

**NPL REPORT IR 37**

**Novel neutron detectors: Looking for an improved dose response in active personal dosimeters**

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**JANUARY 2017**



## Novel neutron detectors: Looking for an improved dose response in active personal dosimeters

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### ABSTRACT

Existing sensors in active personal neutron dosimeters are invariably based on silicon semiconductors. These are compact and provide a signal that is easy to interface to the processing electronics. However, their response as a function of neutron energy differs considerably to that of body tissue, causing such dosimeters to read incorrectly when used in fields for which they have not been calibrated.

A literature study identified several novel neutron detector technologies that offer the potential for an improved dose response. Those technologies are set out and discussed in this report. It is intended that, in a future project, samples of devices from among these technologies will be obtained and tested experimentally.

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ISSN 1754-2952

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## 1. INTRODUCTION

It has long been realised that there is a need for a reasonably-priced, active (battery-powered) neutron personal dosimeter. Most radiation workers who are exposed to neutrons still wear passive devices so that the dose received is not known until the dosimeter is processed, and this can be a month or more after an exposure. This is far from ideal particularly for the sorts of exposure that can occur when, for example, high-output neutron sources are being handled and the neutron dose rate is high. Active dosimeters, by contrast, can give a reading at any time, and can sound an alarm if a dose or dose rate threshold is exceeded. However, they will usually give the wrong reading if used in a radiation field other than the one in which they were calibrated. For example, the 2007 EVIDOS project<sup>(1)</sup> reported under- and over-readings by more than a factor of two for the same dosimeter in different workplace fields. This happens because such dosimeters are invariably based on silicon sensor technology, and this material differs significantly from body tissue.

Under- and over-reading are of course both undesirable, the first obviously leading to an underestimate of health risk, and the second to unnecessary curtailment of work. There is therefore a need for new neutron sensors that have a dose response closer to that of tissue. During NPL's 2013 programme of research funded by the National Measurement Office, various novel neutron detector technologies were investigated with this in mind, the results are presented in this report. The state of development of these technologies differs considerably from one to another, some having been demonstrated in prototype, and others existing only as a concept. The intention is that selected detector types will be investigated in more detail in a subsequent research project.

It should be noted that the response of passive dosimeters is also less than ideal, and improvements are needed in this area as well, but the present report is confined to active technologies that can be used to give an immediate reading.

## 2. TECHNOLOGY TYPES INVESTIGATED

The sensor technologies investigated fell into 4 principal categories, as set out in Table 1.

**Table 1 The categories of sensor technology covered in the present study**

	<b>Technology type</b>	<b>Principle of operation</b>
1	Etched, porous or particulate silicon with a hydrogenous converter layer	The response of a conventional silicon P-N junction device is modified by intimate contact with a hydrogen-rich material that "converts" incident neutrons into recoiling protons. Complex 3D geometry may additionally enhance detector efficiency.
2	Low-Z semiconductor	A material such as carbon (diamond), boron carbide, or an organic semiconductor is used instead of silicon, providing a better match to body tissue. A hydrogen-rich converter may provide additional improvements to response and efficiency.
3	Electrometer device	Charged particles produced by neutron interactions are detected by a device sensitive to electric fields, e.g. a very high input impedance electrometer chip or a graphene field-effect transistor.
4	Scintillator	A scintillator in which neutron and gamma signals can be discriminated is coupled to a solid-state photodetector. Spectrometric information from the scintillator pulse height may be used to improve the dose response of the device.

Each of these categories will be discussed in the subsections that now follow.

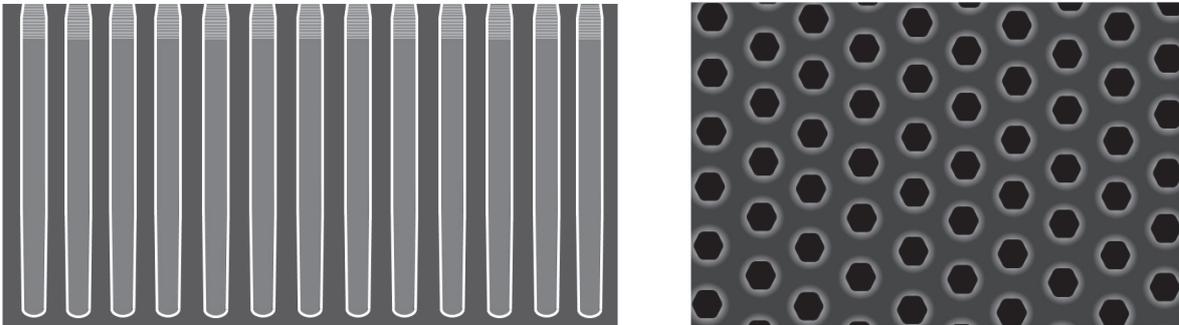
## 2.1 SILICON WITH HYDROGENOUS CONVERTER

In this type of sensor the active part is the depletion region around a conventional P-N junction in silicon. This acts as a solid-state ion chamber for the electron-hole pairs generated by the interactions of charged nuclear particles.

Neutrons are not detected directly, but via the charged particles they produce through elastic or inelastic scattering, or nuclear reactions such as  $^{28}\text{Si}(n, p)$ . To enhance neutron detection, the silicon is typically coated with a hydrogen-rich material such as polyethylene or parylene-N (one of a group of polymers designed to be applied via chemical vapour deposition). Neutrons interacting within this converter material produce recoiling protons that are then detected conventionally in the Si depletion region. This allows the device to be sensitive to intermediate energy ( $\sim$  MeV) neutrons that would produce no detectable signal in the Si directly, but limits the efficiency because of geometric effects and the relatively short proton range. The need to set a minimum energy threshold, in order to discriminate against gamma rays, also has an adverse effect. At neutron energies above about 10 MeV, the reactions that occur directly in the silicon typically cause the response to diverge further from that of body tissue.

Currently available active personal neutron dosimeters invariably include a neutron sensor of this type. Early versions comprised a planar Si diode with the converter layer deposited on top, but in more advanced designs the converter fills trenches etched in the silicon. This increases the geometric efficiency for proton detection.

For the novel neutron sensors considered in this report, the principle is extended to complex 3D geometries in which the converter and the silicon are enmeshed at a very small scale, improving efficiency and tissue equivalence still further. This is done either by (a) starting with etched or porous silicon (Figure 1) and then filling the pores in this with the converter material, or (b) by embedding silicon particles within a matrix of hydrogen-rich material which is then heated and compressed so that the particles form a bulk silicon sponge with converter-filled interstices.



**Figure 1. Porous silicon made by etching a grid of hexagonal wells into a silicon wafer. Left, slice through the wafer; Right, top surface of the wafer. (Sketch based on Figure 3 of Ref. 2.)**

For the former approach, it is necessary for the organic converter material to wet the silicon pores efficiently. For the latter, the silicon sponge needs to be able to form a depletion region (or large number of small regions). It has been suggested<sup>(3, 4)</sup> that printer technology could be used in the manufacture of this second type, using two inks made from suspensions of oppositely-doped silicon particles. Alternate layers would be printed from each ink and then heat and pressure applied to establish and stabilise the sponge structure. It is possible that the final product may remain mechanically flexible, which would have advantages in certain applications.

Ideally one would also optimise the composition of the converter material to include elements that are particularly important in the transport of neutrons and the deposition of dose in tissue. Nitrogen is a case in point because the  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction is exothermic and so has no energy threshold, which is unusual for (n,p) reactions.

## 2.2 SEMICONDUCTOR MADE OF A LOW-Z MATERIAL

In all the technologies described in this section, the aim is to create a region free of mobile charge carriers (usually by forming a depletion layer) within a semiconductor material that has a lower atomic number than silicon and is therefore closer to the composition of tissue. In cases where this material is hydrogen-rich, and there is no need for a converter layer, the efficiency is also improved because the recoil protons are created within the detector itself instead of outside it.

### 2.2.1 Organic diodes

It has recently become possible to produce semiconductors, including components such as photocells and light-emitting diodes, from organic materials. These devices have clear potential as neutron sensors because they are close to tissue in composition and cheap to produce in bulk. However, although X-ray induced photocurrents have already been demonstrated in such devices<sup>(5)</sup>, the observation of signal pulses from neutron interactions has not so far been reported. It is understood that this is because the charge carriers have low mobilities and a high probability of recombination<sup>(6)</sup>. Accordingly, this technology is not now considered the most promising (although worthy of careful monitoring for future developments).

### 2.2.2 Boron Carbide diodes

Pure boron carbide diodes<sup>(7,8)</sup> can be produced by chemical vapour deposition of two different isomers of carborane, one of which produces P-type boron carbide and the other N-type (Figure 2.) Note that the 12 hydrogen atoms associated with each carborane molecule are lost during deposition, so that the diode contains only boron and carbon.



**Figure 2. A pure boron carbide diode can be fabricated by chemical vapour deposition of two different isomers of carborane,  $C_2B_{10}H_{12}$ . (a) Closo-1,2-dicarbododecaborane (orthocarborane) produces P-type boron carbide; (b) closo-1,7-dicarbododecaborane (metacarborane) produces N-type. In the diagrams, blue circles represent boron atoms and black circles carbon. Each atom is bound to a hydrogen atom (not shown).**

Devices of this type are very promising as thermal neutron detectors, because of the large cross section for thermal neutron capture in  $^{10}B$ , which produces energetic charged particles directly inside the detector. The efficiency can be improved further if the boron is enriched in  $^{10}B$ . Furthermore, the gamma sensitivity is low, the power requirement is small, and the devices are likely to be cheap to manufacture in bulk.

Unfortunately the prospects for fast neutron detection are less good. With no hydrogen present in the diode, the most important reactions for low energy (sub-  $\sim$  MeV) neutrons are scattering on B or C, and as these targets are both substantially more massive than a neutron the maximum energy that can be transferred to them is only small. Some thermal neutrons will inevitably be present, and the signal from these may obscure the fast signal.

### 2.2.3 Diamond detectors

Single-crystal diamonds can be grown by chemical vapour deposition to produce high quality radiation detectors<sup>(9, 10)</sup>. They offer a wide band gap (5.5 eV), high electron-hole mobility, high radiation hardness and good mechanical properties. Currently the maximum crystal size is somewhat limited (about 5 mm x 5 mm in area by 0.5 mm thick), but several could be combined if necessary. Much larger samples are available in polycrystalline form, comprising large numbers of small randomly-orientated diamond crystals, but these offer very poor energy resolution.

Fast neutrons are detected via scattering and nuclear reactions on carbon, e.g.  $^{12}\text{C}(n, \alpha)$  which occurs for  $E_n \geq 6.2$  MeV and deposits  $(E_n - 5.7)$  MeV in the detector. Below this energy the most significant detection process is  $^{12}\text{C}$  elastic scattering, which can deposit anything between zero and  $0.30 E_n$ , i.e. a relatively small signal. Neutron response functions for a single crystal diamond detector have been reported by Pillon *et al.*<sup>(11)</sup> for neutron energies between 5 and 20.5 MeV.

Carbon-based detectors are clearly closer than silicon-based ones to tissue equivalence, although the absence of hydrogen obviously constitutes a significant difference relative to tissue. This could be partly addressed, and the low energy response of the diode improved, by including a hydrogen-rich converter layer similar to that already discussed in section 2.1 for silicon diodes. Complex 3D geometries that enhance the recoil proton detection efficiency would be applicable here also, provided appropriate masking or etching techniques are available during the production of the diamond crystal.

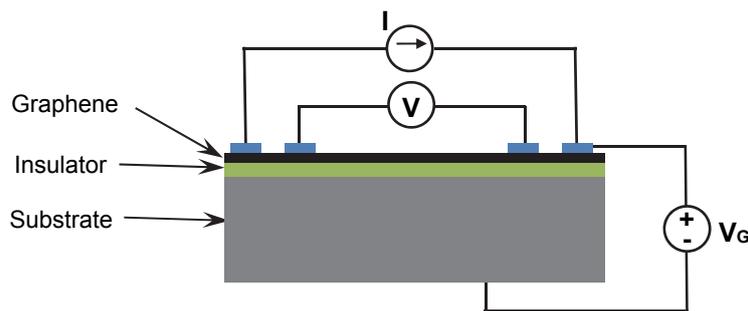
## 2.3 ELECTROMETER DEVICES

The technologies in this section are based on detecting charged particles via the changes they produce in an electric field. Such devices can be thought of as electrometers, in the sense that the emphasis is on the measurement of electric field rather than current.

### 2.3.1 Graphene Field Effect Transistors (GFETS)

Graphene is an allotrope of carbon in the form of a single flat sheet, one atom thick. The atoms in the sheet are arranged in a repeating hexagonal pattern that can be described as atomic-scale chicken wire.

Under suitable conditions, the electrical resistance of a graphene sheet is very sensitive to the local electric field. This can be exploited<sup>(12)</sup> to detect electric charges, such as those produced by ionising radiation, as illustrated in Figure 3.



**Figure 3. The basic principle of using a Graphene Field Effect Transistor (GFET) as a radiation detector. The resistivity of the graphene layer is measured using a current source  $I$  to establish a voltage drop  $V$  (measured by a volt meter). A gate voltage  $V_G$  is adjusted so that the graphene resistivity is maximally sensitive to the electric field. When charges are generated inside the substrate by ionising radiation, the difference in electric field changes the resistivity, and this is detected by a change in the volt meter reading.**

Neutron detection is facilitated by means of an appropriate converter, e.g.  $^{10}\text{B}$  for thermal neutron sensitivity, or a hydrogen-rich layer for fast neutrons.

The basic principle illustrated in Figure 3 can be developed (as described in Ref. 12) to include additional structures and electrodes so that the charges in the substrate can be marshalled into a particular location, allowing the total charge to be deduced reliably from the change in resistance. Arrangements can also be made to clear the charges away after the measurement. If an organic semiconductor could be used as the substrate, this would potentially allow the same material to function both as a fast neutron converter layer and also as a means of marshalling and clearing the charges.

Irradiation tests<sup>(13)</sup> using a  $^{252}\text{Cf}$  neutron source have demonstrated that graphene is sufficiently robust for use in a neutron dosimeter: Raman spectrometry showed no degradation in a graphene sample after exposure to  $7 \times 10^{12}$  neutrons  $\text{cm}^{-2}$ .

### 2.3.2 EPS (EPIC) device

The Electric Potential Sensor (EPS)<sup>(14)</sup> or Electric Potential Integrated Circuit (EPIC), developed at the University of Sussex, is an electric field sensing device with a very high input impedance ( $>10^{13} \Omega$ ). It has proven applications<sup>(15)</sup> in the detection of macroscopic objects via the changes they induce in the ambient electric field. In the light of this, it is apparent that it may also have a role to play as a neutron sensor, detecting neutron-generated charges in the same way as the GFET described above.

### 2.3.3 Direct ion storage (DIS) device

The RADOS (Mirion Technologies) direct ion storage system<sup>(16, 17)</sup> is based on broadly similar principles. Here, charge produced in an ion chamber by ionising radiation is stored on the floating gate of a modified EEPROM memory cell. Currently the commercially-available DIS-1 product is not intended for use with neutrons, and is a passive system with an external reader. However, the readout process is non-destructive so appropriate circuitry could be included. Also, a neutron dosimeter can be constructed using a dual-chamber system, in which the two chambers have different neutron sensitivities in order to distinguish the neutron and gamma components. Careful tailoring of the chamber materials is necessary in order to achieve good photon discrimination over a wide energy range.

## 2.4 SCINTILLATOR-BASED DETECTORS

Scintillators have not conventionally been favoured for use in personal neutron dosimeters, because of the multiple disadvantages that have applied up to now:

- An organic liquid, possibly flammable, has been needed in order to achieve the necessary neutron / gamma discrimination.
- A photomultiplier (PMT), typically rather larger than one would want in a dosimeter, has been required for detecting the scintillations (or an expensive miniature version has been necessary), and high voltage has been required to run it.
- Bulky electronics has been needed to implement neutron / gamma discrimination.

However, recent developments are starting to allow each of these problems to be addressed, as discussed in the following sections.

### 2.4.1 Novel scintillator materials

Plastic scintillators with neutron / gamma pulse shape discrimination (PSD) properties have now been reported<sup>(18, 19)</sup>. These are a great deal easier to shape and contain than a liquid, especially one that expands significantly with temperature. Some at least of these PSD-capable formulations retain the narrow pulse width (and therefore high count rate capacity) of conventional non-PSD plastics.

The novel scintillator  $\text{Cs}_2\text{LiYCl}_6:\text{Ce}$  (CLYC)<sup>(20, 21)</sup> has been exciting considerable interest recently. It is capable of gamma spectrometry and also detects thermal neutrons via capture in  $^6\text{Li}$ . Furthermore it

can detect fast neutrons via reactions in the chlorine (e.g.  $^{35}\text{Cl}(n, p)$ ). These give rise to a peak in the pulse height spectrum that is easier to interpret in terms of neutron energy than is the recoil edge that occurs in conventional organic scintillators. CLYC also allows neutron and gamma events to be distinguished by pulse shape discrimination (albeit with a wider pulse than for organics), because the fastest luminescence component is only present in gamma excitations.

#### 2.4.2 Alternative photosensors

Alternatives to PMTs are now becoming available. These include silicon photomultipliers, which comprise a large number of individual avalanche photodiodes. Each operates in Geiger mode, i.e. it is either triggered or not triggered by the light signal, but the total number of triggered channels provides an amplitude readout. These devices operate at much lower voltages than PMTs and are insensitive to magnetic fields. They have been successfully used in conjunction with CLYC<sup>(22)</sup>, although their characteristics are not (yet) as good<sup>(20)</sup> as PMTs. If used with a conventional fast organic scintillator, it is not clear whether they would be able to follow the time profile of the pulse well enough to allow PSD.

#### 2.4.3 Signal acquisition and processing

As recently as a few years ago, PSD processing for fast scintillators typically required one or more analogue electronics modules in a mains-powered crate. Now, however, detailed pulse shapes can be acquired via fast digital sampling ICs, and data processing can be carried out by customised hardware such as field-programmable gate arrays and microprocessors. Consequently it now seems likely that PSD could be implemented in a wearable battery-powered device.

### 3. OTHER CONSIDERATIONS

#### 3.1 USE OF SPECTROSCOPIC INFORMATION

Considerable computing capacity is now available in compact battery-powered devices, as evidenced by the increasing capabilities of mobile phones. It is therefore plausible that spectroscopic information derived from the neutron signal (by spectrum unfolding, for example) could be used to compute a dose figure for the specific radiation field in which the dose meter was being used. The calculation would need to be entirely automatic (no operator intervention) and repeated sufficiently frequently to provide real-time dose data. This represents a considerable challenge in electronics design and programming, but if successfully implemented would allow the dose meter to retain accuracy irrespective of the details of the actual workplace field. The degree of tissue equivalence of the sensor itself would no longer be an issue.

#### 3.2 LINEAR ENERGY TRANSFER (LET) DEVICES

For some of the semiconductor technologies discussed, the current state of the art provides a sensitive region that is a maximum of a few microns thick, i.e. much less than the typical range of the neutron-generated charged particles one is trying to measure (e.g. several mm for ~20 MeV protons in plastic). This need not rule such technologies out, however. Firstly, several devices could be stacked on top of one another to increase the sensitive thickness. Secondly, the energy deposited by a charged particle in a thin region provides useful information on the particle's dosimetric properties. This effect is exploited in the Tissue Equivalent Proportional Counter (TEPC)<sup>(23)</sup>, which is capable of measuring doses in complex radiation fields.

TEPCs are gas-filled proportional counters with low gas pressure, so that the amount of material in the chamber corresponds to a few microns of tissue (i.e. about the size of a cell). The energy deposited by a particle crossing the chamber is related to its Linear Energy Transfer ( $dE/dx$ ), which in turn determines the radiation quality  $Q$ . Accordingly, an Ambient Dose Equivalent value can be calculated from a spectrum of energy deposition values, even if the radiation field is a complex mix of particle types.

A neutron sensor with a thin sensitive region could therefore be used as a miniature solid state TEPC (see, for example, Ref. 24). With several such sensors stacked together, the detection efficiency would be correspondingly increased, and additional energy information (including possibly the original particle energy) would also be available for use in the dose calculation.

#### 4. NEXT STEPS

The next stage in this project is to obtain prototypes of these technologies and measure their responses to various neutron fields. The facilities of the NPL Neutron Metrology Group, which include well-characterised monoenergetic and radioisotope neutron sources, are well suited to this.

The technology closest to practical implementation is probably the diamond detector with sculpted 3D converter layer. Detectors based on monocrystalline diamond have already been demonstrated, and the sculpting of diode surfaces followed by deposition of a converter layer has also been carried out, albeit on silicon rather than diamond.

The technology with greatest potential for tissue equivalence is that of organic semiconductors, but a breakthrough in the mobility and lifetime of the charge carriers appears to be necessary before this potential can be realised.

#### DISCLAIMER

The mention of a commercial product in this report does not imply endorsement of that product.

#### ACKNOWLEDGEMENTS

This work was funded by the UK government's Department for Business, Energy and Industrial Strategy (BEIS). The author is grateful to David Thomas and Graeme Taylor of the NPL Neutron Metrology Group for their helpful comments on the text.

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