

## Fine Powder Proportional Counters for Neutron Detection

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### Abstract

A novel neutron detection approach is presented, consisting of a neutron sensitive aerosol obtained by dispersing B<sub>4</sub>C fine powder in a gaseous proportional counter operating in continuous flow, which proved to effectively detect thermal neutrons. This technique shows potential to be an affordable alternative to <sup>3</sup>He neutron detectors, providing an additional mechanism to overcome the <sup>3</sup>He shortage crisis that compromises the deployment and maintenance of neutron detectors in research facilities and security applications. The effect of the B<sub>4</sub>C nanoparticles in the charge gain and energy resolution of a proportional counter filled with Ar-CH<sub>4</sub> (90%-10%), by irradiation with x-rays from a <sup>55</sup>Fe source shown an average gain loss of 36% and increase in energy resolution (FWHM) by 15% (absolute value). Results obtained by thermal neutron irradiation are also presented, showing neutron captures in which the simultaneous energy deposition of both reaction products from the <sup>10</sup>B neutron capture reaction was achieved.

**Author Keywords.** Neutron Detection, <sup>3</sup>He Alternative, Proportional Counter, Fine Powder.

**Type:** Research Article

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### 1. Introduction

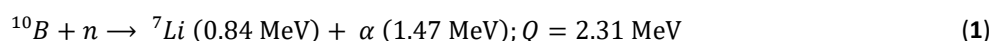
The use of proportional counters as radiation detectors is a massively deployed and well-known technology, introduced in the late 1940s (Knoll 2010). In its simplest geometry, a proportional counter is a gas-filled cylinder with a thin wire on its centre to which a high voltage is applied, resulting in an electric field established between the wire (anode) and the detector walls (cathode). When ionizing radiation passes through the gas, it interacts with some of its atoms, creating a number of electron-ion pairs proportional to the energy lost by the incoming radiation. The ionized electrons are then accelerated to the anode, by means of the electric field established between the cathode and the anode of the proportional counter. In the close proximity of the anode, the electric field reaches values above the ionization potential of the gas, allowing the amplification by impact ionization of the drifting electrons, in a chain process known as avalanche multiplication. The final charge collected at the anode is thus proportional to the energy deposited in the counter by the ionizing radiation, allowing the identification of the ionizing particle.

If a suitable filling gas is used, proportional counters can be used in the detection of neutrons, which despite having a neutral charge, undergo nuclear capture reactions with certain isotopes which results in the emission of secondary heavy charged particles. This neutron capture reaction only occurs with a few isotopes and the ones with practical interest for neutron detection are <sup>3</sup>He, <sup>10</sup>B and <sup>6</sup>Li (ordered by decreasing cross-section). Of these, only

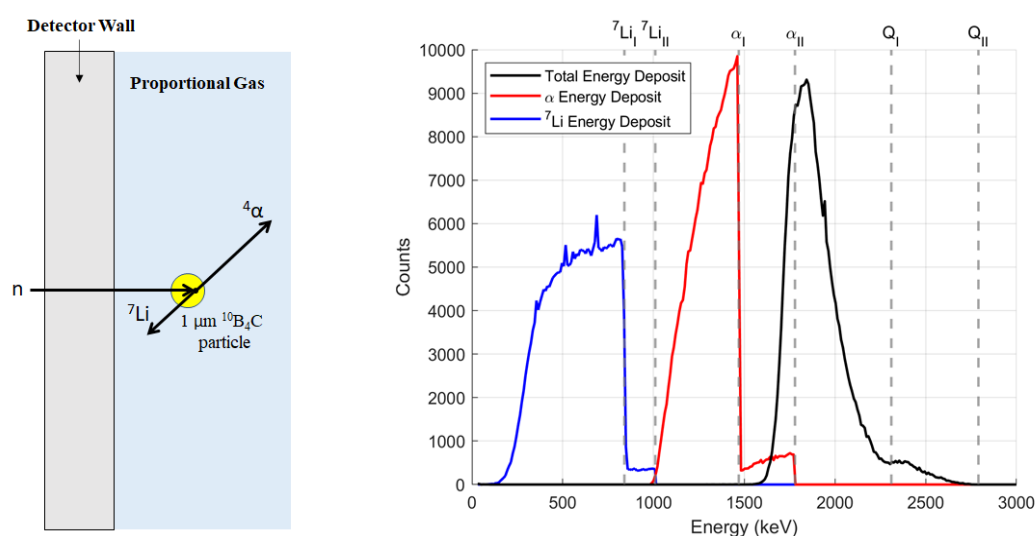
$^3\text{He}$  and  $^{10}\text{B}$  can be found in gaseous form. The latter exists only as the molecular gas  $\text{BF}_3$ , which is extremely toxic and consequently not a viable option to be used in detectors. As for  $^3\text{He}$ , it performs remarkably well in most neutron detection applications. In fact, due to their excellent detection efficiency, good gamma-ray discrimination and non-toxicity,  $^3\text{He}$  proportional counters are currently the most deployed neutron detectors. However, an insurmountable drawback of  $^3\text{He}$  proportional counters is that this gas has become extremely scarce and expensive.  $^3\text{He}$  natural isotopic abundance is only 0.0002% and its reserves come mainly from the fact that this gas is a by-product released during the nuclear warhead production and storage. With the end of the Cold War and the adoption of non-proliferation treaties, the production of this gas dropped significantly (Cho 2009). Finding adequate  $^3\text{He}$ -free neutron detection alternatives has become an imperative that motivated a great amount of research and development over the last decade (Hurd and Kouzes 2014). In this work, we present the new developments of an innovative  $^3\text{He}$ -free neutron detection concept that consists of a gaseous proportional counter in which a boron-based fine powder is dispersed, forming a neutron sensitive aerosol.

## 2. Operation Principle

In the neutron sensitive aerosol detector (Amaro et al. 2017),  $\text{B}_4\text{C}$  microparticles are suspended in a proportional counter by an appropriate gas flow which counter-acts the gravity force. As an incoming neutron interacts with the  $^{10}\text{B}$  atoms of the particles, two secondary products are released in opposite directions: a  $^7\text{Li}$  ion (0.84 MeV) and an  $\alpha$  particle (1.47 MeV):



An advantage of having the neutron capture occurring in nanoparticles is the possibility of simultaneously capturing the energy of both secondary particles that arise from the neutron capture reaction due to the nano/micrometric size of the suspended particles (Figure 1 - left). This is currently unachievable using detectors that rely on boron coating, and a particularly beneficial feature in environments with high gamma-ray interaction rates that require a cut-off energy that must in some cases extend to several hundred keV (Choi et al. 2019). This allows for a peak shape response in the pulse height distribution of the detector (Figure 1 - right).



**Figure 1:** Neutron capture reaction in the aerosol neutron detector (left) and detector response obtained with Monte Carlo Simulation for a single suspended 1  $\mu\text{m}$  diameter  $^{10}\text{B}_4\text{C}$  particle (right)

### 3. Methods and Materials

The selected  $B_4C$  powder used was provided by PlasmaChem GmbH (non-enriched, with >97% purity). A sample of it was previously subject to characterization by laser diffraction (Beckman Coulter Inc, LS 13 320) and the mean particle diameter value was determined to be  $d_{50} = 1.03 \mu m$ , with  $d_{10} = 0.55 \mu m$  and  $d_{90} = 1.60 \mu m$ . Approximately 1 gram was placed in the disperser cone of the detector (Figure 2) which was then tightly sealed with a copper gasket on the bottom flange. The top and bottom gas entrances were equipped with  $0.5 \mu m$  filters (SS-4VCR-2-0.5M) to prevent the powder from escaping the detector. The dispersion of the powder is done by the gas flow itself, which consisted of  $Ar/CH_4$  in 90/10 % proportion continuously flowing from the bottom inlet to the top outlet of the detector.

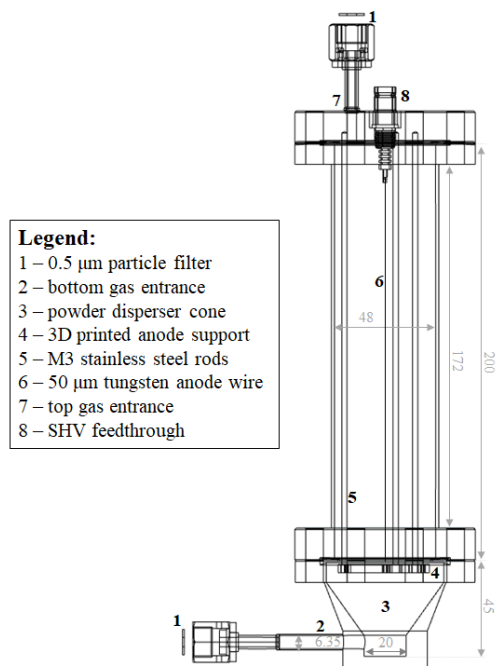
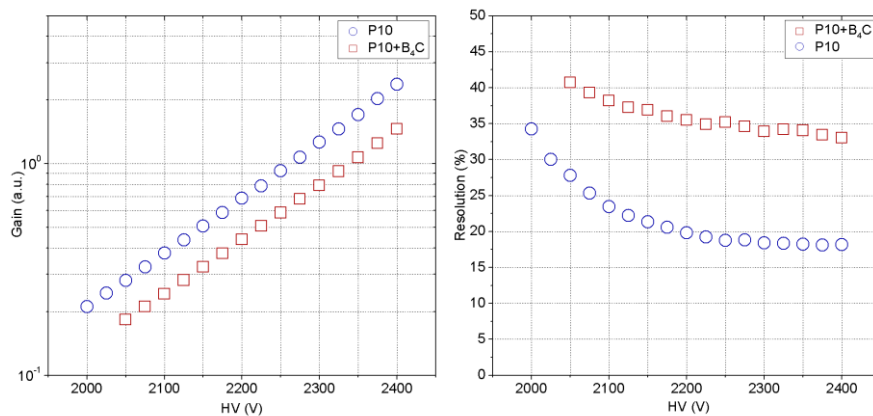


Figure 2: Technical design of the aerosol detector (dimensions in mm)

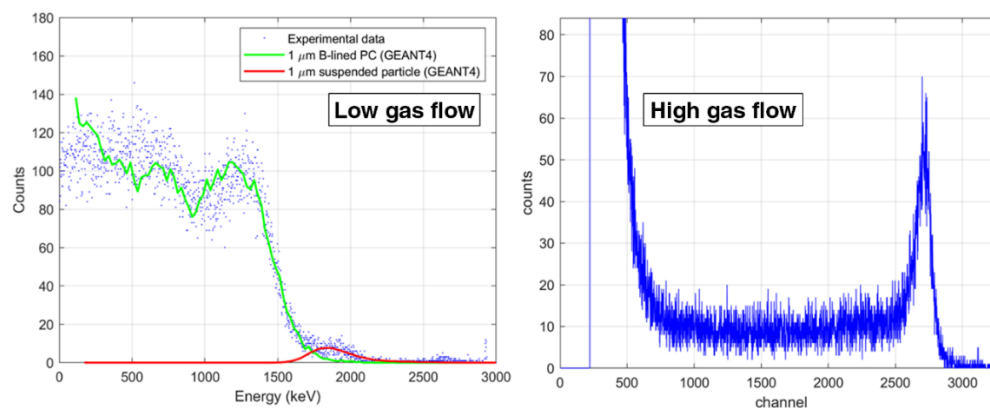
### 4. Results with X-rays and Neutron Irradiation

The operational properties of the aerosol detector were studied by irradiation with soft x-rays from a  $^{55}Fe$  isotropic source (5.9 keV). To allow the penetration of the x-rays, a 10 mm diameter window was made, using a  $50 \mu m$  thick aluminized Mylar film, glued to the detector with conductive epoxy. The detector gain (Figure 3 - left) and energy resolution (Figure 3 - right) were compared before and after the insertion of the  $B_4C$  microparticle. The fine powder did not compromise the detector operation by causing electrical discharges or drastically affecting its charge gain, which is an essential aspect for its stable long-term operation. A gain decrease by a factor of 36% and an energy resolution increase by 15% (absolute value) were observed (Duarte et al. 2019). This can be attributed to the fact that in the close proximity of the anode, where the electron avalanche multiplication takes place, the nanoparticles create inhomogeneities in the electric field and a consequent rise in the fluctuations of the avalanche charge gain, increasing energy resolution.



**Figure 3:** Effect of the presence of the B<sub>4</sub>C nanoparticles in the charge gain (left) and in the energy resolution (right) of the aerosol detector

Irradiation measurements with thermalized neutrons from a moderated <sup>241</sup>AmBe source were conducted at Institute Laue Langevin. After inserting the nanoparticles in the dispersion cone and sealing the detector, Ar/CO<sub>2</sub> gas in 90/10 % proportion was opened at a very low flow to purify the gas inside the detector without immediately dispersing a large number of nanoparticles. A spectrum was acquired in these conditions, for a 300 second acquisition time (Figure 4 - left). A detector response similar to that of conventional boron coated detectors was obtained, arising from neutron captures in nanoparticles attached to the walls. A small peak centred at a higher energy, approximately 1800 keV is also present, attributed to neutron captures occurring in the suspended nanoparticles. Although the gas flow was low, because the nanoparticles are extremely light weighted, a fraction of them will inevitably be dispersed. The spectrum was calibrated in energy taking by reference the two visible count slumps, which correspond to the energies of the secondary particles of Equation (1). The experimental data agrees with GEANT4 (Agostinelli et al. 2003) simulations of a detector coated with 1 μm layer of B<sub>4</sub>C, combined with a detector with suspended 1 μm B<sub>4</sub>C particles (Figure 1 - right). By significantly increasing the gas flow, an increase of the peak response of the detector was briefly observed (Figure 4 - right). However, a leakage current inhibited the detector operation for longer periods of time. This leakage current was attributed to the fact that the suspension of the B<sub>4</sub>C powder was creating an electrically conductive path between the top of the anode wire, soldered to the SHV feedthrough and the surface of the cathode flange. This issue can be resolved by redesigning the top flange, to increase the distance between the flange surface and the conductive end of the feedthrough.



**Figure 4:** Pulse height spectrum of the aerosol neutron irradiated by thermal neutrons, initially with a low gas flow circulating (left), and later with a high gas flow (right)

## 5. Conclusions and Future Work

By irradiation with x-rays from a  $^{55}\text{Fe}$  source (5.9 keV), the effect of the  $\text{B}_4\text{C}$  nanoparticles in the charge gain and energy resolution of a proportional counter shown an average gain loss of 36% and an increase in energy resolution (FWHM) by 15% (absolute value). Through thermal neutron irradiation with an  $^{241}\text{AmBe}$  source, we observed neutron capture reaction occurring in suspended  $\text{B}_4\text{C}$  particles, which resulted in the presence of a peak in the pulse height spectrum of the detector, which is not achievable by typical boron coated detectors, and may be an important feature to improve gamma-ray discrimination in environments with high gamma-ray interaction rates. Further developments are being carried to stabilize the currents in the detector to allow for longer acquisition periods, namely increasing the distance between the flange surface and the conductive end of the feedthrough. To optimize the fine powder dispersion, strategies to reduce the particle attachment will be investigated, namely the effectiveness of polishing the detector inner walls. After these developments, new neutron irradiation experiments will be carried out.

Finally, the described detection technique can, in principle, also be used to increase the detection efficiency of gaseous detectors to hard x-rays and gamma-rays using adequate micro/nanoparticles, made of high Z number materials, such as Bi.

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