe neutron source, is shown.

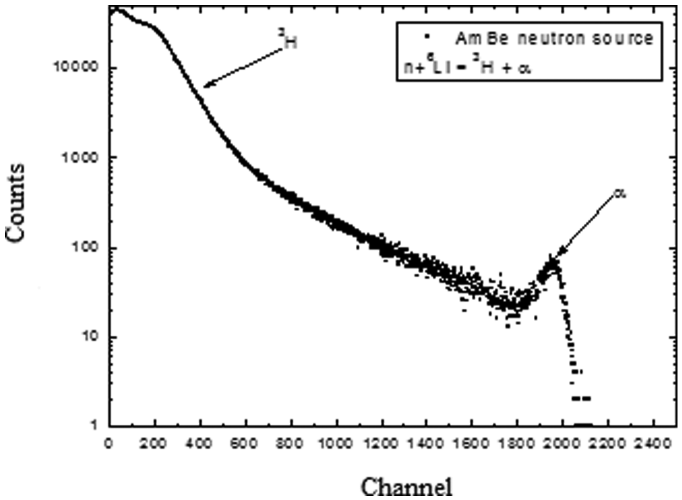

Fig. 2. Pulse height spectrum of the CsI:Li inorganic scintillator from the AmBe neutron source.

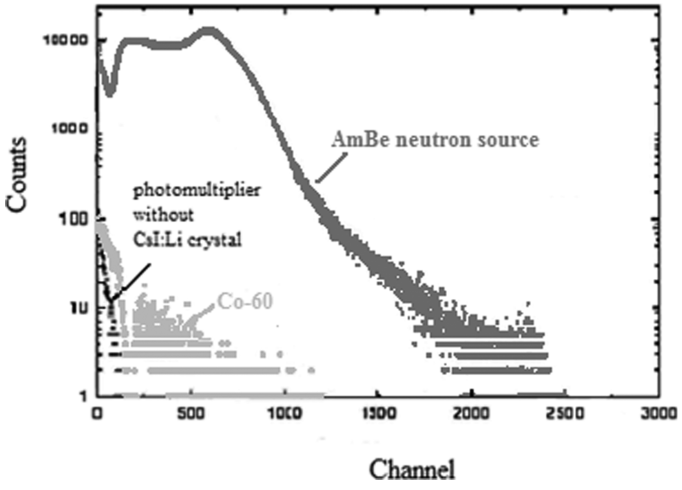

Fig. 3. Pulse height spectra of the CsI:Li inorganic scintillator, from radiation of ^{60}Co and AmBe sources.

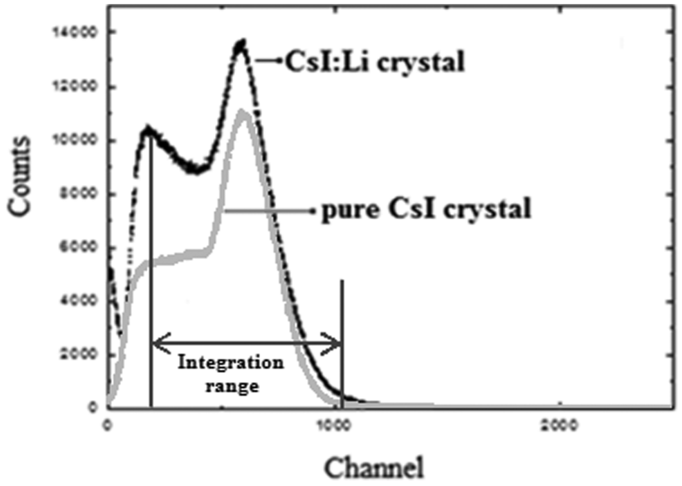

Fig. 4. Pulse height spectrum of the CsI:Li inorganic scintillator and the pure CsI.

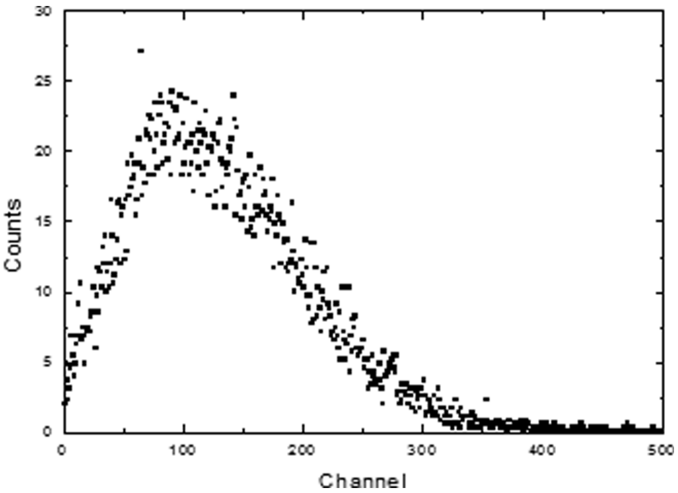

Fig. 5. Pulse height spectrum of the CsI:Br inorganic scintillator, with paraffin for neutron attenuation.

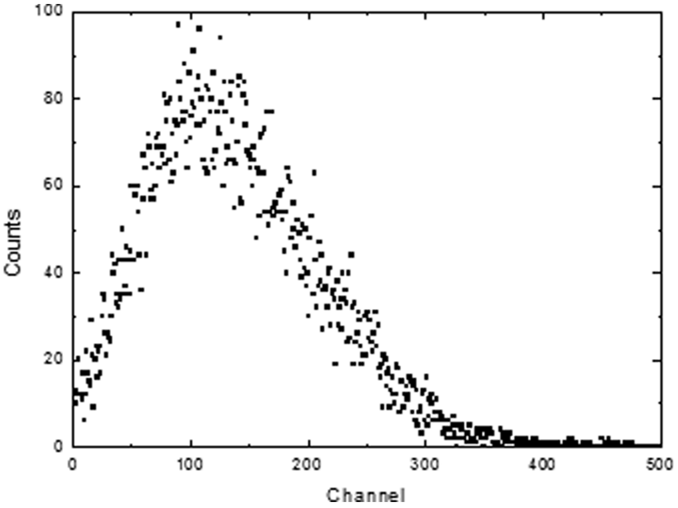

Fig. 6. Pulse height spectrum of the CsI:Br inorganic scintillator, without paraffin.

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. Integrating the stated range of the two spectra, we have increased efficiency of approximately 10 times.

In Figs. 5 and 6, the neutron radiation spectra using CsI:Br crystals, with and without paraffin, are shown.

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 [10]–[12].

In Figs. 7 and 8, the neutron radiation spectra using pure CsI crystals, with and without paraffin, are shown.

As it may 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.

The main difference between Li and Br, in addition to the cross section, is the reaction type with neutron: Li has (n, α) and Br has (n, γ) reaction, this allows a better responses of the detector doped with Li when irradiated with neutrons.

The small size of this detector reduced the response functions of gamma rays. The small-sized detector based on cesium

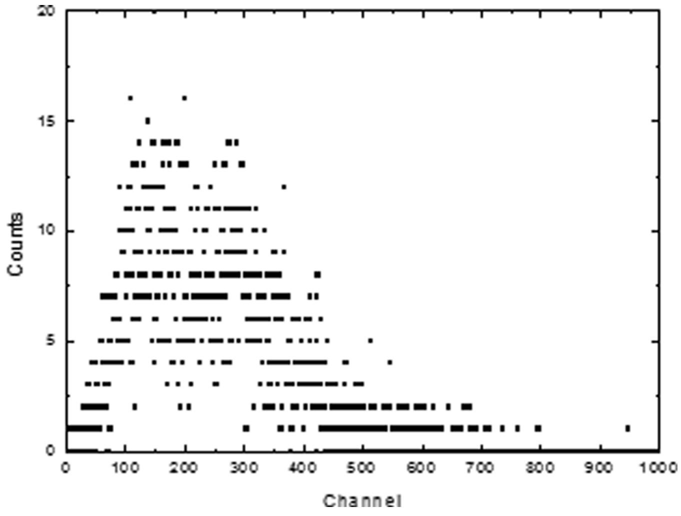

Fig. 7. Spectrum of the pure CsI crystal from radiation of an AmBe source, with paraffin.

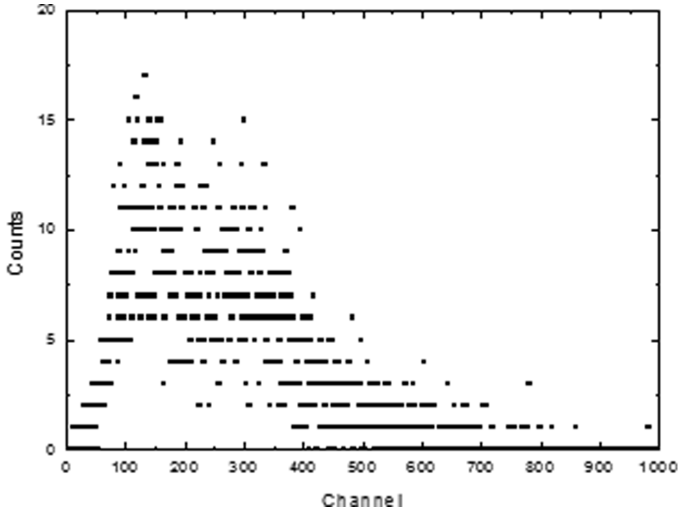

Fig. 8. The spectrum of the pure CsI crystal from radiation of an AmBe source, without paraffin.

iodide scintillator doped with natural bromine, does not show a good response to neutrons due to its small cross section (both isotopes).

IV. Conclusion

The addition of Li and Br to 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 occurrence of dopants concentration 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 $(\mathrm{n} +^{6} \mathrm{Li} \rightarrow^{3} \mathrm{H} + \alpha)$ 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 is not fully absorbed in the crystalline volume.

References

[1] M. N. Thompson and J. M. Taylor, "Neutron spectra from Am-a-Be and Ra-a-Be sources," *Nucl. Instrum. Methods Phys. Res.*, vol. 27, pp. 305–308, 1965.

[2] G. F. Knoll, *Radiation Detection and Measurement*, 3rd ed. Hoboken, NJ, USA: Wiley, 2000.

[3] M. J. Weber, "Inorganic scintillators: Today and tomorrow," *J. Luminescence*, vol. 100, pp. 35–45, 2002.

[4] A. J. Peurrung, "Recent developments in neutron detection," *Nucl. Instrum. Methods Phys. Res.*, vol. 443, pp. 400–415, 2000.

[5] T. Matsumoto, H. Harano, A. Masuda, J. Nishiyama, Y. Sakurai, and A. Uritani, "New idea of a small-sized neutron detector with a plastic fibre," *Radiat. Protection Dosimetry*, pp. 1–4, 2011.

[6] T. M. Filho, R. B. Lima, H. Yoriyazi, and A. C. Hernandes, "Experimental and Monte Carlo evaluation of the neutron flux of an assembly with two AmBe sources," *Radiat. Protection Dosimetry*, vol. 115, no. 1–4, pp. 412–414, 2005.

[7] R. B. Lima, *Avaliação experimental do fluxo de nêutrons de um irradiador com fontes de AmBe e sua possibilidade de uso em análise de materiais*, M.S. thesis dissertation, IPEN, São Paulo, Brazil, 2003.

[8] C. B. Zamboni and T. M. Filho, *Fundamentos da Física de Nêutrons*, 2nd ed. São Paulo, Brazil: Livraria da Física, 2007.

[9] C. W. E. Eijk, "Inorganic-scintillator development," *Nucl. Instrum. Methods Phys. Res.*, vol. 460A, pp. 1–14, 2001.

[10] C. W. E. Eijk, "Inorgancic scintillators for thermal neutron detection," *IEEE Trans. Nucl. Sci.*, vol. 59, no. 5, pp. 2242–2247, 2012.

[11] C. M. Bartle and R. C. Haight, "Small inorganic scintillator as neutron detectors," *Nucl. Instrum. Methods Phys. Res.*, vol. 422 A, pp. 54–58, 1999.

[12] C. W. E. Eijk, "Neutron detection and neutron dosimetry," *Radiat. Protection Dosimetry*, vol. 110, no. 1–4, pp. 5–13, 2004.