



Development of self-powered neutron detectors used in nuclear medicine for the measurement of neutron flows during treatment of boron neutron therapy

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Abstract

Purpose The Self Powered Neutrons Detectors (SPND) have the advantage of not requiring a high voltage power supply for their operation and are small in size, enhancing the interest of these detectors in medicine.

Methods In this context, we have developed a thermal neutron detection system based on SPND. This detector was placed in the thermal channel of our nuclear research reactor; where the values of the current for each detector have been recorded as a function of time, with a chain in a current mode where electrometers without HV were used.

Results We performed the real-time measurement of neutron flux during boron neutron capture therapy or boron neutron therapy, the different materials constituting the SPND detectors have been carefully chosen for this application. These detectors were tested at a power of four MW corresponding to a neutron flux of $10^9 \text{ n cm}^{-2} \text{ s}^{-1}$.

Conclusions The usefulness of ^{103}Rh -SPND is for online measurement of thermal neutron flux on BNCT patients has been demonstrated based on an appropriate calibration of the thermal neutron spectrum.

Keywords SPND · Thermal neutron · Capture therapy · Flux · Detection · Boron

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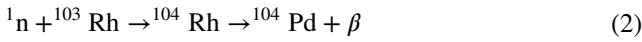
Introduction

Boron neutron capture therapy (BNCT) or boron neutron therapy is a technique that requires simultaneous presence of neutron fluxes with adequate energy and a neutron sensor, the boron-10 (^{10}B) [1]. Boron and neutrons interact to attack tumor cells without causing significant damage to healthy cells because the destructive effect occurs mainly in tumor cells that have selectively accumulated boron, the capture reaction is as follows [2]



The path of alpha (^4He) and lithium (^7Li) particles in tumor tissue is around the diameter of the tumor cell ($\sim 10 \mu\text{m}$) [3–5]. Therefore, the destructive action of the capture reaction occurs mainly in those cancer cells that have accumulated boron, while the normal cells with a low concentration of boron are not attacked. In order to optimize treatment by the BNCT technique, we are interested in determining the heat flux of neutrons in specific areas of patients. A few activation methods are currently used

for this purpose, but they are not able to provide real-time information. Many authors have developed "in real-time" thermal neutron SPNDs using fast response time materials with neutron capture cross sections to supply current [6–8]; for example, the signal rhodium-based detector is the beta-current type of self-powered detector, which uses the activation reaction below to produce a current that can be measured according to Eq. (2).



When the neutron is captured by ${}^{103}\text{Rh}$, a ${}^{104}\text{Rh}$ nucleus is formed. Then, it decays into ${}^{104}\text{Pd}$ and e^- by decay β^- . Thus, high-energy electrons are generated in the emitter. Figure 1 shows the ${}^{103}\text{Rh}$ reaction chain.

A lot of self-powered neutron detectors were designed, developed and tested around the world [9]. Recently, many new self-powered neutron detectors types have been developed and tested for medical applications [10, 11]. Those detectors are now considered one of the most robust and reliable choices of in-core flux-detectors owing to their successful performance for decades.

In this work, we present the design and realization of new SPND detectors that we have developed in our measurement detectors laboratory in the nuclear research center of Birine, in order to use it in the measurement, online, of the thermal neutrons flux incident during therapy by neutron capture (BNCT). Compared to other in-core detectors, BNCTs feature many advantages, those types of detectors are small size, which can be placed directly under the skin or inside the patient’s brain, for processing to determine exactly

the actual flux used in the irradiated area; in addition, they are simple and robust structure. In addition, they show a good stability under temperature and pressure conditions. Another advantage of this type of detector is no high voltage is applied, which is very important for medical applications.

Conception, realization

Principle

The self-powered detector SPND is a coaxial cylindrical assembly of different materials, as shown in Fig. 2, which consists of an emitter, a center conductor with a material of relatively high cross section, an insulator and an external conductive sheath (collector). In this type of detector, the central emitter picks up the neutrons and emits the high-energy electrons, which reach the outer cladding after passing through the insulator. In the SPND detector, the current is proportional to the neutron flux that can be measured using an electrometer connected between the emitter and the sheath (Fig. 2). Table 1 shows the labels of our SPND prototype.

These kinds of detectors have a probability for a response to gamma radiation due to compton or photoelectric effects which can produce free electrons with enough energy to pass from the collector to the emitter or vice versa. This probability of occurrence of the two effects increases with Z and the mass of the electrode materials [2]. When designing the detector, the appropriate materials must be well chosen

Fig. 1 Rhodium reaction chain

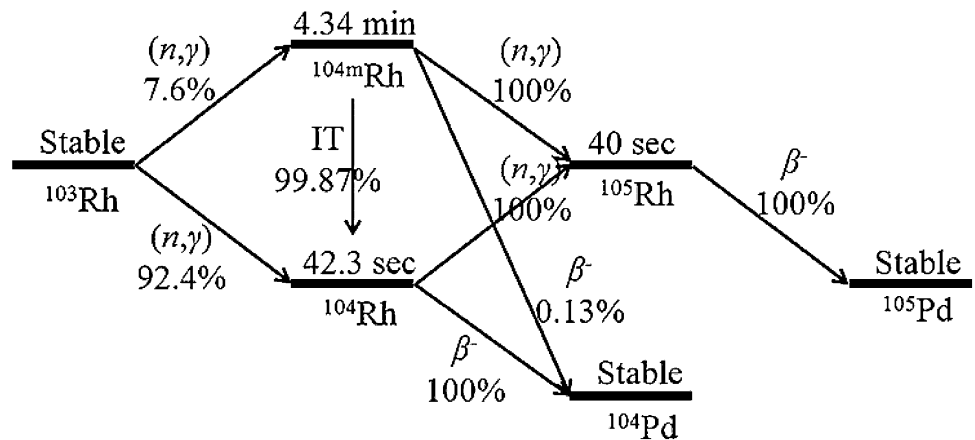


Fig. 2 The schematic diagram of self-powered neutron detector SPND

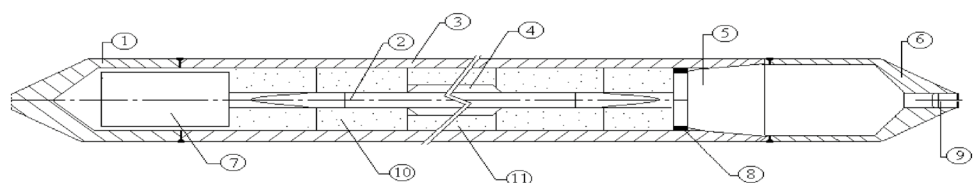


Table 1 Labels of prototype self-powered neutron detector SPND

No.	Designation	Material
1	Plug 1	Inconel 600
2	Central bar	Inconel 600
3	Outer tube (collector)	Inconel 600
4	Emitter	Vanadium
5	Copper plug	Copper
6	Plug 2	Inconel 600
7	cylinder	Copper
8	Washer	Copper
9	Washer	Inconel 600
10	Insolation pad	Magnesium oxide (MgO)
11	Insolation pad	Magnesium oxide (MgO)

[13–16], so that the gamma response is as low as possible to eliminate the effect of gamma radiation [12, 17].

Material selection

In this work, several prototypes that aimed to optimize our detector design have been realized, then study and characterizing its response, and the materials of the components of the developed detector are chosen as follows:

Emitter: must have an effective neutron capture section, with a low mass, would provide a good level of signal at the output with a flux to be measured greater than 10^8 n cm⁻² s⁻¹. The current level (I) is being estimated according to the following formula [2]:

$$I = C \cdot q \cdot \sigma \cdot N \cdot \Phi \quad (3)$$

where C is constant relating to geometry and efficiency; q is charge released by absorbed neutron; σ is effective activation section of the emitter; N is number of atoms of the emitter; Φ is neutron flux.

Among all the materials analyzed, rhodium 10^3 (10^3 Rh) was the appropriate choice because this element has a fairly high capture cross section: 134 barns [18, 19], $q = e$ (electron charge), $C = 1$, $\Phi = 10^8$ n cm⁻² s⁻¹ in the worst case, and by determining N for a cylindrical emitter of dimensions (length = 1 cm and diameter = 1 mm), the current levels were obtained of the order picoammeter. In this picoammeter order, current levels can be easily measured with instrumentation very similar to low current measurement. In addition, although the rhodium signal is delayed due to the beta decay half-life of 42.8 s [18, 20], this time is negligible compared to the irradiation time of the treatment (more than 30 min); therefore, in these cases, in practice, the signal can be considered "online." For high-intensity beams or if several irradiation steps are used and the irradiation times

Table 2 Characteristics of detectors SPND1, SPND2 and SPND3

	SPND1 and SPND2	SPND3
Emitter material	Rhodium	Zircaloy-4
Insulation material	Acrylic	Acrylic
Collector material	Zircaloy-4	Zircaloy-4
Sensitive length	10 mm	10 mm
External diameter	1.9 mm	1.9 mm

may therefore be only a few minutes, it will be necessary to use dynamic compensation to correct for the delay.

Insulation: we have chosen acrylic as the insulating material in all designs; it is generally used in all detectors for applications in the medical field. It is a tissue-equivalent material for neutrons, which does not increase the radiological risk during or after irradiation, because it does not alter the treatment conditions and does not activate after the irradiation is terminated. In addition, the properties of acrylic, for this type of application, do not present any risk of appreciable modification under gamma or neutron irradiation up to 50 kGy [21, 22] and 10^{17} n cm⁻² [23–25], respectively. Therefore, with the typical fluxes used for BNCT treatment, the acrylic insulation shows no significant degradation even in the long term.

The collector (outer sheath): the sheath material must not be toxic to avoid contamination of the patients during medical application, must be sterilized before each use, must have a very low effective capture section, and have the lowest possible residual radioactivity. For that, two materials were evaluated: graphite and Zircaloy-4 [25–27], and the main advantage of using graphite is the possibility of obtaining very thin conductive layers, but it is a water-soluble material, which complicates its implantation in tissues. Zircaloy-4 is a zirconium-based alloy that must be subjected to a purification process to reduce impurities present in the base metal and which can have a high effective cross section. In addition, zirconium is a biocompatible metal [28, 29], used in biological prosthesis. With this material (zircaloy), we cannot get thin conductive layers like with graphite, and this will result in a larger outside diameter of the detector [30, 31]. Table 2 shows the characteristics of detectors SPND1, SPND2 and SPND3.

Characterization

For the neutron characterization on the nuclear research reactor, the equipment used is as follows: three detectors: SPND1, SPND2 and SPND3, acrylic support, thin activation sheets for measuring neutron flux, electrometer model Keithley 602, electrometer model Keithley 617 and the last electrometer model Keithley 602 and PC for data acquisition. The detectors are placed on acrylic support as described

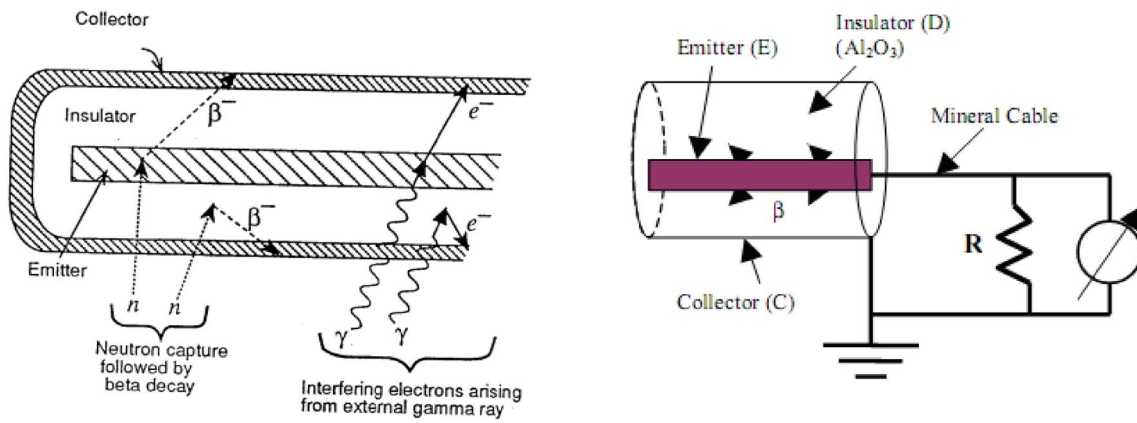


Fig. 3 Self-powered neutron detector SPND operation and measurement scheme [12]

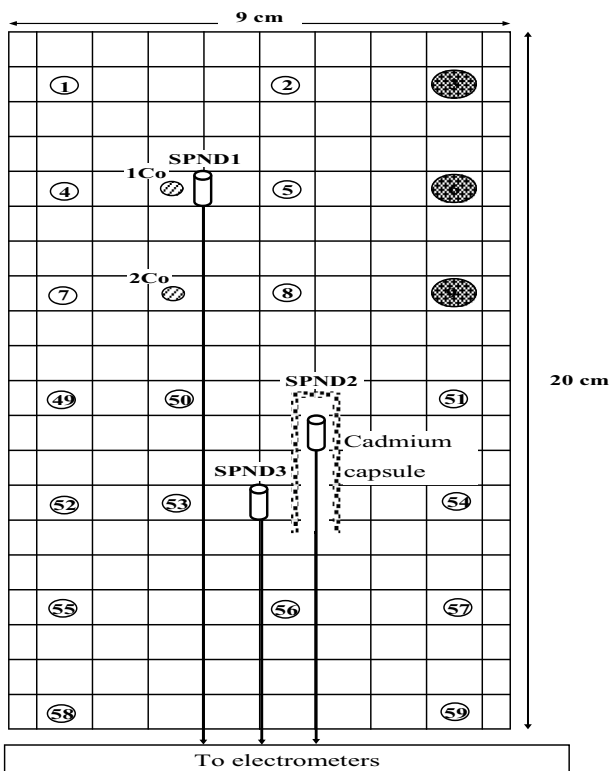


Fig. 4 Diagram of the irradiation device

in the diagram of the irradiation device and the device for irradiating SPNDs (Figs. 3, 4 and 5). In order to measure the heat flux in the areas considered, very thin sheets (activation detectors) are placed in the irradiation area. One of the detectors (SPND2) was placed in a cadmium capsule to estimate the contribution of the entire signal of thermal neutrons for comparison with the naked detector (SPND1) without a capsule. The device was placed in the thermal column of the reactor. We recorded the value of the leakage current, I_0 , produced by each detector in the absence of radiation.

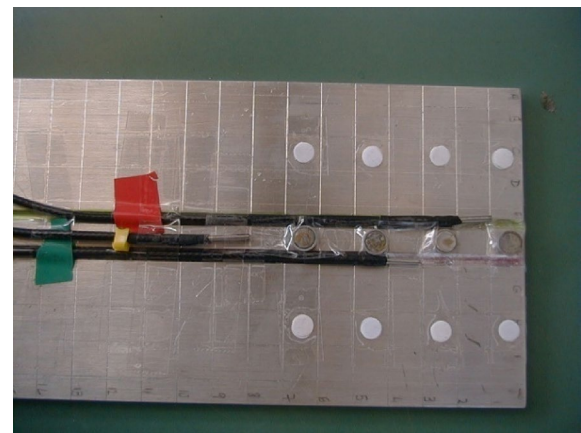


Fig. 5 Device for irradiating SPNDs and gold and copper sheets

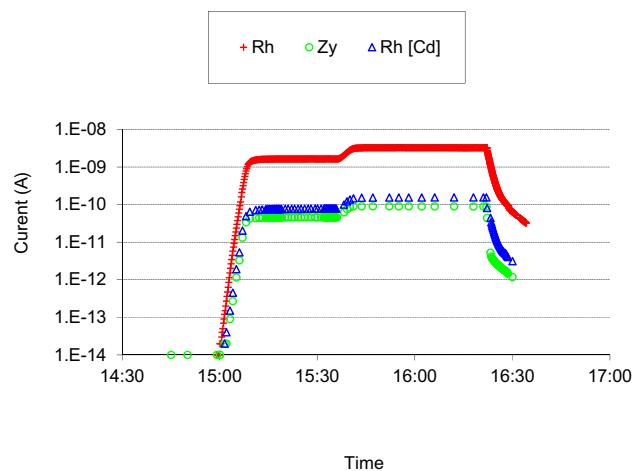


Fig. 6 Temporal evolution of SPNDs Signals

We performed the irradiation on our SPND detector and by recording the results of the current produced by these

detectors as a function of time (Figs. 6, 7). The first level of the curve corresponds to the intensity of the heat flow for a power of 2 MW and the second level to a power of 4 MW.

Results

Table 3 shows the currents measured under neutron irradiation at powers 2 MW and 4 MW, Table 4 shows the neutron flux for the various 4 MW detectors in $n\text{ cm}^{-2}\text{ s}^{-1}$, and the neutron flux values were calculated by our reactor physics group.

Fig. 7 Time evolution of the signal in percentage

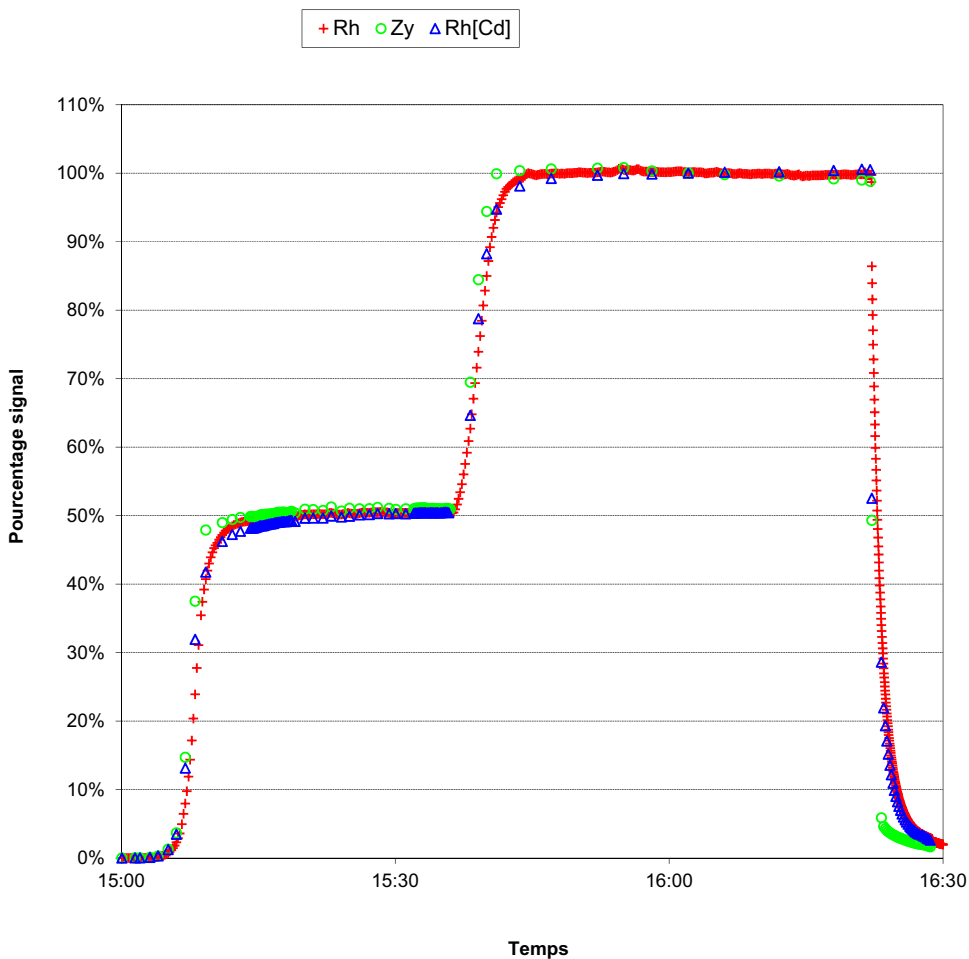


Table 3 Measured neutron currents (A)

Reactor power	SPND1 (Emitter Rh) naked	SPND2 (transmitter Rh) under Cd	SPND3 (Emitter Zy-4) naked
$P=2\text{ MW}$	$(1.62 \pm 0.01) \times 10^{-9}$	$(7.6 \pm 0.1) \times 10^{-11}$	$(4.52 \pm 0.04) \times 10^{-11}$
$P=4\text{ MW}$	$(3.24 \pm 0.01) \times 10^{-9}$	$(1.54 \pm 0.06) \times 10^{-10}$	$(8.91 \pm 0.06) \times 10^{-11}$

Table 4 Values of neutron fluxes ($n\text{ cm}^{-2}\text{ s}^{-1}$)

Reactor power	SPND1 (transmitter Rh) naked	SPND2 (transmitter Rh) under Cd	SPND3 (transmitter Zy-4) naked
Thermal neutron flux	$(1.39 \pm 0.02) \times 10^{12}$	$(1.10 \pm 0.02) \times 10^{12}$	$(1.03 \pm 0.01) \times 10^{12}$
Flux $E >$ Thermal neutron flux	$(2.49 \pm 0.03) \times 10^{10}$	$(1.57 \pm 0.03) \times 10^{10}$	$(1.35 \pm 0.02) \times 10^{10}$

Discussion

Estimation of the gamma rate

Our prototype SPND detectors have been calibrated under gamma radiation from a ^{60}Co source, we deduced the sensitivity of each prototype, and the results obtained are:

$$S_{\text{ySPND1 and SPND2}} = 2.0 \times 10^{-14} \text{ A/mGys}^{-1}$$

$$S_{\text{ySPND3}} = 5.12 \times 10^{-15} \text{ A/mGys}^{-1}$$

We notice that the results of the sensitivity of detector SPND1 and SPND2 are five times higher than SPND3, this is mainly due to the composition of the material of the transmitter, and as the main material of the SPND3 transmitter being Zircaloy (Zy4). This material has a low effective capture section for thermal neutrons, so its low contribution is expected. However, if we assume that the signal from this detector comes from gamma radiation, the value obtained is:

$$I_{\text{net SPND3}} = I_{\text{tot}} - I_0 = S_{\gamma} \cdot \Phi_{\gamma}$$

$$\Phi_{\gamma} = I_{\text{net}}/S_{\gamma} \text{ the value of } \Phi_{\gamma} = 17.4 \times 10^4 \text{ Gy/s} = 6.264 \times 10^6 \text{ R/h}$$

However, the gamma flux cannot be as high in this irradiation position, which is why we consider that the signal produced by the SPND3 can come mainly from the contribution of the coaxial cable.

Evaluation of the neutron sensitivity of SPNDs with Rhodium emitter

Simple model

Suppose that the signal of each SPND is of the form:

$$I^{\text{tot}} = S_{\text{nt}} \cdot \Phi_t + I_0 \quad (4)$$

where S_{nt} is the sensitivity to thermal neutrons, Φ_t is the heat flux, I_0 is the leakage current. The net current of the detectors is equal to the total current measured minus the leakage current, namely:

$$I^{\text{net}} = I^{\text{tot}} - I_0 = S_{\text{nt}} \cdot \Phi_t \quad (5)$$

The values obtained during the experiment are $\Phi_t^1 = (1.39 \pm 0.02) \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$, $I^{\text{net}} = (3.24 \pm 0.01) \times 10^{-9} \text{ A}$, and the sensitivity is therefore equal to $S_{\text{nt}} = (2.33 \pm 0.03) \times 10^{-21} \text{ A/n cm}^{-2} \text{ s}^{-1}$.

General model

In this model, we assume that the signal of each SPND is of the form:

$$I^{\text{tot}} = S_{\text{nt}} \cdot \Phi_t + S_{n>t} \cdot \Phi_{>t} + S_{\gamma} \cdot \Phi_{\gamma} + I_{\text{cable}} + I_0 \quad (6)$$

S_{nt} is the sensitivity to thermal neutrons, Φ_t is the heat flux, $S_{n>t}$ is the sensitivity to neutrons of energy greater than the thermal, $\Phi_{>t}$ is the neutron flux of energy greater than thermal, S_{γ} is the gamma sensitivity, Φ_{γ} is the gamma flux, I_{cable} is the response of the cable, which behaves like an SPND with a different emitter material than the active area and I_0 is the leakage current. The net current is therefore the difference between the total current and the current of escape that is to say:

$$I^{\text{net}} = I^{\text{tot}} - I_0 = S_{\text{nt}} \cdot \Phi_t + S_{n>t} \cdot \Phi_{>t} + S_{\gamma} \cdot \Phi_{\gamma} + I_{\text{cable}} \quad (7)$$

After calculations, we get the S_{nt} sensitivity:

$$S_{\text{nt}} = (2.09 \pm 0.03) \times 10^{-21} \text{ A/n cm}^{-2} \text{ s}^{-1} \quad (8)$$

Conclusion

In this work, an implantable SPND-based system was developed in compliance with all design requirements (materials, dimensions and sensitivity). It can be used to obtain online the thermal neutron doses delivered to patients, and to recalculate treatment parameters for their optimization and correction during irradiation. From the results, it can be seen that SPND1 exhibited a strong neutron response, comparing to the response of SPND2 and SPND3, and this leads to the conclusion that most signal comes from the interaction between neutrons and rhodium, compared to Zy-4 that has a low interaction contribution with neutron flux. In addition, it is shown that the level of leakage current of the two detectors SPND1 and SPND2 was less than 1% of that of the neutrons, which is low enough to be considered negligible. This leakage is mainly due to electronic noise, the cable and also depends on the temperature of the medium. The designed detectors provide a stable performance and high confidence that meets the requirements needed for the medical applications tests. This holds promises for developing industry-compatible sensitive self-powered detectors.

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