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Boron-10 lined proportional counter development for thermal neutron detection

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Abstract

Purpose Neutrons detection is of paramount importance in nuclear physics experiments, reactor physics, monitoring and control of nuclear reactors and in radiation protection. Among the neutron detectors used are Proportional Counters (PC to BF_3, boron deposition, ^3He), Our Detection and Measurements laboratory has embarked on a project to develop, design and build proportional counters with boron deposition mainly for gas filling, several prototypes of which have been produced.

Method This work presents the Boron-10 Lined Proportional Counters (PC), developed and produced in the Detection and Measurements Department (DDM19). The PC was designed by adopting, for the electrodes, a stainless steel cathode 0.5 mm thick and a gold-plated tungsten anode with diameter of 25 μm. The detectors are filled with an $Ar-CH_4$ mixture (10%) at a pressure of 20 cmHg. The boron deposition is performed by the technique of electrophoresis.

Result The results obtained using the DDM19 prototype PC (Detector produced in our Detection and Measurement laboratory, 2019), show the advantage of electrophoresis as a better boron deposition technique, and the limitation of the surface mass of ^{10}B to optimal values is around 0.5 at 0.6 mg/cm^2.

Conclusions The characteristics of the gas amplification in the mixture ($Ar-CH_4$ (CH_4 10%)), were measured by different methods and compared with the values of the proportional reference counter LND232. The counting plates for the two reference meters LND232 and our DDM19 prototype from 200 to 300 V have slopes ranging from 6.5 to 10%/100 V, and gas amplification factors from 1436 to 1380, respectively.

Keywords Neutron detector · Boron-10 lined proportional counter · Reference LND232 · Boron deposition technique

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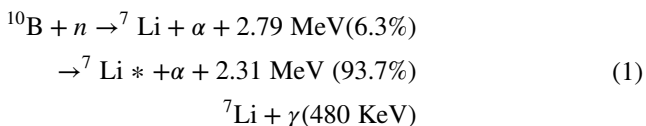
Introduction

Neutrons are relatively massive particles that make up the nucleus with essentially the same mass as the proton. As they are not electrically charged, they do not react with the electrons of atoms and can only be detected through their interaction with a converter material during which charged particles are produced and detected.

These ionizing secondary particles will degrade their energy in the counter by interaction the electrons located near their orbits, based on the medium crossed, it produces pairs of ions in a gas; then they collected as an electrical signal. The famous reactions used in neutron detectors are: the ^{10}B (n, α) ^7Li, which interact in proportional counters of BF_3, proportional counters with boron deposition and ionization chambers. In pulse mode neutron detectors, the count rate is proportional to the neutron flux, where the discrimination of

gamma radiation takes place in pulse amplitude, generally, they operate from 10 to 10^6 c/s [1–9].

The sensitivity of a neutron detector depends on the longitudinal section with the converter material, on the quantity actually used and on the way in which the reaction is operated in the detector. The detectors operating in pulse mode are generally more sensitive to neutrons than ionization chambers with direct collection of average electric current. In this work, several processes have been developed (boron deposition, degassing and filling, etc.), all the electrical and nuclear tests were carried out in the DDM and sub-critical reactor laboratory AURES laboratory, their characteristics were noted and compared with those of the LND232 reference detector. In the nuclear detectors designing process, many criteria and operating requirements should be taken into account, among them: the type of thermal neutron flux in the range of 0.1 to 10^6 n/cm^2 s of radiation to be measured, the range of flux or power, maximum temperature and relative temperature variations, the response time required for control and safety, the size of the detector (to avoid disturbance of flow in the reactor), resistance to shocks, vibrations and humidity; radiation resistance, the precision required for the measurement and the reproducibility of the measurements, Cost, availability and other economic factors [10, 11].

In this work, the thermal neutron capture reaction by the ^{10}B isotope; was taken into consideration the most used conversion reaction to thermal neutron, as well known, the cross section of this capture is around 4010 barns, the reaction proceeds according as the following [1]:

$$^{10}B + n \rightarrow ^7Li + \alpha + 2.79 \text{ MeV}(6.3\%)$$
$$\rightarrow ^7Li * + \alpha + 2.31 \text{ MeV } (93.7\%) \quad (1)$$
$$^7Li + \gamma (480 \text{ KeV})$$

The rules of energy conservation and momentum cause an energy distribution $E_\alpha = 1.47$ MeV and $E_{Li} = 0.84$ MeV in the most probable mode (93.7%) [11]. Note that boron 10 only constitutes barely 18.8% of natural boron [12]; however, it has the advantage of not being toxic or radioactive. It is separated from natural boron by very complicated methods and techniques, among these techniques the fractional distillation of an etherate of boron trifluoride to obtain enrichments of about 95% in ^{10}B. It is used either in solid form in a crystalline state or in amorphous powder with a particle size of the order of a micron.
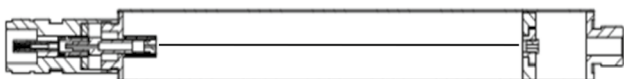

Fig. 1 Sectional view of PC with boron deposition designed

Materials and methods

Technology adopted

The *DDM19*/1 prototype counter, produced by our Detection and Measurement Laboratory was designed by using gold-plated tungsten anode, (electrodes, a 0.5 mm thick stainless steel cathode and a 25 μm diameter) [13]. The detectors are filled with an Ar-CH_4 mixture (10%) at a pressure of 20 cm Hg, after a long degassing under secondary vacuum at a temperature of 80 °C [11, 14]. The deposit of natural boron with ^{10}B at 18.8% was carried out by the electrophoresis technique. The thickness of the deposit is 0.6 to 0.5 mg/cm^2 [10, 11, 14]. Figure 1 shows a sectional view of PC with boron deposition designed, Figs. 2, 3 and 4 show the different stages of the realization of our prototype PC.

Boron deposition by electrophoresis

The choice of electrophoresis was done due its advantages compared to the other methods, the most important advantages of deposition by electrophoresis are the quality of the deposit (uniformity and adhesion), ease of implementation,
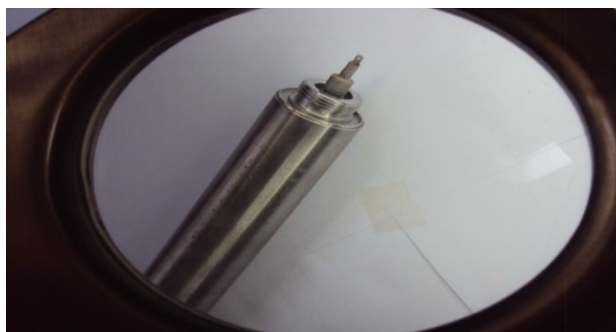

Fig. 2 Photo of PCDB-DDM19 signal part

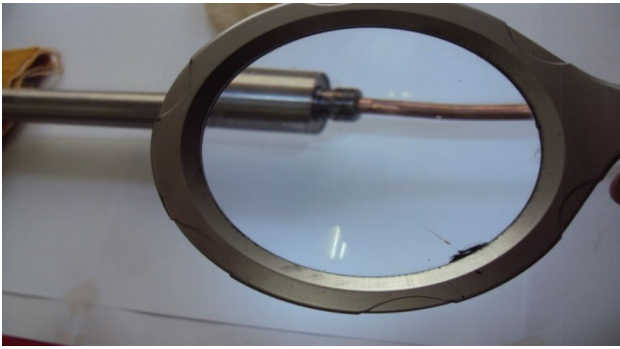**Fig. 3** Photo of PCDB-DDM19 signal and filling tubes part

Fig. 4 Photo of PCDB-DDM19 part of filling tubes

reproducibility, high deposit rate and low product consumption [4, 5].

Experimental protocol of electrophoresis

Figure 5 shows a general diagram of the experimental device for depositing boron by electrophoresis; this device includes (high voltage–power supply, a stainless steel or aluminum cathode $\Phi = 2.44$ cm and $L = 15$ cm, a stainless steel or copper anode (rod). The chemicals used in this experiment are natural amorphous boron (from LABOSI), distilled isopropyl alcohol, pure crystallized magnesium chloride (Mgcl$_2$) and pure methanol (CH$_3$OH), nitric acid (HNO$_3$), acetone (C$_3$H$_6$O) were used for the cleaning, degreasing and stripping. The quantity of boron was calculated taking into account the thickness of the deposit (surface density), the surface of the cathode (internal surface of the tube) and the loss of a certain quantity of boron (approximately 20%). Otherwise, the coated boron weight can also be estimated
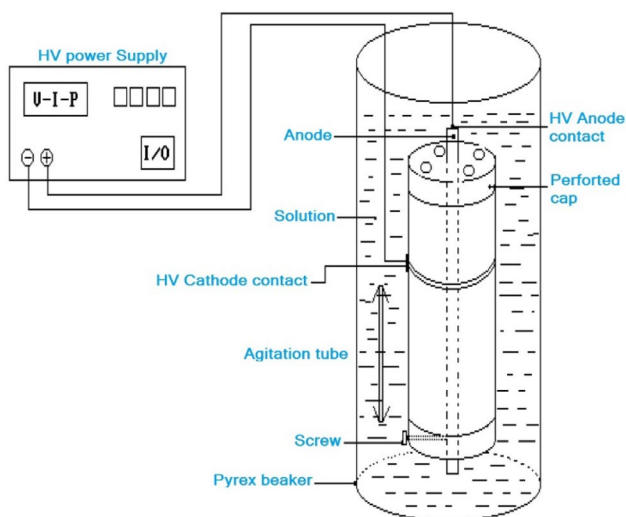
by weighing the coated samples before and after coating (Fig. 6). 0.5 mg/cm^2 of boron surface mass was taking because the best sensitivity, good quality of spectrum with boron deposition are obtained with this thickness [7, 9], the mass of boron is calculated as follows:

$$m_B = m_s \times S(1 + 0.20) \tag{2}$$

where m_B: mass of boron, S: the internal surface of the tube, m_s: mass per unit area.

The boron 10 should be diluted to 0.3 mg/ml; the volume of isopropyl alcohol should be mB/0.3, because MgCl$_2$ acts as the source of electrical charge for the boron particles, for this, 23 ppm MgCl$_2$ is needed in the solution. The methanol (CH$_3$OH) must have 0.1% from the boron mass. In this work, we have followed the next procedure, firstly, we weighing the mass of boron mB and the mass of Mgcl$_2$, then measure the volume of methanol and the volume of isopropyl alcohol, after that mix Mgcl$_2$ with methanol. Add this mixture to the boron and mix in the mortar. Isopropyl alcohol is added in a test tube; it should be able to contain the electrophoresis solution and the electrodes with the possibility of the stirring. Put the contents of the mortar in the test tube and then rinse the mortar with isopropyl alcohol. After that mount the two perforated caps on the electrodes and connect the electric wires, finally, place the electrodes in the test tube. At the end of this experiment, we have obtained the tube with a thin layer of boron deposit (Fig. 6).

Experimental

The electrical and nuclear tests of the PC prototype and reference were carried out under ideal conditions, corresponding to low-intensity neutron flux, room temperature, without vibrations, shocks and electromagnetic disturbances, these conditions are obtained in our sub-critical reactor laboratory (AURES), using a source of ^{241}Am-Be (2Ci) (Fig. 7) [4].

Fig. 5 Diagram of the experimental device of the Boron deposition by electrophoresis

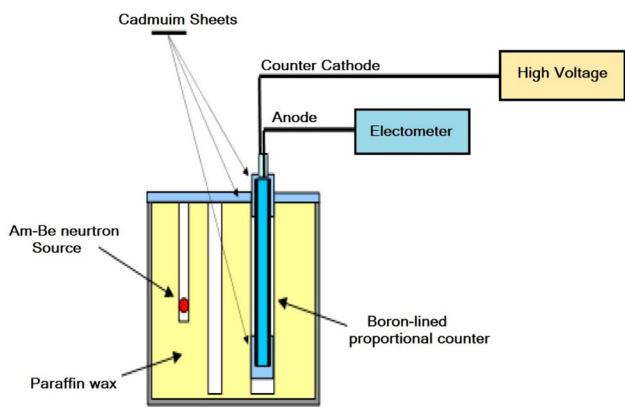**Fig. 6** Thin layer of boron deposit inside the tube of detector

Fig. 7 Experimental device for nuclear measurements in pulse and current mode

The high-performance electronic pulse measurement chain consists of a 2006E Canberra type charge preamplifier (PA), selected for its variable gain (1 or 5), its low noise and its speed (Rise Time < 20 ns). The search amplifier used (A450 EGG) helped to optimize the formatting constant suitable for this type of experience. The discrimination and plateau curves are deduced using a single-channel analyzer type SCA 550 EGG. The Canberra Dual Counter Timer 2070 type counting scale and a multi-channel analyzer (MCA 35 + Canberra) are used to collect energy spectra. These are systematically transferred to a computer for processing and analysis. Two high voltage precision modules (TENNELEC) were used for the polarization of the counters. The PC-PA connecting cable has a length of around 70 cm. The signal, high voltage and test cables, connecting the PA to the rest of the chain, installed in the AURES control room (Table 1).

The data obtained for the DDM/19 prototype proportional counter show very interesting results compared to the reference LND232. Figures 8 and 9 show the evolution of plateau curves and discrimination plateau of CP with boron deposition prototype DDM19. The counting plateau at $S = 200$ mv is 300 V and a slope of 6.53%/100 V (400–"700 V) therefore the plateau is located between 400 and 700 V of length 300 V, for the reference detector the slope is 5%/100 V < 10% 100 V. For the discrimination curves, two curves were recorded, one at 700 V for the prototype counter and the other at 750 V for the reference counter, on the first and second, a discrimination level does not appear, the discrimination threshold could be set at 200 mv for the two prototype and the reference LND232 detectors.

Distribution of energy pulses in PCBD

In this work, the average of recoil particles in solid boron ^{10}B (density = 2.45 g/cm^3), were 0.88 and 0.47 mg/cm^2 for α ($E_\alpha = 1.49$ MeV) and ^{7}Li ($E_{Li} = 0.85$ MeV), respectively,

Table 1 Characteristics of boron deposition proportional counters DDM 1″/19 and LND 232

Characteristics	PC DDM 1″/19	PC LND 232
Diameter (cm)	2.54	2.54
Total length (cm)	35	20
Useful length (cm)	20	10
Sensitive area (cm^2)	154	77
Deposition Thickness (mg/cm^2)	0.5	–
Sensitive material	Natural boron	B10
Cathode	SS	1100 Al
Anode (μm)	W (50)	W (25)
Insulating	Al$_2$O$_3$+PTFE	Al$_2$O$_3$
Gas (Pressure: 20 cmHg)	Ar-CH$_4$ (10%)	Ar-CO$_2$ (5%)
Capacity (pF)	4.6	4
Plateau (V)	300	200
Slope: %/100 V	6.5	10
Insulation resistance (*)	>10E +12	>10E + 14

noted that, the optimal thickness for better conversion of α and ^{7}Li particles were found in the range of 0.40–8 mg/cm^2 [5, 7].

From Fig. 10 we noticed that proportional counter PC DDM19 presents a better quality of energy spectrum compared to the standard counter (LND232), Fig. 11 represents the Amplitude displayed by the oscilloscope of PC DDM19.

Gas amplification

The gas amplification is measured first by the technique in current mode I (V), the measurement of the gas amplification by I (V)/I_s was carried out by polarizing the detector negatively in 10 V steps on the cathode of the PC and by measuring on the anode, the current was generated using a
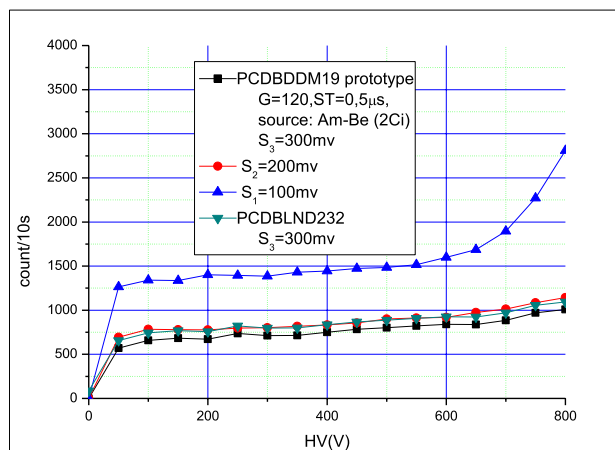

Fig. 8 Plateau curves of the prototype boron deposition DDM19 and LND232 at different thresholds
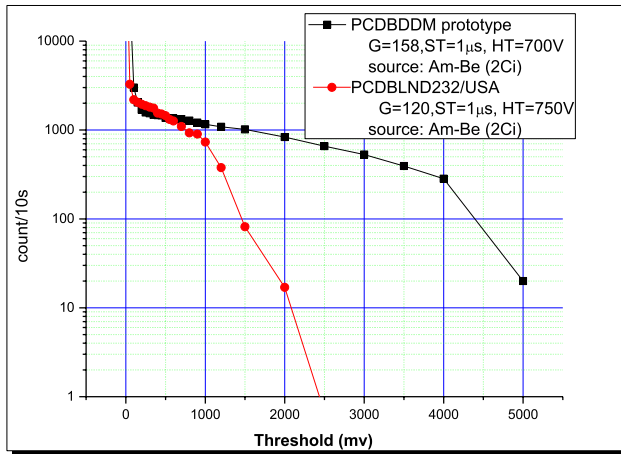
Fig. 9 Discrimination threshold plateau of the prototype DDM19 PC with boron deposition and PCDB LND232
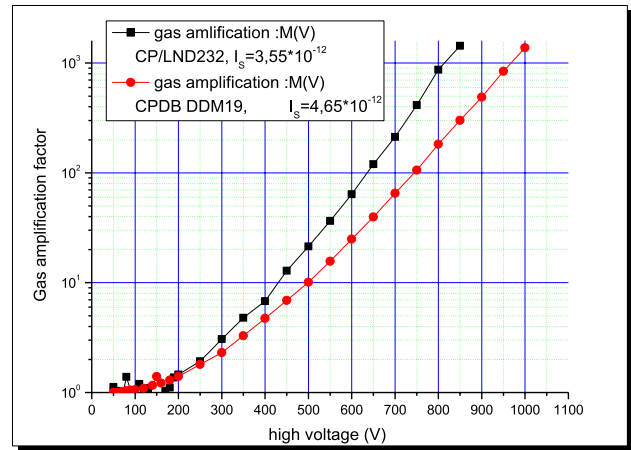

Fig. 10 Energy spectrum of PC with prototype boron deposition and PC LND232/USA

Fig. 11 Amplitude displayed by the oscilloscope (GWIMSTEK) of PC with boron deposition DDM19

programmable digital electrometer (Keithley617). The precision measurement of the gas amplification obtained during the digital acquisition of the currents at different voltages

Fig. 12 Curves gaseous amplification in proportional counters to deposit boron prototype and reference

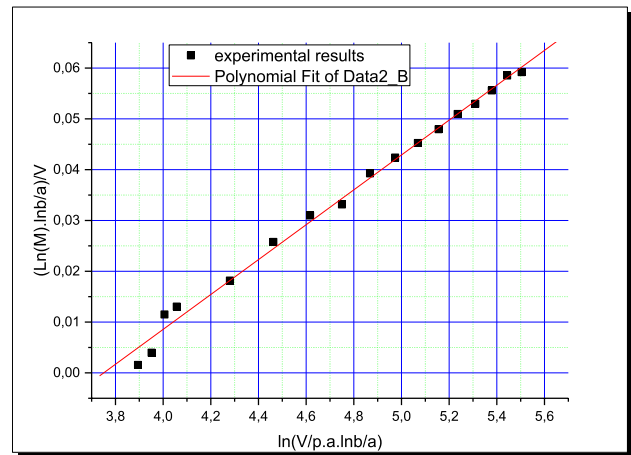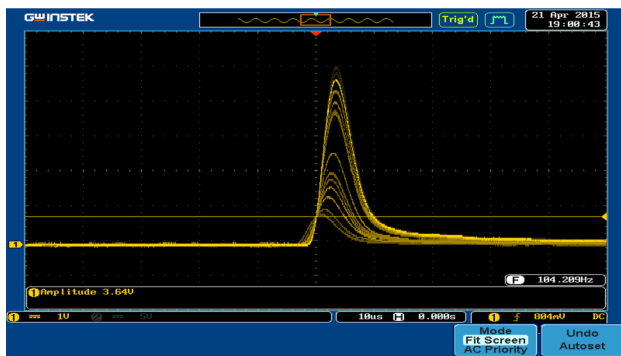**Fig. 13** Analyses of the experimental points according to the formula of DIETHORN Formula of the PCDB LND232

was excellent; the reproducibility reached during the realization of the PCDB DDM19, was evaluated by the PCDB LND232. Figure 12 shows the results of measuring the characteristics of the gas amplification of two detectors (DDM19 and LND232 according to the formulations of DIETHORN [6] and SHALEV & HOPSTONE [2] (Figs. 13, 14).

Table 2 gives a comparison of the characteristics of the two proportional counters (PCs), measured for amplification factors ranging from 1 to 1436 (LND232 detector), and from 2 to 1380 for (DDM19 detector). The characteristic (ΔV) of the formulation of DIETHORN represents the potential difference allowing an electron to cover an average path [6], it must be close to the necessary energy required to produce a pair of ions. K represents the critical value of the field strength per unit of pressure (E/p). The comparison of the characteristics (ΔV, K) of DIETHORN [6] and (A6, B6) of SHALEV et al., shows the excellent reproducibility between
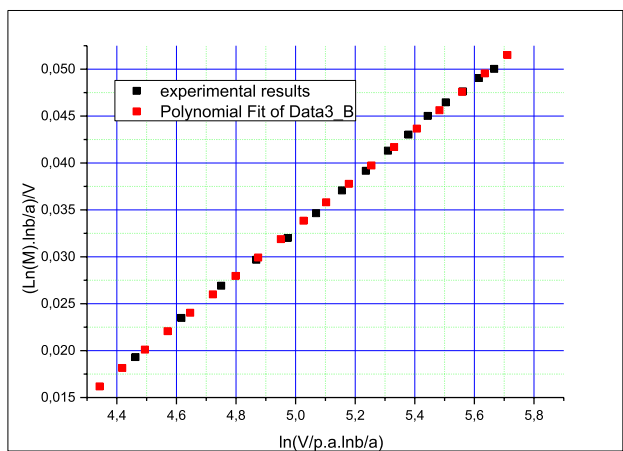
Fig. 14 Analysis of the experimental points according to the formula of SHALEV Formula of the PCDB DDM19

the two counters LND232 and our prototype DDM19 (Figs. 12, 14) (Table 3).

The gradient of the gas multiplication curve (dM/dV) is inversely proportional to A_6 and high values of this term would mean a slow increase in gas multiplication, as a function of the anode voltage [3], compared to PC (90% Ar + 10% CH$_4$) for example (A_6 and B_6 is 110, 2.8, respectively) [3]. Figure 12 represents the evolution of the gas amplification factor as a function of high voltage.

Discussion

We can conclude from the general results obtained for PCDB-DDM19 (terms of plateau curves, neutron-noise signal, discrimination curves and spectral energy quality), the feasibility of producing this type of detector in our laboratories. The discrimination characteristic allows the appropriate choice of the threshold compared with the reference detector $S = 300$ mv, in order to obtain a good signal-to-noise ratio without sensitivity losing (Figs. 6 and 7). The energy spectra measured show a low thermal neutron flux of natural boron, despite the low counting statistics compared to the reference detector, for this reason, it is necessary to increase the useful surface of the detector. The rise times of the pulses delivered by the PCDB-DDM19 were estimated at 200 ns, the discrimination curves obtained using the PCDB-DDM19 and for the standard detector are similar (Fig. 7), the energy spectrum presents a good quality compared to the spectra of the PC LND232. The optimal product (pressure-inter-electrode distance) was determined to be approximately 33.86 K Pascal cm. The Ar-CH4 (10%) or P10 mixture, showed some superiority in terms of charge collection speed, compared to the mixtures (Ar-N$_2$ (4%), Ar-CO$_2$ (10%) [7, 10], the optimal conditions determined correspond to a filling pressure of Ar-CH$_4$ (10%) or Ar-N$_2$ (4%), of 26.66 K Pascal (200 torr), a bias voltage of 700 V, collection of charges is then less than 200 ns.

Table 2 Characteristics of the gas of PCDB DDM19 and LND232 according to the formulations of DIETHORN

Counter type	ΔV	DIETHORN formulation	
		K(V/cm mm Hg)	Gas gain range validity
LND232 (our result)	20.19 ± 0.47	43.27 ± 2.99	170 ($M = 1.03$) 850 ($M = 1436.5$)
Gas (75%Ar + 15%Xe + 10% CO$_2$)	20.2 [4]	37.5 [4]	
DDM$_{19}$ (90%Ar + 10%CH$_4$) (our result)	26.85 ± 0.53	41.73 ± 2.45	300 ($M = 2.3$) 1000 ($M = 1380$)
PC (90%Ar + 10%CH$_4$)	27.9 [4]	44.9 [4]	$10 < M < 10^5$
PC (90%He + 10%CH$_4$)	26.01 [4]	32.4 [4]	$1.5 < M < 10^6$

Table 3 Characteristics of the gas from PCDB DDM19 and LND232 according to the formulations of SHALEV et al., measured in the AURES laboratory

Counter type	Formulation of SHALEV and al		
	A6 (V/cm mmHg)	B6 (V)	Gas gain range validity
LND232/USA (our result)	206 ± 1.4	2.5 ± 1.99	170 ($M = 1.03$) 850 ($M = 1436.5$)
S. Shalev	180 [3]	2.7 [3]	
DDM19(90%Ar + 10%CH$_4$) (our result)	207.3 ± 1.3	2.05 ± 2.6	300 ($M = 2.3$) 1000 ($M = 1380$)
PC(90%Ar + 10%CH$_4$)	110 [3]	2.8 [3]	$10 < M < 1$ Ã— 10^5

Conclusions

The quality of the results obtained in this work allowed us to confirm that our designed detectors meeting the quality criteria required by the control and monitoring requirements of nuclear reactors, and on the other hand, the possibility of its development. The characterization work on prototype detectors was done under high neutron flux, in the laboratory of the sub-critical assembly, we have a source of ^{241}Am-Be neutron with an activity of 2Ci, it is considered as a mixed neutron-gamma field, this is for the confirmation of the absence of influence of gamma radiation effect on the performance of these prototype counters. All this for the aim to obtain better accuracy in the evaluation of the characteristics. In the future, we will be working on producing other prototypes (deposit of 90% enriched boron) and improving the sensitivity of PCDB prototypes, ensuring reproducibility in the technological realization of this type of detector and the reliability of its response in actual operating conditions, parallel to that, launching of program continuity intended for the development of meters operating under working conditions (temperature, humidity, vibration, etc.) under intense gamma flux and provided with integrated cables with mineral isolation and high noise immunity.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

References

1. G.F. Knoll, *Radiation Detection and Measurement* (Wiley, New York, 2010)
2. A. Williams, R.I. Sara, Parameters affecting the resolution of a proportional counter. Int. J. Appl. Radiat. Isot. **13**, 229–238 (1962)
3. S. Shalev, P. Hopstone, Empirical expressions for gas multiplication in 3He proportional counters. Nucl. Instrum. Methods **155**, 237–247 (1978)
4. M. Baaliouamer, C. Belaragueb, D. Mazed, Gas gain measurement in helium-isobutane mixtures filled-proportional counters. Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. **382**, 490–494 (1996)
5. D. Mazed, Experimental gas amplification study in boron-lined proportional counters for neutron detection. Radiat. Meas. **42**, 245–250 (2007)
6. W. Diethorn, *A Methane Proportional Counter System for Natural Radiocarbon Measurements* (United States Atomic Energy Commission, Technical Information Service, Washington, 1956)
7. P.M. Dighe, D.N. Prasad, K.R. Prasad, S.K. Kataria, S.N. Athavale, A.L. Pappachan, A.K. Grover, Boron-lined proportional counters with improved neutron sensitivity. Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. **496**, 154–161 (2003)
8. P.M. Dighe, D. Das, D.N. Prasad, L.P. Kamble, C.P. Nagaraj, R.K. Kaushik, S. Sarkar, S.S. Taliyan, P.P. Selvam, K. Binoy, Design and development of high temperature 10B coated proportional counters for PFBR. BARC Newsl. **2015**, 22–32 (2015)
9. P.M. Dighe, D. Das, Performance studies of boron lined proportional counters for reactor applications. Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. **770**, 29–35 (2015)
10. C. Guo-Yun, W. Zhi-Yong, X. Yong, L. Sheng-Jie, H. Fu-Cheng, H. San-Bo, Z. Li, Z. Jing-Wu, M. Jia-Yi, A boron-lined proportional counter (BLPC) used in a mixed field of reactors. Chin. Phys. C **36**, 434 (2012)
11. K.S. McKinny, T.R. Anderson, N.H. Johnson, Optimization of coating in boron-10 lined proportional counters. IEEE Trans. Nucl. Sci. **60**, 860–863 (2012)
12. G.F. Knoll, *Radiation Detection and Measurement* (Wiley, New York, 1979)
13. M. Fares, A. Messai, S. Begaa, M. Messaoudi, K. Negara, M.Y. Debili, Design and study of the characteristics of a versatile ionization chamber for gamma-ray dosimetry. J. Radioanal. Nucl. Chem. **326**, 1405–1411 (2020). https://doi.org/10.1007/s10967-020-07446-5
14. P.M. Dighe, D.N. Prasad, K.V. Rao, C.K. Pithawa, R.K. Patil, Anode wire mounting technique for high temperature boron-10 lined proportional counter. Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. **621**, 713–715 (2010)