Development of neutron detector using the surface barrier sensor with polyethylene (n, p) and ^{10}B (n,α) converters

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Abstract

A Si semiconductor detector, surface barrier type, with a slim film of a converter material, capable to produce charged particles, was used as a sensor of neutrons to be used in an environment of a zero power reactor. Two types of converters were used to improve the detection efficiency: (1) polyethylene, $n(CH_2)$, which produces recoil protons from the (n,p) interaction and, (2) ^{10}B which generates α particles from the (n,α) reaction. The optimal thickness of these converters was determined experimentally. Specifically for the polyethylene, a mathematical model was developed to fit the experimental data. For an AmBe source the optimum polyethylene converter thickness was of 5.8×10^{-2} cm ($62.64 \, \mathrm{mg \, cm^{-2}}$), while for the ^{10}B it was equal to 6.55×10^{-4} cm ($1.54 \, \mathrm{mg \, cm^{-2}}$). The converters of polyethylene and ^{10}B improved the detection efficiency to a factor of 4.7 and 3.0, respectively, compared to measurements without converter. Comparing the three spectra of the background radiation, the polyethylene recoil protons and the ^{10}B α radiation, it was concluded that the polyethylene presented better performance than the ^{10}B converter. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Neutron detector; Surface barrier detector; Polyethylene and ^{10}B converters

1. Introduction

The nuclear fission reactor is primarily a potent neutron generator and the development of the neutron detectors is essential in the nuclear reactors technology. However, the neutron sources generate γ radiation, which can interfere in their measurement. Then, it is necessary that the detecting system be capable to discriminate the γ interference. Several techniques have been proposed, among them two are commonly used: (i) pulse-height analysis applying threshold electronic discriminator and (ii) pulse-shape discrimination [1–3]. The choice between these alternatives depends fundamentally on the pulse height generated from the γ photon and the neutron particle. If they are similar and overlapping each other, then the pulse-shape discrimination can be used, otherwise the technique of electronic discrimination would be preferable.

The main types of sensitive neutron detectors are gaseous, scintillator, semiconductor and self-power. These detectors must have a neutron converter internally to produce ionising radiation. The

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converter is fundamental in the nuclear reactions of the type: neutron + converter → detectable radiation [2,4–6].

In the field of reactor research, the dimension of the detector and its materials are important parameters in order to avoid interferences or deformations in the flux measurements. By reducing the size of the detector, the deformation of the flux can be decreased. When that is not possible, it is necessary to use complex mathematical processes to overcome the problem, for example, Dalton [7] developed theoretically a study of the transport equation to evaluate the neutron population in a detector position in order to correct that influence. Among other factors, this is the cause of the continuous effort to project small and sensitive neutron detectors or to improve their characteristics.

In the nuclear fission reactors, the neutrons generated in the primary process have energies around 20 MeV, which are classified as fast neutrons [8]. Other devices, such as the particle accelerators and sources of AmBe, also produce fast neutrons. However, in the projects of detectors the most used nuclear converters, such as ^{10}B and ^{7}Li, are not efficient in that energy level. This limitation can be overcome by using a moderator material to reduce the neutron energy level. Materials with high hydrogen concentration such as paraffin, polyethylene or polystyrene are considered good moderators [8,9]. The efficiency of these moderators is due to the similarity between the neutron and the hydrogen nucleus, favouring the energy transfer from the neutron to the hydrogen.

The silicon detectors are efficient for charged particles measurements and they could be also used for neutron measurements using a converter [5,9,10,11]. The advantageous use of the Si detector is due to its relatively high density ($2.33 \, \text{g cm}^{-3}$) compared to gaseous detectors. Furthermore, the ionising particles lose all their energy in a short path length and, as a consequence, it is possible to reach high-detection efficiency with a small detector volume. The α particles, produced in the neutron reaction with the ^{10}B converter, have in the Si a stopping-power that depends on the initial energy of the particle. For α particles ranging from 3 to 50 MeV, the path length varies from 0.01 to

1 mm [12]. Thus, a thin layer Si detector, of about 1 mm thick is sufficient to stop α particles close to 1.5 MeV, as the particles generated in the ^{10}B (n,α) ^{7}Li reaction. The silicon also has a high sensitivity for proton detection. Analogously, the range of the proton in the silicon varies approximately from 0.09 to 1 mm for particles ranging from 3 to 12 MeV [12]. Another advantage of the silicon detector is due to the high mobility of the electrons and holes. This characteristic and the small dimension of the detector allow the charge collection in a short time and therefore, a high resolution can be reached in a time of nanoseconds.

In this work, neutron detectors using silicon semiconductor, surface barrier type, using ^{10}B as (n, α) reaction and polyethylene as (n, p) recoil protons generation converters were developed. In such design the optimum thickness of the converters is an important parameter to be established. The optimum thickness was determined experimentally and theoretically. The ability of the detector to act as controller of a reactor of zero-power-type was evaluated.

2. Experimental procedures

The Si surface barrier detectors were made from wafers (Topsil), with resistivity of 50 kΩ cm, 2.54 cm in diameter — total area $5 \, \text{cm}^2$ and 1 mm in thickness, according to Takami et al. [4]. Approximately, $3.14 \, \text{cm}^2$ sensitive surface was covered with $80 \, \mu\text{g cm}^{-2}$ Au and the rear face with $40 \, \mu\text{g cm}^{-2}$ Al in order to form the ohmic electrode, using the vacuum deposition technique. Such assembly presents depletion layer depth of $420 \, \mu\text{m}$ at 40 V bias voltage and specific capacitance of $70 \, \text{pF mm}^{-2}$ [4].

To determine the optimum thickness of the ^{10}B (90.1% enriched and 91% chemical purity) acting as a (n,α) converter, different ^{10}B thicknesses were covered in the radiation sensitive face of the detector, ranging from 0.0 to $2.0 \, \mu\text{g cm}^{-2}$. Fig. 1 shows the schematic assembly and the picture of the detector with the ^{10}B converter. The ^{10}B converter presents a larger cross-section for thermal neutrons than for fast neutrons [13]. So, in order to thermalize the fast neutron flux from the AmBe source, 7 cm paraffin block was used.
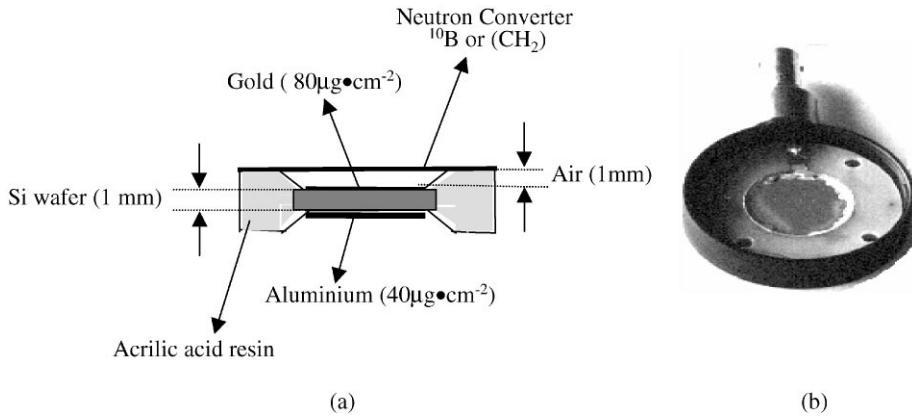
Fig. 1. Assembly diagram of the Si detector with converters (a) and illustration of the developed surface barrier detector (b).

The optimum ^{10}B converter thickness was assumed being the result that presents the best efficiency ε, that is, the ratio:

$$\varepsilon = \frac{\text{Counting rate (ips)}}{N_0 \text{ Incident neutrons (n s}^{-1})}.$$

For a full-pulse amplitude scale of 10 V a threshold of 0.6 V was used. This level was capable to cut signals from noise and γ radiation. For α particles from ^{10}B$(n,\alpha)^7$Li, the peak was located around 2.4 V in a spectrum ranging from 0.6 to 7.5 V.

The polyethylene converter was used as recoil proton generator. The same approach described above was used to establish the best thickness of the polyethylene. Experiments were carried out using different thicknesses of the polyethylene ranging from 0 to 1.2 mm. To fit the thicknesses X (cm) and counting rate R (ips), a mathematical model was proposed in this work, which was defined as

$$R(\text{ips}) = \varepsilon_p N_0 (1 - e^{-\Sigma X}) e^{-\mu \cdot X} + \varepsilon_n N_0 e^{-\Sigma X}$$

where ε_p is the intrinsic efficiency for recoil protons detection, assumed as equal to 1 (all the protons reaching the detector generate a signal), ε_n the intrinsic efficiency for direct neutron detection, which was assumed as equal to

$$\frac{\text{Counting rate without converter (ips)}}{N_0 \text{ (ns}^{-1})} = 0.00165$$

where N_0 is the 29,600 (n s^{-1}). This AmBe neutron emission rate was measured by activation of the

gold foil technique [14] and depends on: (i) the solid angle between the neutron source and the detector geometry [15] and (ii) the AmBe source activity/geometry, Σ (cm^{-1}) the mean macroscopic cross-section to the incident neutrons in the converter and

$$\mu = \frac{1}{0.00235\, E_{\text{proton}}^{1.8}} (\text{cm}^{-1})$$

the proton absorption coefficient in the polyethylene converter with E_{proton} in MeV [16,17].

The parameters Σ and E_{proton} are regression parameters determined by non-linear least square.

The detectors were studied in two different situations: (1) in the laboratory, using an AmBe source and (2) in the zero power nuclear reactor (IPEN-MB-01), at a radial distance of 360 mm and half height from the reactor core.

Both detectors, (n, p) and (n,α) converters, were placed around the reactor core, then the reactor IPEN/MB 01 was operated in different powers. The slope and interception of the experimental points [power (W) versus counting rate (ips)] were determined by the least-squares method.

3. Results and discussion

Fig. 2 shows the response of the two proposed Si surface barrier detectors using ^{10}B and polyethylene converters and that without a converter. As it
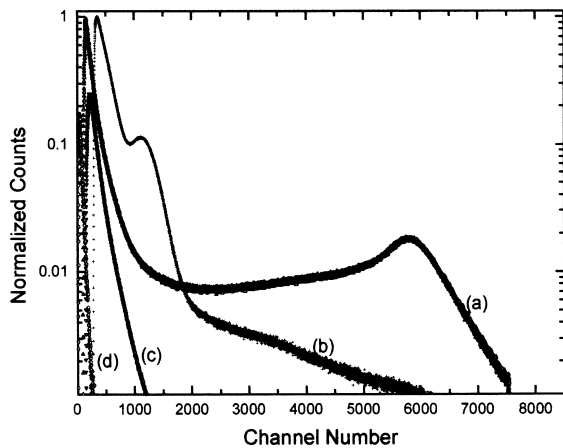
Fig. 2. AmBe spectra of the Si detectors with polyethylene converter (a), [10]B converter (b), without converter (c) and laboratory radiation background (d).

can be observed in the figure the neutrons could generate small signals in the detector even without converters. Fast neutrons can produce charged particles elastically scattered. Like a standard "kinematical billiard ball" calculation it is possible to predict the recoil atom energy, without relativistic considerations. In this way, the neutron energy fraction transferred to the recoil atom can be predicted by $4x/(1 + x^2)$ [18], where x is the ratio of the mass of the recoil atom to that of the neutron. For silicon ($x \cong 28$), this fraction is ≈ 0.143. For instance, for a neutron energy of 4 MeV, which is representative of the AmBe source, the maximum recoil energy will be only ≈ 0.572 MeV. Therefore, small signals without converters shown in Fig. 2 may be explained. However, the use of a film capable to convert neutrons in ionising radiation (α or protons) increased the efficiency of the detection significantly with the advantage of generating high signals far from interference of the noise and γ radiation. The use of a converter implies in losses and benefits due to the effect of the self-absorption of the produced particle inside the converter material itself [19]. A very slim thickness of the converter is not efficient, because it inserts a little layer of atoms for interaction with the incident neutrons. On the other hand, with thicker films, the interaction probability increases and, consequently, a larger amount of charged particles is produced. However,

since these particles (α or protons) have little range in the materials [12], they can be absorbed or it disappears in the converter itself.

In the detector set up with a [10]B converter it was observed that the optimum thickness of the [10]B film was of 6.55×10^{-4} cm (1.54 mg cm^{-2}), while the ideal thickness for a polyethylene converter was of 5.83×10^{-2} cm (62.64 mg cm^{-2}), as shown in Figs. 3a and b, respectively. Using that ideal thickness for the converters, the detection efficiency increased 3 times for the [10]B and 4.7 times for the polyethylene compared to that measured without a converter. For polyethylene, the macroscopic cross-section $\Sigma = 0.338 \pm 0.010$ cm^{-1} ($= n_H \cdot \sigma_H + n_C \cdot \sigma_C$) was estimated by the mathematical model, where n_H, n_C are the number of atoms per cm^3 in the polyethylene; σ_H and σ_C are
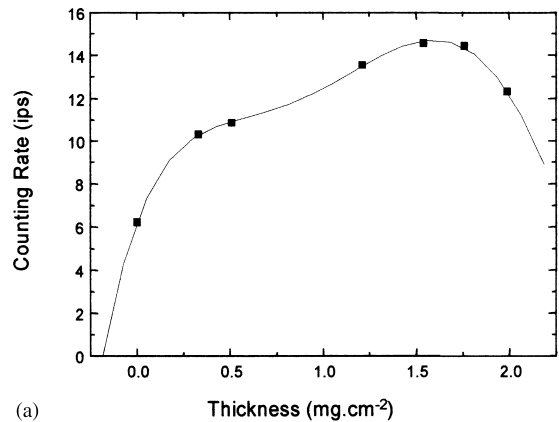
(a)
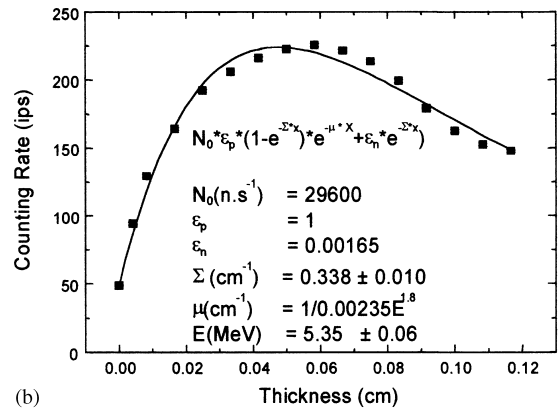
(b)

Fig. 3. Detection efficiency of Si detector as a function of the converter thickness of: (a) [10]B(n,α) and (b) polyethylene (n,p).

the microscopic cross-section in barns for H and C, respectively. Using $\Sigma = 0.338 \pm 0.010\,\text{cm}^{-1}$ and microscopic cross-section σ (barns) values, which is a function of energy E from Jaeri data [13], the representative energy of the neutrons from the AmBe source was estimated as $E = 8.88$ (8.74 to 9.01) MeV. This energy is in agreement with the neutron energy range for the AmBe source described by Thompson & Taylor [20] and Marsh et al. [21]. From the mathematical model, proposed in this work, the mean energy of the recoil protons, E_{proton}, was of $5.3 \pm 0.010\,\text{MeV}$. This value is also in accordance with the range from 1.5 to 6.0 MeV described by Salgir and Walker [6].

Practically, the radiation background level in the laboratory, did not interfere in the neutrons measurement, mainly for the polyethylene converter, which generates spectra relatively far from those produced by the radiation background, as it can be observed in Fig. 2.

The use of the detector in a reactor environment can be promising. Fig. 4a shows spectra of the detector for recoil proton, for measurements carried out in the IPEN/MB-01 zero power type reactor, operated at 1, 2, 3, 4, 5 and 10 W. As shown in this figure, the detector with polyethylene showed a good response to the power variation in the operation of the IPEN-MB01 reactor. The linear correlation between the nominal power of the IPEN/MB-01 reactor operation versus the pulse count rate (ips) of the detector with the polyethylene showed coefficient $r^2 = 99.93\%$ (Fig. 4b). Consequently, there is an excellent correlation of the linear adjustment model for the studied powers. The counting rate values used in the linear adjustment were obtained cutting the γ interference by using the pulse rate threshold technique. This was possible due to the difference between the pulse height generated by the γ radiation and the recoil protons.

Fig. 4c shows the spectra of the detector with ^{10}B in the IPEN/MB-01 reactor, operated at 10, 50 and 100 W. After the reactor had been operating for 2 h, it was shut down and the spectra of the γ radiation were measured. With the use of the ^{10}B converter a good linear correlation was not obtained between the nominal power and the pulse count rate. That occurred because the pulse height gener-
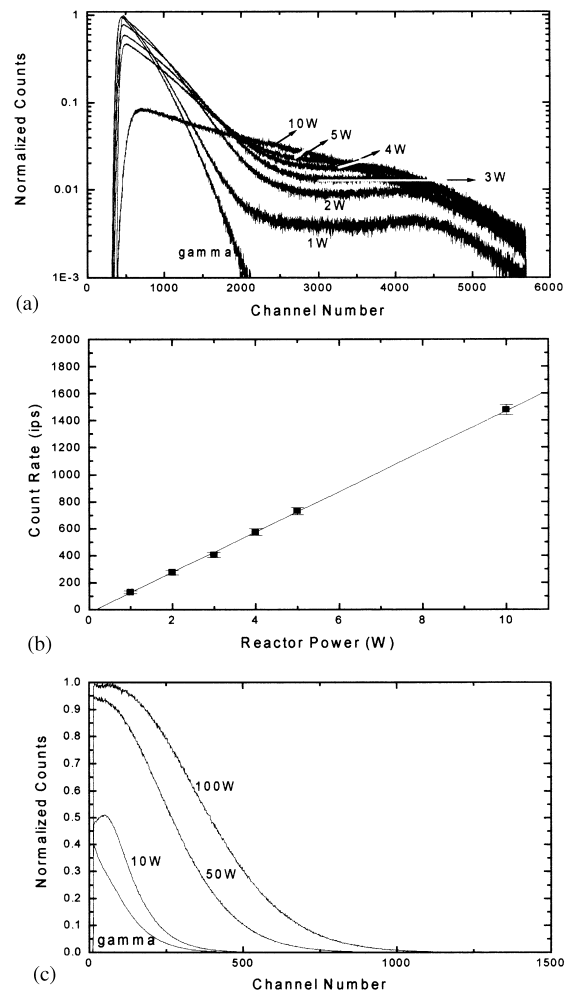

Fig. 4. (a) Spectra of the developed detector using polyethylene converter, in environment of the reactor, operating at 1, 2, 3, 4, 5 and 10 W and γ radiation spectrum after shutting down the reactor. (b) Correlation between the power and the counting rate. The predicted power (W) is $0.17 + 0.0067 \cdot R$ (ips). (c) Spectra of the detector using ^{10}B convert for 10, 50, 100 W and the γ-ray residual.

ated by the energy of 1.47 MeV of the particle of the reaction ^{10}B(n, α)^{7}Li is close to the pulses height of the γ radiation existent in the reactor. Even obtaining significant differences of pulses for the several operation powers of the reactor IPEN/MB-01, the discrimination of the γ interference was critical (Fig. 4c). With the use of that detector in more potent reactors, or more subject to high flows, irradiation damages could occur [5], and that

would limit the sensor to be used as a power controller.

The detection efficiency depends on the size, format and intrinsic efficiency of the detector [2]. The efficiency (ε) of detection is defined as the ratio between the detected value and the amount of neutron incident in the converter–detector, i.e.

$$\varepsilon = \frac{\text{Counting rate (ips)}}{N_0 \text{ Incident neutrons (ns}^{-1})}.$$

The incident neutrons (N_0) can be estimated, theoretically or experimentally, in function of the used source. The gold foil activation technique [14] is a reliable experimental choice. Applying this technique to the AmBe source used in this work, it was found that an incident fast neutrons flux of $N_0 = (9422 \pm 471)$ ns^{-1} cm^{-2}. On the other hand, N_0 estimated geometrically from the solid angle of a source-detector [15] results in a value of $N_0 = 7958$ ns^{-1} cm^{-2}. The similarity between these two values allows us to adopt the experimental value as the flux of fast neutrons from AmBe source used in this work. Consequently, the activity of incident neutrons in the detector (3.14 cm^2) could be estimated as being $(2.96 \pm 0.13) \times 10^4$ ns^{-1}. So, for the polyethylene detector version using the optimised thickness $(5.8 \times 10^{-2}$ cm) the counting rate was ≈ 225 ips (Fig. 3b), then the efficiency was $\varepsilon \cong 0.8\%$. Similar considerations are used for the ^{10}B detector, except for the implication that a ^{10}B conversion is not efficient for fast neutrons, implying a thermalization step with enriched hydrogen material. Once more, the gold foil activation technique can be used in order to estimate the neutron flux from an AmBe source thermalized with 7 cm of paraffin block. In such a case, the thermalized flux was equal to (361 ± 18)ns^{-1} cm^{-2} meaning that (1134 ± 57)ns^{-1} reach the ^{10}B converter. Hence, for optimised ^{10}B layer thickness $(6.55 \times 10^{-4}$ cm) showing a counting rate of 14.6 ips the intrinsic efficiency was of 1.29%.

4. Conclusion

It could be shown experimentally that, for fast neutrons detection, polyethylene is more efficient than ^{10}B as a converter, considering the efficiency increase and the overlapping of its spectra with the radiation background spectrum. The detection efficiency of the fast neutrons increased 4.7 times, using a polyethylene thickness of 5.8×10^{-2} cm $(62.64$ mg cm$^{-2})$, while for a ^{10}B thickness of 6.55×10^{-4} cm $(1.54$ mg cm$^{-2})$, the efficiency was increased 3.0 times, compared to the detector without a converter. The results show the capacity of the detector to act as the controller of a reactor of zero power type. The mathematical model R (ips), as a function of the parameters $\varepsilon_n, \varepsilon_p, \mu, \Sigma, N_0, E_n$, used to explain the intensity of protons that emerge from the outer surface of the recoil proton converter, was able to estimate the physical parameters realistically.

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