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^{10}B+ZnS(Ag) as an alternative to ^{3}He-based detectors for Radiation Portal Monitors

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Abstract. Typical radiation portal monitor systems, RPM, deployed to detect illicit trafficking of radioactive materials include a set of gamma-ray detectors and neutron detectors. Usually the employed neutron detectors are pressurized ^{3}He-based neutron detectors tubes. Due the shortage of ^{3}He reported since 2009, the amount of ^{3}He available for use in gas proportional counter neutron detectors has become limited, while the demand has significantly increased, especially for homeland security applications. For this reason, many different alternatives are being investigated for its use in RPM systems. The aim of this work is to study a scintillation detector ZnS(Ag) mixed with highly enriched ^{10}B, ^{10}B+ZnS(Ag). Using Monte Carlo methods, MCNPX code, the response of two neutron detectors based on ^{10}B+ZnS(Ag), manufactured by BridgePort Instruments LLC with different geometries, were estimated by calculating the number of ^{10}B(n,α)^{7}Li reactions for 29 monoenergetic neutron sources. Measurements and models were made, and both detectors were compared. The importance of the distance with respect to the ground was studied. The response with a ^{252}Cf moderated neutron source (0.5 cm lead and 2.5 cm polyethylene) was calculated in order to compare with other studied alternatives in the USA by Pacific National Northwest Laboratory, PNNL. With these results we conclude that neutron detectors using ^{10}B+ZnS(Ag) are an interesting alternative for replacing ^{3}He detectors. From the analysis with MCNPX we propose an improvement in the detector design.

1 Introduction

Since September 11th, 2001 the security systems in the United States of America improved significantly, adding radioactivity detection systems with neutron detectors, which until then only had gamma-ray detectors [1].

Currently the Radiation Portal Monitors (RPM) used for interception of illicit materials at borders are a set of gamma-ray detectors also including highly sensitive ^{3}He-based neutron detection systems. The detection of neutrons in the international trade of commodities is a serious concern, since a neutron signature from a vehicle may indicate the presence of SNM [2].

The main reason for having neutron detection capability is the need to detect fission neutrons from Special Nuclear Materials, SNM, like ^{239}Pu. Plutonium itself is a significant neutron source, while uranium in large quantities can be detected by its neutron signature. Since the shielding of neutrons can be difficult, neutron detection is an important means to finding SNM.

There are three basic requirements for neutron detectors for safeguards applications involving detection and measurements of SNM: 1) High absolute detection efficiency; 2) detector's low intrinsic gamma ray sensitivity; and 3) maintaining neutron detection efficiency when simultaneously is exposed to high gamma ray exposure rate. ^{3}He proportional counters clearly fulfil these requirements [3].

The use of ^{3}He as a neutron detector material has the great advantage that ^{3}He is only sensitive to neutrons and its sensitivity in proportional counters to gamma ray is negligible (pileup effects only become a problem in radiation fields of ~1 R/h). The proportional counters tubes containing ^{3}He are very simple in design, mechanically robust over a wide range of environmental conditions, and do not degrade over years of operation [4].

Sources of most concern include: complete weapons of mass destruction (WMD), improvised nuclear devices (IND); Special Nuclear Material (SNM) for weapons

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production, including plutonium and highly enriched uranium (HEU); and material or assemblies for radiological dispersal devices (RDD), usually known as "dirty bombs" [5].

The amount of nuclear material needed to manufacture a weapon of mass destruction is relatively small. In Figure 1 the radius of a sphere of bare fissile materials (Godiva) necessary to have a critical system are shown. Above 6.2 cm-radius of pure ^{239}Pu or war grade ^{239}Pu (Pu WG) becomes critical.

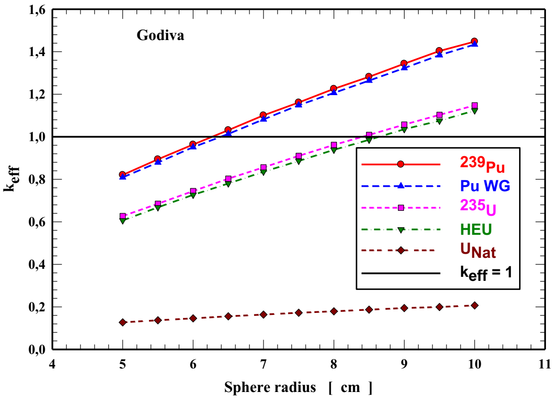

Figure 1. Effective multiplication factor of spheres of different radius with different nuclear materials (PuWG, HEU and ^{235}U).

Due the shortage of ^{3}He, reported since 2009, several efforts have been carried out to find alternative detectors for RPM, seeking similarity to those already installed with ^{3}He features [6].

The aim of this work was to study the properties of scintillation ^{10}B+ZnS(Ag) detectors to detect neutrons in RPMs. This type of detector has a scintillator ZnS(Ag), mixed with enriched ^{10}B. The neutron response of two ^{10}B+ZnS(Ag) detectors, with different geometries, were calculated using Monte Carlo methods with the MCNPX 2.7.0 code [7]. The neutron response was assessed from the number of ^{10}B(n,a)^{7}Li reactions for 29 monoenergetic neutron sources whose energies were between 10^{-9} to 20 MeV. Also, the actual neutron responses were evaluated through experimental measurements with small ^{252}Cf sources.

2 Materials and methods

2.1 Description of the N-15 and N-48 neutron detectors of ^{10}B+ZnS(Ag)

The N-48 and N-15 were manufactured by BridgePort Instruments LLC [8, 9]. Both use a mixture of ZnS(Ag) with ^{10}B, as neutron detection and screens on polymethyl methacrylate (PMMA) that guide the light pulses to a photomultiplier tube (PTM). The PTM has an embedded high voltage supply and multichannel analyzer eMorpho® digital electronics. The external size of the N-15 detector is 23 x 36 x 4 cm, and the external dimensions of the N-48 detector are 141.5 x 16.7 x 6.35 cm. Figure 2 shows the overall geometry of both detectors.

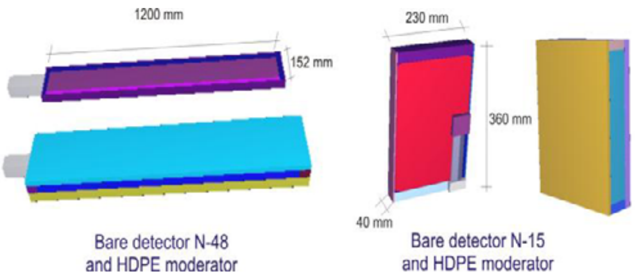

Figure 2. N-15 and N-48 geometries (MCNPX model)

In both cases, the detector's sensitive area is composed of five transparent ~ 0.017 cm-thick layers of a mixture of ZnS(Ag) and 95% ^{10}B enriched boron. These layers are arranged in four plates of PMMA sizing 23 x 36 x 0.635 cm for the N-15 and 120 x 15.2 x 0.635 cm for the N-48 detector. The PMMA function is twofold, as light guide and as neutron moderator. All is surrounded by ~8μm thick aluminium mylar as light reflector. The detector configuration is displayed in Figure 3.

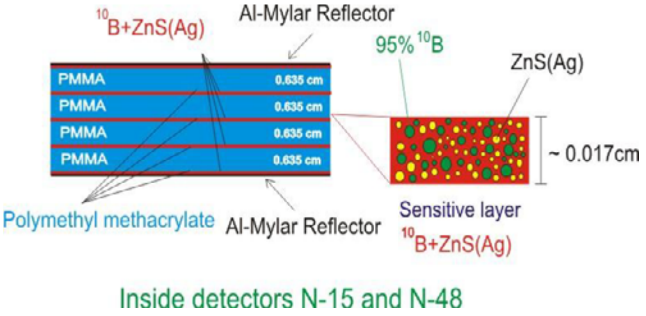

Figure 3. Internal configuration of the detectors.

Each detector has an outer moderator made of high density polyethylene (0.94 g cm^{-3}), HDPE.

For the N-15 detector the moderator thickness is 24 mm in the front, lateral faces, and top, bottom, 36 mm while 48 mm-thick in the back (24+36+48 mm). In the N-48A detector the moderator is 25 mm-thick in front, top, bottom, and lateral faces, while 50 mm-thick in the back (25+50 mm). The HDPE moderator on both detectors can be seen in Figure 4.

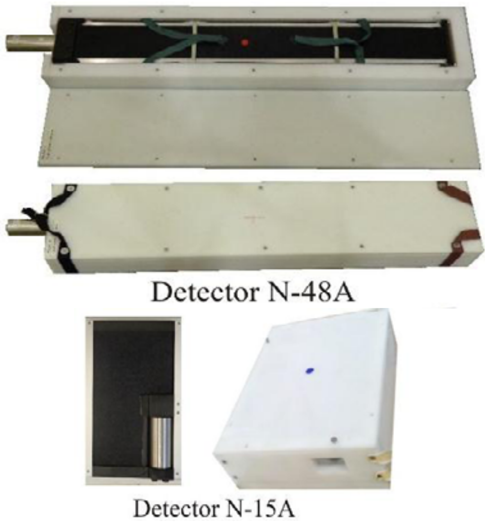

Figure 4. N-15 and N-48A detectors with HDPE moderators.

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2.2 Monte Carlo calculations

2.2.1 Detectors response

Using the MCNPX 2.7.0 code, models of both detectors were built including all the detectors details such as: the sensitive layers, the PMMA, the PMT and the moderators. Here, the PMT was modelled as an empty cylinder of glass. For each detector the response was calculated using 29 monoenergetic neutrons with energies from 10^{-9} to 20 MeV. The response was estimated for the bare detector (without moderator) and with the HDPE moderator. The response was estimated using tally f4 [10, 11] where the number of $^{10}B(n,\alpha)^{7}Li$ reactions were calculated.

2.2.2 Indoor response

The detector models were also coupled to the model of the Neutron Measurements Laboratory of the Universidad Politécnica de Madrid (LMN-UPM) [12] and the response against $^{252}Cf_{UPM}$ neutron sources to 200 cm were calculated in similar conditions to those in the experimental measurements, Figure 5.

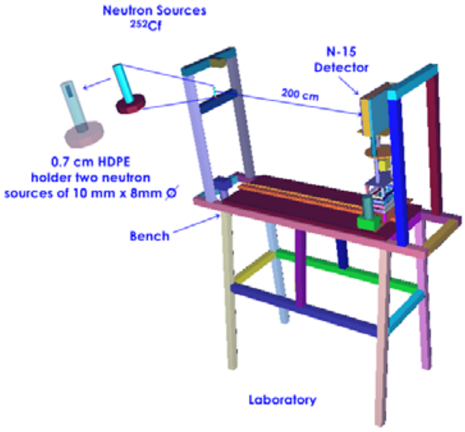

Figure 5. Configuration of the in-laboratory model (with N-15detector).

2.2.3 Outdoor response

A model was made with two N-15 detectors, in similar conditions to the outdoors measurements, without walls from a laboratory, representing normal operation conditions. Here, the detector response to neutrons produced by $^{252}Cf_{UPM}$ located 220 cm from the detector and 115 cm above the floor was calculated. In Figure 6 an scheme of the model is shown.

2.2.4 Effect on the response due to floor-to-detector distance.

Both, N-15 and the N-48 detectors, were positioned 200 cm from the $^{252}Cf_{UPM}$ source and the response was calculated varying the distance between the detector's centres and the floor, as it is shown in Figure 7. Here, the $^{252}Cf_{UPM}$ source was fixed at 100 cm above the floor. This response was in the aim to study the room-return effect.

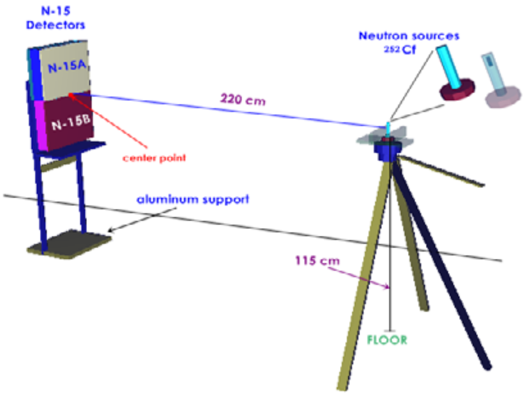

Figure 6. Configuration of the outdoors model (N-15 detectors).

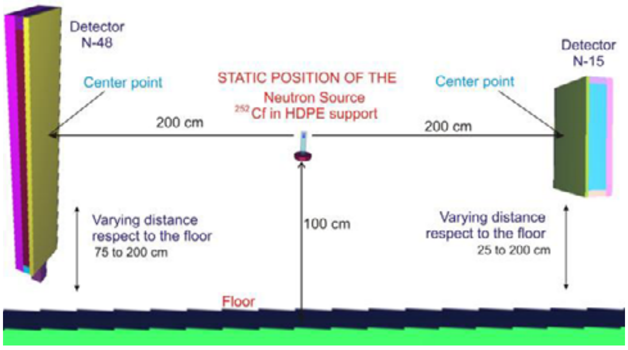

Figure 7. N-15 and N-48 outdoors (MNCPX model) (varying distances of detectors with respect to the ground).

2.2.5 Response with a moderated neutron source of ^{252}Cf

Different studies of alternatives to ^{3}He neutron detectors have been made in the USA by the Pacific Northwest National Laboratory, PNNL [13]. A ^{252}Cf neutron source was used for the tests of neutron sensors; in order to reduce the gamma-ray flux from the ^{252}Cf, the source is surrounded by at least 0.5 cm of lead, and to moderate the neutron spectrum the neutron source is surrounded with 2.5 cm of Polyethylene.

They recommended that the absolute detection efficiency for such the ^{252}Cf source, located 200 cm perpendicular to the geometric midpoint of the neutron detector sensor, shall be greater than 2.5 cps/ng of ^{252}Cf [13] and in accordance with the ANSI requirements [14].

Three different models for the N-15 neutron detector were made, using the neutron source described above (0.5 cm lead and 2.5 cm polyethylene) at a distance of 200 cm, in order to compare some previous results with others alternatives studied by PNNL.

The three different models were as shown in Figure 8, for the N-15 detector; A) an array like in Figure 6, with the only difference in the neutron source $^{252}Cf_{PNNLL}$ and at 200 cm respect to the neutron source; B) only one neutron detector, N-15 at 200 cm respect to the geometric midpoint of the detector in horizontal position, and C) one N-15 detector at 200 cm in vertical position.

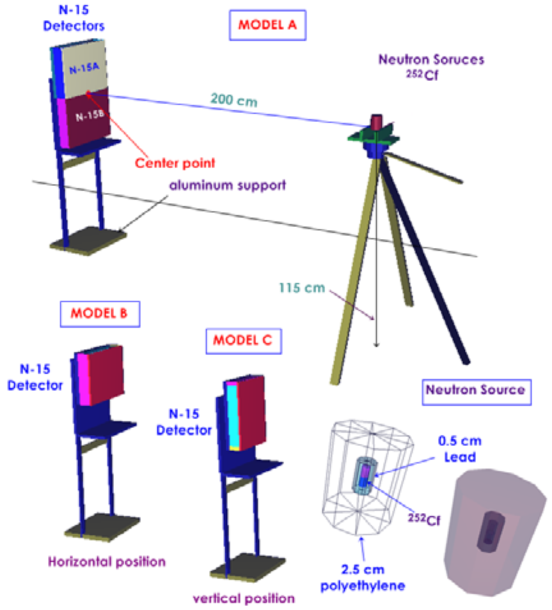

Figure 8. Configuration of the three models with different moderated neutron source conditions (2.5 cm polyethylene and 0.5 cm lead)

An extra model was made to determine the response in cps/ng of $^{252}Cf_{PNNL}$ at 200 cm in this conditions for the N-48 neutron.

In all the Monte Carlo calculations the number of histories was large enough to obtain uncertainties less than 5%. For the calculations, the cross sections were taken from the ENDF/B-VI library, where the S(α, β) treatment was included to take into account the thermalized neutron interactions [10].

2.3 Measurements

Measurements were carried out using a $^{252}Cf_{UPM}$, the experimental array was the same used in the Monte Carlo modelling. Here, the count rates per ng of ^{252}Cf was determined.

Also, measurements were carried out outside the laboratory, emulating the actual conditions of detectors in the checking points at borders, where the detectors were outside the laboratory.

The measurements were carried with two small $^{252}Cf_{UPM}$ neutron sources; these sources are double encapsulated within two steel cylinders of 1mm thickness with dimensions 10 mm x 8 mm Ø, which are moderated with 0.7 cm thickness of HDPE, 0.94 gr/cm^3. The HDPE is also the support for positioning the sources in the bench or in the irradiation point as shown in Figure 9.

For the laboratory measurements, the $^{252}Cf_{UPM}$ source was located at 200 cm from the detectors, as shown in Figure 10.

For the outdoor measurements, the $^{252}Cf_{UPM}$ source was located at 220 cm from the N-15 neutron detectors, and these were 115 cm above the floor respect to the centre point, Figure 11.

Figure 9. Neutron sources of Laboratory of the Universidad Poltecnica of Madrid, UPM.

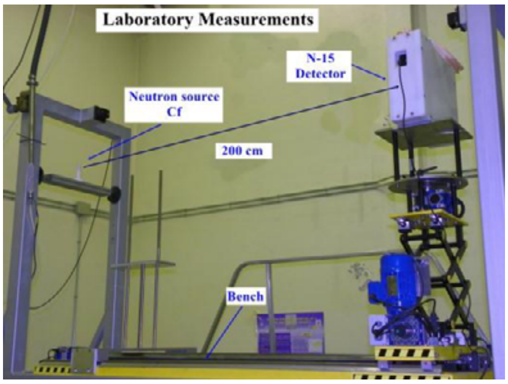

Figure 10. Indoor measurements with the N-15 detector at the LMN-UPM facility.

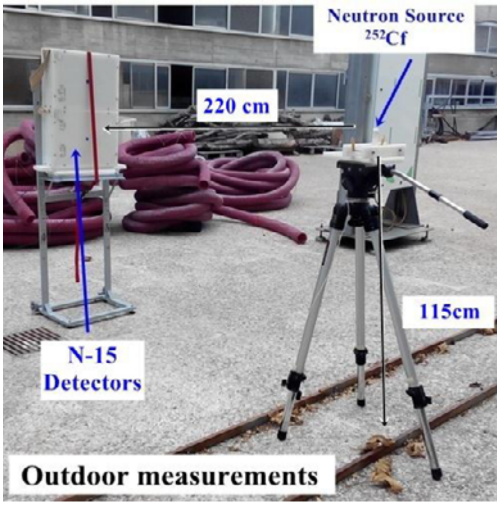

Figure 11. outdoor measurements with the N-15 detector

3 Results

3.1. Calculated responses

Figure 12 shows the response of both detectors, bare and with the HDPE moderators. Responses are the amount of $^{10}B(n,\alpha)^7Li$ reactions per neutron from the source, in a range of monoenergetic neutrons from 10^{-9} to 20MeV.

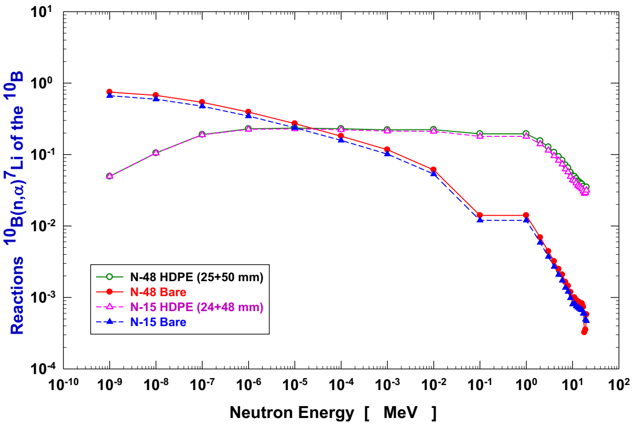

Figure 12. N-48 and N-15 detectors response, $^{10}B(n,\alpha)^7Li$ reactions per neutron emitted from the source

Both bare detectors have approximately the same response, being slightly larger for the N-48 detector. Without moderator, both detectors have a large response for low energy neutrons decreasing for larger energy neutrons. When the moderators are included both detectors have roughly the same response, being smaller for low energy neutrons (<0.1 eV) and when the energy is > 1 MeV. From 0.1 eV to 1 MeV the response is almost flat, being almost independent from neutron energy.

3.2. Detection capability in cps/ng ^{252}Cf to 200 cm and 220 cm

Table 1 shows the calculated and measured count rates per ng of $^{252}Cf_{UPM}$ at 200 cm indoors for both neutron detectors, N-15 and N-48, and outdoors at 220 cm, N-15 detectors. The calculated values were scaled up to neutron source strength, where 1 ng of ^{252}Cf produces 2340 n/s [15].

Table 1. Count rate per ng of ^{252}Cf.

Detector	**Indoors**	
	Measurements [cps/ng ^{252}Cf]	**MCNPX [cps/ng ^{252}Cf]**
N-48 **[200 cm]**	1.77± 0.10 [7]	1.76 ± 0.01
N-15 **[200 cm]**	0.76 ± 0.20 [15]	0.73 ± 0.01
	Outdoor	
N-15 **[220 cm]**	0.61 ± 0.20	0.68 ± 0.02

Measured and calculated count rates were corrected by the detector efficiencies that were previously determined [7, 16]. For both conditions, the measured results are in good agreement with the MCNPX calculations. By comparing the indoor and outdoor results for the N-15 detector, the differences are not relevant. Comparing both detectors under indoor conditions the detector N-48 show a better response per ng of ^{252}Cf, probably due to the detector size.

3.3. Detection capability in cps/ng ^{252}Cf to 200 cm, varying the position respect to the ground

Table 2 shows the calculated detectors response, in cps per ng of ^{252}Cf, under outdoor conditions with the centre of both detectors located at varying distance from the floor ($^{252}Cf_{UPM}$). Uncertainties of these results are only those from the Monte Carlo calculations.

Table 2. Importance of the position of detectors above ground for outdoor measurements at 200 cm from the ^{252}Cf source (MCNPX calculations).

Distance above ground (cm)	**Detector N-48 cps/ng $^{252}Cf_{UPM}$**	**Detector N-15 cps/ng $^{252}Cf_{UPM}$**
200	1.16 ± 0.06	0.57 ± 0.04
175	1.33 ± 0.06	0.66 ± 0.04
150	1.48 ± 0.07	0.70 ± 0.04
125	1.60 ± 0.07	0.81 ± 0.05
100	1.67 ± 0.07	0.85 ± 0.05
75	1.70 ± 0.07	0.86 ± 0.05
50	-	0.83 ± 0.05
25	-	0.78 ± 0.04

The detectors' efficiency varies depending on the position with respect to the floor. Due to the size of N-48 detector, the minimum allowable distance between the centre of the detector and the floor was 75 cm. In all cases, the count rate of N-48 detector is larger than the count rate of N-15, due to the detector size. Nevertheless, the source-to-detector's centres distance is the same and both detectors reach the largest count rate when their centres are allocated at 75 cm above the floor, probably due the amount of scattered neutrons from the floor.

The efficiency of both detectors changes with their position with respect to the floor. Therefore, it is concluded that the use of these detectors in actual RPM should be characterized in situ according to their position above ground, thereby selecting the best efficiency in each case in situ; that is, to calibrate the detector.

3.4. Detection capability in cps/ng ^{252}Cf to 200 cm with a moderated neutron source

Table 3 shows the calculated count rates per ng of ^{252}Cf at 200 cm for outdoors conditions, in three different models for the N-15 (Figure 8) neutron detector and one for the N-48, with a moderated neutron source with 2.5 cm of HDPE and gamma-shielded with 0.5 cm thickness of lead, $^{252}Cf_{PNNL}$. The calculated values were scaled up to neutron source strength, where 1 ng of ^{252}Cf produces 2,340 n/s [15].

The N-15 detectors response with the UPM sources is ~ 0.81± 0.05 cps/ng $^{252}Cf_{UPM}$ and for the model neutron source similar PNNL sources is ~ 0.93 ± 0 .04 cps/ng ^{252}Cf$_{\text{PNNL}}$

Table 3. Count rate per ng of ^{252}Cf at 200 cm (MCNPX calculations, with the ^{252}Cf$_{\text{PNLL}}$).

	Two detectors	
Model	**N-15A [cps/ng ^{252}Cf] ^{252}Cf$_{\text{PNLL}}$**	**N-15B [cps/ng ^{252}Cf] ^{252}Cf$_{\text{PNLL}}$**
Model A	0.90 ± 0.05	0.94 ± 0.05
	N-48A [cps-ng^{252}Cf] ^{252}Cf$_{\text{PNLL}}$	
Model N-48	1.71 ± 0.07	

The N-48 detector response with UPM sources is ~ 1.60 ± 0.07 cps/ng ^{252}Cf$_{\text{UPM}}$ and for the model neutron source similar PNNL sources is ~ 1.71 ± 0 .04 cps/ng ^{252}Cf$_{\text{PNNL}}$. These results correspond to the neutron detectors in outdoor conditions and the same distance above floor in order to compare with other alternatives previously studied by the PNNL.

4 Conclusions

Using Monte Carlo methods the response functions of two different ^{10}B+ZnS(Ag) detectors were calculated, as well their performance against ^{252}Cf neutrons. Measurements were carried out indoors under laboratory conditions and outdoors emulating their use in border checking points.

The response increases significantly in efficiency for both detectors, when calculation is based on the PNNL neutron source ^{252}Cf$_{\text{PNNL}}$ configuration, probably due to the larger moderation of the source which increases the thermal neutron flux, thus obtaining a better efficiency than that reported with the sources used in UPM.

The ^{10}B+ZnS(Ag) detectors are an alternative to replace ^{3}He detectors in RPMs. The N-15 detectors are considered suitable for portable backpack detectors. The N-48 detector is close to be considered a replacement for ^{3}He detectors. An improvement in the geometry of the detector increasing the amount of ^{10}B could increase the detector efficiency aiming to reach 2.5 cps/ng ^{252}Cf, defined as a goal to use this type of detectors as an alternative in RPMs [13].

Regardless the type of detector, the use of moderator allows to have a flat response in a wider energy range.

The position of the detector with respect to the ground is an important feature due to the detector response to scattered neutrons [17].

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