Study of a ^{10}B+ZnS(Ag) neutron detector as an alternative to ^{3}He-based detectors in Homeland Security

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HIGHLIGHTS

- A detailed study of ^{10}B+ZnS(Ag) neutron detector is presented using MCNPX and experimental measurements.
- The neutron detector response, bare and with different moderator configurations was estimated using Monte Carlo methods.
- The ZnS(Ag) efficiency was determined from the comparison of calculated and measured response.
- The detector performance was evaluated for ^{241}AmBe and ^{252}Cf sources.

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abstract

The response of a scintillation neutron detector of ZnS(Ag) with ^{10}B was calculated, using the MCNPX Monte Carlo Code. The detector consists of four panels of polymethyl methacrylate (PMMA) and five thin layers of ''' 0.017 cm thick ^{10}B þ ZnS(Ag) in contact with the PMMA. The response was calculated for the bare detector and with different thicknesses of High Density Polyethylene, HDPE, moderator for 29 monoenergetic sources as well as ^{241}AmBe and ^{252}Cf neutrons sources. In these calculations the reaction rate ^{10}B(n, α)^{7}Li and the neutron fluence in the sensitive area of the detector ^{10}B þ ZnS(Ag) was estimated. Measurements were made at the Neutron Measurements Laboratory, Universidad Politécnica de Madrid, LMN UPM, to quantify the detections in counts per second in response to a ^{252}Cf neutron source separated 200 cm. The MCNPX computations were compared with measurements to estimate the efficiency of ZnS(Ag) for detecting the α that is created in the ^{10}B(n, α)^{7}Li reaction. After validating new models with different geometries it will be possible to improve the detector response trying to achieve a sensitivity of 2.5 cps ng^{252}Cf comparable with the response requirements for ^{3}He detectors installed in the Radiation Portal Monitors, RPMs. This type of detector can be considered an alternative to the ^{3}He detectors for detection of Special Nuclear Material, SNM.

1. Introduction

Radiation portal monitor (RPM) systems are installed at international border crossings to detect illicit trafficking of radioactive materials. They are also used for safeguards and for scrap metal screening. At the borders RPMs help to detect special nuclear materials (SNM) such as ^{239}Pu. Here, neutrons are detected with ^{3}He proportional counters because they have high detection efficiency and an excellent gamma discrimination (Kouzes et al., 2008, Shea and Morgan, 2010, Kouzes et al., 2010).

In 2009, a shortage of ^{3}He was reported, and various investigations have been carried out with the goal to find alternative neutron detection technologies having similar characteristics to ^{3}He detectors in RPMs (Cooper et al., 2009; Zeitelhack, 2009). ^{3}He, ^{6}Li and ^{10}B are widely used for neutron detection due to their high capture cross sections.

The natural abundances of ^{6}Li and ^{10}B are 7.5% and 19.8% respectively. In order to have a larger amount of these isotopes, for

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practical neutron detectors in RPMs, an enrichment processes is required (McMillan and Marsden, 2010). ZnS(Ag) is an inorganic scintillator widely used to detect α particles. It has a high scintillation efficiency but has a long light decay constant and absorbs its own luminescence. Thus it cannot be used in layers thicker than 25 mg/cm^2. By adding ^{10}B to the scintillator the ^{10}B+ZnS(Ag) could be a feasible option to replace the ^{3}He in the RPMs (Koontz et al., 1955; Lee et al., 2011).

Two detectors manufactured by BridgePort Instruments, LLC, commercially referred as "nDet Brick" N 15 (BridgePort, 2013), were tested by the Universidad Politécnica de Madrid (UPM), These detectors use plates of polymethyl methacrylate (PMMA) with very thin layers of ^{10}B+ZnS(Ag). For both detectors, moderators were designed to increase the neutron detection efficiency. The aim of this work is to determine the neutron response of the nDetBrick^{10}B+ZnS(Ag) under different moderating conditions. The response was determined with Monte Carlo methods using the MCNPX 2.6.0 code (Pelowitz, 2008). Calculations were validated through measurements carried out with ^{252}Cf neutron sources.

2. Materials and methods

2.1. Description of the nDetBrick detector (N 15)

The N 15 detection system is manufactured by BridgePort Instruments, LLC (BridgePort, 2013). It uses ^{10}B+ZnS(Ag) neutron detection screens, a photomultiplier tube and embedded high voltage supply, and Multichannel Analyzer eMorpho® digital electronics. The detector and the photomultiplier tube measure 23 × 36 × 4 cm.

The sensitive area of the detector is formed by 5 transparent ~0.017 cm thick layers of a mixture ZnS(Ag) and Boron enriched to 95% of ^{10}B. The layers are deposited on 4 plates of PMMA of 23 × 36 × 0.635 cm acting as light guide and moderator, surrounded by ~8 μm thick aluminum mylar as light reflector.

Two moderators made of high density polyethylene, (HDPE), 0.94 g/cm^3, were designed to host the detector and the photomultiplier tube. One moderator is 12 mm thick in the front, top, bottom, and lateral faces the back of the moderator is 24 mm thick (12+24) mm. Another moderator is 24 mm thick in the front, top, bottom, and lateral faces on its backside the moderator is 48 mm tick (24+48) mm, as is shown in Fig. 1.

2.2. Monte Carlo calculations

Using the MCNPX code, a model was built including all the detector details such as: four plates 0.635 cm thick made of PMMA with 1.19 g/cm^3 density, the photomultiplier tube was modeled as an empty cylinder, the aluminum supports, the 5 layers of ^{10}B+ZnS(Ag), with 95% of ^{10}B and 5% of ^{11}B, and the ZnS(Ag) (50% Zn and 50% S). The Ag was not included in the model. Fig. 2 shows the cells used in the model.

The detector response was calculated for 29 monoenergetic neutrons sources ranging from 10^{-9} to 20 MeV in different conditions: unmoderated or bare, and encasing the detector in HDPE with thicknesses from 10 to 100 mm. In addition, the responses were calculated for the actual moderators 12+24 and 24+48 mm HDPE. Finally, responses were also calculated for ^{241}AmBe and ^{252}Cf neutron sources. The response was estimated as the number of ^{10}B(n, α)^{7}Li reactions using the tally F4 (Vega Carrillo et al., 2014a, 2014b). The neutron fluence was estimated for each cell used to define the detector as is shown in Fig. 2.

The detector model was inserted in the model of the Neutron Measurements Laboratory of the Universidad Politécnica de Madrid (LMN UPM) (Vega Carrillo et al., 2012) where the tests were performed with two small ^{252}Cf neutron sources. Using MCNPX code the response was estimated when the detector was located at 50, 75, 100, 125, 150, 175 and 200 cm from the neutron sources ^{252}Cf. The number of (n, α) reactions were compared with the count rates measured and the detector efficiency was calculated.

Fig. 1. Configurations of "nDetBrick" detector. Inside view section.

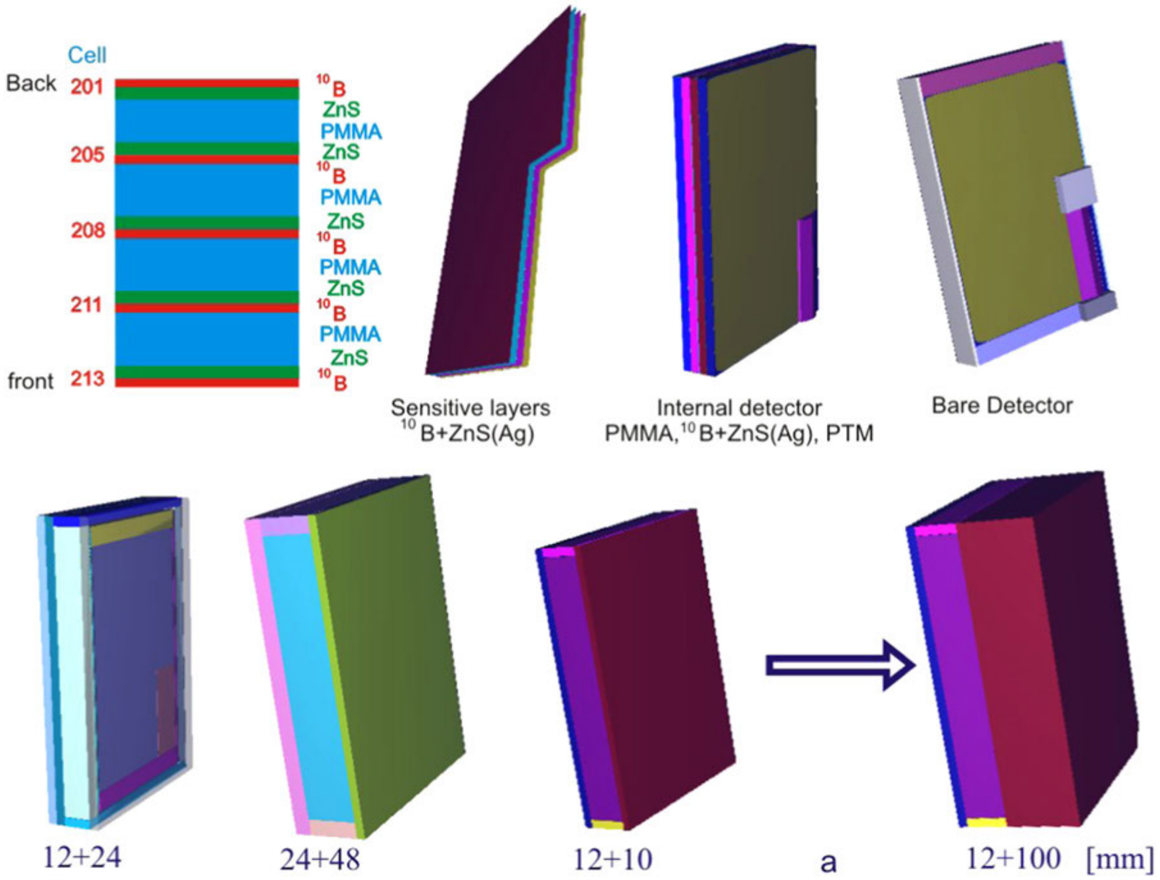

Fig. 2. MCNPX model of Detector N-15, bare and with different moderator configurations.

The same procedure was used coupling the MCNPX models of the detector and the CIEMAT Neutron Standards Laboratory (LPN CIEMAT) (Guzmán García et al., 2015), with the detector located at 200 cm from the ^{241}AmBe and ^{252}Cf neutron sources.

In all Monte Carlo calculations sample statistics was large enough to obtain uncertainties less than 3%. In the calculations the cross sections were obtained from the END/B VI library, also the S (α, β) treatment was included (Vega Carrillo et al., 2014b).

2.3. Measurements at the LMN UPM

The measurements were performed using two 37 kBq ^{252}Cf sources taking into account the corresponding decay corrections for the date of each test. Each source is cylindrical (7.8 Ø × 100 mm) with double steel encapsulation. Both sources are kept in a polyethylene support for handling and are placed into a polyethylene cylindrical container for storage, as shown in Fig. 3.

Measurements were made on the irradiation bench of the laboratory (LMN UPM) with the neutron sources at distances 50, 75, 100, 125, 150, 175 and 200 cm from the detector. For each position the neutron background was also measured in order to obtain the net count rates (Fig. 4).

3. Results and discussion

3.1. Detector response MCNPX

Fig. 5 shows the responses (^{10}B(n,α)^{7}Li reactions) for 29 monoenergetic neutron sources occurring with the bare detector case and with two HDPE moderators (12+24 mm and 24+48 mm HDPE) and it clearly shows the effect of moderation. The best moderator thickness, 24+48 mm HDPE, causes a more linear response, i.e. a relatively constant sensitivity from 10^{-7} to 1 MeV. For a large number of neutron energies it provides better performance, clearly showing that it is the preferable of the two moderator configurations.

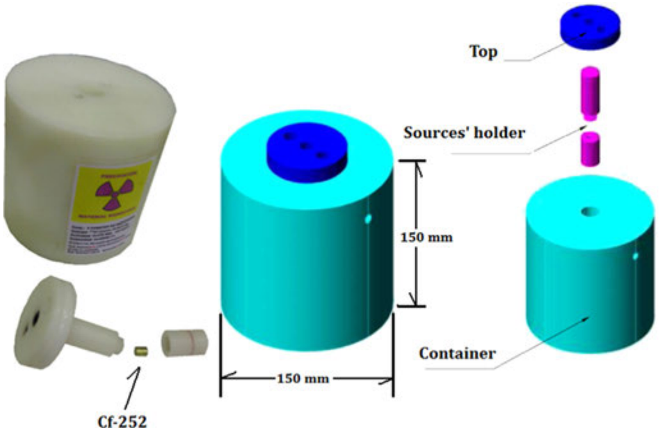

Fig. 3. ^{252}Cf Neutron sources, support and container (photo and 3D scheme).

In Fig. 6 the neutron fluence response in the sensitive areas of the detector (bare and with HDPE moderators of 12+24 and 24+48 mm) is shown. The graphs highlight the moderating quality of the HDPE.

For slow neutrons, ($E < 1E(-2)$ MeV), the largest response is from the bare detector (0 mm HDPE). For the moderated detector the highest sensitivity is achieved for neutrons of approximately 10^{-7} MeV energy. In a wide range of energies (10^{-6} to 1 MeV) the

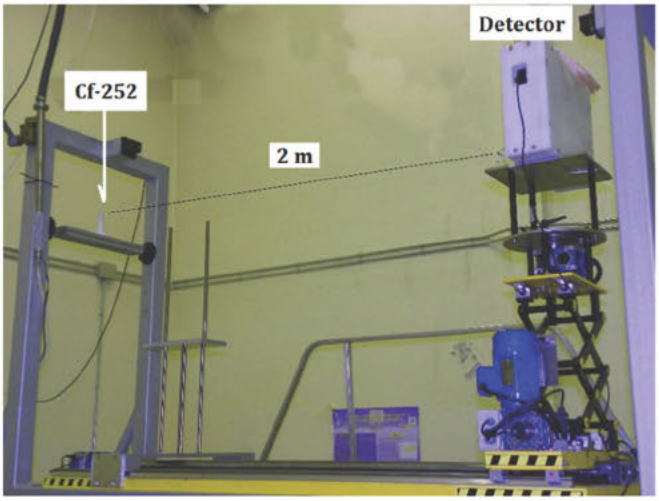

Fig. 4. Overall view of the experimental set with the ^{252}Cf sources and the N-15A detector at 200 cm on the irradiation bench.

response of the moderated detector is almost constant, and large for the thickest moderator in comparison with the thinnest moderator.

Fig. 7 shows the detector response (in ^{10}B(n,α)^{7}Li reactions) for 10 different thicknesses of HDPE. In Fig. 8, the fluence responses are shown.

These reactions results show consistency in the range from 10^{-7} to 1 MeV for thicknesses between 30 and 40 mm HDPE, giving more constant results for the response to higher neutron energies. It is noted that as the thickness increases, the response to low energy neutron decreases, but it increases for large energies. This behavior is similar to the Bonner sphere spectrometer due to the moderation of neutrons in the moderator (Mazrou et al., 2010). In the fluence response the moderator of 10 mm seems to have an almost constant fluence for all energies; this is most noticeable above 10^{-7} MeV.

Results for irradiation with ^{241}AmBe and ^{252}Cf neutrons are shown in Fig. 9. There the total number of reactions ^{10}B(n, α)^{7}Li, occurring in the detector, are plotted as a function of the HDPE thickness.

We note that, for the neutron source ^{241}AmBe the highest reaction number occurs for a moderator thickness of 50 mm, while for ^{252}Cf neutrons the maximum occurs at 40 mm of HDPE. This is because the average energy of each source is different, 5 MeV for the ^{241}AmBe and 2 MeV for the ^{252}Cf. Hence the neutrons from ^{241}AmBe require a greater thickness of HDPE to thermalize, as can also be observed in Fig. 7. The energy distribution function for fission neutrons from ^{252}Cf peaks between 1 and 1.2 MeV (Vega Carrillo et al., 2005). The neutrons are moderated in the HDPE hosting the detector reaching the sensitive layers and inducing (n, α) reactions in ^{10}B. When the moderator thickness is reduced, the neutron moderation is not sufficient to induce the (n, α) reactions.

3.2. MCNPX model and LMN UPM measurements, model validation

The decay corrected intensity of both neutron sources was 5.31E(4)s^{-1} which corresponds to a mass of 22.6E(−9) g of ^{252}Cf (Vega Carrillo, 1988). MCNPX output reports the number of ^{10}B(n,α)^{7}Li reactions occurring in the detector per neutron emitted from the source; it must be then multiplied by the intensity of the neutron sources ^{252}Cf. Model results show a difference with measurements ranging from 48.13% to 55.52%. Knowing that not all the occurring reactions in the ^{10}B+ZnS(Ag) can be detected by the photomultiplier and correctly recognized by the digital algorithms in the detector, we estimate an efficiency factor of (53.03 ± 0.4)% or (0.5303 ± 0.04).

Table 1 shows the counts per second (cps) of the measurements on the bench and the MCNPX model response, adjusted for the efficiency factor ZnS(Ag) of (0.5303 ± 0.04). Fig. 10 shows the correlation plot of the results with admissible correlation in the comparison, inside the uncertainties limits.

ANSI N42.35 standard (ANSI, 2006) sets a requirement for the absolute detection efficiency such that at 200 cm from a ^{252}Cf source the relationship between the counts per second detected and the californium mass in nanograms be greater than 2.5 cps/ng of ^{252}Cf.

In our measurements at 200 cm from the sources, the detector records 17.43 ± 4.18 cps, thus the absolute detector efficiency is 0.77 ± 0.17 cps/ng ^{252}Cf. This value can be justified by the small size of the N-15 detector. The effective cross section, σ, is 164.97 cm^{2} determined by the Eq. 1.

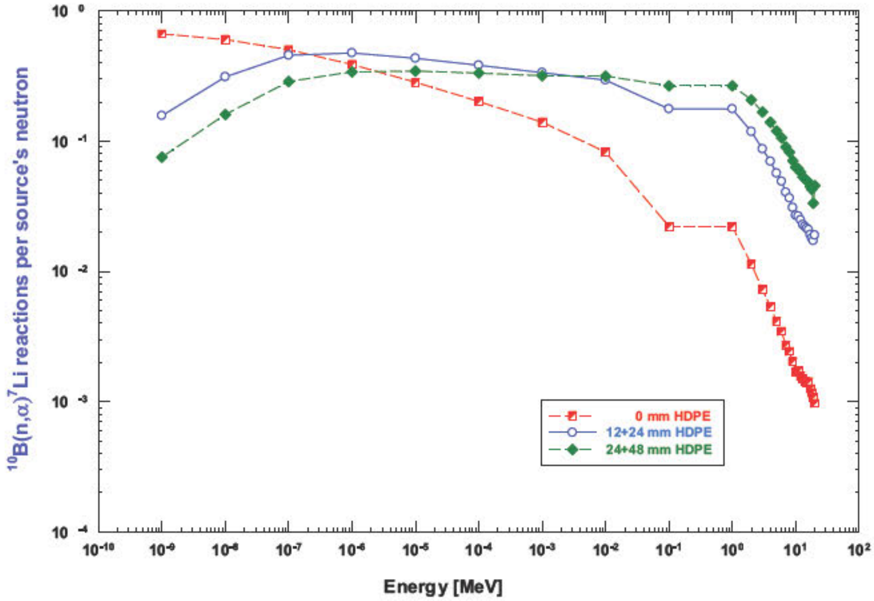

Fig. 5. N-15 detector response, ^{10}B(n,α)^{7}Li reactions (per neutron emitted from the source).

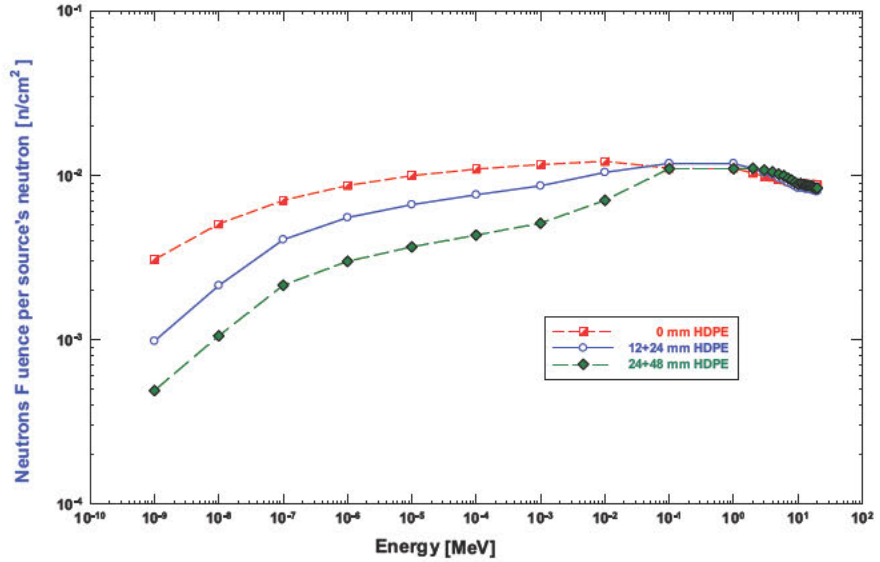

Fig. 6. N-1A detector response, neutron fluence reactions (per neutron emitted from the source), bare and moderated with 12+24 and 24+48 mm, HDPE.

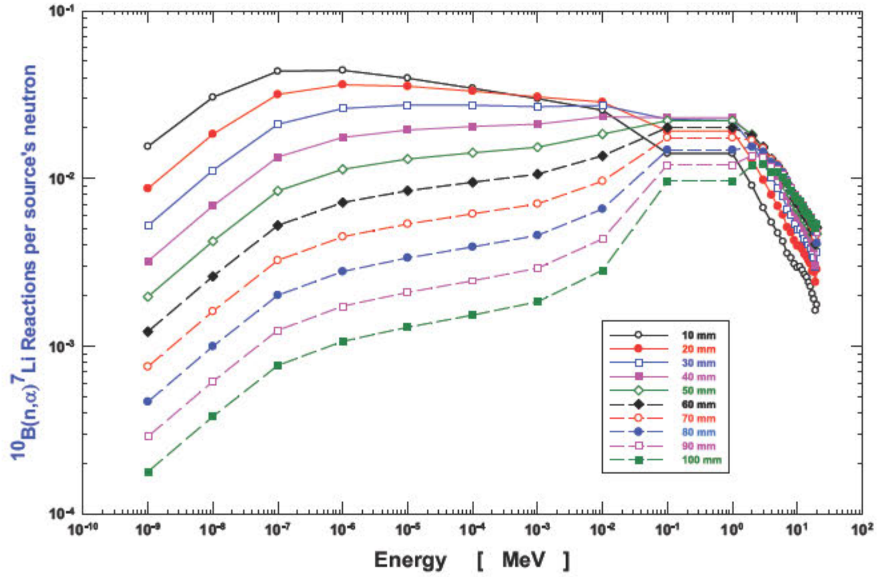

Fig. 7. N-15 detector response, $^{10}B(n,\alpha)^7Li$ reactions (per neutron emitted from the source) for variable moderator thickness, 12+10 mm to 12+100 mm, HDPE.

$$\sigma = 4\pi R^2 \varepsilon_{tot} \tag{1}$$

where R is the distance from the source, and the ε_{tot} is the total efficiency counting for the ^{252}Cf source.

3.3. MCNPX model of LPN CIEMAT

Table 2 shows the number of reactions per neutron emitted from neutron sources of ^{241}AmBe and ^{252}Cf at LPN CIEMAT. The detector counting rate in cps can be estimated multiplying it times the intensity of the sources, taking into account their respective decay, and using the efficiency factor of ZnS(Ag) as estimated above.

The responses to both models (LMN UPM and LPN CIEMAT) for ^{241}AmBe and ^{252}Cf neutron sources are very similar; the difference depends of the respective laboratories conditions, which influence the neutron room return contribution (Vega Carrillo et al., 2007).

4. Conclusions

The intensity of neutron capture reactions in the ^{10}B as well as the neutron fluence in the detector was calculated using Monte Carlo methods for 29 monoenergetics neutrons sources, for bare and moderated detector (12+24 and 24+48 mm HDPE). Of then, the 24+48 mm moderator provided a rather constant response for the greatest range of neutron energies, and a greater number of capture reactions occurring in the sensitive areas of the detector.

The calculations for monoenergetic sources were extended to different HDPE moderators with 12 mm thickness on the sides while the front face thickness was varied from 10 to 100 mm HDPE. The greatest number of reactions for neutrons with less than 10 ^{7} MeV energy occur for the bare detector. But this configuration also has the lowest number of neutron reactions for neutron energies greater than 2E(2) MeV. As the thickness of HDPE increases the response to low energy neutrons decreases while it increases for higher energy neutrons. We conclude that

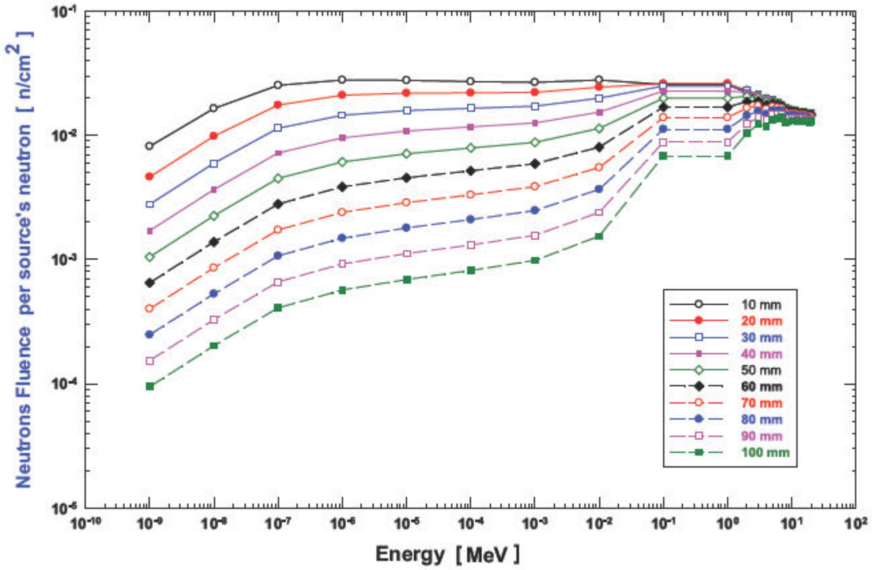

Fig. 8. N-15 detector response, neutron fluence reactions (per neutron emitted from the source) for variable moderator thickness, 12 + 10 mm to 12 + 100 mm, HDPE.

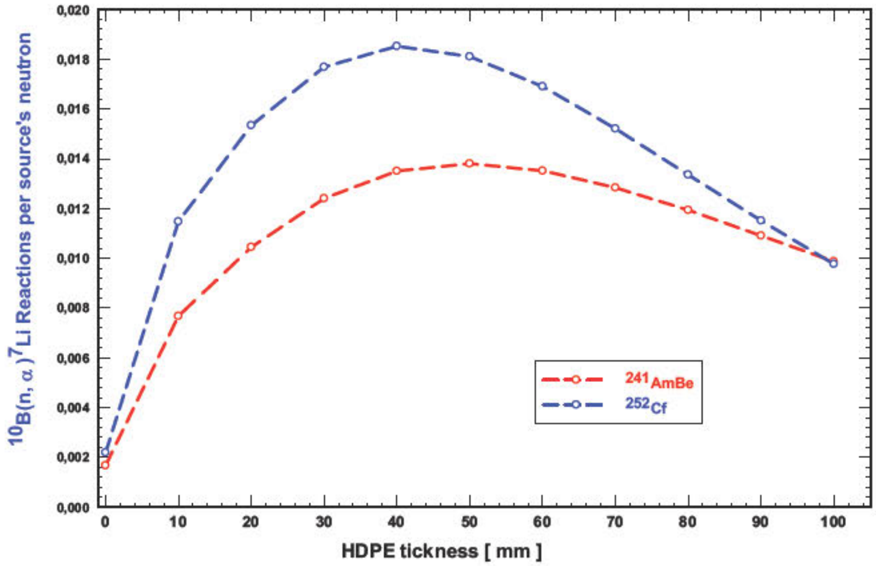

Fig. 9. N-15 detector response to ^{241}AmBe and ^{252}Cf neutron sources [$^{10}B(n,\alpha)^{7}Li$ reactions per neutron emitted from the source], for variable moderator thickness, 12 + 10 mm to 12 + 100 mm, HDPE.

thicknesses of 30 and 40 mm HDPE offers responses that are more relatively constant, over a broad range of neutron energies, indicating an optimum thickness to achieve highest reaction rates in these energy zones.

Table 1
Counting rates measured and calculated corrected by the efficiency factor for ZnS (Ag).

Distance [cm]	Measurements [cps]	MCNPX [cps]
50	208.69 ± 14.45	228.6 ± 3.20
75	100.87 ± 10.04	103.02 ± 2.14
100	57.82 ± 7.61	58.92 ± 1.61
125	39.17 ± 6.25	38.46 ± 1.30
150	27.84 ± 5.28	27.45 ± 1.10
175	21.90 ± 4.67	20.91 ± 0.96
200	17.43 ± 4.18	16.66 ± 1.07

The detector response was also simulated for neutron sources of ^{241}AmBe and ^{252}Cf, bare detector and with different thicknesses of HDPE moderators, thereby obtaining the optimal thickness to improve the detector response: 50 mm for ^{241}AmBe and 40 mm for ^{252}Cf, respectively.

Measurements and detailed MCNPX calculations were performed at the LMN UPM for validation of the model. Combining these data an efficiency factor of 0.53 ± 0.4 was determined for the ZnS(Ag).

Neutron detectors using ^{10}B + ZnS(Ag) are an interesting alternative to replace the ^{3}He detectors installed in RPMs. An improvement in geometry can increase the detection capability aiming to meet the standard of 2.5 cps/ng ^{252}Cf necessary to be considered as an alternative in RPMs (Woodring et al., 2010). Given its small size, the studied N 15 detector reaches an efficiency of 0.77 ± 0.17 cps/ng ^{252}Cf, small if used alone for RPMs. However, it can be used in backpacks for mobile neutron detection with an

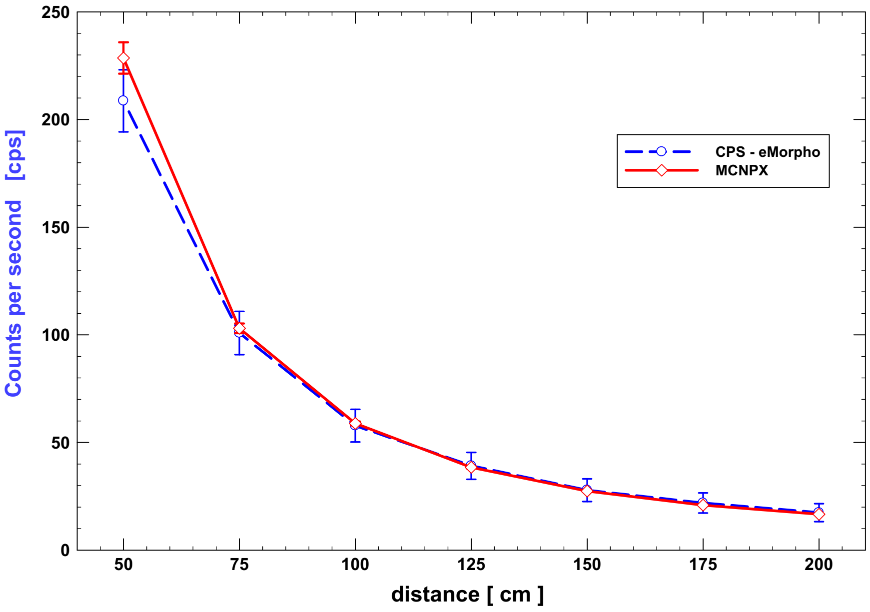

Fig. 10. Calculated and measured counting rates of the N-15A detector on the irradiation bench of LMN-UPM at different distances (^{252}Cf sources).

Table 2
Calculated ^{10}B(n,α)^{7}Li reactions of the N-15 detector for ^{252}Cf and ^{241}AmBe sources at LMN-UPM and LPN-CIEMAT irradiation benches.

Source	MCNPX LMN-UPM ^{10}B(n,α)^{7}Li/n	MCNPX LPN-CIEMAT ^{10}B(n,α)^{7}Li/n
^{252}Cf	(59.15 ± 2.0)E-05	(59.95 ± 3.3)E-05
^{241}AmBe	(42.84 ± 2.3)E-05	(47.41 ± 3.0)E-05

effective cross section to ^{252}Cf neutron source at 200 cm of 164.97 cm^2.

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