Design and Development of High Temperature $^{10}$B Coated Proportional Counters for PFBR

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Abstract

High sensitivity High Temperature Boron-10 Coated proportional Counters (HTBCCs) which can work in 250°C environment are developed for Fast Breeder Reactor. HTBCCs with sensitivity of 12 cps/nv, are used in Control Plug during initial core loading and first approach to criticality experiments to enhance the core monitoring, as Instrumented Central Sub-Assembly (ICSA) is lifted up and moved along with control plug away from the core region, during fuel loading. In case of an unforeseen long shutdown for more than 4 months, Shut Down Count Rate (SDCR) may become < 3 cps. For the subsequent flux monitoring during fuel loading and start-up, it is required to use three boron coated counters (BCCs) with a sensitivity of 4 cps/nv. The detectors will be placed side by side at the spare detector location in Control Plug. HTBCCs of neutron sensitivity 12 cps/nv and one assembly containing three numbers of 4cps/nv detectors are developed and characterized for reactor applications. The functional tests and qualification tests were carried out on these detectors and the design specifications were established.

Introduction

Neutron detectors capable of operating in high temperature environment are required in fast breeder reactors for reactor control and safety as part of nuclear instrumentation. The high temperature neutron detectors are used at above-the-core locations in the sodium pool for measurement of neutron flux from the fuel loading stage to the reactor power operation. The flux monitoring during fuel loading and approach to first criticality in fast breeder reactors is carried out using neutron detectors with sensitivity more than 10 cps/nv operating at 250°C continuously. $^{10}$B coated proportional counters are best suited for this requirement since compared to other detectors like $^3$He and $^{10}$BF$_3$ proportional counters, $^{10}$B coated proportional counters have better tolerance to ambient gamma radiation, operate at lower bias voltages, and are non-corrosive in reactor environment. Earlier, high sensitivity (~30 cps/nv) $^{10}$B coated proportional counters has been developed but their maximum operating temperature is limited to about 120°C. The challenges of detector operation at high temperatures are overcome by advanced mechanical design and proper selection of construction material for long term continuous operation. $^{10}$B
coated proportional counters with a sensitivity of 12 cps/nv are required for use at Control Plug locations of Prototype Fast Breeder Reactor (PFBR) for monitoring the flux during fuel loading / handling, approach to first criticality. In case of an unforeseen long shutdown for more than 4 months after source activation, the subsequent flux monitoring during reactor start-up is proposed to be carried out using boron coated counters with a sensitivity of 4 cps/nv operating at 250°C. The present article describes design, development and characterization of 108 coated proportional counters of 12 cps/nv and 4 cps/nv thermal neutron sensitivity capable of operating upto 250°C developed for PFBR.

**Design**

Conventional boron coated proportional counters developed for reactor applications are of cylindrical shape as shown in Fig. 1. The outer cylinder acts as cathode. 94% enriched $^{10}$B powder is mixed with binder and thinner and homogenous solution is prepared. Thin layer of this solution is coated on the inner surface of cathode and dried at 250°C. The process is repeated till desired coating thickness on the cathode surface is obtained. A very small diameter (thin) anode wire is mounted axially over insulators in the geometric centre of the detector. Heat shrinkable polyethylene sleeve is provided over the detector to isolate detector ground from the local ground.

For development of high temperature boron coated proportional counters, the following special design features have been incorporated.

- In conventional boron counters, polyethylene sleeve is provided over cathode tube for ground isolation. In the present detectors, cathode is encased in SS cylindrical housing insulated using alumina ceramic spacers for high temperature operation.
- Spring assembly made of spring steel is provided at one end of the detector assembly to absorb dimensional changes during temperature variation.
- Alumina ceramic spacers and feed throughs have been introduced as insulators instead of Teflon, Mylar insulators.
- The detector is constructed out of SS 304 L to minimize corrosion at weld joints.
- The detector is joined with triaxial mineral insulated cable for taking out the signal.

Incorporating above design modifications, two types of High Temperature Boron Coated proportional Counters (HTBCCs) have been developed. A 12 cps/nv HTBCC with 100 mR/h gamma tolerance is developed for measuring neutron flux during fuel loading operations. After long operation and long shut down of more than 4 months, for subsequent startup, high sensitivity boron counters for redundant safety channels are required with 4 cps/nv neutron sensitivity and 200 R/h gamma tolerance. For such requirements, three numbers of 4 cps/nv HTBCCs assembled in single housing have been developed. The fabrication of the detectors was carried at ECL, Hyderabad. The main specifications of the detectors are given in Table 1. Fig. 2 and Fig. 3 give the construction schematic of the detectors.

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Fig. 1: Picture and schematic of standard boron coated proportional counter
**Table 1: Main specifications of $^{10}$B coated proportional counters**

<table>
<thead>
<tr>
<th>Detector</th>
<th>12 cps/nv</th>
<th>4 cps/nv</th>
</tr>
</thead>
<tbody>
<tr>
<td>Outer housing material, overall dimensions</td>
<td>SS 304L; 1000 mm Long, 63 mm OD</td>
<td>SS 304L; 735 mm Long, 63 mm OD</td>
</tr>
<tr>
<td>Sensitive length</td>
<td>700 mm</td>
<td>378 mm</td>
</tr>
<tr>
<td>cathode</td>
<td>OD 54 mm, ID 51.3 mm</td>
<td>OD 25.4 mm ID 23.8 mm (single detector)</td>
</tr>
<tr>
<td>Anode wire</td>
<td>25 µm dia. tungsten</td>
<td></td>
</tr>
<tr>
<td>Filling gas</td>
<td>Ar (95%) + CO2 (5%) at 18 cm Hg</td>
<td></td>
</tr>
<tr>
<td>Cable and End connector</td>
<td>12 m long tri-axial Mineral Insulated cable</td>
<td></td>
</tr>
<tr>
<td></td>
<td>having Triaxial bulkhead receptacle</td>
<td></td>
</tr>
<tr>
<td>Neutron sensing material &amp; coating thickness</td>
<td>94% $^{10}$B enriched, 0.55 mg/cm²</td>
<td>94% $^{10}$B enriched, 0.88 mg/cm²</td>
</tr>
<tr>
<td>Charge collection time</td>
<td>400 ns</td>
<td>200 ns – 350 ns</td>
</tr>
<tr>
<td>Operating voltage</td>
<td>800 V - 900V DC</td>
<td>750 V - 850 V DC</td>
</tr>
<tr>
<td>Measurement range</td>
<td>0.3 nv – 5x10^3 nv</td>
<td>1 nv – 5x10^4 nv</td>
</tr>
<tr>
<td>Operating temperature</td>
<td>250 °C</td>
<td></td>
</tr>
<tr>
<td>Influence of gamma</td>
<td>upto 0.1 R/h without count loss</td>
<td>upto 200 R/h without count loss</td>
</tr>
</tbody>
</table>
**Design of cathode dimensions**

The requirement of neutron sensitivity in boron coated proportional counters governs the cathode dimensions. Elemental $^{10}$B is coated on the internal surface of the cathode. Initially neutron sensitivity increases with increase in coating thickness. However, above an optimum coating thickness, the neutron sensitivity decreases due to self shielding effect. Therefore in boron coated proportional counter, the neutron sensitivity is directly proportional to boron coated surface area. The neutron sensitivity of boron counters can be derived from the following equation:

$$S_n = N \sigma \phi C$$  \hspace{1cm} (1)

where $N = \text{number of } ^{10}\text{B atoms which is given as;}$

$$N = S A t / a$$ where $A$ is Avogadro number $6.023 \times 10^{23}/\text{mol}$, $S$ is boron coated surface area, $t$ is coating thickness in mass per unit area and $a$ is atomic weight of boron, $\sigma = \text{reaction cross section}$, $\phi = \text{neutron flux}$ and $C = \text{counting efficiency}$ which depends upon the coating thickness. The efficiency $C$ can be estimated from the following equations $^1$:

$$C_a = \frac{1}{2} \mu \cdot \left( T - \frac{T^2}{2R_a} \right)$$  \hspace{1cm} (2)

$$C_{Li} = \frac{1}{2} \mu \cdot \left( T - \frac{T^2}{2R_{Li}} \right)$$  \hspace{1cm} (3)

where $C_a$ is efficiency for alpha particles emitted, $\mu$ is attenuation coefficient, $T$ is coating thickness, $R_a$ is range of alpha particles in the boron coating, $C_{Li}$ is efficiency for lithium particles emitted, $R_{Li}$ is range of lithium particles. The total efficiency $C$ is the sum of efficiencies of both the particles and is given as

$$C = (C_a + C_{Li})$$  \hspace{1cm} (4)

For cylindrical counters, if some neutrons have not interacted on one side of the coating, they may interact on the opposite coating while going from first coating surface to the second. Therefore, the efficiency in case of cylindrical counters is given by $^2$

$$C_{cylindrical} = 2C - C^2$$  \hspace{1cm} (5)

Substituting numerical values, the neutron sensitivity of boron counters can be estimated and cathode dimensions are derived.

**Design of anode dimensions**

Neutrons interact with $^{10}$B isotope ($\sigma$ for thermal neutrons approximately 3836 barns) of boron coating and produce $\alpha$ and lithium particles, which interact with gas and produce ionization.

$$^{10}\text{B} + ^{1}\text{n} \rightarrow ^{7}\text{Li}^+ + ^{4}\text{He} + 2.31 \text{MeV}$$

The energy per neutron interaction produced is not sufficient to produce measurable pulse output. Therefore, it is required to amplify the primary charge produced by charge multiplication. This requires high electric field gradient. The high electric field in the cylindrical geometry detectors is produced by selecting very thin diameter anode wire. The charge multiplication coefficient, $M$, increases with increase in voltage applied to the anode. The total charge $Q$ generated by $n_o$ original ion pairs is $Q = n_o e M$ and the pulse amplitude $V$ is given as

$$V = Q / C$$  \hspace{1cm} (6)

where $C$ is the capacitance of the detector.

Diethorn derived a widely used expression for $M$ and is given as $^3$

$$\ln(M) = \frac{V}{\ln \left( \frac{V}{\Delta V} \right)} \left( \ln \frac{V}{p a \ln \left( \frac{b}{a} \right)} - \ln(k) \right)$$  \hspace{1cm} (7)

where $p$ is gas pressure $k$ is a Diethorn constant and $\Delta V$ is ionization potential. The equation indicates that smaller is the anode wire diameter, greater
is the charge multiplication and hence the pulse output.

**Design of anode wire mounting assembly**

The anode wire chosen in proportional counters is always of a very small diameter to generate high electric field gradient for adequate charge multiplication. However, any small drift in anode wire position causes reduction in the gap between the cathode and anode. This happens especially at the ends of anode wire, where it is mechanically mounted over the insulator or at the centre, due to slackening. This position shift gives rise to generation of random breakdown pulses. Therefore, in order to avoid the breakdown to occur, it is very essential to have a very rugged anode wire mounting mechanism with appropriate insulator design for the proportional counters operating at elevated temperatures. For high temperature boron counters, a spring assembly using alumina ceramic spacers is designed. The details of the spring mounting assembly are given in Fig. 4. The anode wire is kept at tension with the help of spring. The anode wire is enclosed with ceramic spacers and bushings and therefore even at elevated temperatures, the occurrence of breakdown phenomenon is avoided due to any differential dimensional variations.

**Detector design analysis for seismic event**

The mechanical integrity of the boron counters during specified seismic event is carried out by analysis using NISA finite element software version 11. The allowable initial tension in 25µm diameter anode wire was computed to be less than 20 gm at operating temperature of 250°C.

**Detector component cleaning procedure**

The cleaning of SS 304 L components is carried out using Trichloroethane, Perchloroethylene, Isoproyl Alcohol or Ethyl Alcohol and Acetone. After cleaning, the components are baked at 400°C. The ceramic components are cleaned in mild alkaline solution, heated upto 80°C and then rinsed in demineralized water. Cleaning is also done in ultrasonic cleaner in Acetone or aviation grade spirit without chlorides or fluorides. The ceramic components are then baked in oven up to 400°C for 2 hours duration just prior to taking up the assembly.

**Procedure for gas filling and gas filling tube pinching**

The welded detector is leak tested at 1.5 kg/cm² (abs) by pressure test and helium leak test up to $10^{-9}$ std. cc /s. The detectors are evacuated and degassed by baking at 250 °C till vacuum of the order of $10^{-6}$ torr is achieved and maintained. After baking and degassing, it is ensured that the vacuum is maintained over a period of 12 hours before the gas mixture is filled in the detector. After gas filling, the filling tube is crimped with pinching tool and the pinched end is welded. The detector has all welded construction and all the weld
joints are Helium leak tested except for the pinched welded end of gas filling tube. Since the detector is filled at sub-atmospheric pressure, there is no known method to check the leak tightness of the crimped and welded end of gas filling tube. Therefore the leak tightness of the pinched end of gas filling tube has been ensured only by standardizing the crimping procedure. The gas filling tube made of SS 304L are annealed at 400°C for 90 minutes to normalize the stressed grains. The tubes are cooled in open furnace. Special pinching tool is designed which exerts a maximum torque of 10 kg-m. After pinching, the pinched end is inspected for uniformity of pinching as per standard procedure.

Tests and results:

Large sets of experiments were conducted on High Temperature Boron Coated Counter (HTBCC) to evolve data on the operation of detectors at 250°C.

**Insulation Resistance and Capacitance**

The bare 12 cps/nv HTBCC (without MI cable) showed 10 pF capacitance and $-10^{12}\Omega$ insulation resistance at 1kV DC at room temperature. No change in the insulation resistance and capacitance was observed at 250°C. The bare 4 cps/nv HTBCC (without MI cable) showed 8 pF capacitance and $-8\times10^{12}\Omega$ insulation resistance at 1kV DC at room temperature. No change in the insulation resistance and capacitance was observed at 250°C. 12 cps/nv and 4 cps/nv HTBCCs were then connected with 12 meter long integral triaxial mineral insulated cable. 12 cps/nv HTBCC with integral cable assembly showed 2.2 nF capacitance and $-10^{11}\Omega$ insulation resistance at 1kV DC at room temperature. At 250°C, the capacitance remained unchanged however the insulation resistance reduced to $-10^{10}\Omega$ at 1kV DC. 4 cps/nv HTBCC with integral cable assembly showed 2.2 nF capacitance and $-10^{12}\Omega$ insulation resistance at 900 V DC at room temperature. At 250°C, the capacitance remained unchanged however the insulation resistance reduced to $-8\times10^{11}\Omega$ at 900 V DC.

**Output pulse characteristics, neutron sensitivity and influence of gamma radiation at 250°C**

The measured charge collection time for 12 cps/nv HTBCC ranges between 300 ns – 400 ns and for 4 cps/nv HTBCC ranges between 200 ns – 350 ns at room temperature. No measurable change in the charge collection time is observed while operating at 250°C. The neutron sensitivity of 12 cps/nv HTBCC and 4 cps/nv HTBCC was measured using a standard source of 27 nv thermal neutron flux. The average neutron sensitivity of 12 cps/nv HTBCC is 11.25 cps/nv ($\pm 10\%$). The average neutron sensitivity of 4 cps/nv HTBCC is 3.7 cps/nv ($\pm 10\%$). 12 cps/nv HTBCC was tested at 250°C with neutron source and in mixed radiation of neutron and 100 mR/h gamma radiation (Fig. 5 and Fig. 6). The variation in the count rate in plateau region is within 6 %. The voltage plateau data and discriminator bias data overlapped for room temperature and for 250°C. The variation in the count rate is within 10 % for 850 V and 900V HV. The observed discriminator bias plateau slope and voltage plateau slope was 1.3%/mV and 1%/V respectively for 12 cps/nv HTBCC.

4 cps/nv HTBCC was tested at 250°C with neutron source and in mixed radiation of neutron and 200 R/h gamma radiation (Fig. 7 and Fig. 8). The variation in the count rate is within 5 % for 850 V HV and 80 mV discriminator bias. The voltage plateau data overlapped below 850 V at 250°C and 200 R/h gamma radiation. However after increasing the operating bias to 900 V, because of 200 R/h gamma radiation, increase in the count rate was observed. At room temperature, at 900 V HV the increase in count rate was 100 % and at 250°C at 900 V HV the increase in count rate was 376 % compared to the count rate without gamma radiation background.
This increase in count rate is attributed to the excess ionization produced by gamma radiation which increases magnitude of avalanches at higher operating voltages. Therefore it is recommended to operate the detectors at and below 850 V HV.

The discriminator bias curves plotted at 850 V show 5% variation in the count rate at 80 mV discriminator bias at room temperature and at 250°C in 200 R/h gamma radiation. The gamma radiation produces additional ionization in the detector volume. This ionization creates space charge effects in the detector and due to this overall pulse amplitude reduces. However, in the plateau range the change in the count rate is within acceptable limit. The observed discriminator bias plateau slope and voltage plateau slope was 1.6%/10mV and 3.4%/10V respectively for 4 cps/nv HTBCC.

![Voltage plateau curve of 12 cps HTBCC in 100 mR/h gamma and 250°C](image1.png)

**Fig. 5: Voltage plateau curve of 12 cps HTBCC in 100 mR/h gamma and 250°C**

![Discriminator bias curve of 12 cps HTBCC in 100 mR/h gamma and 250°C](image2.png)

**Fig. 6: Discriminator bias curve of 12 cps HTBCC in 100 mR/h gamma and 250°C**
Count rate linearity w.r.t. neutron flux (in reactor)

12 cps/nv and 4 cps/nv HTBCCs were tested for signal linearity (Fig. 9 and Fig.10) in AHWR-CF. 12 cps/nv signal linearity is within 10% upto $1.5 \times 10^3$ nv neutron flux and within 30% upto $5 \times 10^3$ nv thermal neutron flux. 4 cps/nv HTBCC signal linearity is within 2% upto $6 \times 10^4$ nv.
Qualification tests conducted on 12 cps/nv HTBCC

Sample detectors from the production prototype lot of 12cps/nv HTBCC was subjected to various qualification tests viz. 12 number of thermal cycle tests at 250°C as shown in Fig. 11, vibration tests at 1-33 Hz and damp heat cycle tests as shown in Fig. 9 (vide ref. PFBR/60510/SP/1008/Rev. 0). The detector was tested for functionality using 27 nv thermal neutron flux before and after the detector was subjected to qualification tests. Fig.13 and Fig. 14 give voltage plateau and integral bias curves of the detector and it was observed that the detector performance remained unchanged even after undergoing the above qualifications tests.
Fig. 12: Damp Heat Cycle Test profile

Fig. 13: Voltage plateau curves of 12 cps/nv HTBCC before and after subjecting to Functional & qualification tests

Fig. 14: Discriminator bias curves of 12 cps/nv HTBCC before and after subjecting to Functional & qualification tests
Summary

12 cps/nv and 4 cps/nv High Temperature Boron Coated Counters (HTBCC) have been designed and fabricated. The fabrications of the detectors have been carried out at M/s ECIL, Hyderabad. The functional tests and qualification tests were carried out on these detectors to establish the design specifications. The performance tests conducted on the detectors showed that insulation resistance of the cable and detector upto 250 °C remains of the order of $10^{10}$ ohms at 1 kV DC as required. The operating voltages of the detectors are observed to be as specified. The change in the neutron sensitivity up to 250 °C at the plateau range is negligible. The detectors’ signal linearity is within ±10% in the required range of operation. The detector performances remained unchanged after subjecting to qualification tests. After qualification of prototype detectors, 8 no. of 12 cps/nv and 6 no. of 4 cps/nv HTBCCs integrated with hangers have been fabricated and supplied to PFBR.

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References